PROCEEDINGS OF THE THIRD IEA INTERNATIONAL WORKSHOP
ON BERYLLIUM TECHNOLOGY FOR FUSION
OCTOBER 22-24, 1997, MITO CITY, JAPAN

January 1998

(Eds.) Hiroshi KAWAMURA and Makoto OKAMOTO*
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Proceedings of the Third IEA International Workshop on Beryllium Technology for Fusion  
October 22-24, 1997, Mito City, Japan

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This report is the Proceedings of the Third International Energy Agency International Workshop on Beryllium Technology for Fusion. The workshop was held on October 22-24, 1997, at the Sangyou Kaikan in Mito City with 68 participants who attended from the Europe, the Russian Federation, the Kazakstan, the United States and Japan.

The topics for papers were arranged into 9 sessions; beryllium applications for ITER, production and characterization, chemical compatibility and corrosion, forming and joining, plasma/tritium interactions, beryllium coating, first wall applications, neutron irradiation effects, health and safety. The issues in these topics were discussed intensively on the bases of 48 presentations.

To utilize beryllium in the pebble type blanket, a series of discussions were intensified in multiple viewpoints such as the swelling, He/T release from beryllium pebble irradiated up to high He content, effective thermal conductivity, tritium permeation and coating, and fabrication cost, and so on. As the plasma facing material, life time of beryllium and coated beryllium, dust and particle production, joining, wastetreatment, mechanical properties and deformation by swelling were discussed as important issues. Especially, it was recognized throughout the discussions that the comparative study by the different researchers should be carried out to establish the reliability of the data reported in the workshop and in others. To enhance the comparative study, the world wide collaboration for the relative evaluation of the beryllium was proposed by the International Organization Committee and the proposal was approved by all of the participants. The details will be informed by the International Organization Committee in near future. The next workshop will be held in Europe in 1999.

Keywords : Beryllium Applications, ITER, Production, Chemical Reaction, Joining, Plasma/Tritium Interactions, Beryllium Coating, First Wall, Neutron Irradiation Effects, Safety Issues

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I.E.A 主催「第 3 回核融合炉ベリリウム技術国際会議」報文集
1997年10月22～24日、水戸市

日本原子力研究所大洗研究所材料試験炉部
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(1998年1月5日受理)

本報文集は、I.E.A 主催の「第 3 回核融合炉ベリリウム技術国際会議」の報文を収録したものである。本国際会議は、1997年10月22日から24日まで、水戸市の茨城県産業会館において、欧州、ロシア、カザフスタン、米国及び日本から68名の出席のもとに開催された。

本国際会議の発表件数は48件であり、ITER に於けるベリリウムの利用、製造と特性評価、両立性と腐食、成形と接合、プラズマ/トリチウム相互作用、ベリリウムコーティング、第一壁への応用、中性子照射効果及び安全取扱い等の多岐にわたる議論がなされた。

会議の要旨としては、ブランケット用関連では、スクリング、ベリリウム及びトリチウムの放出挙動、充填層の有効熱伝導率、トリチウム透過とコーティング等の研究の重要性が指摘された。プラズマ対向材としては、寿命評価、ダストの生成量評価、接合技術、廃棄物、機械的特性及びスクリーングによる変形などの研究の重要性が指摘された。特に、研究者間のデータが矛盾していることが明らかとなったため、材料を共通化させた比較研究の重要性が指摘され、国際的な協力体制のもとに共同で実験を行うことがベリリウムワークショップ国際組織委員会から提案され、全参加者によって了承された。また、次回は1999年に欧州で開催されることが決定した。
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H.Kawamura, H.Sagawa, M.Nakamichi

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G.I.Pepekin, A.B.Anisimov, A.S.Chernikov, S.I.Mozherinn, A.A.Pirogov

11. 結言

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Appendix II 第3回核融合炉ベリリウム技術国際会議参加者リスト

Appendix III 第3回核融合炉ベリリウム技術国際会議参加者写真
International Organizing Committee

H. Kawamura, JAERI, Japan (Chairman)
M. Dalle Donne, FZK, Germany
G. Longhurst, INEL, USA
V. Shestakov, SRIEP, Kazakhstan

Local Organizing Committee

M. Okamoto, Tohoku University (Chairman)
T. Terai, University of Tokyo
N. Ooka, JAERI
T. Iwadachi, NGK Insulators, LTD.
1. Preface

We have only 92 elements on our hand, the elements have their specific natures given by God. Not only human, every thing on the earth are the daughters of the 92 elements.

As accumulating the knowledge on the properties of the elements, and the accumulation of the techniques of the utilizations of the elements, we have distinguished the elements to be good or bad according to our aspiration. The elements which were distinguished to be bad have never done anything by their will, they gave us negative impacts by the wrong handling of us. The neutrons, as you know, they can give very intense energy from the fission reaction with uranium, resulting the artificial radioactive nuclides. Even these nuclides are useful to keep our lives. Unfortunately, from the radioactivities, the utilization of the atomic energy are facing the strong against wind. Mercury Hg is the only metallic element in liquid phase in wide range of temperature with good electric conductivity and suitable vapor pressure and other specific properties, so Hg has been used as the most relevant element for many technologies, such as the electrode for the production of NaOH which is one of the most important materials for chemical industry. After the MINAMATA, Hg has been listed up as the most strong enemy of the human being. We have many similar examples including chemical compounds of artificial and natural. Products designed to destroy us have to be denied in principle. Almost of the reasons by which the elements and the compounds to be denied by the people exist in How to Use them for human life.

The unfavorable results found in the uses of the risky elements and the compounds caused from our poor knowledge of these elements and compounds. We have learned many things from the history of technologies. We should know all of the properties and the behaviors of the risky elements and the compounds, before uses of these for human life. Not only Be but also every elements have their specific properties, these properties become favorable or unfavorable by our hands. They do not change their properties by themselves.

We should know all of the Be properties for its utilization without any negative impacts. To realize the magnetic fusion devices, Be has been recognized as one of the key elements, and studied intensively. In these situations, it is pleased to hold the 3rd International Workshop on Beryllium Technology for Fusion in Japan. We will have about fifty papers and more than seventy participants in the Be-Workshop from many countries.

I hope that you all of the participants will be able to enjoy the discussions and find many fruits in the Workshop.

Dr. Makoto Okamoto  
Chairman, Local Committee  
Prof. of Tohoku University
2. Beryllium Applications for ITER
2.1 Beryllium Assessment and Recommendation for Application in ITER Plasma Facing Components

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The design status of the ITER Plasma Facing Components (PFC) is presented. The operational conditions of the armour material for the different components are summarized. Beryllium is the reference armour material for the Primary Wall, Baffle and Limiter and the back-up material for the Divertor Dome. The activities on the selection of the Be grades and the joining technologies are reviewed.

1. INTRODUCTION

Since the beginning of the ITER EDA (Engineering Design Activities), beryllium has been one of the candidate armour materials for in-vessel components of ITER because it has a low atomic number, relatively good thermal and mechanical properties, and an oxygen gettering effect. However, beryllium has also a number of problems which need to be addressed. They include heat loading limits arising from temperature and stress constraints under steady state conditions, armour lifetime including the effects of sputtering erosion as well as vaporisation and loss of melt layer during disruption events, tritium inventory and permeation, and chemical hazards [1]. Other issues such as fabrication and the possibility of in-situ repair have an important impact on the overall assessment of Be as PFC armour [1].

This paper describes the present status of the application of Be in the design of ITER PFCs. First, a general description of the first wall and divertor is given, and the operational conditions of PFCs are summarised. Then the description of the results of the activities on the selection of the Be grades and the development of the Be/Cu alloy joining technologies are reviewed.

2. ITER PLASMA FACING COMPONENTS

2.1 First Wall

The First Wall (FW) is that part of the Blanket System providing the first protection against plasma interaction [2]. The Blanket System for the Basic Performance Phase (BPP) consists of modules with integrated FW and neutron shield mounted on a support structure called a back plate (BP). The FW includes primary wall, limiter and baffle modules, Fig. 1. The port mounted limiter defines the plasma boundary during plasma start-up and shut-down. The baffles maintain a relatively high density of neutral particles in the divertor region.

The FW/shield is composed of 739 modules mounted on the BP. The toroidal length of the module varies from 1.5 to 2 m, while the poloidal length varies from 0.8 to 1.1 m. Each module is an integrated structure of a 27-30 mm deep FW section comprising plasma facing armour and heat dissipation layer, and a variable depth shield section. The primary wall must dissipate a steady state heat flux of 0.5 MW/m², the upper region of the baffle
Beryllium is used as the plasma facing material at the primary wall (10 mm thick), the limiter (4 mm thick), and in the upper region of the baffle (10 mm thick), because of its low atomic number and its good compatibility with the plasma [2]. Fig. 2 shows the FW and shielding cooling layout, the structures of the primary wall and the upper part of the baffle. Fig. 3 shows the heat removal structure of the limiter [2]. Tungsten is used in the lower baffle near the divertor because of the elevated particle flux in this region where only W armour can provide the necessary erosion lifetime. The use of W also results in a considerable reduction of the tritium inventory [2].

2.2 Divertor

The divertor is designed to receive 300 MW of surface heat load from the plasma plus approximately 100 MW of neutron bulk heating. The cross section through the divertor is presented in Fig. 4, [3].

The divertor consists of 60 cassettes, each of which is approximately 2 m high, 5 m long and 0.5 - 1 m wide. Each cassette is mounted on inboard and outboard toroidal rails which are fixed to the vacuum vessel.

The PFCs of the divertor are vertical targets (VT), a dome and liners. PFCs are mounted onto the cassette body, and are demountable in a hot cell. The design lifetime of the divertor PFCs is 3000 full power discharges, requiring the divertor to be exchanged three times during the BPP.
Table 1 summarises the operation conditions of the PFCs of the FW and divertor.

### 3. BERYLLIUM SELECTION AND RECOMMENDATION

The justification of the selection of armour and structural materials for applications in ITER in-vessel components will be presented in the ITER Final Design Report (FDR) in the Materials Assessment Report (MAR) [4]. In the following part of the MAR related to the Be assessment is briefly summarised. Some other issues such as erosion lifetime, tritium inventory, and safety considerations, are included in [1].

#### 3.1. Selection of Beryllium grades

Issues related to the application of Be as armour material have been extensively discussed in [1, 5]. Some issues addressed at first sign, do not depend on the material grade (joining to the Cu heat sink, erosion, behaviour of melt layers, etc.). The selection of the optimum grade is driven by those properties which are very sensitive to the impurity levels, grain size, methods of production, thermomechanical treatment, and which usually differ for the different Be grades, such as thermal fatigue resistance, mechanical properties, swelling, neutron irradiation response, T-retention.

For the armour, thermal fatigue resistance is the most important factor because cracking could not only lead to enhanced armour erosion,
but also to crack propagation in the permanent heat sink structure.

The commercially available Be grades (S-65C vacuum hot pressed (VHP), S-65C HIP, S-65 cold isostatic pressed (CIP), S-200F HIP, S-200F VHP - Brush Wellman, US; DShG-200, TShG-56, TR-30, TGP-56, TShGT, TShG-200 - Russian Federation) were evaluated as potential candidate materials at the beginning of the EDA. First screening experiments based on thermal fatigue resistance [6] and thermal shock experiments [7] indicated that the most resistant grades are S-65C VHP and, to a lesser degree, DShG-200. Therefore S-65C VHP was selected as the reference grade, also on the basis of its availability, better database, and previous experience in JET. DShG-200 was selected as the back-up. The R&D activities were focused on the characterisation of the selected grades for ITER-relevant conditions.

The chemical composition of the selected Be grades [8,9] is given in Table 2.

Table 2. Chemical composition of S-65C and DShG-200 Be.

<table>
<thead>
<tr>
<th>Element, wt. %</th>
<th>S-65C</th>
<th>DShG-200</th>
</tr>
</thead>
<tbody>
<tr>
<td>Be (min)</td>
<td>99</td>
<td>99.34</td>
</tr>
<tr>
<td>BeO (max)</td>
<td>1.0</td>
<td>0.79</td>
</tr>
<tr>
<td>Al (max)</td>
<td>0.06</td>
<td>0.0045</td>
</tr>
<tr>
<td>C (max)</td>
<td>0.1</td>
<td>0.077</td>
</tr>
<tr>
<td>Fe (max)</td>
<td>0.08</td>
<td>0.024</td>
</tr>
<tr>
<td>Mg (max)</td>
<td>0.06</td>
<td>-</td>
</tr>
<tr>
<td>Si (max)</td>
<td>0.06</td>
<td>0.013</td>
</tr>
<tr>
<td>Others (max)</td>
<td>0.04</td>
<td>0.04</td>
</tr>
</tbody>
</table>

The chemical composition of S-65C in the greater detail needed for the safety assessments, is presented in [4]. S-65C is produced by vacuum hot pressing from impact ground beryllium powder. DShG-200 is produced from distilled ~ 200 μm Be powder, hot pressed to partial density and then punched to full density.

3.2. Data base on thermomechanical properties

**Thermophysical properties:** The available data on thermophysical properties of Be needed for the design assessment have been collected, assessed and included in the Materials Properties Handbook (MPH) [10]. The database is still incomplete (e.g. Poisson's ratio, elastic modulus, emissivity, etc.), but at least the main baseline properties needed for the preliminary assessment of the design are now available.

**Tensile properties:** As the result of the ITER R&D program significant amount of new data for the reference grades has been generated. The mechanical properties of S-65C, DShG-200 and other grades have been studied. Some scatter in the values of mechanical properties measured on different batches by different laboratories has been observed. Sources of this scatter are the difference in specimen geometry and size, lot to lot variation of microstructure and chemical composition, and strain rates during testing.

For both grades, there is some anisotropy measured in the direction parallel (L) and perpendicular (T) to the hot pressing direction, this is particularly evident in the ductility values.

The tensile properties of Be S-65C are given in Fig. 5-7. The ultimate and yield strengths are in relatively good agreement among different laboratories. The strength anisotropy is small, especially above 200°C. The strain rates used for testing are shown in Table 3. The scatter of ductility data is larger than that of the strength data, but in all cases the ductility of S-65C is good in the temperature range ~ 200-600°C.

Table 3. Tensile Test Conditions of S-65C Be.

<table>
<thead>
<tr>
<th>Source, Reference</th>
<th>Strain Rate</th>
</tr>
</thead>
<tbody>
<tr>
<td>M. Smith, 1985, [11]</td>
<td>8.3x10⁻⁵ s⁻¹ up to 1%, 8.3x10⁻⁴ s⁻¹ after 1%</td>
</tr>
<tr>
<td>F. Moons, 1995, [12]</td>
<td>1.2x10⁻⁴ s⁻¹</td>
</tr>
<tr>
<td>D. Dombrowski, 1996, [13]</td>
<td>8.3x10⁻⁵ s⁻¹ up to 1%, 8.3x10⁻⁴ s⁻¹ after 1%</td>
</tr>
<tr>
<td>S. Goods, SNL, CA, 1996, [14]</td>
<td>1.1x10⁻⁴ s⁻¹, 2.2x10⁻⁵ s⁻¹</td>
</tr>
</tbody>
</table>
Tensile properties of DShG-200 are shown in Fig. 8-10, [9, 15-19]. The strain rate typically used in the RF is much higher than the strain rate used in US and EU ($\sim 1.3 \times 10^{-3} \text{s}^{-1}$).
DSA-200 is characterised by a larger anisotropy in the strength data, which are also slightly lower than for the S-65C VHP. The anisotropy in the ductility is also larger.

**Creep and Fatigue:** There are no published data on fatigue on reference grades.

**Fracture toughness:** Fracture toughness data for S-65C VHP and some other grades are shown in Fig. 11. Neglecting the scatter due to the different grades and to the different laboratories, there is a common trend of increased fracture toughness with increasing test temperature.

![Fracture toughness data for Be grades](image)

Fig. 11. Fracture toughness of some Be grades vs. temperature. [20-22]

### 3.3. Influence of in-service conditions

#### 3.3.1. Thermal fatigue/thermal shock resistance

The resistance to thermal shock/thermal fatigue is the decisive criterion for the selection of the reference grades. More than 15 different Be grades were tested at SNLA using a novel technique for investigating the thermal fatigue resistance [6]. The samples were heat flux loaded by an electron beam, producing a localised temperature spike and, as a result, plastic strain. These conditions are much more severe than those expected in ITER and this test is considered to be a screening for different Be grades. The results show that S-65C VHP, DShG-200, TShG-56 are the grades with the highest resistance to crack initiation and with the lowest crack propagation rate.

Different Be grades were also tested in conditions simulating the disruption heat loads [7]. One of the goals of the test was to compare the surface changes of Be samples after heat loads of 1 - 7 MJ/m². For Be S-65C, all cracks stopped in the melted zone, whereas for TShG-200 (analogue to DShG-200), the cracks propagated to the bulk of the sample. The worst performance was observed in the TR-30 grade (Be with small grains and high level of BeO, ~ 2.5%), where severe cracking was observed.

In addition to these tests, Be grades (mainly S-65C and TGP) have been tested at high heat flux ~ 1-10 MW/m² as armour of actively cooled mock-ups. The main goal, here, was to test the quality of joining technologies, but also some information on Be behaviour can be inferred from these experiments. A direct comparison of these experiments is difficult, because of the different Be thickness, different stress and temperature distribution, and different joining cycles. Nevertheless, it turns out that in almost all the experiments, performed in a wide range of heat fluxes and for duration up to several thousand cycles, no fatigue damage has been observed in the Be armour.

#### 3.3.2. Neutron irradiation effects

A good deal of data on radiation damage of beryllium is available in the literature, but because of the large variety of forms and grades with widely different grain sizes, impurity content and mechanical properties, it is difficult to generalise them and to infer from them what will be the behaviour of beryllium armoured plasma facing components under ITER operating conditions. Displacement damage leads to hardening and embrittlement, while the helium production by gaseous transmutation results in high levels of gas-driven swelling and grain boundary embrittlement at high temperature. Some comprehensive reviews of neutron irradiation effects on Be were available in the literature at the beginning of the EDA [23,24], not including S-65C VHP and DShG-200.
Strength and ductility changes, as well as swelling, are strongly dependent on the irradiation temperature. Four regimes of temperature can be identified:

1. Low temperature (<20°C), where point defects are created, but mobility is so low that coalescence is rare;
2. Intermediate temperatures (20-300°C), where point defects are mobile, but gas atoms are practically immobile and remain in supersaturated solid solution;
3. High temperatures (300-600°C), where gas atoms become mobile (4), very high temperatures, where gas driven swelling becomes dominant (>600°C).

**Physical properties:** There is no effect of neutron irradiation on physical properties of Be in the dose range of interest (thermal conductivity, thermal expansion coefficient, elastic modulus etc.).

**Swelling:** The available information on swelling of different Be grades at low and elevated irradiation temperatures is shown in Fig. 12 (including the predictions from existing correlations) and Fig. 13 as a function of He content and as a function of neutron fluence.

It is evident that, in the range of irradiation temperatures relevant for the Be primary wall armour, swelling is not a problem, (for the BPP fluence). For the upper Baffle and the Dome, a region near the surface of the Be armour could experience irradiation at temperatures in the range ~500-700°C, where swelling approaches or even exceeds 1%. However, the phenomenon is limited to a narrow layer near the surface, which will be progressively eroded by the interaction with the plasma, before swelling becomes a real problem. The occurrence of He swelling during thermal excursion due to VDEs or plasma disruptions cannot be excluded, even if the transient time is very short.

For the beryllium armour of the start-up limiter, the irradiation temperature is below 300°C. However, for ~ 100 s at the beginning and at the end of each operational cycle, the surface temperature reaches ~650-750°C, a temperature and time sufficient for He migration to the grain boundaries to form He bubbles. In these circumstances, a part of the beryllium armour is subjected to He swelling and embrittlement. The extent of this phenomenon and its possible influence on the limiter performance have still to be analysed.

---

**Tensile Properties:** Data on the influence of n-irradiation on mechanical properties of S-65C VHP and DShG-200 are still very limited. As shown in Fig. 14, low temperature (230°C) irradiation at ~ 2 dpa leads to an increase of strength and to severe embrittlement. At higher irradiation temperature (300 - 500°C) ductility improves and up to ~ 1-5 dpa remains above a few % in the longitudinal direction, [14].

Still there is no data on the mechanical properties of Be at low irradiation
temperatures (100-270°C) and at a dose of ~ 1 dpa.

Fracture toughness. Neutron irradiation decreases the fracture toughness of S-65C VHP and of other grades [20]. As shown in Fig. 11, fracture toughness increases with increasing irradiation temperature, but less than for unirradiated material.

3.4. Results of the development of the Be/Cu joining technologies

An extensive R&D program is carried out in the ITER Home Teams to develop reliable Be/Cu joining technologies for application in ITER PFCs. The main problem of bonding Be to Cu alloys is that Be reacts with almost all metals and forms brittle intermetallic phases that are detrimental for the joint reliability and the fatigue lifetime. Only a few metals do not form stable beryllides below 760°C, i.e. Al, Si, Zn, Ag and Ge. Some of them, Ag and Zn, are not permitted for use in high vacuum systems.

The goal of the joining technology developments is to avoid or control BeCu intermetallic formation for an increased reliability and lifetime of the joints [27]. There are a few possible ways to solve this problem, for example the use of Al or Si, as a filler material, to use diffusion barriers which could protect against the formation of intermetallics, to use low temperature joining processes (less than 500°C at this condition the formation of the BeCu intermetallic is low). The last joining process also could allow CuCrZr to be used as heat sink without overageing of the alloy during joining.

The different joining technologies have been proposed and studied: low and high temperature HIP with or without interlayers, brazing (CuMnSnCe, Cu-Mn, CuInSnNi, Ti-Zr), low and high temperature diffusion bonding, plasma spray, diffusion soldering, explosion bonding, joint rolling etc. Some of them were abandoned due the bad results before high heat flux testing.

More than 30 mock-ups were fabricated and 25 of them have already been tested.
Table 4 Testing conditions and results of the HHF tests of Be/Cu mock-ups.

<table>
<thead>
<tr>
<th>Armour/Heat Sink</th>
<th>Technologies/parameters</th>
<th>Cu Alloy</th>
<th>Coolant T, °C</th>
<th>Heat Flux, MW/m²</th>
<th>Cycles</th>
<th>Observation</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>PS Be 16x16x(5-10)</td>
<td>Plasma Spray, knurled surface</td>
<td>CuNiBe</td>
<td>160</td>
<td>1 - 3</td>
<td>3000</td>
<td>No damage</td>
<td>Tiles cracked</td>
</tr>
<tr>
<td>S-65C, 16x16x(5-10)</td>
<td>HIP (625°C, 1h, 155 MPa) [(Be+PVD Al)&amp;Cu] + (Expl.Bond. AlBeMet, Expl.BondTi onto CuNiBe)]</td>
<td>CuNiBe</td>
<td>20</td>
<td>5</td>
<td>1000</td>
<td>No damage</td>
<td>29</td>
</tr>
<tr>
<td>S-65C, 16x16x(5-10)</td>
<td>HIP, (625°C, 1 h, 155 MPa) [(Be+PS Al) + Al-12%Si+ (Al/Ti Expl.Bonded onto CuCrZr)]</td>
<td>CuCrZr</td>
<td>20</td>
<td>5</td>
<td>2870</td>
<td>No damage</td>
<td>29</td>
</tr>
<tr>
<td>S-65C 25x12, 10</td>
<td>Monoblock, Brazing, InCuSil</td>
<td>OFHC Cu</td>
<td>RT</td>
<td>10</td>
<td>160</td>
<td>No damage</td>
<td>30</td>
</tr>
<tr>
<td>S-65C 25x31x8 castellated</td>
<td>HIP, interlayers, 800°C, 1h, 100MPa;</td>
<td>DS Cu/SS</td>
<td>RT</td>
<td>2.5</td>
<td>5-7</td>
<td>1000</td>
<td>No damage</td>
</tr>
<tr>
<td>S-65C 25x31x8 castellated</td>
<td>Brazing, CuMnSnCe, (~720°C, few min);</td>
<td>CuCrZr</td>
<td>RT</td>
<td>5.4</td>
<td>1000</td>
<td>No damage</td>
<td>30</td>
</tr>
<tr>
<td>TGP 40x40x10</td>
<td>Brazing CuInSnNi, (~820°C, few min)</td>
<td>CuCrZr</td>
<td>RT</td>
<td>1.5</td>
<td>3000</td>
<td>No damage</td>
<td>31</td>
</tr>
<tr>
<td>TGP 10x10x(5-10) or 20x20x10</td>
<td>Brazing CuInSnNi, (~820°C, few min)</td>
<td>CuCrZr</td>
<td>RT</td>
<td>7</td>
<td>10³</td>
<td>No damage</td>
<td>31</td>
</tr>
</tbody>
</table>

In Table 4 a short description of the joining technologies which have demonstrated the best thermal performance and the test conditions are presented.

Promising thermal fatigue results were obtained with flat tile mock-ups HIPed at intermediate temperature (625°C) with a complex interface including a Ti diffusion barrier and an AlBeMet layer. Mock-ups with an AlBeMet layer withstood 1000 cycles at 10 MW/m² without damage. This technology is the only one up to now which demonstrated that Be/Cu joints are able to withstand the heat flux of the port limiter.

A Be/Cu/SS mock-up produced by HIPing (with interlayers) at 800°C withstood 1000 cycles at 2.5 MW/m² without any visible damage [10]. The failure limit for this type of joining is ~ 5 MW/m². This method seems very promising for the fabrication of the low heat flux components with large curved surface.

Two silver-free brazing alloys (CuInSnNi, CuMnSnCe) have been developed, which also shown promising thermal fatigue performance of the joints. This technology could be applied to low and intermediate heat flux components, but the scaling of brazing to the manufacture of large and complex components is still to be demonstrated.

In addition to the high heat flux testing AlBeMet and Al-Si joints survived 10 shots at
200-250 MJ/m², simulating multiple VDEs at the same location. Although the Be tile almost completely melted, the joint itself did not fail.

Fig. 16 shows the ITER requirements in terms of heat flux and number of cycles and the achievements of the screening phase. It can be seen that still more development and testing are needed to support the selection of the base technologies to be used.

Fig. 16. Results of HHF testing of actively cooled Be-Cu mock-ups.

The neutron irradiation influence on the durability of the Be/Cu alloy joints is an additional issue which is also being addressed by the on-going R&D program.

3.5. Development of the Be plasma spray.

One of the advantages of using Be in the PFCs is the possibility of in-situ or in hot cell repair, which would save maintenance time and reduce the amount of radioactive waste. Low Pressure Plasma Spray (LPPS) is the most suitable coating technique for in-situ repair and is recommended as the reference beryllium coating technology [4]. Coatings approaching the thermal conductivity of bulk beryllium have been produced [32].

The result of high heat flux test on repaired ISX-B beryllium limiter tiles indicates that LPPS is suitable for the repair of the PW [33]. However, the thermal fatigue resistance of the coating is marginal for the baffle and certainly not adequate for the limiter and the dome, at this stage of development.

LPPS can be used also for the initial manufacturing of the primary wall armour. A 10 mm thick Be armour was tested and had a thermal fatigue limit of ~ 2 MW/m².

Preliminary coating trials show that a better coating microstructure can be obtained, which could possibly extend the use of LPPS, as a manufacture and a repair method, to higher heat fluxes.

4. CONCLUSIONS

Beryllium is the reference armour material for the ITER First Wall. The issues related to the application of Be in ITER PFCs have been determined during design and R&D activities and the general level of knowledge on the beryllium behaviour under ITER conditions has been significantly improved during recent years. At this stage, based on the analysis and experimental results some conclusions can be made:

- S-65C VHP is suitable reference material for ITER Plasma Facing components. DShG-200 is retained as back-up. The performance of both materials looks similar.
- Although the general database on beryllium properties has been significantly improved during the EDA, in particular for S-65C grade, more information on some properties of unirradiated Be and on temperature and dose dependence of mechanical properties, and on the swelling behaviour under thermal transients is still needed.
- During operation, part of the armour tile will be irradiated at temperatures below 300°C. These regions will progressively embrittle. The behaviour of the brittle materials as a part of the design has to be studied. For the current design it is recommended to avoid any stress raiser (e.g. castellation, diagnostic holes) and to give
preference to an armour and joint design which minimises thermal stresses.

- The question of whether the correlation between fusion and fission neutron spectra used in many of the above measurements is valid needs to be discussed. For high temperature swelling the value of the helium generated is the best correlation parameter. Comparison of changes to mechanical properties, especially at low temperature, need to be made with the same He/dpa ratio to ensure the results will be valid for ITER.

- Preliminary recommendations can be made for Be/Cu joining technologies:
  - high temperature HIPing and LPPS are recommended as the most promising methods for the large surface and the low heat flux of the Be Primary Wall, both giving adequate performance and ease of fabrication.
  - for the Be Port Limiter, it seems that HIP joining with an AlBeMet compliant layer is a viable technique. Brazing technologies have demonstrated promising results. The development need to continue.

- This recommendation on joints has to be confirmed on larger and more representative mock-ups and the influence of neutron irradiation has to be studied. For the final selection, cost and reliability will also have to be considered.

- Low Pressure Plasma Spray can be used as a repair method and also for the initial manufacture of the primary wall armour.

5. Acknowledgements

This report was prepared as an account of work undertaken within the framework of the ITER EDA Agreement. The views and opinions expressed herein do not necessarily reflect those of the Parties to the ITER Agreements, the IAEA or any agency thereof. Dissemination of the information in this paper is governed by the applicable terms of the ITER EDA agreement.

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2.2 Status of Experimental Data Related to Be in ITER Materials R&D Data Bank

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To keep traceability of many valuable raw data that were experimentally obtained in the ITER Technology R&D Tasks related to materials for In-Vessel components (divertor, first wall, blanket, vacuum vessel, etc.) and to easily make the best use of these data in the ITER design activities, the "ITER Materials R&D Data Bank" has been built up, with the use of Excel™ spreadsheet. The paper describes status of experimental data collected in this data bank on thermo-mechanical properties of unirradiated and neutron irradiated Be, on plasma-material interactions of Be, on mechanical properties of various kinds of Be/Cu joints (including plasma sprayed Be), and on thermal fatigue tests of Be/Cu mock-ups.

1. INTRODUCTION

To keep traceability of many valuable raw data that were experimentally obtained in the ITER Technology R&D Tasks related to materials for In-Vessel Components (Divertor, First Wall, Blanket, Vacuum Vessel, etc.) and to easily make the best use of these data in the ITER design activities, the "ITER Materials R&D Data Bank" [1] has been built up, with the use of Excel™ (a software for personal computer) spreadsheet.

This data bank is unique compared with the conventional materials data bank in the following respects; 1) In addition to thermo-mechanical properties of single materials (Be, W, carbon-based-materials, Cu alloys and SS), the experimental data for joining technologies (Be/Cu, W/Cu, CFC/Cu, Cu/SS, Cu/Cu and SS/SS joints, thermal bond layers, Be coating and W coating) and thermal fatigue test of mock-ups fabricated using the developed joining technologies are collected. 2) For plasma facing materials (Be, W and carbon-based-materials), experimental data on plasma-material interactions such as sputtering, disruption erosion, and hydrogen-isotope trapping and release are collected.

The present paper describes status of experimental data related to Be, which have been collected so far in this data bank.

2. STATUS OF COLLECTED DATA

2.1. Data on thermo-mechanical properties of Be

Table-1 summarizes the status of collected data on thermo-mechanical properties of unirradiated and neutron irradiated Be.

Data on thermal conductivity of unirradiated and neutron-irradiated S-200-F, tensile properties of unirradiated and irradiated S65-C (reference grade of Be) and S-200, and of unirradiated DSh-G-200 and TShG-56 are available in the data bank. Figure 1 shows a map of irradiation temperature vs. dose of neutron irradiation for collected data on tensile properties of Be. As can be seen from the figure, tensile properties of unirradiated Be have been collected in the temperature range from 20°C to 700°C, and those of neutron-irradiated Be at temperatures from 106°C to 616°C with doses up to 3 dpa.

Table-1: Status of collected data on thermo-mechanical properties of unirradiated and neutron-irradiated Be

<table>
<thead>
<tr>
<th>Property</th>
<th>Unirradiated Be</th>
<th>Neutron-irradiated Be</th>
</tr>
</thead>
<tbody>
<tr>
<td>Material Properties</td>
<td>S-200 F</td>
<td>S-200 F</td>
</tr>
<tr>
<td></td>
<td>S65-C HIP</td>
<td>S65-C HIP</td>
</tr>
<tr>
<td>Tensile Properties</td>
<td>S-200 F</td>
<td>S-200 F</td>
</tr>
<tr>
<td></td>
<td>S65-C HIP</td>
<td>S65-C HIP</td>
</tr>
<tr>
<td></td>
<td>S-200 F, DSh-G-200</td>
<td>TShG-56, HPA/P, Be</td>
</tr>
</tbody>
</table>

Table-1: Status of collected data on thermo-mechanical properties of unirradiated and neutron-irradiated Be

- 14 -
Fig.1: Map of Tirr vs. dose of neutron irradiation for collected data on tensile properties of Be

Shear strength of unirradiated and irradiated S65-C and B-26D, hardness of unirradiated and irradiated S65-C, swelling of S65-C, B-26D, S-200-F and SP-200-F and HIPped high purity beryllium powder (HIPed H.P. Be), and specific heat of unirradiated and irradiated S-200-F are also collected.

2.2. Data on plasma-material interactions of Be

Many experimental data related to plasma-material interactions (PMI) of Be have been obtained during ITER EDA. Table-2 summarizes the status of collected data on PMI of Be. The yield of Be self-sputtering (dependence on energy, incident angle and temperature) and the sputtering yield of Be by D⁺ and He⁺ ions and that (including carbon poisoning effect) by D plasma produced in linear plasma device (PISCES-B), erosion of Be by disruption simulating electron beams (JUDITH), tritium release from neutron-irradiated Be, H/D/T retention of unirradiated and neutron-irradiated Be by gas charging/atom implantation/ion impla./plasma impla., codeposition of D on Be, and permeation and diffusion of H/D in Be are collected.

2.3. Data on mechanical properties of Be/Cu joints

Table-3 summarizes the status of collected data on mechanical properties of Be/Cu joints. To join Be with Cu alloys/OFHC Cu various kinds of joining techniques (Ag brazing, Ag free brazing, HIP, diffusion bonding, joint rolling, electroplating, explosive bonding, friction welding, etc.) have been attempted during ITER EDA. Tensile properties, shear strength and bending strength of these Be/Cu joints have been collected in the data bank.

Table 3: Status of collected data on mechanical properties of Be/Cu joints

<table>
<thead>
<tr>
<th>Properties</th>
<th>Be/CuJoint</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tensile</td>
<td>B2015 by Ag braze</td>
</tr>
<tr>
<td>Shear</td>
<td>B2015 by HIP</td>
</tr>
<tr>
<td>Bonding</td>
<td>B2015 by Ag braze</td>
</tr>
</tbody>
</table>

Data on Be plasma spray (for example, technique of Be coating on Cu by plasma spray, thermal conductivity of plasma sprayed Be, bending strength of Be/Be joint by plasma spray, fracture toughness of Be/Cu joint by plasma spray, etc.) are predominantly obtained from Los Alamos National Laboratory in US and are also available in the data bank.
2.4. Data on thermal fatigue tests of Be/Cu mock-ups

As progress was made in the technology for joining Be and Cu alloys, the power handling capability of Be/Cu mock-up, which simulates the heat removal structure of the first wall for ITER, has increased year by year.

Thermal fatigue tests are conducted in such a way that mock-up(s) cooled with water flowing through channel(s) is (are) heated repeatedly from one side by a heat source (electron beam, ion beam or lamps) at a constant heat flux and a fixed heat loading time (longer than thermal equilibrium time of the mock-up) and a fixed dwell time, until the planned number of cycles is reached or any failure occurs prematurely.

Table-4: Status of collected data on thermal fatigue tests of Be/Cu mock-ups.

<table>
<thead>
<tr>
<th>Be/Cu mock-up</th>
<th>Incident or absorbed heat flux (MW/m²)</th>
<th>Heat loading time (sec)</th>
<th>Number of attained cycles</th>
</tr>
</thead>
<tbody>
<tr>
<td>EU</td>
<td>90000</td>
<td>900</td>
<td>800 on</td>
</tr>
<tr>
<td>US</td>
<td>600</td>
<td>600</td>
<td>800 on</td>
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<tr>
<td>BS</td>
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<td>2000</td>
<td>2000 on</td>
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<td>BS</td>
<td>1000</td>
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<td>1000 on</td>
</tr>
</tbody>
</table>

Table-4 summarizes the results of thermal fatigue tests for Be/Cu mock-ups. Three main parameters of thermal fatigue tests: 1) the incident or absorbed heat flux (in MW/m²), 2) the heat loading time (in sec) for each cycle and 3) number of attained cycles without any failure on the mock-up, are shown in the table. In addition, the type of joining (Ag brazing, Ag-free brazing, diffusion bonding, hot isostatic pressing (HIP), vacuum plasma spray (VPS), plasma vapour deposition (PVD), etc.) are also shown.

3. SUMMARY

As a result of ITER Technology R&D Tasks related to materials for In-Vessel components, many experimental data on thermo-mechanical properties of unirradiated and neutron-irradiated Be, on plasma-material interactions of Be, on mechanical properties of various kinds of Be/Cu joints (including plasma sprayed Be), and on thermal fatigue tests of Be/Cu mock-ups have been obtained and collected in the ITER Materials R&D Data Bank. These data have been effectively used in the comparative assessment of plasma facing materials, and in the selection of Be grade and Be/Cu joining technology for ITER.

ACKNOWLEDGEMENT

This paper was prepared as an account of work performed under the Agreement among the European Atomic Energy Community, the Government of Japan, the Government of the Russian Federation, and the Government of the United States of America on Co-operation in the Engineering Design Activities for the International Thermonuclear Experimental Reactor (ITER EDA Agreement) under the auspices of the International Atomic Energy Agency (IAEA).

REFERENCES

3. Production and Characterization

3.1 Measurement of the Thermal Conductivity and Heat Transfer Coefficient of a Binary Bed of Beryllium Pebbles

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The four ITER partners propose to use binary beryllium pebble bed as neutron multiplier. Recently this solution has been adopted for the ITER blanket as well. In order to study the heat transfer in the blanket the effective thermal conductivity and the wall heat transfer coefficient of the bed have to be known. Therefore at Forschungszentrum Karlsruhe heat transfer experiments have been performed with a binary bed of beryllium pebbles and the results have been correlated expressing thermal conductivity and wall heat transfer coefficients as a function of temperature in the bed and of the difference between the thermal expansion of the bed and of that of the confinement walls. The comparison of the obtained correlations with the data available from the literature show a quite good agreement.

1. INTRODUCTION

The Forschungszentrum Karlsruhe has accumulated a considerable experience in the field of the heat transfer in pebble beds /1-8/. The Helium Cooled Pebble Bed (HCPB) DEMO relevant blanket, which is being developed within the European Community, is based on the use of ceramic breeder pebble beds and of beryllium pebble beds as neutron multiplier and the study of the heat transfer in these beds is essential for the assessment of the blanket temperatures during the operation of the blanket. The EU Home Team has put forward a design of the ITER Breeding Blanket, which is based on the HCPB DEMO blanket and uses the beryllium in form of a binary pebble bed of large (2 mm) and small (0.1-0.2 mm) beryllium pebbles. Recently the ITER Joint Central Team (JCT) has accepted this European proposal for the ITER Breeding Blanket. The use of the binary bed offers the advantage to have a higher pebble bed packing factor (from 63% for a single size bed to 81%) and improves the heat transfer parameters of the bed. The same binary bed of beryllium pebbles is used for the HCPB as well. The correlations of the two heat transfer parameters of this bed, namely the effective thermal conductivity of the bed and the heat transfer coefficient between bed and containing walls have been obtained with experiments performed within the European R. and D. program for the HCPB DEMO blanket.

This paper describes the experiments performed for the binary beryllium pebble bed, explains how the correlations were obtained and compares them with the data available from the literature.

2. THE HEAT TRANSFER EXPERIMENTS

The binary bed of beryllium pebbles is formed by pebbles of two different sizes, namely:

- pebbles of 2 mm ± 0.3 mm in diameter, made by the firm Brush Wellman as an intermediate product in the production of beryllium with the method of Mg reduction of BeF$_2$ (average density of the pebble material = 1.824 g/cm$^3$). These pebbles were very cheap;

- pebbles of 0.1 - 0.2 mm in diameter made by the firm Brush Wellman with the atomisation method (melting and spraying with an inert gas). The average density of these pebbles is 1.791 g/cm$^3$. These pebbles are more expensive than the larger ones.
2.1 Experimental Apparatus

Fig. 1 shows schematically the experimental apparatus to measure the heat transfer parameters of the bed. The pebble bed is contained between two concentric tubes. The inner tube contains an electrically heated rod. Helium can flow in the axial direction through the bed.

The radial distribution of the temperature in the bed is measured either in one or in two axial positions in the central region of the test section where the axial temperature gradients are negligible in comparison to the radial ones, thus ensuring that the heat produced in the inner rod moves in the radial direction only. Banks of 32 thermocouples each, placed at various radii at four different azimuthal angles are used for the measurements. For the present experiments only one bank of 32 thermocouples placed at the middle axial position of the test section was used. Furthermore the temperatures on the inner and outer tube surface are measured by 40 and 36 thermocouples respectively placed in the walls to control the axial temperature distribution in the tubes.

The filling of the test section with the pebbles was made in the following way:

- filling of the annulus between the two concentrical tubes (outer radius of inner tube = 0.8 cm, inner radius of the outer tube = 5.1 cm) with 6280 g of the 2 mm pebbles; strong vibration of the pebble bed by means of a compression hammer applied to the outer tube through a wood plate; this allowed to achieve a packing factor of the 2 mm pebbles of 64.5 % for an height of the bed of 67 cm
- afterwards, filling of the annulus with 1554 g of 0.1 - 0.2 mm pebbles, corresponding to a packing factor of 16.3 %; the small pebbles flowed relatively well in between the bigger ones, so that only a gentle vibration was required to make the filling up to the same level of the bigger pebbles; at the end of the operation the level of the binary bed was controlled showing that the height of the pebble bed did not increase and thus the contact between the bigger pebbles was maintained.

The resulting packing factor of the binary bed is 80.8 %.

A porous steel plate was placed just in contact with the pebble bed upper surface and kept in place by a piston pressed on its surface by maintaining a pressure of 2 bar on the piston. This was done to avoid any demixing of the bed during the experiment.

Besides the measurements with a total of 108 thermocouples, the mass flow of the helium flowing through the bed and the helium pressure at the inlet and outlet of the test section were measured. The measurements
were performed with helium flowing at very low velocity so that the heat transfer parameters are not affected by the helium convection. This is also the case for the pebble bed in the blanket, where the velocity of the purge helium flow is very small (10-30 cm/sec).

2.2 Experimental Results

Two kinds of experiments were performed:

- a series of experiments (altogether 17 test runs) with a thermal insulation on the outside surface of the outer tube, to ensure that the thermal expansion of the outer tube is such that no constraint is exerted on the pebble bed;

- a series of experiments (altogether 7 test runs) with a water cooling on the outside surface of the outer tube to ensure that various levels of constraint are exerted on the pebble bed.

Fig. 2 shows a typical radial temperature distribution in the bed at the measurement axial position along the test section, for an experiment performed with the thermal insulation at the outside tube. Also shown are the measured temperatures at the inner tube surface (r = R1) and at the outer tube surface (r = R2). With a constant radial heat flow and bed thermal conductivity, the theory predicts a straight line in a semilogarithmic diagram temperature versus log r/R2. Fig. 2 shows that the experimental points are correlated quite well by a straight line in such a diagram. This was always the case for all the tests performed during this experiment. The slope of the temperature line allows the calculation of the effective thermal conductivity of the bed. The difference between the measured temperature on the inner tube wall and the extrapolated temperature from the bed to the wall allows the calculation of the heat transfer coefficient at the wall. (Essentially, the heat transfer coefficient accounts for the greater void fraction of the bed near the wall.) This temperature difference is always smaller at the outer tube surface, so that the obtained heat transfer coefficients for the outer tube scatter considerably more than those for the inner tube and were not used for the correlation. (For a more detailed description of the evaluation of the experiments see Ref. /4/).
Fig. 3 shows the radial temperature distribution for a test of the second series of experiments, with water cooling of the outer tube. The heat flux at the inner tube and the radial temperature gradient in the bed are much higher than for the experiment of Fig. 2 leading to considerably higher thermal conductivity and wall heat transfer coefficient than in the previous case. This phenomenon was already observed for a pebble bed of aluminum and a mixed bed of aluminum and Li₄SiO₄ pebbles /5/ and for a mixed bed of beryllium and Li₄SiO₄ pebbles /7/, but not in the case of a bed formed of Li₄SiO₄ pebbles only /5/. The increase in the heat transfer parameters is due essentially to the compressive load that the better cooled outer tube wall is exerting on the pebble bed.

The differential thermal expansion between bed and bed containment walls ("interference") produces a compression of the bed which increases the contact surface area of the pebbles. This of course has a much greater effect on the heat transfer parameters of the bed in the case of the highly heat conducting metallic pebbles than in the case of ceramic pebbles which are harder and have a much lower thermal conductivity.

Fig. 4 and 5 show the bed thermal conductivity and the heat transfer coefficient at the inner tube wall of the pebble bed without constraint ($\Delta \ell/\ell = 0$) versus the average temperature and versus the temperature of the inner tube wall respectively.

![Fig. 4 Bed thermal conductivity as function of temperature (case without constraint: ($\Delta \ell/\ell = 0$)).](image)

![Fig. 5 Heat transfer coefficient at the inner tube wall (case without constraint: ($\Delta \ell/\ell = 0$)).](image)
The bed thermal conductivity varies very little with bed temperature. This is because the beryllium thermal conductivity decreases with temperature, whereas the thermal conductivity of helium increases with temperature and the two effects somewhat compensate each other. Fig. 6 and 7 show the results of the experiments with water cooling, i.e. with pebble bed constraint ($\Delta \ell / \ell \neq 0$), in the plots $k/k_{\ell}$ and $\alpha/\alpha_{\ell}$ respectively, versus the percental ratio of the interference $\Delta \ell$ to the width of the pebble bed $\ell = R_2 - R_1$. As expected, the effect of $\Delta \ell / \ell$ is linear, as the increase of the ratio of the contact surface to the cross section of the pebbles is also linear. This increase is quite small in comparison to the pebble diameter and thus, neglecting a second order contribution, it is proportional to $\Delta \ell / \ell$. Also for the heat transfer coefficient the effect of $\Delta \ell / \ell$ is linear, however only for $\Delta \ell / \ell \geq 0.015$ (see Fig. 7), the increase for $\Delta \ell / \ell < 0.015$ being much stronger. The reason for this is probably given by the fact, that for low values of $\Delta \ell / \ell$, which forces the small pebbles to go in the region near the wall where the bed void fraction is considerably higher than in the middle of the bed. This increase of the number of pebbles near the wall produces a strong increase of $\alpha$, as the wall heat transfer coefficient is essentially proportional to the number of contact points of the pebbles to the wall. This
relocation effect is not evident in the thermal conductivity results (Fig. 6). This is probably due to the fact that this effect is smaller than in the case of the heat transfer coefficient. The thermal conductivity is determined by the bulk of the bed where the void fraction is smaller than at the wall. Thus this effect is covered by the bigger effect due to the increase of the contact surface of the pebbles. Thus the equations correlating the experimental results are:

\[
k \left[ \text{W/mK} \right] = \left( 7.3145 + 1.00652 \times 10^{-4} T_m \right) \left( 1 + 7.259 \frac{\Delta \ell}{\ell} \right) \tag{1}
\]

\[
\alpha \left[ \text{W/cm}^2 \text{K} \right] = 6.138 \times 10^{-2} \cdot f \cdot 0.00333327 T_w \tag{2}
\]

with \( f = 4.023 + 54.63 \frac{\Delta \ell}{\ell} \) for \( \frac{\Delta \ell}{\ell} \geq 0.015 \)

and \( f = 1 \) for \( \frac{\Delta \ell}{\ell} < 0.015 \)

These experimental results are valid for \( T_m \) or \( T_w \) in the temperature range 130 - 600 °C and \( \Delta \ell/\ell \) [%] in the range 0 - 0.1 %.

Fig. 8 and 9 show the fitting of equation (1) and (2) to the experimental points. The standard deviation of the experimental points are 2.1 % and 4.7 % for the thermal conductivity and for the heat transfer coefficient respectively.

3. COMPARISON WITH LITERATURE DATA

3.1 Data from UCLA

The University of California Los Angeles has been very active in the field of pebble bed heat transfer /9,10,11,12/. Generally the UCLA data on the thermal conductivity of beryllium pebbles agree quite well with the FZK data. Indeed, the thermal conductivity data for a binary bed of beryllium pebbles without constraint obtained by UCLA /9/ agree quite well with our correlation (eq. (1)). Also measurements of the thermal conductivity for a single size bed of 2 mm beryllium pebbles at room temperature and without constraint obtained at UCLA /11/ agree relatively well with the data at FZK /6/.

An important work performed at UCLA was the experimental study of the effect of the constraining pressure of the containing walls on the thermal conductivity and wall heat transfer coefficients /11,12/. The experiments are performed with cylindrical pebble beds with the heat flux parallel to the cylinder axis, rather than radially as in the case of the FZK experiments. The constraining effect of the containing walls is simulated by a piston which compresses the bed in axial direction. The gas pressure on the piston is the pressure which causes the increase of the heat transfer parameters. The results are correlated using the external pressure rather than \( \Delta \ell/\ell \), therefore they cannot be directly compared. However, one can see that they are qualitatively in agreement. For instance, a pressure of 0.7 MPa increases the thermal conductivity of a single size bed of 2 mm beryllium pebbles in helium by a factor two. Furthermore, UCLA finds, in agreement with the FZK data /5,6,7/, that these effects hold only for large ratios of solid to gas thermal conductivity (Al/He, Be/He) but not in the case of low ratios (ceramic breeder /helium). This holds both for the thermal
conductivity and for the wall heat transfer coefficient.

Another interesting consideration resulting from the comparison of the UCLA and FZK data is that, although the geometries were different (annular cylinder and full cylinder for FZK and UCLA respectively) and the heat fluxes were in different directions (radial for FZK and axial for UCLA), the comparable results (i.e. those without wall constraint) agree quite well. This shows that the effective thermal conductivity of a pebble bed, as the thermal conductivity of a material, can be applied to bodies of quite different shapes.

3.2 DATA from JAERI

Data of the thermal conductivity of single size beryllium pebble beds in helium have been published in Ref. /13/ and /14/. The results of /14/ supersede the previous of Ref. /13/ because the experiments of /14/ have been obtained with a greater amount of beryllium pebbles (about 1 kg). The thermal conductivity data of the 2 mm beryllium pebbles without constraint can be directly compared with those of FZK /6/. In the temperature range of interest the JAERI data are 10% higher than those of FZK.

4. CONCLUSIONS

The heat transfer parameters of the binary beryllium pebble bed, on which the HCPB DEMO relevant blanket and the ITER breeding blanket are based, have been obtained by experimental investigations and correlated as a function of temperature and of the interference between bed and constraining walls, due to the differential thermal expansion. The correlations allow an easy application for the calculations in the blanket.

However, these correlations have been obtained for a very stiff containment, so that they can be directly applied only in cases where very small deformations of the containing walls due to the pressure exerted by the bed are expected. This is the case, for instance, where containing plates have similar pebble beds on both sides. In case considerable deformations are expected, also the pressure effects have to be investigated experimentally. The results of these investigations, together with the present correlations (possibly extended to higher values of Δℓ/ℓ) would allow to solve the problem also in the more general case. Experiments with the measurement of the pressure on the containing walls are at present on the planning stage at FZK.

ACKNOWLEDGEMENT

This work has been performed in the framework of the Nuclear Fusion Project of the Forschungszentrum Karlsruhe and is supported by the European Communities within the European Fusion Technology Program.

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List of Symbols

\( k = \) heat effective thermal conductivity of the bed \([\text{W/mK}]\)

\( \ell = \) thickness of the bed in the direction of the heat flow \(= R_2 - R_1 \) [cm]

\( p = \) helium pressure [bar]

\( Q_o = \) heat flux at the inner wall \([\text{W/m}^2]\)

\( R_1 = \) outer radius of the inner heating tube [cm]

\( R_2 = \) inner radius of the outer containing tube [cm]

\( T = T_{\text{m}} = \) average temperature of the bed \([^\circ \text{C}]\)

\( T_w = \) temperature of the outer surface of the inner tube \([^\circ \text{C}]\)

\( T_o = \) room temperature = 20 \(^\circ\)C

\( \alpha = \) heat transfer coefficient between bed and containing wall at the outer surface of the inner tube \([\text{W/cm}^2{\circ \text{C}}]\)

\( \alpha_{pe} = \) thermal expansion coefficient of beryllium at \(T_m\) \([\text{K}^{-1}]\)

\( \alpha_{st} = \) thermal expansion coefficient of the containing walls of stainless steel \([\text{K}^{-1}]\)

\( \Delta \ell/\ell \) [%] = percental difference between the thermal expansion of the bed and of the confinement walls referred to the thickness of the bed =

\[
= 100 \left[ \alpha_{pe} (T_m - T_o) + \frac{\alpha_{st} R_2 - \alpha_{st} R_1}{R_2 - R_1} T_o - \frac{\alpha_{st} R_2 T_w - \alpha_{st} R_1 T_w}{R_2 - R_1} \right]
\]

Subscripts:

\( m = \) mean value

\( w = \) wall

\( 1 = \) inner wall \((r = R_1)\)

\( 2 = \) outer wall \((r = R_2)\)

\( o = \) at room temperature
3.2 Inherent Structure Features of Beryllium and Their Influence on the Performance Polycrystalline Metal under Different Conditions

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...The anisotropy of physical properties of beryllium single crystals resulting from covalent bonds in crystal lattice leads to significant residual thermal microstrains (RTM) in the polycrystalline metal...

...It is demonstrated experimentally that there is a simple linear dependence between the magnitude of RTM and the ultimate tensile strength. The factors controlling RTM are analysed and in the framework of powder metallurgy process the technological methods of producing beryllium with the needed properties are recommended.

...Primarily it is necessary to control the quantity and extent of dispersity of intergranular oxide inclusions and mean grain size in combination with the high degree of macro- and microhomogeneity of the structure. The requirements to beryllium microstructure for different operating conditions including neutron fluxes and transient temperature fields are formulated.

...In the framework of the concept under development one can explain formerly not fully understandable effects, which are characteristic of polycrystalline beryllium such as unexpected Pitch-Stro curve, the role of twinning etc., and predict new ones. In particular, it can be possible to expect the growth of ductility of high strength beryllium grades as neutron irradiated.

1 INTRODUCTION

Nowadays the limited plasticity that traditionally restrains the wide usage of Be as a structural material is not considered to be a serious obstacle due to developed design methodology and problems of high reliability have become of primary importance. In each of the known fields of Be utilization besides traditionally high requirements for its mechanical properties, the specific requirements are produced to such properties as dimensional stability and isotropy (optical systems), mechanical characteristics (gyrodevices), thermal shock resistance (aviation brake systems) and some other. One fails to ensure some combinations of its properties in the framework of the existent technology what caused the compelled compromises. In some extent it caused by historically based development of the Be science and technology that have two parallel directions of development that can be conditionally divided into a "fundamental" one - the systematic study of the nature of processes taking place in pure and superpure poly- and single crystal Be and an "applied" one - work in the field of Be powder technology aimed at maximally complete resolution of important practical goals related to specific semi-products and items having specified properties.

Under those conditions generalizing investigations of powder metallurgy produced (PMP) Be were relegated to the background, while the direct transfer the results of fundamental studies performed using as cast Be to the PMP technologies had a limited success, because in many cases one failed to predict or at least explain effects observed in PMP metal. As a result, new process developments in a significant extent were based on an empirical approach, which did not make one to be sure of the optimized character of specific technical solutions. The resultant gap between the fundamental theoretical and experimental work in the Be powder metallurgy field is not compensated by a great number of investigations that accompany process developments. The more so, an increased number of investigations in this field not only failed to clear the specific properties of PMP Be, but, on the contrary, made the concepts more sophisticated and the viewpoints of different scientists more contradictory. Now, as a result the turned out situation is the following: seeming conflict of the data available does not allow their unambiguous interpretation based on one of the known concepts, to systematize the data a unifying idea is needed.

2 EXPERIMENTAL RESULTS

As applied to the most of metals this idea is conventionally a concept that polycrystal behaviour under load is governed by single crystal flow and fracture stresses taking account of specific translation of slipping over grain boundaries. From this viewpoint Be is not an exception. However, it is very likely that in Be polycrystal leading
role can be played by effects related to the unusuality of its physical properties that are absent or secondary in most other metals. One of these properties is anisotropy of the coefficient of thermal expansion (CTE) parallel or transverse to the hexagonal axis which together with high elastic moduli and melting temperature is a cause of high internal microstresses in a polycrystalline metal. In PMP Be significant thermal stresses are also to be generated at oxide inclusion-matrix interface, as their (CTE) differ by a factor of 2. Preliminary calculations show that the magnitude of the indicated thermal stresses if relaxation is not taken into account can exceed ultimate strength. The fields of residual thermal microstresses through interaction with the field of macrostresses can significantly distort the pattern of stress distribution both in a grain volume and near grain boundaries and secondary phase inclusions. In other words, the stressed state at each point of a polycrystal volume is determined by the algebraic sum of macro- and microstresses and external load induced stresses. This results in a non-uniform and not simultaneous plastic strain effected by external load at different points of a strained polycrystal. Taking account of significant magnitude of RTM their contribution to the total pattern of stress distribution in a material volume can prove to be decisive. It is likely that the neglect of this circumstance leads to the fact that the classic method of studying Be mechanical properties vs its grain size, impurity content, heat treatment etc does not allow reliable correlations to be derived in many cases. Therefore, it seems most important to elucidate all aspects of interaction between Be polycrystal RTM magnitude and plastic strain and fracture properties as well as to study factors that control RTM. The anisotropy of Be single crystal thermal expansion is such that all grains in polycrystal as cooled from some equilibrium temperature prove to be compression stressed parallel to the hexagonal axis and tensile stressed in azimuthal directions; in absolute value tensile stresses being half as much as compressive ones. The maximum stress value is defined by expression (1):

\[ \tau_{\text{max}} = \int_{-\Delta \alpha}^{\Delta \alpha} \frac{1}{2} E(T)(1-\nu) A_{\alpha} \left( T - T_{0} \right) \alpha(T) \left( 1 + \nu \right) dT \]  

(1)

where \( E \) and \( \nu \) are Young modulus and Poisson coefficient of a matrix, \( \Delta \alpha \) is a difference between CTE parallel and transverse to the hexagonal axis.

Compressive stresses block the feasible nucleation of fracture on basal planes until stresses induced by external loads exceed their magnitude. Since the indicated type of fracture is the main one for Be, it follows that with RTM increasing the strength must grow. The experimental check-up of this statement is very simple and consists in X-ray measurement of RTM magnitude in materials with known mechanical properties. In this case one should take into account or reduce to a minimum the influence of other factors capable of affecting strength. They primarily include texture, mean grain size and structural uniformity.

These requirements were largely complied by hot pressed blocks selected for studies: their mean grain size varied within 18 - 30 \( \mu \) and the ultimate tensile strength varied from 250 to 500 Mpa in the direction transverse to the pressing axis. Their microstructure was adequately uniform which is confirmed by both visual and metallographic controls and the small scatter of the results of mechanical testing. The other factors that certainly influence the strength, viz., contents of oxide and metallic impurities, composition and condition of solid solution etc are suggested to have a direct effect only on relaxation properties that define RTM. It should be pointed out that a change in mean grain size from 18 to 30 \( \mu \) cannot induce significant changes in strength, as according data \( [2,3] \) the value of the constant \( k \) in Peth - Stroh equation for hot pressed Be is within 2.3 - 3.6 \( \text{kg/mm}^{3/2} \) and strength changes with mean grain size variations in the indicated range must be about 40 - 60 Mpa. Thus, according to the assumptions the range of alterations the mechanical properties of the selected materials are governed by RTM, while the influence of the mean grain size and texture is at minimum.

The measured results are given in Fig. 1. It can be seen that the experimental points are adequately described by the line \( Y = 174 + 1.06 X \) at the correlation coefficient \( R = 0.97 \).

Thus, in the first approximation one can assume that the measured ultimate strength can also serve as an integral measure of the ability of a particular material for relaxation of RTM.

As far as the dependence of percent elongation on RTM is concerned it has a maximum at 200 - 250 Mpa. Taking account of a significant scatter in the points and a small height of the maximum one should make sure of the statistical credibility of this result. With this aim in a view a large data array was employed on the results of many year mechanical testing of hot pressed Be blocks.
Figure 1. Room temperature experimental dependence between RTM and mechanical properties under tension for different grades of hot-pressed beryllium.

Figure 2. Statistical relation between UTS and elongation for existing commercial and experimental grades of PMP beryllium at room temperature.

This approach based on the following considerations. Assume the relationship between elongation (δ) and RTM as well as the linear dependence of UTS on RTM to exist in reality. Then, since UTS and δ are the functions of the same argument (RTM) and one of them is linear, there should also be a functional relation between UTS and δ of the type similar to that of δ - RTM. Taking in account this assumption, plot the results of mechanical testing in the δ - UTS coordinates. As a result we have some non-uniform set of points confined by a fitting line (Fig. 2). The number of points exceeds 10". The indicated fitting line is an approximate graphic depiction of the functional relation between δ and UTS for PMP beryllium prepared by the existing commercial process. The points below the curve should be considered to fit materials having different kinds of defects leading to a premature fracture.

3. SOME CONSEQUENCES

3.1. Explanation of δ - RTM curve

As it seems to us we have convincingly corroborated existence of extreme dependence between δ and RTM. Now let us consider how in the framework under development one can explain the observed course of the δ - RTM curve. With this aim one should follow changes in the characteristics of the 'elementary' processes of plastic strain and fracture in individual polycrystalline grains with RTM growing from the minimum to some maximum value.

In the main slipping systems, viz., basal and prismatic ones, flow stress does not essentially change which follows from the experiments on the influence of hydrostatic pressure on the above characteristics [4].

Fracture stress on the basal plane grows with RTM. Fracture stress on prismatic planes of the second kind is decreased: the rate of decrease being a factor of two lower than the growth rate for basal planes. As distinct from fracture stresses for basal planes, for prismatic planes this parameter was not found experimentally. One can but assume that it is rather high, as the strength of Be highly textured by extrusion can reach 1000 MPa at zero residual elongation in tests in the direction of extrusion (in this case the hexagonal axis are normal to the load applied).

The strength of grain boundaries grows at boundary areas parallel to basal plane and degrades at areas parallel to the hexagonal axis.

The twinning stress grows with RTM since the latter inhibits the positive dilatation of a grain in the hexagonal axis direction.

The discussed relationships are presented as a diagram in fig. 3. Twinning is a factor which role not quite comprehensible in the process of Be polycrystal strain. On the one hand, twinning can to some extent compensate for the deficiency of slipping systems, thus, increasing plasticity [5,6]. According to another viewpoint [2] twins, being stress concentrators lead to a premature fracture. However, each of these mutually exceptional conclusions is supported by the results of theoretical and experimental studies therefore it will apparently be correct if one assumes that we deal with different aspects of the same phenomenon. Indeed, on the diagram of fig. 3 with the growth of RTM one can discern at least three areas within which the competition between the processes of plastic strain and fracture can be significantly different. In the first area at low RTM the fracture takes place after an insignificant strain,
Figure 3. Influence of RTM on main deformation systems in Be (scheme).

As here the slipping stress along prismatic planes is higher than the stresses of twinning and fracture on the basal plane. The resultant twins will be the stress concentrators and will increase the probability of a brittle fracture although on the whole strength and plasticity in that area grow with RTM. In the second area the pattern is altered because here stresses of twinning and prismatic slipping are close. As a result in a formed twin prismatic slipping can develop, as it is favourable oriented what must reduce stress concentration at twin boundaries and cause the plasticity growth. Plasticity can be also increased due to the improvement of conditions of strain compatibility between the resultant twin and adjacent grains. Since twinning is only experienced by grains the direction of hexagonal axes of which is close to the direction of applied tensile stress, this process can be qualitatively compared with the extrusion texture formed during tension; which, as known, has a higher plasticity compared to the chaotic orientation of crystals.

When going to the third area this process is gradually attenuated due to the fact that twinning stresses exceed prismatic slipping stresses, strain compatibility conditions are degraded and plasticity is reduced.

Thus, the role of twinning in the first and second areas is opposite: in the former one it favours brittle fracture and in the latter one it increases plasticity. However, in the third area with RTM growth the contribution of twinning to the strain process is gradually reduced which leads to the observed external dependence of percent elongation vs RTM magnitude.

3.2. Pettich - Stroh equation

One more aspect of the RTM vs ultimate strength relationship is that the RTM concept allows the refinement of the mechanism of influence exerted by a Be polycrystal grain size on its fracture stress. For the most materials, Be metal included, the strength vs grain size dependence is adequately described with Pettich - Stroh equation that establishes UTS - $d^{-1/2}$ linear dependence. However, since the linear RTM - UTS relation is characteristic of Be it follows that RTM parameter is also to depend on a grain size. The mode of the dependence can be revealed by studying RTM distribution within a grain and boundary adjacent areas in real structure. It is apparent that microstresses in a boundary adjacent region depend on the mutual orientation of contacting grains and quickly diminish with an increase of the distance from the boundary to attain some constant value. This is shown schematically in fig. 4.

When going to the third area this process is gradually attenuated due to the fact that twinning stresses exceed prismatic slipping stresses, strain compatibility conditions are degraded and plasticity is reduced.

Thus, the role of twinning in the first and second areas is opposite: in the former one it favours brittle fracture and in the latter one it increases plasticity. However, in the third area with RTM growth the contribution of twinning to the strain process is gradually reduced which leads to the observed external dependence of percent elongation vs RTM magnitude.

Figure 4. Schematic presentation of correlation between mean grain size and X-ray measured value of RTM.

It can be seen that as the grain size is decreased the relative contribution made by intergranular microstresses to the X-ray measured RTM magnitude is increased and that from the beginning of some critical grain size $d_0$ its further decrease will not result in RTM growth. It follows from this schema that the functional RTM vs $d^{-1}$ dependence is linear for $d > d_0$. No special experiments are needed to check-up this conclusion. It is adequate to make use of the available results of the works studying Pettich - Stroh dependence for isotropic Be [7-10] by reconstructing them in the UTS - $d^{-1}$ coordinates (see fig. 5).

One can readily notice that in the grain size range of 4 - 7 $\mu$ there is a departure from the linear dependence and with the further grain refinement the growth of strength is slowed down or terminated. Based on this fact the mean effective depth of the grain boundary stress concentration zone can be assessed, which is 2 - 3.5$\mu$. One
more independent estimation of this value can be received from the experiments on low temperature neutron irradiation [11] when along grain boundaries of irradiated beryllium the 1-2 μ areas are seen free of helium bubbles. The connection of this event with grain boundary stress concentration proved by such areas absence along the boundary of the bicrystal, i.e. in system without microstresses.

It is apparent that the contribution of the described mechanism will increase with a grain refinement. Then, if an adequately wide grain size range is considered one obtains that in case of very coarse grains the classic equation of Petch - Stroh "operates" while with going to a fine grain region the RTM related mechanism starts prevailing. It is also apparent that an excessive grain refinement in Be does not effect a significant growth of strength while the plasticity is reduced. Therefore, the value of 4 - 7 μ should apparently be considered to be the ultimate minimum grain size of PMP Be.

3.3. Relaxation mechanisms

The simple linear RTM vs UTS dependence motivates the need of analyzing possible mechanisms of RTM relaxation since the calculation in an elastic approximation of cooling induced stresses gives values in excess of not only the yield strength, but also the ultimate strength. In actual process and materials those stresses do not develop at a moment but they grow with time. As they grow different relaxation mechanisms become activated leading to a lower grain boundary concentration of stresses.

The stressed state of boundary adjacent regions is highly non-uniform. For instance, on boundary areas parallel to the basal plane primarily tensile stresses (on boundary plane) are developed while compressive stresses develop in the transverse direction. In the boundary regions that are normal to the basal plane the pattern is reversed. Each grain is in contact with many differently oriented grains and the stress mode in some boundary area also depends on the adjacent grain orientation.

In this situation on a microvolume scale the appearance of the rotational moments is inevitable [12]; their relaxation is to be accomplished by a microvolumes rotation in combination with intragranular dislocation slipping.

Besides, when considering relaxation of RTM one should take in account os the fact that it proceeds at the background of residual thermal macrostresses that are essentially always available in adequately bulky bodies and have their own relaxation mechanisms as well as at the background of stresses effected by external loads. Hence, in different grains or within a single grain but in different areas of boundary surface and its adjacent regions relative contribution of the known mechanisms of stress relaxation can be different, therefore it is probably senseless to speak about the prevailing mechanism as applied to a polycrystal as a whole. It is expedient to assess the feasibility itself and conditions of action of RTM relaxation mechanisms in a Be polycrystal.

The relaxation mechanisms altogether can be conventionally divided into the following groups:

- conservative dislocation processes;
- rotational modes of deformation;
- non-conservative (diffusion - dislocation) processes;
- diffusion processes.

From the practical viewpoint it is important to point out the relaxation mechanisms that can be governed by varying process parameters. It is apparent that diffusion and diffusion - dislocation mechanisms proceed at an appreciable rate at an elevated temperature, i.e. when RTM themselves are not high. Therefore, their contribution to RTM identified at room temperature is not decisive. As far as dislocation slipping combined with rotational dislocation modes is concerned these processes theoretically are feasible in any temperature range. The more so, since RTM is a driving force of those processes their role is increased with a temperature decrease.

One of the possible ways of realizing rotational deformation modes is sliding along grain boundaries that indeed, observed in as cast high purity Be at room temperature but is absent in PMP Be [2]. It is likely that grain boundary oxide inclusions that are always present in the structure of PMP Be inhibit grain boundary sliding thus...
resulting in higher RTM in PMP Be compared to that of as-cast one. It is also apparent that the amount and extent of dispersion of intergranular oxide inclusions should significantly affect the room temperature identified magnitude of RTM and hence, ultimate strength.

3.4. Grain boundary strength

Another aspect of the influence exerted by intergranular oxide inclusions on the ultimate strength and fracture mode of PMP beryllium is due to a difference between CTE of Be and its oxide and as a result an oxide inclusion in the structure experiences a hydrostatic compression. With the growth of oxide dispersity the fraction of a grain boundary surface occupied by inclusions increases what should weaken the boundaries and increase the fracture of intergranular fracture with the simultaneous drop of plasticity. This fact was pointed out by some investigators [8] who observed the increase of the fraction of intergranular fracture during growing the strength of PMP Be.

The presented approach to the mechanism of oxide inclusions influence is also applicable to intermetallic inclusions (simple and complex beryllides) usually present in the structure of PMP Be since CTEs of beryllides are lower than those of Be. Taking account of this circumstance one can comprehend the unambiguous effect of similar heat treatments on the mechanical properties of different grades of PMP Be. Actually, depending on the quantity and the particular distribution of oxide inclusions by size, the intermetallic precipitates, appeared as a result of heat treatment, can both improve or degrade the properties of Be. This effect is only inherent in PMP metal and is not observed in its cast Be without intergranular oxide inclusions.

4. DISCUSSION

All the abovementioned prove that the grain boundary oxide inclusions definitely influence the characteristics of PMP Be. And two parallel mechanisms can be distinguish here. From the one hand, oxide inclusions lead objectively to the RTM growth and correspondingly to UTS and G changing (curve fig. 2) by preventing of the grain boundary sliding. From the other hand, with inclusions dispersity growth the grain boundaries coverage share increases that leads to the weakening of the last. The both mechanisms lead to the intergranular fracture growth while the RTM increasing. The difference is in the 1st case UTS increases with the RTM growth but in the 2nd - decrease with simultaneously plasticity drop.

It's obvious that for the each very grain size D and common oxygen concentration C the optimal size of oxide inclusions d exists. The desirable value interval of \(d_{\text{min}} < d < d_{\text{max}}\) can be estimated having analysed the idealized structure consists of the same-sized equaized grains with the diameter D covered with inclusions with the diameter d. The interval lower boundary \(d_{\text{min}}\) equals the case when inclusions touch each other, and the upper \(d_{\text{max}}\) - when inclusions stop hindering the grains growth during recrystallization. The results of not-complex geometrical estimation for several real D and C values are shown into the table 1. The table analysis leads to the unexpected conclusion that in spite of assumptions lead to \(d_{\text{min}} - d_{\text{max}}\) interval widening were made during calculation, the interval appears suddenly very narrow.

The grain boundary inclusions producing mechanism comes out from destroy and further coagulation of the oxide-film coverage powder within the process of consolidation. This process details and kinetics investigated by D. Webster et al [14, 15]. The fig. 6 contains the main results of D. Webster on the influence of consolidation temperature & fusible impurities content on the average size of oxide inclusions. The main tendency is that the inclusions average size increasing with the temperature growth and total content of Al, Si & Mg.

![Fig 6. Dependence between mean oxide inclusions size and low melting impurities content for different consolidation temperatures (D. Webster)](image)

Just summing up: the real possibility exists to regulate purposefully the size of oxide inclusions and, therefore, the RTM magnitude by control of the initial ingot chemical content and choice of method and temperature of consolidation. One should keep in mind that in real PMP Be which is produced from powders of wide range particles size is impossible to provide the optimal size.
Table 1.
Calculated intervals of admissible grain boundary oxide inclusions sizes \( d_{\text{min}}, d_{\text{max}} \) (in \( \mu \)) for different combinations of mean grain (particle) size and oxide content.

<table>
<thead>
<tr>
<th>Oxide content, ( C ), vol.%</th>
<th>Mean grain (particle) size, ( D ), ( \mu )</th>
<th>( 5 )</th>
<th>( 20 )</th>
<th>( 50 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td>0.024-0.038</td>
<td>0.10-0.15</td>
<td>0.24-0.38</td>
<td></td>
</tr>
<tr>
<td>1.0</td>
<td>0.052-0.075</td>
<td>0.20-0.30</td>
<td>0.52-0.75</td>
<td></td>
</tr>
<tr>
<td>1.5</td>
<td>0.076-0.11</td>
<td>0.30-0.45</td>
<td>0.76-1.1</td>
<td></td>
</tr>
</tbody>
</table>

of oxide inclusions in the whole material volume, as the oxide-film coagulation process goes identically for the all volume, while for the each particle size the individual optimal oxide inclusions size exists. In connection with that one should give more preference to those methods of powder producing and classification, that allow to get the powder with the maximal narrow particles size interval. Generally, one can get the metal with the properties in accordance with fitting line on fig. 2 just by increasing of the homogeneity of PMP Be on the all levels.

In general case, the microstresses source is not the CTE anisotropy only but the anisotropy of several other physical properties. For example, Be behaviour during deformation under hydrostatic pressure is also stipulated by intergranular microstresses due to anisotropy of the pressure coefficient. Anisotropy of self-diffusion and diffusion can also influence on the microstresses picture. The Be radiation growth anisotropy estimation can be helpful for the analysis of neutron irradiation influence to the mechanical properties of Be. By the way, it's obvious [13] that Be monocystal swelling anisotropy under neutron irradiation is approximately the same value (and the same mark) as the CTE anisotropy. That means the additionally appeared microstresses within the irradiation process should decrease the RTM value. In such a case the character of changes of the irradiated material mechanical properties will depend on the initial RTM (or UTS) value. Particularly, should initial Be - high-strength (from field 3, fig. 2) one could expect even several plasticity increase while the RTM lowering under irradiation process. The quantitative value of resulting effect is hardly possible as the "usual" embrittlement mechanism takes place simultaneously and the experimental results on low-temperature of high-strength beryllium grades are needed.

5. CONCLUSION

In the whole, the discussed approach to the problem of PMP Be strength and plasticity is, essentially, a model that allows an interpretation of properties features taking account of the inherent specificty of this metal. With this approach the task of managing the properties is reduced to a search for ways of controlling RTM the nature of initiation and relaxation of which is all known and mainly possible to be regulated within technological processes of powder metallurgy of the nowadays.

REFERENCES


3.3 Production of Various Sizes and Some Properties of Beryllium Pebbles by the Rotating Electrode Method

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The particle size distribution of beryllium pebbles produced by the rotating electrode method was investigated. Particle size depends on some physical properties and process parameters, which can practically be controlled by varying electrode angular velocities. The average particle sizes produced were expressed by the hyperbolic function with electrode angular velocity.

Particles within the range of 0.3 and 2.0 mm in diameter are readily produced by the rotating electrode method while those of 0.2 mm in diameter are also fabricable.

Sphericity and surface roughness were good in each size of pebble. Grain sizes of the pebbles are 17 \( \mu \) m in 0.25 mm diameter pebbles and 260 \( \mu \) m in 1.8 mm diameter pebbles

1. INTRODUCTION

Several production methods for beryllium pebbles have been proposed for use as neutron multipliers in pebble-bed-type tritium breeding blankets in thermonuclear fusion reactors. The rotating electrode method (REM) was anticipated to be a suitable method for impurity control and mass production.

There are some design concepts that will determine the beryllium pebble’s size in the breeding blanket. The binary size pebble concept is one of them and has been proposed to ensure thermal conductivity in breeding blankets. In this concept, large size pebbles and small size pebbles will be mixed. Therefore, it is very important to understand the capability of production and the quality of various sizes of beryllium pebbles produced by the rotating electrode method.

This process uses vacuum cast ingots as the raw material and there is no need for refractory materials, so their purity is excellent. Furthermore, beryllium pebbles produced by this method have a smooth surface and good sphericity, while quality reproducibility by this method is good.

Previous studies have found that large 2 mm, 1.6 mm diameter beryllium pebbles, which were the intermediate product after the magnesium reduction process have been investigated and large scatter was found in their microstructure and mechanical properties. \[1\]

Furthermore, it was noted that the levels of impurities such as magnesium and fluorine were high.

Microstructure, impurity levels, and mechanical properties’ reproducibility are important because they may influence the behavior of neutron irradiated beryllium such as the tritium inventory, swelling properties, etc.

Beryllium pebbles produced by the rotating electrode method are excellent in their reproducibility, have good
sphericity and are of low impurity.

As before, the main evaluation of pebbles produced by this method was conducted using those of 1 mm diameters.

In this study, attempts were made to produce various sizes of beryllium pebbles and an investigation into the effects of process parameters on the pebbles' sizes was conducted.

2. EXPERIMENTAL PROCEDURE

2.1 Trial Production of various sizes of beryllium pebble

The rotating electrode method is a centrifugal atomizing method which involves rotating consumable beryllium electrodes in a vacuum chamber filled with inactive gases such as helium. A schematic view of this method is shown in Figure 1. The production flow of beryllium pebbles using this method is shown in Figure 2.

The sizes of the beryllium pebbles produced by the rotating electrode method depend on the physical properties of beryllium and the process parameters such as the electrode's angular velocity (rotating speed), electrode diameter, charged electric power and so on.

Electrode diameters can be selected such as 15mm, 20mm, 35mm, 65mm. In this study, the results using electrode of 35 mm in diameter are described. After air evacuation, the chamber was filled with helium. Then the electrode was rotated and DC arc power was provided. Angular velocity is changed from 209 radian/sec to 1256 radian/sec. Particles produced under each condition were collected and particle size distribution was analyzed by sieving.

2.2 Sphericity and surface roughness measurement

Measurements are conducted with pebbles of over 0.8 mm in size. Five pebbles were picked at random after sieving. Sphericity was evaluated by measuring roundness of each size pebbles using Tayler-Hopson TALYROND200. Surface roughness is measured in three pebbles using Tayler-Hobson FTS-S4C.
2.3 Microstructure observation

Each size pebble's shape and surface condition was observed by SEM, and their microstructure was observed by using polarized light microscope.

3. RESULTS

3.1 Particle size distribution

Typical particle size distribution plots are shown in Figure 3. In this figure, the effects of the electrode's angular velocity on particle size distributions are shown. As angular velocity increases, the particle size become smaller, and on the contrary, as angular velocity decrease, the particle size becomes larger. In this experiment using electrodes of 35 mm in diameter, sharp particle size distribution with average particle size from 0.3 mm to 2.0 mm diameter was gained.

3.2 Sphericity

Results of roundness measurement for each size range of pebbles are shown in Table 1. Roundness is good in all ranges, and the value is in the range of 10 to 30 $\mu$m.

3.3 Surface roughness

The surface roughness of each size of pebbles is shown in Table 2. Surface roughness is about the same and the values is in the range of 0.25 $\mu$m to 0.65 $\mu$m in Ra.

---

**Table 1**

<table>
<thead>
<tr>
<th>Pebbles diameter- sieve size range (mm)</th>
<th>Roundness data ($\mu$m)</th>
<th>Average roundness ($\mu$m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>+1.7 - 2.0</td>
<td>15.6, 22.0, 24.2</td>
<td>20.6</td>
</tr>
<tr>
<td>+1.4 - 1.7</td>
<td>18.1, 17.4, 14.5</td>
<td>16.7</td>
</tr>
<tr>
<td>+1.18 - 1.4</td>
<td>29.1, 11.1, 17.6</td>
<td>19.3</td>
</tr>
<tr>
<td>+0.85 - 1.0</td>
<td>26.9, 14.7, 21.4</td>
<td>21.0</td>
</tr>
</tbody>
</table>

**Table 2**

<table>
<thead>
<tr>
<th>Pebbles diameter- sieve size range (mm)</th>
<th>Surface roughness</th>
<th>Ra ($\mu$m)</th>
<th>Average surface roughness</th>
<th>Ra ($\mu$m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>+1.7 - 2.0</td>
<td>0.334, 0.561, 0.439</td>
<td>0.445</td>
<td></td>
<td></td>
</tr>
<tr>
<td>+1.4 - 1.7</td>
<td>0.267, 0.500, 0.257</td>
<td>0.341</td>
<td></td>
<td></td>
</tr>
<tr>
<td>+1.18 - 1.4</td>
<td>0.644, 0.259, 0.519</td>
<td>0.477</td>
<td></td>
<td></td>
</tr>
<tr>
<td>+0.85 - 1.0</td>
<td>0.613, 0.279, 0.276</td>
<td>0.389</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
3.4 Microstructure

The SEM micrographs of pebbles of about 1.8 mm in diameter and 0.3 mm in diameter are shown in Figure 4 and Figure 5 respectively. The particles are well formed and spherical.

Polarized light micrographs of about 1.8 mm diameter and 0.25 mm diameter are shown in Figure 6 and Figure 7. Each pebble is multigranular and grain size is about 26 μm in 1.8 mm diameter and 17 μm in 0.25 mm diameter. Grain size increases with particle size.

Figure 4. SEM micrograph of pebbles of about 1.8 mm in diameter

Figure 5. SEM micrograph of pebbles of about 0.3 mm in diameter

Figure 6. Polarized light micrograph of pebbles of about 1.8 mm in diameter

Figure 7. Polarized light micrograph of pebbles of about 0.25 mm diameter
4. DISCUSSION

In the rotating electrode method, molten metal must fly off in the form of spherical particles from a rotating electrode and freeze in flight.

In this case, the centrifugal force caused by the metal rod rotation must be equal to or exceed liquid surface tension. Centrifugal force $F_c$ is expressed as follows:

$$F_c = (4 \pi r^3 \rho/3)R \omega^2$$  

where $r$ is the particle radius in cm, $\rho$ is the liquid density in g/cm$^3$, $R$ is the radius of electrode in cm and $\omega$ is the angular velocity of the electrode in radians/sec.

The liquid surface tension $F_s$ is expressed as follows:

$$F_s = 2 \pi r \gamma$$

where $\gamma$ is the surface tension of the liquid.

The condition of liquid drop formation is as follows:

$$F_c \geq F_s$$  

So, next relation can be obtained: 

$$r \geq 1.225 (\gamma/R \rho)^{1/3}/\omega$$

$\gamma$ and $\rho$ are inherent property of a certain metal.

In practice, particle size may be controlled primarily by the rotational speed of the electrode and the electrode radius in certain metals.

Regression analysis of various metals' particle size distributions by the rotating electrode method was conducted as a function of a metal's physical properties and using process parameters of B. Champagne et al. [3]. The next relation of average particle diameter by weight was obtained:

$$d_{so} = 3.65 \times 10^{-6} \omega^{-1} (\gamma/\rho)^{0.46} D^{-0.9} Q^{0.56}$$

where $\gamma$ is the surface tension in N/m, $D$ is the electrode diameter in m, and $Q$ is the melting ratio in m$^3$/sec.

After evaluating $d_{so}$ related to the angular velocity of the electrode from Figure 3, the data was compared with the calculated results of equation (4) and (5). Physical properties and used process parameters are given in Table 3. The results are shown in Figure 9.

![Figure 8](image-url)

**Table 3**

<table>
<thead>
<tr>
<th>Physical property</th>
<th>Process parameter</th>
</tr>
</thead>
<tbody>
<tr>
<td>Surface Tension $\gamma$</td>
<td>Electrode Diameter</td>
</tr>
<tr>
<td>Density $\rho$</td>
<td>Angular Velocity</td>
</tr>
<tr>
<td>1.390 (N/m)</td>
<td>0.035 (m), 0.065 (m)</td>
</tr>
<tr>
<td>1690 (kg/m$^3$)</td>
<td>209~1256 rad/sec</td>
</tr>
<tr>
<td>Melting Rate $Q$</td>
<td>$1.74 \times 10^{-7}$ (m$^3$/sec)</td>
</tr>
</tbody>
</table>
It was found that the experimental data of this project corresponds well fitted with the curve calculated from equation (5), and the average diameter of beryllium pebbles produced by the rotating electrode method can be calculated by the hyperbolic function of angular velocity and electrode diameter. The calculated results at 65 mm diameters are also shown in Figure 8.

5. CONCLUSION

The production of various sizes of beryllium pebble by the rotating electrode method was tried and the following conclusions were derived:

(1) Beryllium particle size is easily controllable by varying the electrode's angular velocity in the rotating electrode method. The particle size and electrode angular velocity relation can be expressed by their hyperbolic function.

(2) Particles within the range of 0.3 mm and 2.0 mm in diameter are readily produced by the rotating electrode method while those of 0.2 mm diameter are also fabricable.

(3) Sphericity and surface roughness was good in the evaluated pebbles and their crystal grains varied from 17 \( \mu \) m to 47 \( \mu \) m as particle size varied from 0.25 mm to 1.8 mm.

REFERENCE

3. B. Champagne and R. Angers, Size Distribution of Powders Atomized by the Rotating Electrode Process, 6th International Powder Metallurgy Conference, Washington,
3.4 Mechanical Properties of S-65C Grade Beryllium at Elevated Temperatures

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Tensile property measurements and fractographic analysis of S-65C beryllium are reviewed. Tests were performed on specimens oriented in the longitudinal and transverse directions with respect to the direction of vacuum hot-pressing. Specimens were tested in air at RT, 100°C, 200°C, 300°C, 415°C and 500°C at an initial strain rate of $1.1 \times 10^{-4}$ sec$^{-1}$. Ductility of the material was strongly affected by the test temperature, exhibiting a peak ductility at 300°C. The material displayed a yield point phenomenon which was most pronounced at this same temperature. Scanning electron microscopy was performed on the resulting fracture surfaces and observations are reported.

1. INTRODUCTION

Mechanical properties of beryllium are sensitive to a variety of factors including impurity content and test temperature. Here we report the results of a study in which the tensile properties of grade S-65C beryllium were measured at temperatures between 25°C and 500°C. This grade is produced via vacuum hot pressing of Be powder according to standard powder metallurgy processes. The S-65C grade is of higher purity than other commercial Be grades (S-200F, for example), particularly with respect to BeO and Al.

Borch states that the decrease in beryllium elongation at elevated temperatures is due to the formation of low melting point grain boundary phases [1]. S-65C differs from other commercial beryllium grades in that it has a lower total amount of metallic impurities (including Al), has a closely controlled Fe/Al ratio, and undergoes a special heat treatment to maximize conversion of all aluminum impurities from low melting point metallic aluminum to the higher melting point intermetallic compound AlFeBe$_4$.

1.1 Alloy processing

Processing of the S-65C begins with vacuum cast ingots that are about 0.9 m in length and 0.4 meters in diameter. These ingots are then cut into large flat chips using an engine lathe with a seventeen tool cutter. The chips are ground into powder to achieve a fine grain size (7 - 11 μm).

Anisotropy is minimized by randomizing the orientation of the basal plane. This is done by impact grinding the chips into smaller and more isotropic particles. Impact grinding yields minus 325 mesh powder particles which have a cube-like morphology. The impact ground powder is subsequently vacuum hot pressed in cylindrical dies at 1050 - 1150 °C and at a pressure of about 7 MPa. Duration of pressing depends on the size of billet. Vacuum hot pressing of the impact ground powder yields good mechanical properties with much reduced anisotropic mechanical behavior compared to previous grades made by attrition grinding.

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1 Conducted for the United States Department of Energy under Contract DE-AC04-94AL85000
The vacuum hot pressed (VHP) billet is then skin cut by lathe and given a 870 °C heat treatment and step cool to force all aluminum impurities into an iron-aluminum-beryllium intermetallic compound \((\text{AlFeBe}_x)\). The billets are then machined into the required shapes by conventional methods (turning, milling, EDM, drilling, etc.) using carbide tools. Machining beryllium is similar to machining heat treatable aluminum alloys. Furthermore, beryllium has a microscopic layer of damaged metal after machining. This damage affects mechanical properties, particularly ductility. Maximum mechanical properties can be recovered after machining by etching 0.08 - 0.13 mm of metal off each surface using a 2% HF - 2% HNO\(_3\) -2% H\(_2\)SO\(_4\) solution. The surface then has a matte appearance and finish. If etching cannot be done or is not required, the beryllium can be machined to a 125 rms microinch surface finish and given a stress relief heat treatment to minimize the effect of machining damage on properties.

2. EXPERIMENTAL

2.1 Materials
The composition and principal impurities of Lot 4971 are shown in Table 1 below. It was from this lot of alloy that all tests were performed. These specimens were etched after machining as described in the previous section.

As indicated above, vacuum hot pressing of beryllium results in a material with some degree of anisotropy in mechanical properties. Strength and ductility measured parallel to the pressing direction (longitudinal) are generally less than those measured in the perpendicular (transverse) orientation. Typical room temperature mechanical properties are shown in Table 2.[2]

<table>
<thead>
<tr>
<th>ELEMENT</th>
<th>wt %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Be</td>
<td>99.54</td>
</tr>
<tr>
<td>BeO</td>
<td>0.60</td>
</tr>
<tr>
<td>C</td>
<td>0.012</td>
</tr>
<tr>
<td>Al</td>
<td>185 ppm</td>
</tr>
<tr>
<td>Fe</td>
<td>620 ppm</td>
</tr>
<tr>
<td>Ni</td>
<td>130 ppm</td>
</tr>
<tr>
<td>Si</td>
<td>160 ppm</td>
</tr>
<tr>
<td>Other</td>
<td>&lt;100 ppm ea</td>
</tr>
</tbody>
</table>

Table 1. Composition of S-65C Beryllium Lot 4971; (wt. %, except as noted)

<table>
<thead>
<tr>
<th>ORIENTATION</th>
<th>YIELD (MPa)</th>
<th>UTS (MPa)</th>
<th>DUCT. (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Longitudinal</td>
<td>251</td>
<td>372</td>
<td>3.8</td>
</tr>
<tr>
<td>Transverse</td>
<td>251</td>
<td>397</td>
<td>6.0</td>
</tr>
<tr>
<td>Minimum</td>
<td>207</td>
<td>290</td>
<td>3.0</td>
</tr>
</tbody>
</table>

Table 2. Typical Room Temperature Mechanical Properties S-65C Beryllium

2.2 Testing
Button end tensile specimens were tested in air at 5 temperatures: 25°C, 100°C, 200°C, 300°C, 415°C and 500°C and at a constant displacement rate of 0.004 mm s\(^{-1}\) corresponding to an initial strain rate of 1.1 \(\times\) 10\(^{-4}\) s\(^{-1}\). Tests were performed on a conventional electromechanical test frame configured with a split clam-shell resistance heating furnace. Strain was determined from the output of an extensometer attached directly to the test specimens. High temperature, nickel-based superalloy springs were used to fixture the extensometer to the specimen allowing for direct measurement of strain at elevated temperatures.

3. RESULTS and DISCUSSION

3.1 Mechanical properties
Tensile curves for specimens tested in the longitudinal orientation are shown in Figure 1. It is clear that strength and ductility are strong functions of test temperature. A similar series of tensile curves are shown in Figure 2 for specimens tested in the transverse orientation. Here too, strength and ductility were significantly affected by test temperature. The temperature dependence of the yield (0.2% offset) and ultimate tensile strengths are shown in Figures 3 and 4. The room temperature strength values agree well with those shown in Table 2. There was only a small difference in elevated temperature strength for the specimens tested in the two orientations. As expected, specimens tested with the longitudinal orientation exhibit somewhat lower strength than those tested with the transverse orientation. The effect of orientation on mechanical properties diminishes with increasing test temperature.

Both orientations exhibited yield point phenomena, an example of which is shown in Figure 5 for a specimen oriented in the longitudinal direction. The inset in Figure 5 shows the detail of the yield point behavior. Depending on the test temperature, specimens tested in the transverse orientation exhibited either the same or slightly greater yield point drop than specimens tested in the longitudinal orientation under the same test conditions. The orientation and temperature dependence of the yield point behavior is summarized in Figure 6. In this Figure the "yield point factor" is defined as:

\[
\frac{\sigma_{yp} - \sigma_{pl}}{\sigma_{pl}}
\]

where \(\sigma_{yp}\) is the maximum stress at the yield point, and \(\sigma_{pl}\) is the minimum stress in the plateau region after yielding occurs. The transverse orientation exhibited a yield point behavior at all temperatures. The longitudinal orientation did not exhibit yield point behavior at either room temperature or at 500°C. The maximum yield point for the longitudinal orientation was seen at 300°C while for the transverse orientation, the maximum was observed at 415°C. Yield point behavior is found in metals where precipitate microstructures can initially pin dislocations. The yield point then occurs at high stress levels due to the rapid multiplication of mobile dislocations. In commercially pure beryllium, Floyd [3] concluded that the yield point effect was the result of dislocation pinning by AlFeBe\(_4\) precipitates. Alternatively, Stonehouse [4] found no correlation with increasing aluminum content and suggested that the effect is more likely due to the presence of FeBe\(_{11}\).

Specimens tested at 25°C and 100°C exhibited a brittle fracture with no necking near the fracture surface. Specimens tested at the higher temperatures developed extensive localized deformation prior to fracture. For these specimens, no systematic differences in ductility were observed with respect to specimen orientation. Ductility as measured by fracture strain and reduction in area are shown in Figures 7 and 8. Both Figures show a maximum at 300°C. The S-65C grade of beryllium exhibits significantly greater ductility than the commercially pure S-200E grade tested over the same range of temperature. As reported by Henshall, et. al., [5] tensile ductility of the S-200E grade (commercial purity) peaks at approximately 30% for the transverse orientation tested at 400°C. The longitudinal orientation exhibits only about half that ductility, also peaking at 400°C.

S-65C differs from S-200E in that it is derived from a more isotropic powder, has a lower total amount of metallic impurities (including Al), has a closely controlled Fe/Al ratio, and undergoes a special heat treatment to maximize conversion of all aluminum impurities from low melting point metallic aluminum to the higher melting point intermetallic compound AlFeBe\(_4\). The difference in high temperature elongation is probably due to all these factors.

Henshall and coworkers have also made preliminary measurements on the S-200F
beryllium grade and compared it to S-200E [6]. S-200F elongation to failure at 500°C was reported to be about 25% as compared to the S-200E values, which are about 12%. The S-200F beryllium grade is very similar to S-200E in terms of chemical composition and thermomechanical processing history. However, S-200F is made from the more isotropic impact ground powder while S-200E is made from attrited powder. The influence of the powder type cannot be discounted.

Figures 9-11 compare the present data for longitudinal orientation to other data for S-65C measured by Smith et al [7] and Brush Wellman (B-W) production data. There is overall good agreement between the data sets. As shown in Figure 9, the present UTS data lie between the data by Smith et al and the production data. In Figure 10 it can be seen that the present yield strength data lies below the two other data sets up to 200°C and between them after that. The percent elongation data is shown in Figure 11. The present data lies above the other two data sets up through 200°C but then comes into agreement with the other data sets at 500°C.

All of the mechanical property results described here are summarized in Table 3.

3.1 Fractography

Scanning electron microscopy was used to characterize the fracture surfaces of all of the tensile specimens. At any given temperature, the fracture surfaces of the two different orientation were quite similar in appearance. However, significant differences were noted as a function of test temperature. Specimens tested at room temperature, 100°C and 200°C all exhibited 100% cleavage fracture. Figure 12 shows the typical smooth crystallographic facetting observed for the specimens tested at room temperature and 100°C. The individual facets are well defined. At 200°C, the fracture surfaces had a somewhat different appearance as shown in Figure 13. While the fracture is still entirely cleavage, it is much flatter than that observed at the lower temperatures. Individual facets are much more difficult to define and the lines of cleavage seem to follow a much more macroscopic pattern of river lines which are more easily seen at lower magnifications (not shown).

Specimens tested at 300°C exhibited two distinct fracture morphologies. These fracture morphologies are apparent in Figure 14, a low magnification SEM image of the longitudinally oriented specimen. Approximately 50% of the fracture surface can be characterized as cleavage while the remaining half is ductile rupture and the transition between the two fracture morphologies is quite abrupt. The cleavage portion of the fracture surface is similar to that shown in Figure 13. The ductile fracture morphology is shown in Figure 15 from which it is clear that fracture has occurred via microvoid growth and coalescence. Dimple size varies from a few microns to as large as 20 microns. At very high magnifications there is no evidence of discrete nucleation sites for these voids (i.e., oxides).

At the highest test temperature, fracture was uniformly ductile in character with no evidence of any cleavage fracture. The fracture surfaces for these specimens looked similar to that shown in Figure 15 with the only difference being the presence of fewer large dimples.

REFERENCES


TABLE 3: Summary of Mechanical Properties of S-65C Beryllium

<table>
<thead>
<tr>
<th>ORIENTATION</th>
<th>25°C</th>
<th>100°C</th>
<th>200°C</th>
<th>300°C</th>
<th>415°C</th>
<th>500°C</th>
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<tbody>
<tr>
<td>YIELD* (MPa)</td>
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<td></td>
<td></td>
<td></td>
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<tr>
<td>Longitudinal</td>
<td>232</td>
<td>229</td>
<td>211</td>
<td>190</td>
<td>173</td>
<td>140</td>
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<tr>
<td>Transverse</td>
<td>248</td>
<td>241</td>
<td>224</td>
<td>195</td>
<td>176</td>
<td>146</td>
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<tr>
<td>YIELD PT. Max (MPa)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Longitudinal</td>
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<td>230</td>
<td>220</td>
<td>200</td>
<td>186</td>
<td>---</td>
</tr>
<tr>
<td>Transverse</td>
<td>251</td>
<td>244</td>
<td>232</td>
<td>207</td>
<td>196</td>
<td>145</td>
</tr>
<tr>
<td>YIELD PT. Min (MPa)</td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Longitudinal</td>
<td>---</td>
<td>229</td>
<td>213</td>
<td>185</td>
<td>173</td>
<td>---</td>
</tr>
<tr>
<td>Transverse</td>
<td>246</td>
<td>240</td>
<td>224</td>
<td>190</td>
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<td>140</td>
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<td>UTS (MPa)</td>
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<td>257</td>
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<tr>
<td>Transverse</td>
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<td>381</td>
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<td>261</td>
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<tr>
<td>UNIFORM STRAIN (%)</td>
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<tr>
<td>Longitudinal</td>
<td>4.9</td>
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<td>Transverse</td>
<td>5.0</td>
<td>16.8</td>
<td>11.2</td>
<td>8.8</td>
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<td>6.5</td>
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<tr>
<td>FRACTURE STRAIN (%)</td>
<td></td>
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<td></td>
<td></td>
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<tr>
<td>Longitudinal</td>
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<td>53.9</td>
<td>32.4</td>
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<tr>
<td>Transverse</td>
<td>5.0</td>
<td>16.8</td>
<td>46.3</td>
<td>50.6</td>
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<td>31.5</td>
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<tr>
<td>REDUCTION in AREA (%)</td>
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<td></td>
<td></td>
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<tr>
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<td>12.3</td>
<td>44.2</td>
<td>69.1</td>
<td>65.0</td>
<td>46.3</td>
</tr>
</tbody>
</table>

* 0.2% Yield Strength,
Figure 1. Stress-strain curves for specimens tested in the longitudinal orientation.

Figure 2. Stress-strain curves for specimens tested in the transverse orientation.
Figure 3. Yield strength decreases with increasing test temperature

Figure 4. Ultimate tensile strength decreases with increasing test temperature
Figure 5. S65C exhibits yield point phenomenon (see inset).

Figure 6. The magnitude of the yield point effect is dependent on test temperature.
Figure 7. Ductility maximum is seen at 300 °C for both orientations.

Figure 8. Reduction in area shows the same temperature dependence as the fracture strain.
Figure 9. Comparison of present work (UTS) to previous studies

Figure 10. Comparison of present work (Yield Strength) to previous studies
Figure 11. Comparison of present work (ductility) to previous studies

Figure 12. Fracture morphology for the specimen tested at 100 °C is typical of the 100% cleavage fracture for specimens tested at this temperature and at room temperature.
Figure 13. Cleavage fracture at 200 °C is flatter than that observed at the lower temperatures. Individual facets are much more difficult to define than at the lower test temperatures. The high contrast features are part of river lines which are more easily seen at lower magnifications.

Figure 14. Fracture surfaces at 300 °C exhibit nearly equal fractions for ductile and cleavage fracture.
Figure 15. Ductile fracture morphology is characteristic of the central "core" of the fracture surface shown in the previous Figure.
3.5 The Beryllium Production at Ulba Metallurgical Plant (Ust-Kamenogorsk, Kazakhstan)

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The Report includes data on beryllium production of Ulba metallurgical plant, located in Ust-Kamenogorsk (Kazakhstan). Beryllium production is showed to have extended technological opportunities in manufacturing semi-products (beryllium ingots, master alloys, metallic beryllium powders, beryllium oxide) and in production of structural beryllium and its parts. Ulba metallurgical plant owns a unique technology of beryllium vacuum distillation, which allows to produce reactor grades of beryllium with a low content of metallic impurities. At present Ulba plant does not depend on raw materials suppliers. The quantity of stored raw materials and semi-products will allow to provide a 25-years work of beryllium production at a full capacity. The plant has a satisfactory experience in solving ecological problems, which could be useful in ITER program.

"UMZ" beryllium complete-cycle production established in 50-s includes the following:
- hydrometallurgical production of Be hydroxide from the ore concentrate
- pyrometallurgical production of Be ingots and Be master alloys on Cu, Al, Ni base
- the production of metallic beryllium by vacuum distillation method
- the production of powders from ingots and from distilled beryllium
- the production of compact billets from beryllium powders
- compact billets processing by pressure
- mechanical treatment and manufacturing of final products from Be
- the production of oxide powders from hydroxide
- the production of compact semi-products and final products from BeO

The processing of ore concentrate by hydrometallurgical method is carried out from a well-known sulfate-alkaline process flow sheet. Initially ore concentrate with 2...3% Be is melted in a thermal caisson furnace, the melt is granulated and crushed in a ball mill. After that this crushed granulate is treated by a heated sulfuric acid up to a sulfate beryllium-containing solution. From this solution after a set of dissolving-precipitating operations with acids and alkalis a beryllium hydroxide of a commercial purity is obtained.

Fig 1. The hydroxide shop

Beryllium hydroxide is treated with the hydrofluoric acid and ammonia water for obtaining ammonium beryllium fluoride (ABF) crystals. From this salt crystals beryllium fluoride is produced by a thermal decomposition in the induction furnace. Beryllium fluoride is loaded together with metallic magnesium in the induction furnace and heated, initiating by that an exothermic
reduction reaction which results in forming of metallic Be and magnesium fluoride. Reduced Be has a very low purity and in order to separate magnesium it goes through a refining melting in the vacuum induction furnace. The refined 25....30 kg Be ingots are the raw material for Be powders and master alloys production. Master alloys on Cu, Ni, Al base are produced by a direct melting of beryllium with the main components in routing (Cu, Al) or in vacuum (Ni) induction furnaces.

Distilled Be production poses an objective to produce a reactor beryllium with a lower content of metallic impurities in comparison with refined Be ingots. Distillation is carried out in vacuum electric resistant furnaces. A raw material for distillation is beryllium scrap after it is machined (compact billets cuttings, chips etc.). Scrap is loaded in a graphite crucible and is heated in a furnace up to 1400 ....1600°C until the major portion of Be is evaporated. Evaporated Be is deposited on a graphite cone which is installed over a crucible. About 15....20 kg of distilled Be is produced within one cycle (8...10 days). The cone with condensed solid beryllium is taken out of a furnace, its middle portion is separated and crushed on a press with following its processing to a powder. Distillation shop has 35 furnaces.

The technology of powders production includes ingots reduction to machine chips on lathers or their grinding to grains on a hydraulic press. Then chips or grains is fed to a cone inertial mill for a preliminary grinding and to attrition disks for the final grinding up to a powder with -56 or -30 μ (300...400 mesh) grain size. These powders are the feed material for production of structural compact billets. In the early 80-s "UMZ" had mastered the process of spherical Be powders production by atomization of melted beryllium flow through a gas injector. The same installation was used for a production of Be-Al alloys spherical powders. During that time the researches on production of powders by impact method were carried out. Mills for impact-centrifugal grinding of beryllium which allowed to produce the powders with equiaxial particle form were designed, assembled and delivered to "UMZ", based on these researches.

Compact billets production is carried out in the following process flow sheets:

- cold isostatic pressing of powders accompanied by hot vacuum pressing;
- cold isostatic pressing accompanied by hot isostatic pressing;
- cold isostatic pressing followed by hot forging on a hydraulic press;
- hot forging of powder in a steel can on a hydraulic press.

Cold isostatic pressing of powders loaded in elastic bags is performed on the installation for a cold isostatic pressing of 12000 t force under the liquid pressure 2000.....4000 bar. Porous billets with 1.2....1.25 g/cm³ density, up to 500 mm in diameter and 200 kg weight can be obtained on this installation. Hot vacuum pressing of porous billets is carried out in a graphite press dies at 1050 ...1150°C and 30....50 bar pressure. The hot-pressed billets are 100...450 mm diameter, up to 500mm height and as much as 150 kg weight. A hot isostatic pressing of porous billets loaded in steel cans is carried out the installation for hot isostatic pressing of 5000 t force under 1000...1500 bar pressure at 1000...1100°C. Hot isostatic pressing allows to produce billets with a diameter up to 250 mm height.
up to 600mm, and weighing as much as 60 kg.

Hot forging of billets following a cold isostatic pressure is performed on a hydraulic 600...9600 t force presses. Prior to forging a porous billet is placed in a steel can, heated up to 1000...1100°C and forged in a special-purpose die. As a rule this method is used for production of billets with the shape different from cylindrical. The powders forging technology is used for production a large lots of small-sized billets with less weight and vice versa or production a large-sized billets with 500...1300mm in diameter, weighing more than 150 kg. To do this a powder is loaded in a steel can, packed by vibration and placed in an electric chamber furnace for heating. After heating up to 1050...1100°C cans with powder are forged on a hydraulic press. Small-sized billets are forged on 600 t press, large-sized - on 9600 t force press.

disks as well as to improve mechanical properties, compact billets are subjected to a hot deformation. The main methods of pressure treatment are extrusion and forging which are performed on the above-mentioned presses at 800...1100°C.

After compactization and/or deformation is been accomplished semi-products are subjected to machining either for enhancing of their appearance or for shaping them to a ready product form and dimensions required by a customer. The main machining technique are turning, milling and drilling. Polishing is also widely used. When machining, a high precision machining equipment including digital program controlled machines is used along with universal machines.

Nickel or copper coating with the thickness up to 20 microns can be made upon request of a customer. To obtain the oxide protection coating of black color an anodizing is used as well. For connection of parts in some cases diffusion welding of beryllium is used. Diffusion welding is carried out either with the tentative copper coating onto the welded surfaces or directly onto non coated beryllium surfaces.

Fig 3. The hydraulic press with force 9600 t

If it is required to manufacture long-length products or plane billets like plates,
treated by acid solution for removal of impurities. Following the acid treatment powders are dried and additionally milled in a ball mill to a required size. In what follows then, products are formed from a powder using a routine ceramics technology (dry or semi-dry pressing, slip casting), sintered and if necessary are machined to a required size on polishing and cutting lathes. Depending on a purpose of application, BeO products can be metallized with coatings of various shape. Products are metallized either by vacuum spray or by burning off a metallization paste applied on the surface of BeO item.

An integral part of Be production is quality control of products, which includes the following:

- step-by-step operation control;
- final control of products for compliance with customer's requirements.

Step-by-step operational control of production includes the following:

- of temperature, pressure, duration of a process and other parameters, listed in an in-plant documentation;
- quality of intermediate semiproducts (chemical, physical and mechanical properties, defects, appearance, dimensions, etc.).

Semiproducts obtained from the process with deviations are not allowed to a manufacturing final products.

The quality control of final products which is carried out to confirm their compliance to customers requirements includes the following:

- chemical analysis;
- mechanical testing;
- radiographic testing;
- ultrasonic testing;
- appearance inspection;
- dimensions inspection;
- density testing;
- metallographic inspection.

The quality control is carried out by a special service which is independent from production. This service makes a final report on the compliance of product to requirements of customers and gives a permission for shipment products.

Within the period of the biggest consumption of beryllium in the USSR in the second half of 80-s, "UMZ" produced annually:

- 800...1000 tons of copper-beryllium master alloy with 4...10% Be;
- 80...100 tons of Al-Be master alloy with 4...8% beryllium;
- 80...100 tons of vacuum-melted beryllium ingots with the purity not less than 99.0%;
- 80...100 tons of metallic beryllium powders with the purity not less than 98.0%;
- 30...35 tons of beryllium in form of compact billets and metallic beryllium parts;
- 100...120 tons of BeO powders (30...40 tons of beryllium);
- 20...30 million items of BeO products;
- 1000...1500 tons of magnesium fluoride from wastes produced by beryllium reduction.

In general the maximum annual production of Be at that time was 170...190 tons with the maximum capacity 130 tons of the main hydrometallurgical stage. It was possible to achieve such a production volume due to the processing of 40-60 t of Be including wastes from home production as well as scrap received from Be users. Sales of Be in world prices at that time was USA $ 100...150 million.

At that years the reconstruction of Be production was started which involved the increase of a production capacity up to 250 t of Be and extension of capabilities in production of Be billets of various types, sizes and grades. For this purpose the construction of a new pyrometallurgical production was started and the project for reconstruction of a hydrometallurgical production was worked out. Installations for cold and hot isostatic pressing were purchased for production of large-sized billets with isotropic properties. This installations allowed to produce compact
powders production by a centrifugal atomization. Besides, the production facility for powders preparation to pressing was purchased in Germany including installation for powders mixing, installation for powders degassing and a facility for powders loading in steel cans and their hermetic sealing.

A quick decline of a demand for Be required to make changes in a process flow sheet, productive capacity, number of workers. With that purpose the hydrometallurgical stage of a production was stopped and the processing of ore concentrate was suspended. At present production is supplied with Be stored in the plant warehouses in ingots and beryllium subcarbonate forms.

Owing to changes which took place at Be market and in the "UMZ" production, the plant at present can provide annually the following products in the following volumes:
- beryllium copper master alloy with 4...10% beryllium 500 tons
- Al-Be master alloy with 4...8% beryllium 100 tons
- vacuum-melted Be in ingots or crushed Be 25 tons
- metallic beryllium powder with grain size -56μ 20 tons
- compact structural Be in billets 12 tons
- compact structural Be in products 3 tons
- beryllium oxide powder 25 tons
- products from beryllium oxide 4 mln. items

Nowadays the indicated production volumes could be provided without the main hydrometallurgical production restarting. The existing needs in metallic Be and BeO could be met by processing of beryllium ingots and subcarbonate stored at the plant. The available ingots and carbonate inventory could provide an operation of Be production within 8 years at the maximum presently possible capacity.

Besides, raw materials in form of ore concentrate with beryllium content 1.5...3.0% are stored in the plant.
warehouses. They were stored at "UMZ" as the USSR state reserve. At present Kazakhstan is owned these raw materials. This available amount of stockpile of beryllium starting materials will allow the plant to operate at the designed capacity within 25 years.

Therefore, "UMZ" owns raw materials and processing capabilities, which make it possible to support any program, related to the use of beryllium, including ITER program.

Knowledge of mechanical properties of Be in different temperatures is necessary for ITER program. Therefore, in the given work the following structural grades of beryllium, manufactured by "UMZ", were studied:

- HP-56, manufactured by cold isostatic pressing with subsequent vacuum hot pressing
- HIP-56, manufactured by cold isostatic pressing with subsequent hot isostatic pressing;
- EHP-56, manufactured by hot extrusion of HP-56 billets;
- BHP-56, manufactured by hot forging of HP-56 billets (brake grade).

Chemical composition (Table 1) of given beryllium grades was typical for commercial beryllium, the contents of beryllium and impurities varied in the following ranges ( % weights):

- Beryllium: 98.5...98.7
- Beryllium oxide: 1.2...1.6
- Aluminium: 0.025...0.03
- Carbon: 0.075...0.083
- Chromium: 0.045...0.07
- Copper: 0.004...0.006
- Iron: 0.015...0.18
- Magnesium: 0.005...0.007
- Manganese: 0.025...0.03
- Nickel: 0.025...0.03
- Silicon: 0.015...0.029

The measurement of samples deformation at room temperature was carried out by electromechanical extensometer. The absolute sensitivity of extensometer in relation to displacement on base of length 10 mm was $2 \times 10^{-4}$ mm. At elevated temperatures the deformation was determined by measuring of distance between labels on a working part of a sample before and after tests. The measurements were carried out using microscope with accuracy 0.001 mm.

During researches the temperature dependencies of mechanical properties of these beryllium grades were determined in the temperature range 20...500°C with an interval through 100°C. Experimental data averaged taking into account three parallel samples on mechanical properties for all beryllium grades are shown on Fig. 6...8.

For all the samples metallographic investigations were carried out to determine the average grain size, which was approximately 20 microns for HP-56, 14 microns for HIP-56, 10 microns for EHP-56 in a crosswise direction and 16 microns for BHP-56 in a radial direction.

Except differences in average grain size, owing to the various technological techniques of manufacturing, the specified grades beryllium have various crystallographic structure. Beryllium of a grade HIP-56 is isotropic. Beryllium of a grade HP-56 has structure, close to isotropic. Beryllium of EHP-56 grade is strong anisotropic and has extrusion texture with preferable orientation of basis planes along extrusion direction. Beryllium of a grade BHP-56 has forging texture with orientation of planes of basis being perpendicular to forging direction and parallel to the plane of forged billet.

Beryllium of a HIP-56 grade has best isotropism of mechanical properties, due to the absence of appreciable crystallographic texture in comparison with other beryllium grades. Deformed beryllium EHP-56 and BHP-56 grades have the highest strength and plastic properties at 20...500°C. It is
explained by formation of the crystallographic texture and linear oriented microstructure to be inherent to deformed semifinished items during deformation (extrusion or forging). In the temperature range 600...800°C HIP-56 and HP-56 grades of beryllium have the highest strength properties, while deformed grades EHP-56 and BHP-56 have the highest plastic properties, that is explained by the change of deformation mechanisms at the tests. Mainly dislocation sliding at 20...500°C is being changed by sliding along grain boundaries.

In addition, while a reactor is operating, ecological problems, connected with environment and personnel protection, will inevitably come up due to a high toxicity of beryllium. Such problems are quite similar to those, been appeared in beryllium production.

Extended investigations on development of a small-wastes technology were carried out at "UMZ" in a former times. Development of magnesium fluoride production from slugs of reducing melting of beryllium was a result of such investigations along with involving in a production liquid beryllium-containing wastes, which helped to reduce a water consumption and liquid wastes disposal to a tailings pond. Technologies were developed for production of glazed tiles from hydrometallurgical production wastes (sulfate-alkaline cakes), utilization of burning wastes by pyrolysis, beryllium withdrawal from gas purification systems and the others.

Based on these investigations results either projects were accomplished or technical tasks were made for designing proper processing areas. An experience, gained at "UMZ" could appear to be useful for ITER program, where the problem of beryllium-contained wastes may turn to be one of the major issues.

CONCLUSIONS

1. "UMZ" could execute the following technological tasks within ITER program:
   • delivery of ordinary structural Be, which "UMZ" produces today;
   • development and implementing a technology for high-purity beryllium manufacturing by vacuum distillation;
   • development and implementing a technology for production of bimetallic (Cu-Be) parts for reactor;
   • development and implementing a technology for spherical beryllium powders production by gas atomization of melt for reactor maintenance (replacement of parts in a reactor operating chamber);
2. The UMZ commercial grades of Be have acceptable mechanical properties for ITER.
Fig. 6 Mechanical properties of HP-56 beryllium.
Fig. 7 Mechanical properties of HIP-56 beryllium.
Fig. 8 Mechanical properties of EHP-56 and BHP-56 beryllium.
3.6 Sanitary-Hygienic and Ecological Aspects of Beryllium Production

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The Report describes an organization of sanitary-hygienic and ecological control of beryllium production at Ulba metallurgical plant. It involves: (1) the consideration of main methods for protection of beryllium production personnel from unhealthy effect of beryllium, (2) main kinds of filters, used in gas purification systems at different process areas, (3) data on beryllium monitoring in water, soil, on equipment. This Report also outlines problems connected with designing devices for a rapid analysis of beryllium in air as well as problems of beryllium production on ecological situation in the town.

Due to a negative effect of beryllium and its compounds on human body, on townspeople and on environment, it is necessary to reduce such an effect to a reasonable minimum. That is why a sanitary-hygienic and ecological control, which is carried out by a specific laboratory at Ulba metallurgical plant are of critical importance in beryllium production. Sanitary-hygienic control involves the control over shop workers exposure to Be and reducing to a minimum a release of Be out of the production area with equipment, scrap metal, used overalls. Ecological control involves the environment monitoring (of air, soil, vegetation, snow, water) in sanitary area and in control observation area. The other directions of a control are: monitoring of waste and disposal sources (ventilation system of working area and industrial storm drain); monitoring of tails disposal; control over the quality of overalls and individual protection means laundry.

Monitoring of air in the workplace. Monitoring is carried out by pumping air through a filters followed by filters analyzing in a chemical laboratory.

For the monitoring of air in the workplace there are two ways of sampling: continuous sampling during one shift and taking single samples. One-shift sampling is carried out on a fixed sampling equipment, single samples are taken with portable aspiration devices. Single samples are used for auxiliary rooms control and for workplaces examinations. According to the normative documents a top permissible concentration for Be (TPC) in air of workplace is 0.001 mg/m³ [1...3].

Among the main shortcomings of the existing system of a control of airborne Be in air of the workplace are the following:

• a long time interval between sample taking and result receiving;
• sampled (discrete) information;
• getting information only over a real fact of air contamination with beryllium.

Therefore, repeated attempts were made at Ulba metallurgical plant, aimed at designing an instrument for a rapid determination of beryllium in air.

Spectral analyzer of Be aerosols in alternating current flash was assembled at "LOMO" plant (Leningrad). All-Union R & D
Institute of Chemical Technology (VNIICHT, Moscow) designed a γ-activation analyzer of aerosols deposited on a substrate. But these devices found no application because of a great number of both constructional and methodical shortcomings (big size, heavy weight, poor sensitivity, bad reproducibility of results). From our experts standpoint, a model of α-activation device for aerosol Be concentration measurement designed by Radiation Instrument Making R&D Institute (RNIIRP, Riga) has the most successful design.

Device operation is based on the nuclear reaction $\text{Be}^9(\alpha, n)\text{C}^{12}$. In order to measure Be content, air sample is pumped through filter with following placing of the filter in a specific holder and irradiating it with α-particles. The flow of fast neutrons which appeared as a result of a nuclear reaction, is registered by a counter. The number of neutrons is in proportion to Be content in a sample. The sensitivity of an approach is 1.2 mkg Be on filter within 1000 sec. With sampling of 1 m$^3$ of air a measurement with 1 mg/m$^3$ accuracy can be obtained. The method is rapid as it requires no preliminary chemical preparing of samples. The device could be used in shops for a rapid determination of contamination levels. Testing of this pilot device gave a positive result.

The control over technological equipment surface, ceiling and floor room, workers skin. Surface contamination is monitored by taken wet samples from a specific area followed by its analysis in a Chemical Laboratory. Sanitary regulations recommend the following limits for surface contamination: 2 mg/ m$^2$ for plane surfaces, 5 mg/ m$^2$ for porous surfaces, 0.5 mg/ m$^2$ for rooms in administrative buildings [3].

Surface cleanup is carried out with warm water either using pail, mop, sackcloth, or by hose washing. The utilizing of organic compounds while washing is restricted since a detergent goes back to the technological process after cleanup and has an adverse effect on it. Some of working areas have an ineffective surface cleanup technology, so the level of contamination of equipment and room surfaces with Be compounds considerably exceeds the acceptable limits. Hence, the main objective for the work in this field is to search for an effective and cheap method of surfaces cleanup, including the cleanup of metal scrap.

Within 1987-91 various cleanup methods and devices, developed by All-Union R & D Institute of Energy Equipment (Sankt-Petersburg) were tested:

- steam-emulsion method with steam-eject sprayer;
- jet method with the use of hydraulic monitors;
- film method with steam generators;
- film method with the use of film-forming substances and removable protective coatings on polyvinyl alcohol base;
- mechanized method with floor washing device.

Among the above listed technologies only steam emulsion method with the steam eject sprayers was introduced in Be production, the others were either ineffective or required the production reconstruction.

After staff have passed a sanitary treatment, there is a control over skin contamination at "UMZ" which is carried out according to the Analytical Instruction [4]. A limit for skin contamination is recommended by Sanitary regulations [3]. In case of this level exceeding, a worker is sent to a repeated sanitary treatment. Analysis lasts for approximately 2 minutes.

Individual protection accessories (IPA). According to sanitary regulations [3] and methodical recommendations [5], which were developed by Biophysics Institute of Russia Health Protection Ministry, all workers & visitors of beryllium production buildings must change their clothes in a sanitary room.

All beryllium production shops have such sanitary rooms, which are equipped according to the above-mentioned documents. Staff takes off all home clothes, and puts on
overalls (cotton or lavsan suit, underclothes, socks or footcloth, cap or kerchief, special boots, respirator "Lepestok-200") in the sanitary rooms. After the work of the shift is accomplished staff takes a shower and overalls is sent to a specific laundry, except special boots & respirator. Respirator is being used within the whole shift and after the work is completed is taken away to a special container and then concealed.

Overalls is washed in a special plant laundry, according to "Recommendations on the overall and supplementary IPM cleanup in the UMZ special laundry" developed by Biophysics Institute of Russia Ministry of Health Protection (IBPh). Laundry intervals are determined by the above mentioned sanitary regulations...[3]. A quality of laundry is determined by Be content in the last rinsing water according to the Instruction on washing [6].

In 1990 a short-used overalls made of nonwoven perforated fabric with rayon and polypropylene filaments of nonwoven cloth, was tested. This cloth is recommended for use in emergency situations and for work in areas with a high level of contamination.

For execution of the following operations & works:
- with equipment and air ducts cleanup;
- gas-purifying elements changing;
- equipment opening; gas-arc welding; beryllium, its alloys and master alloys melting;
- beryllium powders & its compounds production

staff uses additional IPA(pneumosuits, pneumojackets, pneumohelmets, pneumomasks).

In order to provide the usage of pipe insulating IPM, all beryllium production buildings are equipped with fresh air pneumolines which have connections to workplaces. The workshops have a special section equipped with washing machines & drying chambers for insulating and plastic compound IPA cleanup.

Water and soil control. Cooling water of all stages of Be production, including Be hydroxide, distilled Be, Be powders, BeO productions are connected up in a water supply circulation system. That means that waters from all production buildings are drained to a water supply circulation system, pumped to a cooler (graduating tower), cooled and than are brought back to a production.

The control over Be content in wasted production waters is carried out by taken a single sample once in a quarter. An average Be content in circulating water is 0.0027...0.0035 mg/l.

At present waters of metallic beryllium ingots and master alloys productions are not connected up. The cooling water of these productions is disposed to a storm drain and goes to a drainage (Ulba river). The control over Be content in drained waters is carried out by taken a single samples once in 10 days according to a schedule, approved by East-Kazakhstan Regional Office of Ecology and Biological resources and by State Sanitary Service.

The control on the plant territory is carried out in the catch pit, where all cooling waters are drained. Water samples are taken daily during 10 days, an average 10-days probe is made on their base and sent to Chemical Laboratory for analysis.

An average Be content in water before throwing off to a river is 0.0019 mg/l (according to standard documents, there must be no beryllium in fisheries ponds).

The evaluation of a level of soil contamination with Be is carried out once in a year during a warm season: samples are taken at the production area and at the key locations 20-30 km far from production area. The level of snow contamination is also determined by sampling once in a year in the last month of a cold season at the same key location as for soil. The Be content in soil of production area is 4.08 mg/g, with the natural Be content 2.0 mg/g; in snow - 0.37 mg/g, at sidewalks - 7.0 mg/g (with the standard 0.5 mg/g). The control over sidewalks and transport ways is carried out at UMZ by taken a wet samples four times in a year from...
the area immediately adjacent to Be production shops.

Gas purification and ventilation.
Beryllium production has 46 sources of a toxic chemical emissions into atmosphere.

Gas, emitted from the technological equipment and production rooms before emitting to atmosphere goes through a multistep purification. In 1990 when Be production worked at a full capacity, the quantity of air emitted into atmosphere was $20 \times 10^9$ m$^3$/year.

Various types of gas-purification equipment are used in Be production, depending on a physical-chemical composition of adverse impurities, the required level of purification and the volume of purified gas.

**Cyclone.** Simple cyclones and cyclones with a reverse cone for the removal of the large particles dust from gas are used. The average actual degree of Be dust removal from gas is 60...85%. Cyclones are used as the first level of purification of gases from furnaces and from machining. It should be marked, that gas decontamination level achieved by Cyclones conforms the present requirements.

**Bag filters** are used in Be production as the second stage of gases purification in ventilation system of metallurgical furnaces, of vacuum hot-pressing furnaces, distillation furnaces and beryllium powders production shop. In the main, the bag filters provide a high level of gas cleaning- 99% and more. But sometimes the level is reduced to 90%, because of short-time modifications in the technology, inopportune changing of bags and ineffective bags recovery. Bag filters developed by "UMZ" designers are used along with the above-mentioned standard bag filters. Filters work constantly and with a high purification level- up to 99%.

**Aerosol filters FPP.** Aerosol filters with FP cloth (Petrianov filters) for fine & superfine decontamination of air and technological gases from solid highly-dispersed aerosol particles are broadly applied in Be production. FP cloth is a nonwoven perchlorovinyl material, resistant to acids and alkalis. The experience of work have showed that the level of purification on FPP filters ranges from 50% to 99.99%; sometimes it is 80-90% and it does not correspond to their possibility. The low efficiency of FPP filters is attributable to a difficult hermetic sealing of filters in the installation as well as to the low inlet concentration of aerosol particles in gases.

**Conic scrubbers with ball nozzle** are used for gas with a high moisture content decontamination. Scrubbers have a purification level from 38% up to 90%. The reduced capacity of purification installations (low then 65%) is attributable to the following:
- low inlet dust concentration;
- inefficient spraying system;
- inefficient drop catching at the scrubber outlet.

**Fibrous printed filters** with laven packing are used in Be production also. The cleaning efficiency is from 60% up to 93.7%. These filters are mainly used as sprays catchers. The reasons of a low filters efficiency are the same as for FPP filters.

The control over emissions into atmosphere has the following objectives:
- determination of the quantity of emitted compounds in order to find out their correlation to norms;
- preventing of a supernormal atmosphere pollution;
- the evaluation of a gas-dust-catching equipment efficiency.

Based on these 3 objectives the control over emissions in beryllium production is so organized, that any deviations from a standard can be prevented in order to protect atmosphere from additional discharges. Determination of a concentration of toxic compounds emitted from the contamination sources is carried out in accordance with the control procedure [7].

It should be marked, that the available sampling and sample testing procedures for determination of the above-mentioned adverse compounds, including
beryllium, does not allow to define rapidly the concentration and to regulate timely emissions into atmosphere. With the devices for a rapid analysis the discharges in atmosphere could be significantly decreased by reducing a time interval between sampling and getting of an analysis result.

Simultaneously with the sampling, the measurements of aerodynamic parameters of gas-air mixture in gas conduits is carried out, including a measurement of ventilation systems capacity, pressure, rarefaction, humidity and temperature. All measurements are carried out in accordance with GOST's requirements [8...11].

The control over atmospheric air, "UMZ" have organized a service, which carries out round-the-clock control over adverse impurities in atmosphere. This control allows to make the objective evaluation of the company role as of the air contamination source as well as to find out areas mostly subjected to contamination in discrete time intervals, and to affect promptly the company technological process for reducing atmosphere contamination levels in case they are identified.

The control is carried out by sampling at key observation stations which are the following:
- stationary (specific rooms);
- control points for sampling at the place (at specific area points).

There are 5 stationary sampling stations and all of them are located at the Company production area. There are 30 control stations, 10 of them are located at the Company production area; and 20 - at the boundary of sanitary area and living area. The way of sampling at the stations is the following:
- sampling at stationary stations is carried out during the working days;
- sampling at control stations is carried out every twenty-four-hour by mobile laboratory.

Meteorological parameters are determined at stations at the same time with the sampling of air:
- wind direction & speed;
- air temperature;
- an atmospheric pressure;
- comparative humidity;
- weather and surface condition.

what is required by normative documents and have an effect upon the rout of a mobile laboratory [12...14].

Today the situation is the following: an average annual and a top single concentration of Be and its compounds in air of production area are being reduced since 1992 as the result of a production capacity decrease. An average annual concentration of Be and its compounds on a dividing line of sanitary area and living area stay relatively stable and has a small fluctuation amplitude. It could be explained by work of a power station located close to a production area which burns off coal with some Be content.

SUMMARY

Ulba Metallurgical Plant has a rich experience in organization of Sanitary-Hygienic and Ecological control of Beryllium at an industrial objects and at out-of-plant area.

This experience of UMZ may be useful for ITER program.

The basic problem connected to a control of Be in air is an absence of an apparatus for express control. At the authors opinion, an α-activation device for measurement of Be aerosol concentration is the most perspective apparatus (or method) of measurement.

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4. Chemical Compatibility and Corrosion
4.1 Investigation of Beryllium/Steam Interaction

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In this report program on investigations of beryllium emissivity and transient processes on overheated beryllium surface attacked by water steam to be carried out in IAE NNC RK within Task S81 TT 2096-07-16 FR. The experimental facility design is elaborated in this Report.

1. INTRODUCTION

From the view point of hydrogen production potential hazard under accidental situation, resulted from exothermic PFC beryllium armor/water coolant reaction, significant attention must be focused on application of beryllium in PFC ITER for providing safety.

Simulation of the model experiments enabling the JCT to validate the computer codes based on experimental data, are of interest for studying the hydrogen production dependency on the PFC mechanical and technological parameters, which determine its operation in ITER.

This work is carried out by IAE NNC RK in accordance with Task S81 TT 2096-07-16 FR as follows:

1. experiments on measuring beryllium emissivity in the wide range of temperatures and surface oxidation degrees (subtask B);
2. study of PFC mock-up behavior under accidental conditions in steam (subtask D).

Presently the experimental facility is under development. Below a brief description of research program and facility design is introduced.

2. EXPERIMENT OBJECTIVES AND CONDITIONS

Objectives of the experiment are:
- to determine experimentally beryllium emissivity as a function of surface temperature and its oxidation degree;
- to determine the dependence of the parameters, characterizing intensity of the beryllium/steam interaction on heat balance and temperature distribution in beryllium samples.

The oxidation occurs under the following conditions. Steam with the temperature of 200°C is supplied into the vacuum chamber to a sample heated in vacuum. Steam pressure in the chamber varies from initial to the extreme values with the velocity of 1...2 kPa/s. Steam/beryllium interaction parameters are determined by measuring the temperature of construction parts of the samples and the rate of hydrogen production.

The initial conditions of the experiments are:
- medium in the chamber steam;
- pressure in the chamber not more than 30 Torr;
- temperature of chamber wall approx. 200°C.
Measured parameters in different experiments have the following values:

- initial temperature of beryllium surface 400...1200°C;
- critical pressure of steam in the chamber 0.1, 0.2 MPa.

While testing the first wall it is required to provide the temperature gradient of not less than 100°C/cm at the beryllium layer and to measure mock-up materials temperature during the transient.

3. TESTING SAMPLES

Articles used in the experiments are manufactured by AOOT «UMZ», Ust-Kamenogorsk, in accordance with Subtask C, by the drawings drawn up in IAE NNC RK.

In Fig.2 sample-PFC mock-up testing diagram is shown. The sample is cylindrical unit, consisting of three layers- beryllium, copper-based alloy and stainless steel. The layers are joint with each other by high temperature soldering. The thickness of the layers are:

- beryllium - 3 mm,
- copper-based alloy - 10 mm.

Stainless steel layer thickness depends on minor diameter and determines temperature gradient at the beryllium layer. XA thermocouples are installed along the layers boundary for measuring temperature profile in the sample.

4. EXPERIMENTAL FACILITY

To carry out the experiments a facility for investigating steam/metal interaction has been designed (FISMI), (Fig. 3).
Fig. 3. Facility for Investigating Steam/Metal Interaction

The schematic diagram of the experimental facility is shown in Fig. 4. The facility consists of a vacuum chamber (VC) for placing studied mock-ups and the systems below:
1. evacuation system;
2. system of steam preparation and supply into the chamber (SPS);
3. parameter measuring and recording system (MRS).

The evacuation chamber is two-layer sealed vessel shaped by outer casing (OC) and inner work area casing (IC). The gap between the casings can be filled with flowing coolant.

In Fig. 5 drawing of a VC for experiments on beryllium emissivity factor is shown.

Fig. 4 Drawing of FISMI facility

k1. 10 - Valves
F 1. 3 - Filters
H 1. 3 - Heaters
n 1. 2 - Nozzles
1 - sample; 2 - inner casing; 3 - outer casing; 4 - steam generator; 5 - injector; 6 - inverted cylinders; 7 - water tank; 8 - natural circulation contour

Fig. 5 Vacuum Chamber

The IC is meant for placing studied objects and providing experimental conditions required. It is a unit, which can be removed and placed into a glove box for dismantling and withdrawal of an oxidized sample and washing.
The IC consists of a casing with a cover with a window for pyrometric measurements. The IC also includes a sample holder and a heater. Lay-out of the black body mock-up in the copper block with stainless steel clad and thermal isolation all over the outer surface is shown in Fig. 6.

![Diagram](image)

**T1...T3 - points of temperature measuring**

1. black body mock-up; 2. copper block, shielded with stainless steel; 3. heater; 4. thermal isolation (kaolinite wool); 5. window for pyrometer; 6. technological plug

**Fig. 6. Lay-out of «Black Body» in the Metallic Block**

Water jacket (WJ) is meant to maintain constant temperature of the vacuum chamber inside wall in the course of experiment.

SPS is meant to control the supply of steam with preset parameters into VC.

SPS consists of a steam generator, producing saturated steam, two control nozzles mounted at the steam generator outlet and at the outlet of the vacuum chamber and steam overeater.

The steam generator is a stainless steel vessel with an electric heater installed. Power of the heater may be smoothly changed in the range of 0 to 2.5 kW, that enables controlling steam flow from 0 to 1 gram/s. Pressure in the vacuum chamber and the steam generator is controlled by controlling nozzles 0.35 mm in diameter. The steam overeater is meant for heating saturated steam produced in the steam generator up to the temperature of 200°C and is located in IC of the vacuum chamber. The steam overeater is a copper pipe 40 cm long with the minor diameter of 2 mm.

MRS consists of a controlling computer, an analogous digital converter and initial temperature and pressure gauges. Sampling rate of the gauges is 1...10 c⁻¹, depending on expected velocity of the processes measured. Higher sampling rate corresponds to higher initial surface temperature.

Hydrogen amount is constantly measured in the course of the experiment by volume. Hydrogen displaces water from under inverted graduated cylinder, its volume is determined by the pressure under the inverted cylinder taking into consideration partial steam pressure and hydrogen solvency in water. Water stream vacuum pump is used for supplying hydrogen in under the inverted cylinder from the low pressure area.

5. **SCOPE AND PROCEDURE OF INVESTIGATIONS**

Objectives of methodical stage, conducted before the investigations elaborated in It. 1, are:

- to study the effects of residence time in oxidized media at low pressure on beryllium surface properties and initial pressure in the vacuum chamber;
- to check if operating characteristics of the facility systems meet those designed.

The scheme of the methodical stage is shown in Table 1.

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Scheme of the Methodical Stage</th>
</tr>
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<tbody>
<tr>
<td>Medium in IC</td>
<td>H₂O</td>
</tr>
<tr>
<td>Pressure, kPa, less than</td>
<td>4</td>
</tr>
<tr>
<td>Sample density</td>
<td>1 dense and</td>
</tr>
<tr>
<td>Beryllium surface temperature, °C</td>
<td>1000</td>
</tr>
<tr>
<td>Residence time, hours, not more than</td>
<td>1</td>
</tr>
</tbody>
</table>

Study of beryllium emissivity includes the following steps:

- preliminary measurement of surface emissivity;
Surface emissivity is measured by comparing with the standard and determined as a surface signal (Fig. 1)/ hole signal (BB mock-up, calculated value of emissivity equal to -99) ratio.

Measurements are taken at constant temperature in the range of 900 to 1200 °C in each 100 °C in vacuum to exclude oxidation while measuring.

Matrix of the conditions of beryllium surface oxidation before measuring beryllium surface emissivity is shown in Tables 2 and 3.

Table 2

<table>
<thead>
<tr>
<th>Conditions of Beryllium Surface Oxidation</th>
<th>&gt;99% of theoretical density</th>
</tr>
</thead>
<tbody>
<tr>
<td>T, °C</td>
<td>400</td>
</tr>
<tr>
<td>0.1 MPa</td>
<td>+</td>
</tr>
<tr>
<td>0.2 MPa</td>
<td>+</td>
</tr>
</tbody>
</table>

Table 3

<table>
<thead>
<tr>
<th>Conditions of Beryllium Surface Oxidation</th>
<th>86.5% of theoretical density</th>
</tr>
</thead>
<tbody>
<tr>
<td>T, °C</td>
<td>400</td>
</tr>
<tr>
<td>0.1 MPa</td>
<td>+</td>
</tr>
<tr>
<td>0.2 MPa</td>
<td>+</td>
</tr>
</tbody>
</table>

Scheme of sample-PFC mock-up oxidation conditions is shown in Table 4.

Table 4

<table>
<thead>
<tr>
<th>Conditions of sample-PFC mock-up oxidation.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Beryllium surface temperature, °C</td>
</tr>
<tr>
<td>Temperature gradient at beryllium layer, °C/cm</td>
</tr>
<tr>
<td>Extreme steam pressure in IC, MPa</td>
</tr>
<tr>
<td>Number of PFC mock-ups</td>
</tr>
</tbody>
</table>

Initial field of temperature is created in PFC mock-up by heating beryllium surface. Two main heating methods are being considered - by direct flashing of high frequency current (frequency of 66 kHz) or induction with the same frequency. Heating method will be confirmed upon experimental check.

Post start-up investigations of samples are conducted in AOOT «UMZ» to study consequences of beryllium/steam interaction and include study of the sample characteristics below:

- change in weight and emissivity;
- roughness and microstructure of surface;
- oxide film thickness.

6. CONCLUSION

Above brief description of the program of investigations and facility design has been given. While fabricating and fine-tuning of the facility its design is subject to changes. In general, the design of the facility enables to gain experimental results provided for in Task S 81 TT 2096-07-16 FR.
4.2 Study of Beryllium Redeposition under Bombardment by High Intensity -Low Energy- Hydrogen Ion Beams

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The results of studying the erosion of beryllium under an effect of intense ion fluxes with the energy of 250 eV, at the fluences ~10^{21} cm^{-2}, at the MAGRAS-stand are given. The operating conditions under which a practically-complete redeposition of the sputtered beryllium upon the target surface were experimentally-realized. A change in the microstructure of a beryllium target under sputtering and redeposition is analyzed. Some technological applications are considered.

1. INTRODUCTION

At present beryllium is considered to be a candidate material for the first wall of the ITER. A great number of studies in various laboratories of the world has been recently done on the plasma and ion beam interaction with a beryllium surface. The process of redeposition - which is necessary to be taken into account under expected fluxes of deuterium and tritium onto the first wall of the ITER - will essentially affect the results of beryllium erosion and hydrogen isotope accumulation in it. In this connection, it is necessary to use the intense hydrogen isotope ion beams - under which the redeposition of a material will take place - for simulating the interaction of the plasma with a surface of ITER plasma facing materials (PFM).

In a given paper the results of studying the erosion of the sputtered target, hydrogen accumulation in the deposited Be-layers, as well as the operating conditions for redeposition of beryllium and an effect of this redeposition on the sputtering of the material as a whole, at the

MAGRAS - facility (magnetron-type) under bombardment of beryllium by hydrogen ion beams of various intensity are given.

2. EXPERIMENTAL TECHNIQUE

The studies were done at the MAGRAS-experimental stand [1]. It is based on the magnetron sputtering system (Fig. 1) in the crossed electric and magnetic fields of which the compensated ion beams are accelerated.

![Figure 1. Magnetron: 1- magnetic material, 2 - coil, 3 - anode, 4 - magnetic force lines, 5 - plasma.](image-url)
The systems of vacuum pumping and gas-puffing (Fig. 2) sustain a dynamic pressure in the chamber within the range $5 \times 10^{-4} - 5 \times 10^{-2}$ Torr, at the hydrogen feed flow rate of 1 - 3 cm$^3$/s. The power supply system sustains the discharge voltage up to 600 V at the current up to 7 A. The design of a magnetron, planar type, disc geometry, with a beryllium target includes two electromagnets changing both the magnetic field configurations and its magnitude. In this case, the magnitude of $B$ is varied in the range 0.02 - 0.1 T within the zone of a maximum ion effect (when $r = 20 - 25$ mm).

Realization of tests is essentially-complicated because of beryllium getter properties and high hydrogen solubility in Be that results in the necessity to degas Be-target carefully before starting the discharge up. The Be-cathode conditioning was done with a high voltage - glow discharge. The main discharge ignition in the magnetron resulted in a noticeable dynamic hydrogen pressure drop in the chamber. Since the energy and the ion flux onto target essentially depend on the pressure, a tough pressure stabilization was used in the experiments with an automatic system having the effective feedback realized with the IBM PC.

Along with hydrogen, a part of the experiments were done within an argon medium for manifesting some technological applications.

A few parameters were varied in the experiments to simulate various operating conditions affecting the wall: electric characteristics (current, voltage), hydrogen pressure, magnetic field induction and a magnetic configuration.

The energy of hydrogen ions entering the cathode surface, as a target, was equal 200 and 250 eV. The ion flux density in the experiments was
equal to $3 \times 10^{17}$ and $3 \times 10^{18} \text{ cm}^2 \text{ s}^{-1}$. The heat flux onto the cathode attained 1.25 MW/m$^2$. The target temperature was sustained at the level of 470 K (in the first experiment) and at that of 570 K (in the second one).

For various magnetic configurations the magnetron discharge looked differently. It was characterized by the ion flux density onto the cathode in the range $3 \times 10^{17} - 3 \times 10^{18} \text{ cm}^2 \text{ s}^{-1}$. The general view of such discharges is illustrated by the photographs given in Fig. 3.

The ring-like electric discharge in the magnetron ignites under ~1 kV at the electrodes and develops into a low voltage mode, when the energy of ions entering the cathode is within 200 - 300 eV. In that case, the current either is circuited to a separately-locate anode, or to the vacuum chamber wall. On the separate table (Fig. 4) located at a distance of 150 - 250 mm from the magnetron cathode, the erosion product collectors, as monocrystalline silicon and a basalt tissue, were used.

The polished removable Be-targets, TGP-56, as thin disks or sectors, were located at the magnetron cathode in the experiment with the fixed temperatures of the very target and of the erosion product collectors. The temperature of the latter could be independently adjusted with a special heater, being equal 470 K in the experiments.

The beryllium erosion product studies were done with scanning and transmission electron microscopies. The density of the particles, $>1 \mu \text{m}$ in diameter was determined with an optical microscope.

The deposited film thickness and composition were determined with a profilometer, as well as by the Rutherford back scattering (RBS) technique in the Van de Graaff accelerator. In those experiments the helium ions with the energy of 1.5 MeV were used. The angle of scattering was equal to 160°. Since the mass number of a collector (Si) exceeded the mass number of the deposited material (Be), the He-ion channeling was used. The elastic nuclei recoil detection technique was used in the same accelerator for measuring the integral hydrogen concentration in the deposited layer and the hydrogen atom distribution in depth.

3. STUDY OF BERYLLIUM SPUTTERING AND HYDROGEN ACCUMULATION IN THE DEPOSITED LAYERS

A polished beryllium, TGP-56 - type, has been used as a cathode of a magnetron in this study. Irradiation doses were $2 \times 10^{21} \text{ cm}^2$ (to target 1M) and $3 \times 10^{21} \text{ cm}^2$ (to target 2M). Under the experiments, the cathode temperature was sustained at about 700 K.
3.1. Microstructure of sputtered beryllium surface and deposited layer

The microphotographs of different irradiated Be-surface areas under various magnifications are given in Fig. 5. The differently oriented Be-grains were manifested as a result of ion etching. The grain size is 8-15 μm. At the grain boundaries some pores, up to 1 μm in diameter, as well as cracks, up to ~12 μm long, were produced. Somewhere the burst bubbles are seen.

3.2. Morphological characteristics of Be-erosion products

The structure of a film deposited on a Si-substrate is similar to the collector surface one. Film chips and particles of different size were detected on the fiber collectors. A few types of particles can be discerned by their morphological properties: spherical, hemispherical, irregularly-shaped and plate-like ones. The size of particles was 0.03-6 μm. Small particles, 0.03 - 1 μm in size, are mainly observed among the erosion products, along with the deposition of a Be-sputtered particle film. The particle densities in the ranges from 0.03 μm up to 1 μm and from 1 μm to 6 μm are (4-5).10^4 cm^-2 and 1.4.10^4 cm^-2, respectively.

The electron-microscopic photograph of fibers with Be-erosion products is shown in Fig. 6.

Figure 4. Layout of targets, magnetron, erosion product collectors in the vacuum chamber.

Figure 5. Microphotographs of irradiated beryllium surface.
Fig. 6. Electron-microscopic photographs of fiber with beryllium particles.

The film chips and the particles of various shapes, observed in the erosion products, can be produced under a burst bubble escape from the H-ion sputtered surface or under beryllium crumbling after its cracking along the grain boundaries.

3.3. Deposited film thickness and composition

The thickness of Be-film deposited on the Si-collectors, measured by a profilometer, is equal to 115 and 140 nm for the targets 1M and 2M, respectively.

The distributions of Be-atoms in depth, within the deposited films, are given in Fig. 7. The film thickness is 113 and ~140 nm, that is in the agreement with the profilometer measurement data. The beryllium and oxygen atom contents in the films are in accordance with the ratio BeO_0.8.

3.4. Hydrogen accumulation in deposited film

The hydrogen distribution profiles in the deposited layers of both targets and in silicon before the deposition are given in Fig. 8.

In the layer, 100 nm thick, hydrogen is uniformly distributed and its concentrations are 1.27x10^{22} and 1.36x10^{22} cm^{-3} for the targets 1M and 2M, respectively. With an increase in depth an abrupt reduction of the hydrogen atom concentration is observed. The integral hydrogen concentrations in the deposited films are 1.7x10^{17} and 2.4x10^{17} cm^{-2} for the targets 1M and 2M respectively.

The concentration of hydrogen atoms in a narrow surface layer of the initial silicon target is close to that observed in the deposited layer. The integral hydrogen concentration in the near-surface silicon layer is equal to 3x10^{16} cm^{-2}. The concentrations of beryllium atoms in the BeO_{0.8} film are 6.4x10^{22} and 7.2x10^{22} cm^{-3} for the targets 1M and 2M respectively, corresponding ratios of atoms H/Be are 0.20 and 0.175.

One should note that the ratio of atoms, H/Be, in the deposited layer at 470 K is in agreement with the results [2], where this ratio at the mentioned temperature is equal 0.18.

An estimate of the beryllium sputtering yield for the conditions of a given experiment - made from the results of weighing the target before and after ion irradiation - has given the value 0.02 ± 0.005, that is in good agreement with the data [3].
4. STUDY OF Be-REDEPOSITION

For studying the redeposition operating conditions some special experiments with sector targets were done. In that case, a magnetron modification with permanent magnets - allowing one to produce the ion fluxes of enhanced density onto the target (up to $6 \times 10^{18} \text{cm}^{-2} \text{s}^{-1}$) at a higher hydrogen pressure in the chamber (~ $5 \times 10^2$ Torr) - has provided an effective scattering of the sputtered particles by the atoms of a gas then practically-total return to the magnetron cathode.

A given facility modification has been used for an analysis of Be (TGP-56 - trade mark) - redeposition processes under ion fluences, arriving at the surface, equal ~ $10^{22} \text{cm}^{-2}$. In those experiments the ion energy was equal 200 eV, average particle flux density, ~ $3 \times 10^{18} \text{cm}^{-2} \text{s}^{-1}$, irradiation time, of about one hour long.

A clearly-pronounced (Fig. 9) ring-like structure of the sputtered target-cathode, having some characteristic axially-symmetric zones, has been observed in the realized experiments:

a) central zone of erosion product deposition (1);

b) zone of sputtering-deposition (2) which undergoes an intense ion bombardment. The peak of sputtering is referred to a narrow zone (3);

c) peripheral zone of sputtering-deposition (4);

d) the zone (5) closed to a plasma effect with structural magnetron elements, where the initial beryllium target structure is retained.

Figure 9. The ring-like structure of the sputtered target-cathode.

Some profiles of the target surface roughness after treatment in the discharge, in the sputtering region near the narrow zone (3) and in the region near the edge of the peripheral zones (4), are shown in Fig. 10a, b. As seen, the maximum of erosion belongs to the very dark ring of the zone (3). The material removal from the target surface in this area results in its deepening down to 78 μm.

The greatest deposition of the sputtered particles is observed in the central zone (1), where the characteristics size of the sprayed layer is about 10 μm. The sprayed layer thickness in the peripheral area is about 2.4 μm. Some estimates of the total amount of sputtered and deposited material have shown that they correspond to each other within the accurate range of 5 %, i.e. the practically-complete return of the sputtered particles to the target surface takes place. This fact is also confirmed by the practically-unchanged mass of a beryllium target before and after treatment and the actually-complete absence of the deposited particles (of the silicon and basalt collectors located in the vicinity to the discharge).

The ring-like structure of Be-target is given in Fig. 11a. The initial beryllium microstructure in the zone (5) is given in Fig. 11b. A change in the initial structure in various zones under ion bombardment and redeposition is shown in Figs. 11c - 11f.

Essential changes took places in the central redeposition zone (1) (Fig. 11c). The increasing whiskers, ~ 10 μm high, are clearly-seen here.

The sputtered surface topography is different for the differently-oriented Be-faces: from the ion polishing (Fig. 11d, e) to the productions of cones (Fig. 11f), emerging as a result of different sputtering coefficient for Be and BeO [4].

5. SOME TECHNOLOGICAL APPLICATIONS

The tasks of realizing the Be-coating upon various materials have also been studied at the MAGRAS-facility, together with the studies on sputtering and redeposition.

Since the vacuum chamber walls were covered with a lavsan jacket because of the safety considerations, the Be-coatings upon the lavsan, produced in the experiments, have been analyzed. It turns out, that they are ~ 1 μm thick and possess of good adhesion. Moreover, the sprayed layers are
Figure 10. Profiles of the target surface roughness after treatment in the discharge.

Figure 11. Microstructures of a Be-target in different zones.
ductile, and they are not destroyed even under bending at the angles greater than 90°.

Some technological experiments were done later on a noble gas - argon, heavier than hydrogen - was used for the plasma production. The current - voltage relations of a magnetron discharge with the Be-cathode in argon have the pronounced hysteresis nature (Fig. 12) that is probably related with different coefficients of the secondary electron emission for pure beryllium and its oxide. The low voltage conditions of operation were realized with a relatively-low ion energy (150 - 200 eV), as well as the high voltage ones, with the ion energy up to 450 eV. Under both operating conditions the plasma luminosity was observed in different ranges of a spectrum.

![Figure 12. Typical current-voltage relation of a magnetron discharge with the Be-cathode in argon.](image)

The rates of Be-sputtering by Ar-ions were essentially-higher than those for hydrogen. Therefore, the coatings of a few μm thick were managed to be done for relatively-short time intervals (15-20 minutes).

The Be-erosion products were deposited upon the samples made of the materials similar to those (metals, alloys and dielectrics) used for a number of elements in some ITER-systems (first wall, inner walls of diagnostics paths, optical elements etc.) located at various distances from the target under sputtering and having different temperatures. Analyzing the nature of the deposited layers, the Be-atom layer thickness and the dispersive structure of the deposited particles were registered. An analysis has shown that the deposition of thin Be-films upon the sample with the good adhesion to the backing is a typical situation for a whole number of materials (silicon, cooper, beryllium, steel, glass, lavsan, paper).

The depositions in the form of Be-scales - mentioned in some papers - were not observed.

The Be-coatings, 2 - 3 μm thick, upon the polished backings of some materials for diagnostics ITER-mirrors were one of the magnetron discharge applications. After developing the technology, we have managed to produce optical coatings upon some materials with reflection coefficients exceeding the similar parameters produced by other authors. So, in Fig. 13, the comparative dependencies of the reflection coefficients on the wave length are given for various glass mirrors with the Be-coatings made by us (solid lines) and by the group headed by Dr. M.Nagatsu from the Nagoya University (dashed lines). As seen, the samples made by us have greater reflection coefficients, in the ultraviolet range, first of all.

![Figure 13. Reflection coefficient of Be-mirrors vs wavelength.](image)

For the diagnostics ITER-mirrors the dynamics of their optical property changes under bombardment by ions arriving from the plasma is of great importance. These problems were studied at the Kharkov Phys. & Techn. Institute.

In the case of beryllium there is an interest to study its prospects as a material for manufacturing the plasma-viewing mirrors (PVM) due to its rather good properties in UV region of a spectrum: the reflectance of beryllium mirrors in the visible region is about 30-40% less than for an aluminium mirror, but important fact is that it is more or less stable (at the level ~50%) through rather broad interval of
wavelengths, even near 200 nm as compared to many other metals. The PVM must be located not far from the hot plasma confinement volume and will be strongly influenced with fluxes of different types of radiation. Therefore the estimation of ITER environment effects on optical properties of Be mirrors is needed. In this paper some results concerning the influence of deuterium ions bombardment on reflectance of beryllium mirrors are presented.

The most significant difference in conditions of PVM operation if beryllium is used for the first wall protection will consist in the difference of the composition of those materials that are mass transferred and redeposited due to erosion of inner components of reactor construction. In operating tokamaks the deposit, called "tokamakium", has very complicated composition and includes elements of those materials that have ever been used in a given machine. But in ITER with Be protection the redeposited layer will consist only of beryllium with some (small) level of deuterium, tritium and, possibly, small amount of oxygen.

It means that, principally, there is a possibility of realization of conditions that the surface of beryllium mirror can be self-sustained during fusion reactor operation. Or, if the locations of plasma-viewing mirrors allow to exclude the high rate of Be deposition, the Be mirrors will be subjected to influence of the same factors as the first wall (i.e., fluxes of neutrons, gammas, soft x-rays, charge exchange atoms and so on) of different quantities. But, probably, the case when the deposition rate will be higher than the erosion rate (mainly due to sputtering with charge exchange atoms) can also be real. It means that there is a need to measure the optical properties of a fresh beryllium film on Be substrate obtained not in an ultra high vacuum (as some experimental points with best reflectance values shown in [5] were obtained) but in conditions typical for a fusion reactor.

The Be mirrors were made of 97.5 wt.% hot pressed beryllium (56μm) with 0.9% oxygen content, made in a form of 17mm or 22mm diameter and 4mm thick discs. One of sample planes was mechanically polished up to the mirror quality. Initial reflectance of samples R was close to values presented in [5] for pressed Be mirrors with some increasing in the UV range and is shown in Fig.14 (curve 1). All measurements of reflectance were carried out at almost normal incidence angle.

Figure 14. Spectral dependence of reflectance of the Be-mirror. 1 - as fabricated and after bombardment with D-ions to difference fluence: 2 - $10^{17}$, 3 - $5 \times 10^{17}$, 4 - $10^{19}$ cm$^{-2}$.

The bombardment of mirror samples (two kinds with some different conditions of pressing) was carried out using ions of a reflex discharge in a longitudinal magnetic field with a deuterium as a working gas. Beryllium samples were placed in turn as one of two cathodes with the second cathode of similar size made of a stainless steel. The voltage between both symmetrical cathodes and the anode (the cylindrical vacuum chamber made of a stainless steel) was chosen to be 800 V, and the mean energy of ions hitting the cathode surfaces was near 650 eV with a half width of the energy distribution around 120 eV.

The fluence of ions was evaluated from measured ion current values to the cathodes with fixed diameter of dielectric diaphragm (15mm) under the suppositions that ions are single charged, the role of a secondary electron emission is negligible and the radial distribution of ion flux to the cathodes near their centers, where reflectance was measured, is close to the flat distribution.

After exposure to a definite fluence of deuterium ions in the range from $3 \times 10^{16}$/cm$^2$ to $3 \times 10^{19}$/cm$^2$ the samples were taken out from the installation and reflectance of their central part was measured in the wavelength range $200 < \lambda < 640$ nm using rather simple scheme of measurement (just this spectral
interval was recently used in JET for diagnosing plasma with charge exchange spectroscopy method.

In addition, the depth distribution of retained deuterium was calculated using results of measurements of proton flux coming out from the beryllium sample due to nuclear reactions between trapped D and $^3\text{He}$ ions bombarding the surface of beryllium mirror. The location of a point of these measurements was chosen near the edge of area bombarded with plasma ions, with the aim not to influence on results of optical measurements. Thus, due to radial nonhomogeneity of deuterium flux distribution, the calculated value of trapped deuterium concentration was estimated to be factor 2-3 lower than near the sample center.

Some spectral dependencies of a reflectance obtained for mirrors made of two kinds of hot pressed beryllium are shown in Fig. 14. It is seen from these data that there is observed very strong degradation of mirror quality after already small fluence of deuterium ions ($\sim 2 \times 10^{17}$ ions/cm$^2$), especially in the ultraviolet range of spectrum.

As it follows from Figs. 14, the every dose dependence of $R$ has some level of saturation which is higher for lower wavelength of light. In spite of low accuracy of $R$ measurements in the region 200 $< \lambda < 250$ nm the degradation of mirror optical quality at these wavelengths is so fast and so high that mirrors made of similar kinds of beryllium probably cannot be used for this part of a spectrum in spite a rather high reflectance value.

To understand the reasons of observed change of reflectance due to deuterium ions bombardment, the surface of several Be samples were analyzed by means of SEM (replica method) and TEM.

Later on, this problem is expected to be studied at the MAGRAS-facility to enhance the absolute values of the reflection coefficients for the mirrors with the sprayed Be-coatings, to study the degradation of such mirrors under hydrogen and deuterium ion bombardment, as well as under deposition of the sputtered Be-particles upon their surfaces.

6. CONCLUSION

1. Under irradiation dose of $\sim 10^{21}$ cm$^{-2}$, separate particles, 0.03 $\mu$m up to 6 $\mu$m in size, are registered in the Be-erosion products. Their emergence is provided by destruction of the blisters produced upon the Be-surface in the process of bombardment by highly-intense hydrogen ion beams.

2. At 470 K the ratio of atoms, H/Be, in the Be-layer deposited upon the surface of a silicon monocrystal is 0.185, on the average.

3. At the MAGRAS-facility, under conditions of a magnetron discharge, the operating conditions under practically-complete redeposition of the sputtered beryllium upon the target surface were realized.

4. In the zone of the target sputtering there is an area with a high rate of the particle removal, where the sputtering coefficient exceeds the average values given in literature for similar conditions. This fact can be explained by an effective ionization of the knocked-out Be-atoms in the zone of the maximal magnetic field induction in the magnetron discharge with the subsequent self sputtering under Be-ion bombardment with the energy of about 200 eV.

5. Some technological applications of a magnetron discharge with the Be-cathode have been studied. The test samples of diagnostic mirror with Be-coatings having high reflection coefficients in a wide range of wavelengths were made for ITER.

A cycle of studies on the sputtering and redeposition phenomena under bombardment of Be by deuterium ions in the expanded range of ion energies as well as the study of mutual redeposition of a few materials (Be with W, Be with graphite), are expected to be realized at the MAGRAS-facility in the nearest future.

REFERENCES


4.3 Steam-Chemical Reactivity for Irradiated Beryllium


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Lockheed Martin Idaho Technologies Company
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ABSTRACT

This paper reports the results of an experimental investigation to determine the influence of neutron irradiation effects and annealing on the chemical reactivity of beryllium exposed to steam. The work entailed measurements of the H\textsubscript{2} generation rates for unirradiated and irradiated Be and for irradiated Be that had been previously annealed at different temperatures ranging from 450°C to 1200°C. H\textsubscript{2} generation rates were similar for irradiated and unirradiated Be in steam-chemical reactivity experiments at temperatures between 450°C and 600°C. For irradiated Be exposed to steam at 700°C, the chemical reactivity accelerated rapidly and the specimen experienced a temperature excursion. Enhanced chemical reactivity at temperatures between 400°C and 600°C was observed for irradiated Be annealed at temperatures of 700°C and higher. This reactivity enhancement could be accounted for by the increased specific surface area resulting from development of a surface-connected porosity in the irradiated-annealed Be.

1.0 INTRODUCTION

Evaluation of the safety hazards for the International Thermonuclear Experimental Reactor (ITER) includes an assessment of the failures of plasma-facing-component (PFC) materials due to various accident scenarios.[1] One scenario is a loss-of-coolant accident (LOCA) in which a water-line break injects steam into the torus vacuum vessel. This type of accident is of particular concern for the current ITER design because steam interactions with hot beryllium, the proposed first-wall PFC material, can produce hydrogen via the following reaction.

\[
\text{Be} + \text{H}_2\text{O} \rightarrow \text{BeO} + \text{H}_2
\]  

(1)

Assessment of the consequences of such LOCAs is typically done by means of accident model-simulations [2] that use experimentally-derived, chemical-reactivity data for the material.

This paper presents the results of chemical reactivity experiments for unirradiated, irradiated and irradiated-annealed Be exposed to steam at elevated temperatures. The primary purpose is to evaluate the influence of neutron irradiation effects and swelling on steam-chemical reactivity behavior for Be. The work also entails: (1) annealing experiments, during which tritium and helium release behavior are measured, and (2) characterization measurements of the bulk densities, swelling, porosity, and specific surface areas for the irradiated-annealed Be. Details of the annealing experiments and material characterization are presented in a companion paper at this workshop [3]. Initial results of part of this investigation have been published previously [4]. The emphasis of the present paper is to relate chemical-reactivity data for irradiated Be to changes in the material porosity and surface area that result from heating. Although many studies have investigated swelling and mechanical properties (5-6), swelling evolution (7-10) and gas-release behavior (11-14), all phenomena that are related to the presence of entrapped tritium and \(^{4}\text{He}\) produced in neutron-irradiated Be via nuclear transmutation reactions, few have studied chemical reactivity for irradiated beryllium.
2.0 EXPERIMENTAL DETAILS

2.1 Specimen Characterization

All of the Be specimens used in this work were fabricated using consolidated powder-metallurgy (CPM) techniques and Brush-Wellman SP-200-F powder (< 44 micron size particles with the following chemical composition in weight percent: Be(99.1%), BeO(0.9%), Fe(0.09%), C(0.08%), Al(0.04%), Si(0.03%), Mg(0.02%), and other metals(< 0.04%). To achieve fully-dense material, the powder was cold isostatic pressed, sintered in a vacuum furnace at 1250°C for 4 hours and hot isostatic pressed at 1000°C for 3 hours with a pressure of 103 MPa. Cylindrical test specimens were made by dry-machining, yielding specimens with a 0.76-cm diameter and three different lengths, 0.635 cm, 2.032 cm, and 3.051 cm. No chemical etching was used to remove surface deformation layers from machining.

Irradiation of 0.635-cm and 2.032-cm long Be specimens took place in position 2B1 (near the core center) of the Experimental Breeder Reactor, EBR-II, a fission reactor with a fast neutron environment. Nominal fast neutron fluences (>0.11 MeV neutrons) were estimated to range from $5.2 \times 10^{22}$ n/cm$^2$ to $6.7 \times 10^{22}$ n/cm$^2$, values that are approximately a factor of ten higher than those expected for Be components in ITER. The nominal irradiation temperature was 400°C.

Irradiated Be specimens were annealed in two campaigns. Small cylinders were annealed at temperatures of 450°C, 600°C, 700°C, 800°C, 1000°C and 1200°C, using a segmented thermal ramp with the thermal history patterned after that used in chemical reactivity tests for the control (unirradiated) and irradiated Be. Approximate annealing temperatures and times were as follows: 450°C (300 minutes), 600°C (1440 minutes), 700°C (260 minutes), 800°C (185 minutes), 1000°C (100 minutes), and 1200°C (80 minutes). These times include ramp-up times of ~60 minutes for the two high temperature anneals and ~30 to 40 minutes for the other anneals. In addition, both long and short cylinders were annealed at a temperature of 1100°C for about 35 minutes, following a linear thermal ramp of 20°C/minute.

Measurements were made to characterize the density, porosity and specific surface area for the test specimens, both before and after annealing tests. Densities were obtained by two techniques: (1) measurements of the masses and of the diameters and lengths of the specimens and (2) water immersion density measurements. Porosity was determined from the water immersion measurements. Specific surface areas for the test specimens were measured using a Kr gas adsorption technique, with analysis of the adsorbed gas data by the method of Brunauer, Emmett and Teller, commonly referred to as the BET method. Detailed results of these characterization measurements are found in the companion paper.

2.2 Steam-Chemical Reactivity System

Steam chemical reactivity measurements for the Be specimens were conducted with an experimental system we developed for measuring hydrogen generation rates and tritium mobilization rates for irradiated Be.[17] The system, illustrated in Figure 1, comprises a flow-through, integrated assembly with components set up in an inert-gas glovebox and in a laboratory hood. Argon carrier gas sweeps reaction gases from the hot sample reaction chamber through a process line equipped with condensers, a cryotrap, instrumentation for gas analyses and an oxidizer and ethylene-glycol traps at the end of the process line. Steam introduced upstream from the sample furnace reacts with the hot specimen inside the quartz reaction chamber and is condensed in two Friedrich condensers downstream from the sample furnace. The argon carrier gas transports the reaction gases through the rest of the system.

Hydrogen generation rates and total quantities of H$_2$ generated were obtained from on-line mass-spectrometer measurements of the gas composition in the system process line. Mass peak signatures were mass-2 for H$_2$, mass-4 for $^4$He and mass-40 for Ar. Based on previous studies of gas release from irradiated Be[14], the contribution of HT to the QMS mass-4 peak is negligible relative to that of $^4$He. Tritium measurements were made with an in-line ion chamber and by post-test analyses (liquid scintillation counting) for tritium in the condensed water and ethylene glycol traps.
Figure 1. Illustration of steam chemical reactivity system for measuring tritium mobilization rates and H₂ generation rates for irradiated Be exposed to steam at elevated temperatures.

A precision balance (0.1 mg sensitivity) was used to measure the specimen mass before and after testing. These weight-gain (WG) measurements of the oxygen uptake by the Be provided a second means to obtain the total hydrogen generated during a chemical-reactivity test.

3.0 MEASUREMENTS AND RESULTS

Chemical reactivity experiments were performed by exposing unirradiated, irradiated and annealed-irradiated Be specimens to steam with experimental system parameters as follows: line pressure (~680 torr or 0.9 MPa), Ar carrier-gas flow rate (100 cm³/min), steam flow rate (2500 cm³/min for a water throughput of 2 cm³/min), steam-generator temperature (350°C). For these conditions, the system response time was about 6 min and the H₂ detection sensitivity was about 3 ppm of H₂ in Ar. The on-line QMS was calibrated using Ar-H₂ standard gas mixtures in which the H₂ contents varied from 50 ppm to 50,000 ppm.

Chemical reactivity experiments were performed for unirradiated Be control specimens at temperatures of 450°C, 500°C, 550°C, 600°C, 650°C, 700°C, 750°C, 800°C, 1000°C, and 1200°C; for irradiated Be specimens, at sample furnace temperatures of 450°C, 500°C, 600°C, and 700°C; and for annealed-irradiated specimens at temperatures from 400°C to 600°C. Steam exposure times for the irradiated specimens tested at 450°C, 500°C, and 600°C experiments were 22.6 h, 24.6 h, and 7.4 h respectively, times that were comparable to those for Be control specimen tests. The steam exposure time for the irradiated Be experiment at 700°C was about 40 minutes because the Be specimen experienced a significantly enhanced reaction rate that nearly consumed the specimen in this time period. A steam exposure time of 400 minutes was used for the chemical reactivity tests.
A summary of the results of these steam reactivity experiments is presented in Figure 2, along with the results of previous measurements by Smolik et al. for fully dense CPM-Be [18] and for porous Be [19]. This figure plots average $H_2$ generation rates for the various experiments as a function of the inverse temperature (10,000/K) along the bottom axis and the corresponding temperature in degrees centigrade along the top axis. Results are presented as average values in that they typically correspond to the total quantities of $H_2$ generated during an experiment divided by the steam exposure time and the geometric surface area of the specimen prior to the chemical reactivity experiment. The geometric surface areas used in these computations correspond to values derived from specimen dimensional measurements of the unirradiated, irradiated and annealed-irradiated Be. For the experiments at 600°C and below, the results are referenced to a time of 400 minutes. $H_2$ generation results based on mass-spectrometer gas measurements are identified with the G and those values derived from weight-gain measurements are designated with a WG. Results corresponding to experiments for unirradiated Be cylinders are identified by INEL96-G2, -WG2 and those for unirradiated Be discs are identified by INEL92-G, -WG. Straight lines are drawn through the data for unirradiated Be to indicate the general trends in
chemical reactivity for three different temperature ranges. The results for experiments on irradiated Be cylinders are designated by INEL96IR-G1, -WG1. Similarly, the results corresponding to experiments with annealed-irradiated Be cylinders are designated by INEEL97IRA-G1, -WG1, -G2, and -WG2. The INEEL97IRA-G1 and -WG1 data represent reactivity trends at 500°C as a function of different anneal temperature, and the INEEL97IRA-G2 and -WG2 data represent reactivity trends as a function of steam-reactivity temperature for specimens annealed at 1100°C. Previous measurements by Smolik et al. for porous Be with a bulk density of 88% are identified as INEL92Porous-G, -WG. A straight line is fit through these data to indicate the magnitude and trend of chemical reactivity for porous Be.

These results show a variation in the observed H₂ generation rates, dependent on the type and condition of the test specimens and the temperature of the steam-exposure experiment. For test temperatures from 450°C to 600°C, the H₂ generation rates for irradiated Be are similar to those for unirradiated Be, although there is some indication that the rates are somewhat higher at 500°C for the irradiated specimen. A significant difference is observed in reactivity of the irradiated Be tested at 700°C. H₂ generation accelerated very rapidly for this specimen and the specimen experienced a temperature excursion to above 1000°C. Experiments for the irradiated-annealed Be gave H₂ generation rates that were significantly higher than those for unirradiated or irradiated Be. However, the rates for the irradiated-annealed Be were much less than those for porous Be.

As shown in Figure 3 for the irradiated specimen exposed to steam at a test temperature of 700°C, the increase in H₂ generation correlates with the rapid mobilization of tritium and helium that were trapped in the material. The rapid release of the tritium and helium are associated with the development of a porosity network that becomes surface-connected to relieve internal gas pressure. The resulting surface area enhancement leads to an accelerating chemical reactivity and temperature excursion as the chemical energy is deposited in the material. High H₂ generation rates continue for tens of minutes after the temperature excursion as steam interacts with a high surface area material.

![Figure 3. Relative tritium and \(^4\)He mobilization rates and H₂ generation rates for an irradiated Be specimen exposed to steam at a test temperature of 700°C. The specimen experienced a temperature excursion between times 8 and 18 minutes.](image)

Irradiated Be that was exposed to steam at a test temperature of 600°C showed a complex reactivity behavior, as presented by the data plotted in Figure 4. This figure shows that, initially, the reaction kinetics were parabolic as indicated by the fast rise and subsequent fall-off in the H₂ generation rate. However, after steam exposure of about 150 minutes, the H₂ rate began to rise, and it rose continuously until the experiment was terminated. The tritium mobilization rates exhibited a similar behavior, indicating a correlation in the steam chemical reaction and the liberation of tritium from the material. It is not clear whether tritium mobilization in this case is indicative of a developing surface-connected porosity, leading to an increase in chemical reactivity and hydrogen generation or to a gradual breakdown in the oxide protective layer. No similar tritium release behavior was observed during the anneal studies [3].

H₂ generation rates for irradiated-annealed Be were much higher than those for unirradiated or irradiated Be. With the exception of the data point corresponding to a specimen annealed at 1000°C, the results (INEEL97IRA-G1,-WG1) in Figure 2 indicate a systematic increase in reactivity for irradiated Be annealed at progressively higher temperatures. The deviation of the 1000°C anneal
result from this trend may be due to the generation of a surface-connected porosity with a higher specific surface area. Results identified by INEEL97IRA-G2, -WG2 in Figure 2, for irradiated Be annealed at 1100°C and tested at 400°C to 600°C, show a systematic increase in reactivity with test temperature. We note, however, that H₂ generation rates for irradiated-annealed Be are much lower than rates observed for unirradiated, porous CPM-Be.

Differences were observed in the reaction kinetics for the various Be specimens tested in steam at 500°C. As shown by the data plotted in Figure 5, the reaction kinetics were parabolic for unirradiated and irradiated specimens and accelerating for the irradiated Be that was previously annealed at 1000°C. The results shown in this figure for the irradiated-annealed specimen bear some resemblance to the data shown in Figure 4 for the irradiated specimen tested at 600°C, indicating the role of porosity and surface area, especially a growing surface area on the hydrogen generation rates.

The observed differences in chemical reactivity of the unirradiated and irradiated Be, compared to the reactivity for irradiated-annealed Be and porous Be, can be accounted for, to a great extent, by the higher specific surface areas for the annealed and porous materials. This is demonstrated by Figure 6 that presents the average H₂ generation rates with the values for the irradiated-annealed Be and the porous Be normalized to BET surface areas. As discussed in our companion paper [3] for the CPM-100%TD Be and for the irradiated Be, we were unable to measure the BET surface area, because it was far below the sensitivity of the BET instrument. Hence, geometric area was chosen to be equivalent to BET surface area for these data. In contrast, the measured BET surface area was much greater than the geometric surface area for CPM-88%TD Be. Similarly measured BET surface areas were used for the data corresponding to irradiated-Be annealed at 1000°C and 1200°C. Measured BET surface areas for the irradiated Be annealed at 1100°C were about a factor of 2 above the sensitivity of the BET instrument. BET surface areas for irradiated specimens annealed at 700°C and 800°C were assumed to be at the sensitivity of the instrument. We note that the surface-area correction brings the H₂ generation data in much better agreement, indicating the significance of surface-area determinations. However, the corrected data for the porous Be data at test temperatures near 700°C are somewhat lower than those for the control material indicating that the surface area correction may become less significant as the test temperature increases above 600°C.
Figure 6. Average $H_2$ generation rates relative to BET surface area for unirradiated (INEL92-G,-WG; INEL96-G2,-WG2), irradiated (INEL96IR-G1, -WG1), irradiated-annealed (INEEL97IRA-G1, -WG1, -G2, -WG2) and porous (INEL92Porous-G) CPM-Be. G and WG identify mass-spectrometer-gas and weight-gain data, respectively.

4.0 DISCUSSION AND CONCLUSIONS

We have highlighted in this paper the results of an experimental investigation to evaluate the influence of neutron irradiation effects and annealing on chemical reactivity of Be exposed to steam at elevated temperatures. The work entailed annealing experiments and material characterization measurements, that were presented in the companion paper [3]. This paper concentrated on the salient features of steam-chemical reactivity experiments.

In this work, we found that neutron irradiation of Be influenced the steam-chemical reactivity, with the magnitude of the effect dependent on the steam exposure temperature. For test temperatures between 450°C and 600°C, the general trend in $H_2$ generation rates for irradiated Be was similar to that for unirradiated Be, although there were indications that the rates for irradiated materials at 500°C and lower could be a little higher. For temperatures of 600°C and below, reactivity kinetics for the irradiated material were parabolic. However, we observed for irradiated Be tested at 600°C that after about 150 minutes of steam exposure, the reactivity...
behavior changed from parabolic to a gradual accelerating behavior. More dramatic increases in the chemical reactivity for irradiated Be were observed at a test temperature of 700°C. At this temperature, the \( H_2 \) generation rate accelerated very rapidly and the specimen experienced a temperature excursion to 1000°C or higher. This behavior is attributed to the development of swelling and a surface-connected porosity network for the irradiated Be, resulting in enhanced reactivity and heat deposition in the material causing the temperature excursion and the accelerating \( H_2 \) generation.

Annealing of irradiated Be at temperatures of 700°C and higher produced enhancements in the steam-chemical reactivity for annealed material relative to unirradiated and irradiated Be. At a test temperature of 500°C, the reaction kinetics were parabolic for unirradiated and irradiated Be, but they were accelerating for irradiated-annealed Be. This resulted in a much higher \( H_2 \) generation rate for the annealed material.

The enhancement in the chemical reactivity for the irradiated-annealed Be could be approximately accounted for by differences in the measured specific surface areas for the tested materials. We note, however, that the surface-area corrections applied to the data for specimens annealed at 1100°C did not produce as good of agreement with the unirradiated data. The measured BET surface areas for the 1100°C-annealed Be were much smaller than expected, although the swelling was comparable to that for specimens annealed at 1000°C and 1200°C, specimens for which the measured specific surface areas were substantial. The measured chemical reactivity data for the tests with the 1100°C-annealed Be indicate that the annealed specimens might have a larger specific surface area than we measured.

Although the measured \( H_2 \) generation rates for irradiated Be annealed at 700°C and higher, were much more than rates for unirradiated Be, they were still smaller than measured rates for porous Be with a bulk density of 88% theoretical. We infer from this that the surface-connected porosity network that develops during annealing of irradiated Be does not create a porosity that exposes all the particles comprising the consolidated Be, in contrast to the case for the 88% density CPM-Be. From this perspective, the reactivity for porous Be serves as an upper bound to \( H_2 \) generation for the forms of Be tested in this work.

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REFERENCES


5. Forming and Joining
5.1 Joining of Beryllium by Braze Welding Technique: Preliminary Results

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Within the framework of some applications, there is a need to join beryllium parts to each other. Gas Tungsten Arc Braze Welds were made in beryllium using 0.3 mm commercially Aluminum (1100) shim preplaced at the joint. The welds exhibited a tendency to form microcracks in the Fusion Zone and Heat Affected Zone. All the microcracks were backfilled with Aluminum.

1. INTRODUCTION

Beryllium has many properties that make it extremely interesting as an engineering material especially for aerospace and many engineering applications. To realize its maximum capabilities and widest applicability, beryllium must be capable of being welded. The combination of the unique properties, high elastic modulus, high thermal conductivity, high strength and limited room temperature ductility, have caused extensive problems in satisfactorily beryllium joining. Thermal and mechanical stresses arising during the welding operation can exceed the intergranular strength in cooling and cause failure. Fusion welding of beryllium is characterized by a tendency to hot crack. Over the years, there has been an ongoing interest in eliminating cracking in fusion welding by the addition of aluminum. The typical rule of thumb is that at least 30-wt% Al (~10at%) must be added to the weld to suppress cracking at room temperature. Thermodynamic calculations suggest that there is a sub-liquidus miscibility gap in the liquid+solid region at high beryllium concentrations in the Al-Be binary system [1]. This suggests the possibility of liquid phase separation occurring in high beryllium compositions. A subsequent rapid solidification could result in intimately, continuous mixed, regions of beryllium and aluminum [2].

The purposes of the present investigation were:

i. Determine the feasibility of welding beryllium by Pulsed Gas Tungsten Arc-Braze-Spot Welding (GTA-B-SW) with preplaced thin shim; ii. To characterize the microstructure of the FZ and HAZ; iii. To determine the dependency of crack susceptibility on the aluminum composition and welding parameters.

2. EXPERIMENTAL

Welding experiments were performed on small diameter (20 mm) thin (2 mm) rings made by fine machining (0.2 \( \mu \)m Ra) of beryllium rods. Joint edges were carefully prepared to remove chipped edges remaining from the machining operation. All the experiments were made in an evacuable controlled chamber under argon atmosphere. A commercially pure Al-1100 (99.0Al+4.5Cu + 1.5Mg+0.6%Mn) shim, 0.3-mm thick, was preplaced at the interface between pieces of beryllium to be joint – Fig 1. Both the beryllium and the shim materials were cleaned, by etching with acid, prior to welding. The beryllium etchant was composed of 2% HF, 48% HNO\textsubscript{3} and 50% H\textsubscript{2}O. The aluminum etchant was composed of 2% HF, 48% HNO\textsubscript{3} and 50% H\textsubscript{2}O. The aluminum etchant was composed of 10% H\textsubscript{2}SO\textsubscript{4} 48 gr. H\textsubscript{2}CrO\textsubscript{4} 1 % HF and 85% H\textsubscript{2}O. Spot-Braze welding weldments, were produced using straight polarity pulsed Gas Tungsten Arc Welding (GTAW) in flat position. Satisfactory welding conditions were developed for partial and full penetration. Radiographic tests were conducted to detect the presence of voids,
inclusions or other weld discontinuities. Following nondestructive testing, the welds were tested destructively by optical and electron (Scanning Electron Microscopy-SEM) metallography. Transverse sections of the weldments were examined metallographically to determine the degree of Al mixing in the Fusion-Zone (F.Z), and to detect and characterized cracking when present. Welding specimens were prepared for metallographic analysis by wet polishing from 240 grit to 0.25 μ diamond paste grit without etching.

3. RESULTS AND DISCUSSION

A schematic sketch of the joint design including the preplaced aluminum shim is shown in fig 1. Figure 2-a shows the typical spot-weld surface macrostructure. Figure 2-b shows enlargement of typical F.Z surface hot cracking, lying in the original Al shim line. Figure 3-(a,b) shows the typical transverse macrostructure of beryllium-aluminum braze weld. The bright phase in all micrographs is aluminum phase as will be shown later. Aluminum channels in the F.Z, lying from the top surface to the bead root, clearly shown in figure 3-a. Variation in aluminum content and aluminum channels clearly seen in figure 3-b. Figure 4-(a,b) shows the F.Z microstructure in two different locations within the joint. Figure 4-a shows the microstructure in the center. Figure 4-b shows the microstructure in the bottom of the bead. The microstructure in the F.Z consists with Like-Eutectic (L.E) microstructure - fine dispersion of primary beryllium and continuous aluminum. The center F.Z solidified in slow rate (coarse microstructure) whereas, the bottom part of the F.Z solidified in rapid solidification (fine microstructure). Figure 5-(a,b) shows aluminum channels near the step area and aluminum channels that extended to the HAZ respectively. Aluminum channels backfilled two kinds of microcracks; i. Microcracks that nucleated in the F.Z during cooling. ii. Prior microcracks in the base metal. Liquid aluminum backfilling accommodate strains and inhibit the growth, branching and propagation of microcracks during cooling. Secondary and Back-Scattered SEM micrographs shows in Figure 6-(a,b) respectively. Beryllium nucleate via dendritic mode while aluminum phase liquates continuously the interdendritic regions. Radiographic inspection was found as an effective tool for determining the quality of the braze welds and the degree of aluminum mixing in the F.Z. Figure 7 shows a typical radiographic micrograph of braze welds. The difference between the absorption coefficients of aluminum and beryllium enables the observation of the following parameters; i. Variation in aluminum concentration in the F.Z. ii. Location of cracks (in aluminum or beryllium regions), iii. Location of aluminum stringers according the following relation:

\[
\mu_{F.Z,\text{Average}} = C_{\text{Be}} \mu_{\text{Be}} + C_{\text{Al}} \mu_{\text{Al}}.
\]

\[
\mu_{F.Z,\text{Average}} = \text{ Average absorption coefficient.}
\]

\[
C_{\text{Be}}, C_{\text{Al}} - \text{ Beryllium and aluminum Concentration, respectively;}
\]

\[
\mu_{\text{Be}}, \mu_{\text{Al}} - \text{ Beryllium and aluminum absorption coefficient, respectively.}
\]

Variation of aluminum content in the F.Z without any discontinuities (inclusions, porosity etc) shown in the radiographic micrograph in figure 7-a. Figure 7-b shown microcracks initiation in the step area which have been backfilled with aluminum. Good aluminum mixing also shown in figure 7-b.

4. CONCLUSIONS

1. Relatively good joint of beryllium parts was achieved.
2. The surface of the FZ is sensitive to hot cracking along aluminum channels.
3. The FZ has a microstructure typical to a pure component eutectic, i.e., very fine dispersion of beryllium primary phase and continuous aluminum as a secondary phase.
4. The beryllium nucleates via dendritic mode with aluminum backfilling along the interdendritic zones.
5. Aluminum from the FZ extends along grain boundaries into the base metal (HAZ) and form "Aluminum Stringers".
6. Aluminum Backfill the “Cracks Like”, absorbed the thermal strain and inhibit crack branching and propagation in the FZ.
7. The radiographic inspection is an excellent technique for beryllium joint when using aluminum foil.
REFERENCES


Figure 1. Schematic sketch of the joint design. Before welding; Enlargement of the Brazed F.Z.
Figure 2. Top surface view of typical spot weld.  a. Macrostructure;  b. F.Z surface hot cracking.
Figure 3. Typical transverse macrostructure of Aluminum-Beryllium braze welds. a. Macrostructure; b. Variation in Aluminum content channels backfilled microcracks in F.Z.
Figure 4.  
a. Center of the F.Z. - Slow solidification;  
b. Fine microstructure in the bottom of the F.Z. - Rapid solidification
Figure 5. a. Aluminum channels near the step area; b. Aluminum channels that extended to the HAZ in the base metal.
Figure 6. F.Z solidification mode.  
a. Secondary emission SEM micrograph showing beryllium nucleation in dendritic mode;  
b. Back-Scattered (Compo) emission showing interdendritic backfilling by aluminum.
Figure 7. Typical radiographic micrograph of braze welds.  

a. Variation in aluminum content without any discontinuities;  
b. Microcracks in the F.Z backfilled with aluminum.

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5.2 Be-Cu Gradient Materials through Controlled Segregation
-Basic Investigations-

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ABSTRACT

The joining of materials has a fundamental problematic nature: Creating a sharp interface between two different materials causes a more or less extreme jump in the properties at this point. This may result in the failure of the component under mechanical or thermal loads. In some cases there are further difficulties caused by using a third component (e.g. the transformation of Ag-lead into Cd by neutron beams). The solution may be the creating of a functionally gradient material (FGM) Be-Cu.

We discuss the advantage of such a FGM and the probabilities of an new procedure for manufacturing 1-dimensional FGMs.

1. INTRODUCTION

We are engaged in the development of Be-Cu gradient materials on a mesoscopic scale, which might be interesting for fusion applications. At present, we are still at the beginning of our investigations and therefore currently trying to answer two basic questions:

1. Why might it be useful to introduce a gradient into the Be-Cu-transition?
2. How can we implement a mesoscopic gradient?

2. BE-CU-GRADIENTS

There are two main reasons for introducing such a gradient:

1. to avoid additional alloying in the region of transition.
2. to reduce residual stresses in the vicinity of the interface zone.

To obtain an idea of the expected macroscopic residual stresses in a functionally gradient material on the beryllium - copper - base, the macroscopic thermoelastic residual stresses were calculated first for the room temperature case by a procedure initiated by Ravichandran [95Rav]. For an applied temperature gradient, a procedure conceived by Wakashima et al. [90Wak] was used.

Figure 1 shows the approximation for the room temperature case: The upper part shows the copper

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Figure 1: Implementation of a mesoscopic gradient (short range gradient)
content in the case of a conventional sharp interface, the lower part shows the corresponding strong shear stress which is responsible for cracking under high thermal and mechanical loads.

If we introduce a short range gradient of a few millimeters, this shear stress can be reduced by a certain amount. But on the Be surface this does not have much influence on the residual stresses. This situation changes if we extend the gradient over a wider depth (figure 2; the upper part shows again the concentration distribution and the lower part the corresponding stress distribution).

If we were able to establish such a smooth gradient, then a significant stress reduction could be expected. Moreover, the stress close to the Be-surface would be accessible to manipulation depending on the gradient function type and even the introduction of compressive stress at the surface could be produced, which might be helpful in preventing crack propagation.

By applying a thermal gradient, as shown in figures 3 and 4 (500 °C on the surface and room temperature 25 mm away from the surface), a significant reduction of the residual stress according to an appropriate gradient function can be chosen.

3. GRADIENT IMPLEMENTATION

 Actually we are concerned in developing a new technique which might be very convenient because it would be self-adjusting if an advanced precursor-powder is available. This technique is dependent on the distribution parameters within this particular precursor powder.

The basis of this technique is the vibration-induced segregation of particles, which is not yet well understood, but there has been ongoing research in the last years. The effect is as follows:

![Figure 5: Sample Configurations for 50 - 50 Binary Distribution of Disks](image)
If particles of different sizes are filled into a box which is under vibration, then as simulated in the Monte-Carlo-Simulation (MCS), larger particles are lifted up because there is a higher probability of smaller particles dropping underneath them during vibration (figure 5), [86Ros].

Following this simulation, we assume that for size ratios above 2.8 the segregation proceeds with time, but at lower ratios, it stops after a small number of vibration circles at a low level of segregation (figure 6), [92Jul].

We carried out some experiments to prove these results of the MCS and found that it is incorrect. The reason for this is, that this process is much more complicated to simulate than assumed in the MCS. This simulation considers neither collective effects nor interactions between particles such as particle rotation, friction of particles which each other, inelastic collisions and surface roughness.

Very recently, Barker and Moreau attempted to improve the understanding of size segregation by using molecular dynamics [93Bar], [93Mor]. They considered cooperative effects with complex functions of both granular structure and coupling with applied force. They showed that the segregation path of a particular particle interact with additional convection rolls of the other particles.

Thus these simulation techniques cannot provide the parameters sufficient for initial powder size and density distribution for optimized vibration stimulation. Experimentally we found that the segregation time is shorter for a larger size ratio.

For a certain stimulation with 7 Hz and 15 mm the segregation is accelerated by more than two orders of magnitude (figure 7). If the size ratio is increased by just 50 %, the segregation time is reduced significantly, if the frequency and amplitude are enhanced, this means the acceleration rises.

However there is a strong limit for increasing the acceleration above a certain limit of g ≈ 1.8 (figure 8): The segregation reaches a maximum value and then falls back to zero [97Hsi].

The reason for this is that particles above this limit are no longer in touch with each other, so that friction forces are reduced and the segregation can be neglected. This can also be seen in the curve below, (figure 8), because the effective density of the ball mixing remains constant until the maximum is reached and then drops down rapidly to a lower level.
Our next aim is to scale down these macroscopic results to a microscopic level in order to find the optimum acceleration for an optimized particle size distribution. At present, after initial investigations, we can assume that the principle also seems to work in microscopic powders (figure 9). Cu-powders of an average grain size of 63 µm were shaken in a cylindrical container and the grain arrangement was fixed. After metallographic preparation an image analysis was performed. It can be observed in these pictures that the larger particles went to the upper region, whereas the smaller particles went downwards. This is confirmed by the grain size plot of both grain distributions.

4. PRECURSOR POWDER

The second question was how to control such a functional gradient, which should be done by coating the beryllium powder grains with copper. By doing this we add a density effect (beryllium has a density of ≈ 1.8 g/cm³ and copper a density of ≈ 8.9 g/cm³) to the discussed size effect. If the layer thickness is equal, as shown in figure 10, the smaller particles will have an effective density closer to the copper density, whereas the density of the larger particles will be closer to that of the beryllium density.

This is an additional driving force for segregation, causing the small Cu-rich particles to move downwards and the big Be-rich particles upwards. This may provide us with a possibility of forming such a mesoscopic gradient.

Beryllium powder coating is not easily accomplished, but we have already succeeded in room temperature deposition of Cu on Be in a hydrous solution by a suitable reduction of Cu-ions. (figure 11).

Last but not least we have to consider the consequences of the unlimited formation of intermetallics like Be₃Cu or BeCu [79Web].

The sintering procedure of the powder should be successful in a dense material but without the huge formation of intermetallics.
The right hand picture (figure 12) shows a region after sintering at 1000 K. The pore was produced because all beryllium diffused away into the copper forming intermetallics. The middle image shows the microstructure after 10 min. of sintering with diffusion zones being observed. On the left hand side, appropriate sintering temperature was chosen for the purpose of avoiding the formation of intermetallics.

5. FURTHER INVESTIGATIONS

In the future we will concentrate on the following topics:

- Coating of beryllium powders
- Calculation of the optimized gradient for fusion applications
- Implementation of the optimized mesoscopic gradient
- Consolidation of the precursor powder
- X-ray measurements of the residual stresses and comparison with calculated data
- Non-destructive testing
- Destructive testing

These future investigations will also include an advanced characterization of the microstructure including orientation imaging microscopy because this method allows us to identify the different phases directly in the metallographic plane, the orientation of the grains as well as the misorientation between the grains.

7. ACKNOWLEDGEMENTS

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6. REFERENCES

5.3 Interface Structure of Be/DSCu Diffusion Bonding

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Abstract

Beryllium is used as plasma facing components of the first wall on ITER. And Dispersion Strengthened Copper (DSCu) is used as heat sink material by joining to Be because DSCu has high thermal conductivity and strength. In this study, Be/DSCu diffusion bonding tests using the interlayer of Al, Ni, Nb, Ti, Zr and Be-Cu alloy have been conducted to choose the suitable interlayer materials. As a result of the shear strength tests, Be/DSCu joints by using Be-Cu alloy interlayer showed the strength of about 200 MPa. Diffusion bonding tests using Be-Cu alloy interlayer or no interlayer (direct bonding) at the range of temperature from 600°C to 850°C have been conducted to identify the effect of bonding temperature and time on interface formation and strength. The thickness of diffusion layer was linear to a square root of bonding time by diffusion controlled process. The shear strength is controlled by the formation of intermetallic layer at Be side.

1. Introduction

Beryllium has been proposed to be used as plasma facing components of the first wall on ITER. And Dispersion Strengthened Copper (DSCu) is used as heat sink material by diffusion bonding to Be because DSCu has high thermal conductivity and strength. In recent research of Be/Cu bonding, Be/Cu diffusion bonding tests using various interlayer have been conducted to choose the suitable interlayer materials[1] and out-of-pile reactivity tests with diffusion couples between Be and Cu were conducted to clarify the generation of reaction products and increasing of the reaction layer.[2] In order to obtain high strength and reliability for high heat flux, the control of interface is important technique.

In this study, Be/DSCu diffusion bonding tests using various interlayer have been conducted to choose the suitable interlayer materials and identify the effect of bonding temperature and time on interface formation and strength.

2. Experimental

2.1. Specimen

Be specimens were hot pressed beryllium produced by NGK INSULATORS, LTD. DSCu were produced by SCM METAL PRODUCTS, INC.. Bonding components of Be and DSCu are the cylinder of 10mm diameter and 20mm length. The interlayer were cut from commercial purity foil of 50 or 100μm thick of Al, Ni, Nb, Ti, Zr and Be-Cu alloy (Cu-1.8%Be).

The jig of diffusion bonding is shown in Fig. 1.
Bonding of Be/DSCu was accomplished by hot pressing technique under a pressure of 50MPa for 1 hour in vacuum. Bonding temperature were 500 °C for Al and 700 °C for Ni, Nb, Ti, Zr and Be-Cu alloy. The specimens were heated in 1hr to the bonding temperature.

![Fig 1 Jig of diffusion bonding.](image)

To make clear the effect of bonding temperature on strength and microstructure of interface, a series of Be/DSCu joints using the interlayer of Be-Cu alloy or no interlayer (direct bonding) was formed at various temperature in the range 600 to 850 °C under a pressure of 50MPa for 1 hour. At 700 °C a series of joints was formed at various bonding time in the range 1 to 6 hours.

2.2. Evaluation of Be/DSCu joint

Interface structure analysis of polished cross sections were carried out for Be/DSCu joints by scanning electron microscope. The thickness of reaction layer was measured by SEM images. The distribution of diffusion element were measured by line profile of energy dispersive spectrometer. The reaction layer were identified by X-ray diffraction.

The quality of Be/DSCu joints was assessed by shear strength testing conducted at the testing rate of 0.5 mm/min at room temperature. Fig.2 shows the jig of shear strength testing. Shear strength was calculated as the failure load divided by the surface area. Fracture path was identified from the fracture surface.

3. Results and discussion

3.1. Interface formation for various interlayer

Fig. 3 shows the SEM micrograph of joint formed at 700 °C using the interlayer of Al, Ni, Nb, Ti, Zr and Be-Cu alloy.

(a) Be/Al/DSCu joint

Diffusion layer about 10 μm were induced at DSCu side. The diffusion layer are consisted of two layers at Al/DSCu interface. η phase formation at Al side and α phase formation at DSCu side were observed. Shear strength was 70MPa. Fracture path was the interface of Be/Al.

(b) Be/Ni/DSCu joint

Diffusion layer were not observed at both Be and DSCu side. Debonding occurred at the interface of Be side.

(c) Be/Nb/DSCu joint

Diffusion layer were not observed at both Be and DSCu side. Shear strength could not be measured because the joint was debonded in making the specimen for shear strength testing.

(d) Be/Ti /DSCu joint

Diffusion layer about 10 μm were induced at DSCu side. TiCu phase was observed. Shear strength could not be measured because the joint was debonded in making the specimen for shear strength testing.
Fig. 3 SEM micrographs of joint formed at 700 °C using the interlayer of Al, Ni, Nb, Ti, Zr and Be-Cu alloy.
(e) Be/Zr/DSCu joint
Be was diffused in Zr interlayer like the island. Ductile intermetallic phase was observed. Diffusion layer about 10 µm were induced at DSCu side. Debonding occurred at the interface of Be side.

(f) Be/Be-Cu alloy/DSCu joint
Diffusion layer are consisted of two layers. Be₂Cu phase formation at Be side and Cu+BeCu(γ) phase formation at DSCu side were observed. Shear strength was about 200 MPa. Fracture path was the interface of Be/5 phase.

3.2. Interface formation for Be-Cu alloy interlayer and direct bonding

Fig. 4 shows the SEM micrograph of joint formed at 700 °C and 850°C by direct bonding. Diffusion layer of these joints are consisted of two layers. Diffusion layer was identified by X-ray diffraction. In the joint formed at 700°C for 1 hr, Be₂Cu (δ) phase formation 6 µm thickness at Be side and Cu+BeCu(γ) phase formation 27 µm thickness at DSCu side were observed. In the joint formed at 850°C for 1 hr or 700°C for 6 hr, diffusion layer had the same structure as the joint formed at 700°C for 1 hr. Diffusion layer generated in comparison with 700 °C for 1 hr. The generation rate of (Cu+γ) phase was larger than that of Be₂Cu (δ) phase.

Fig. 4 SEM micrographs of joint formed at 700°C and 850 °C by direct bonding.
layer with bonding temperature. As increasing bonding temperature, \((Cu+\gamma)\) layer generated several 100 \(\mu\)m. \(Be_2Cu\) layer generated slowly under 50 \(\mu\)m. In diffusion controlled reaction the thickness of diffusion layer is expressed by parabolic law. Fig.6 shows the changes of the thickness of diffusion layer with bonding time at 700°C. The thickness of diffusion layer was linear to a square root of bonding time as same as the case of annealing at 700°C. [2]

3.3. Shear strength of \(Be-Cu\) alloy interlayer and direct bonding

Fig.7 shows the bonding temperature dependence of shear strength. The shear strength of joints formed under 750°C were about 200 MPa in spite of the growth of diffusion layer except joint using the interlayer of \(Be-Cu\) alloy at 600°C. The joints formed up to 800°C showed the degradation of shear strength.
Fracture path were the interface of Be/Be$_2$Cu phase and so the shear strength was not sensitive for the thickness of diffusion layer.

Fig. 8 shows the bonding time dependence of shear strength of joints formed at 700°C. The joint formed for 6 hours did not show the large degradation of shear strength because interface layer at Be side was still Be$_2$Cu phase for 6 hours. From the above it may be concluded that the shear strength is controlled by the formation of intermetallic layer at Be side.

4. Conclusions

Be/DSCu diffusion bonding tests using various interlayer have been conducted to choose the suitable interlayer materials and identify the effect of bonding temperature and time on interface formation and strength. The following conclusions were reached.

1. In the joints using Al, Ti interlayer, diffusion layer were observed at DSCu side. In the joints using Ni, Nb interlayer, diffusion layer were not observed at both Be and DSCu side. In the joints using Zr interlayer, Be was diffused in Zr interlayer like the island and diffusion layer about 10 μm were observed at DSCu side. As a result of the shear strength tests, Be/DSCu joints by using Al interlayer showed the strength of 70 MPa.

2. The diffusion layer of Be/DSCu joints using the interlayer of Be-Cu alloy and direct bonding are consisted of Be$_2$Cu(8) phase at Be side and Cu+BeCu (γ) phase at DSCu side.

3. The thickness of diffusion layer was linear to a square root of bonding time.

4. The shear strength of joints formed under 750°C were about 200 MPa. The shear strength up to 800°C was is degraded. The shear strength was not sensitive for the thickness of diffusion layer.

References

5.4 Investigation of Be/Cu Joints Via HHF Tests of Small-scale Mockups

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Beryllium-copper (Be/Cu) joints in divertor components work under cyclic heat loads. To develop reliable joints small-scale mockups are fabricated by divertor technologies and tested under the divertor conditions. One of the critical damaging factors that exist in the divertor and have to be simulated is thermocyclic heat loads in the range of 1-15 MW/m². This work presents the divertor mockups that have beryllium tiles with different dimensions (5x5 - 44x44) mm² brazed with copper alloy heat sink. The electron beam was used to braze these mockups so as to decrease the formation of brittle intermetallic layers. The description of mockups design, geometry of armour tiles and fabrication techniques are presented in the paper. The results of screening and thermocyclic tests of these mockups in the heat flux range of 2-12 MW/m² with a number of cycles ~10³ are presented. The results of metallographic analysis are also presented. The results of fabrication and testing with small-scale mockups for first wall application are also described.

1. Introduction

For the high heat flux (HHF) components of the divertor beryllium was selected as an armour material and CuCrZr alloy as a heat sink material. Beryllium-copper structures in such HHF components have to operate under the conditions of cyclic heat loads with a number of cycles of 10³-10⁴ [1]. Cyclic stresses in the joints limit the lifetime of HHF components, that is why one of the critical issue is the selection of reliable joining of beryllium with copper alloy. To investigate armour geometry and Be/Cu joints the brazing was selected as one of the joining methods and the cyclic heat loading of small-scale divertor mockups by electron beam as a testing method.

The following beryllium mockups were fabricated and tested: the mockups for divertor components with heat loads $Q = 1-5$ MW/m² (moderate loads) and $Q = 5-15$ MW/m² (high loads). The goal of mockups testing with moderate loads was to examine the reliability of beryllium armoured elements under heat cyclic conditions for the divertor liners, and the main goal of testing with high heat loading was to determine the heat flux limit for joints.

2. Mockups description

Beryllium small-scale mockups were fabricated from the following materials: plasma facing materials- beryllium TGP-56, hot isostatic pressed blocks; heat sink material (HSM)- copper alloy CuCrZr, block with cooling channel; brazing alloy- CuInSnNi amorphous brazing alloy (STEMET trade mark, AMETO).

The chemical composition (wt %) of materials is the following:

- Beryllium TGP-56: Fe-0.016; Cr-0.025; Ti-0.02; Al-0.012; Si-0.013; C-0.056; O-1.2; Be-base;
- Copper alloy CuCrZr: Cr-0.97; Zr-0.073; Cu-base.

Fig. 1. The sketch of Be mockup for moderate heat loads (top total view, bottom-cross-section (mockup #1).

The designs of mockups fabricated for tests are presented in figures 1, 2. Beryllium tiles for plasma facing components (PFC) should be different in geometry depending on the purposes. Thus, the PFC for moderate loads (1-5 MW/m²) such as the first wall, baffle, limiter and divertor liners should have large dimensions of armouring tiles [2] in view of comparatively low stresses in the armour-HSM joint and to make cheaper fabrication, but the highly loaded (5-15 MW/m²) divertor components (vertical target, dome) require that the armouing tiles should be decreased in dimensions so as to eliminate overstrains of the joints.

For the beryllium/copper joints the difference in thermal expansion coefficients is negligible, that is why, direct bonding of beryllium tiles to a flat HSM block was used,
a) The beryllium mockup with lock-type brazing (mockup #2)

b) The beryllium mockup with flat-type brazing, (10x10x5) mm³ tiles (mockup #3)

c) The beryllium mockup with flat-type brazing, (5x5x5) mm³ tiles

Fig. 2. The beryllium mockups high heat flux components (mockup #4)
but the problem is only one of brittle intermetallic layers produced during traditional furnace resistive brazing. To reduce these intermetallic layers attempts were made to realise electron-beam brazing with high heating rate. Another reason for operation of high-rate heating brazing (in the temperature range $T=500-800 \, ^\circ C$) is to preserve strength of CuCrZr copper alloy, as it was measured after such brazing the ultimate strength 360 MPa.

The beryllium mockups were brazed in the vacuum chamber by electron beam heating. This type of heating provides the heating rate in the range of 2-4 degree/sec. These mockups were brazed under the following conditions: pressure in the vacuum chamber - $10^{-4} \, \text{Pa}$; heating time from 450 °C to 800 °C - 2-3 minutes; cooling time from 800 °C to 450 °C - 8-10 minutes; exposure at 800 °C - 5-10 seconds. The typical heating-cooling diagram of electron beam brazing is presented in figure 3.

The typical diagram of brazing for small-scale mockups is shown in figure 3.

Fig. 3. The typical diagram of brazing for small-scale mockups

3. Test facility description

Thermocycling and screening tests for beryllium divertor mockups were performed on the TSEFEY electron beam test facility at the Efremov Institute. The main parameters of the facility are as follows: total power $P_{\text{max}}=80 \, \text{kW}$; electron energy $E_e=25 \, \text{keV}$; time of loading/pause $t=0.1\,\text{stationary}$; loading area $S=20x20400x400 \, \text{mm}^2$; scanning frequency $f=1000 \, \text{Hz}$; background pressure $P=10^{-4} \, \text{Pa}$; water cooling system $P=0.11.6 \, \text{MPa}$, $w=1-10 \, \text{m/s}$, $T=1090\,\text{°C}$; beryllium compatible. The facility is equipped with the following diagnostic instrumentation: heat flux meter (HFM) devices for measurement of incident and absorbed heat flux density; IR-camera for surface temperature measurement; thermocouple devices for calibrations and bulk temperature measurements; devices for measurement of cooling parameters; video camera for testing procession monitoring. For testing the mockups are installed on the special water-cooled target-table shown in figure 4. The target-table is set upon the manipulator that can move inwards and outwards the vacuum chamber trough the sluice. For correct surface temperature measurements the IR-camera is calibrated by e-beam heating of either the mockup ($T=20-300 \, ^\circ C$) or the Be-block ($T=300 \, ^\circ C$). The calibration was repeated few times for each experiment because during cyclic heating emission coefficient (e) of armour material changed.

4. Tests of divertor mockups and experimental results

Four beryllium divertor mockups were tested at the TSEFEY electron beam facility. The testing conditions are listed in table 1. The pictures of the mockups are presented in figure 5.

Mockup #1 with two $(44x44x10) \, \text{mm}^2$ tiles has survived 2000 cycles at a heat flux of $2.0 \, \text{MW/m}^2$ without any changes and damages during the test.

Mockup #2 with 20 $(10x10x7) \, \text{mm}^3$ tiles (lock-type brazing) was tested during 1000 cycles at $7.5 \, \text{MW/m}^2$ without any changes and damages, too.

Mockup #3 with 8 $(10x10x5) \, \text{mm}^3$ tiles was tested in the screening mode up to the 10 $\, \text{MW/m}^2$. The surface temperature of one of tile turned out higher than that of the rest seven tiles. This tile was assumed to have a defect in the brazing zone. The thermocycling test that followed proved this assumption. During the second cyclic shot at a heat flux of $10 \, \text{MW/m}^2$ this tile detached in a brittle manner, as detected by the IR-camera by a low cool-down rate. Figure 6 illustrates the detachment of the tile. The debonded tile was removed and the cyclic test continued during 1000 cycles. The rest seven tiles successfully survived 1000 cycles at $10 \, \text{MW/m}^2$. No changes and damages of armour material and HSM and their joint were observed. Figure 7 (top) shows the smooth beryllium surface after the test.

Mockup #4 with 32 $(5x5x5) \, \text{mm}^3$ tiles was tested at screening mode up to a heat flux of $12 \, \text{MW/m}^2$ without any visible changes and damages. The thermocyclic test was performed during 2000 cycles at a heat flux of $12 \, \text{MW/m}^2$. No damages in the Be/Cu boundary were observed. The surface of the beryllium tiles was covered by grid of micro-cracks, and large beryllium grains became visible after the test (figure 7, bottom). These changes of beryllium surface did not result in a loss of the thermal contact between beryllium and HSM.

Tests were carried out at the following cooling parameters: $W_{\text{flow rate}}=515 \, \text{m/s}$; $T_{\text{water in}}=15\,\text{°C}$, $P_{\text{in}}=0.8 \, \text{MPa}$. 

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Fig. 4. Target-table installation on the manipulating device

Fig. 5. The pictures of the mockups
deposited (PVD) beryllium tiles was fabricated. PVD Be (20x20x10) mm³ tiles consisted of five deposited layers 2 mm thick each. Beryllium was deposited on the SS-grid (SIA "Looch", Podolsk). These tiles were brazed via the (20x20x5) mm³ copper layer to the CuCrZr heat sink using CuInSnNi brazing alloy. The mockup is presented in figure 8 (top).

Each tile was tested separately in the following sequence:
1-st tile: screening test in the range Q=1-2.5 MW/m² (10 cycles per step) - debonding of the upper Be-layer (2 mm) at Q= 2.5 MW/m²;
2-nd tile: 1000 cycles at Q=1 MW/m² - OK and 50 cycles at 1.5 MW/m² - debonding of the upper Be-layer (2 mm); the layer was removed, 1.5 MW/m²-650 cycles, 2.5 MW/m²-500 cycles, 2.5 MW/m²-500 cycles, 3 MW/m²-10 cycles, 4 MW/m²-10 cycles-OK; 5MW/m² - two cycles - slight nonuniform surface temperature was detected and testing was interrupted;
1-st rile: the debonded layer was removed, the similar test (as for second tile) up to 5 MW/m² was continued with the same results.

The performed test with the PVD beryllium mockup has demonstrated the possibility to use this technique for beryllium armoured components that work under heat flux in the range of 2-4 MW/m².

5. Fabrication and testing of small-scale mockups for first wall application

5.1. Be-CuCrZr mockup with PVD beryllium

To develop the repairing technique for beryllium armoured components the small-scale mockup with two physical-vacuum-
Fig. 5. The pictures of beryllium divertor mockups.

Mockup with two tiles (44x44x10) mm²
#1

Mockup with 20 tiles (10x10x7) mm³, lock-type brazing
#2

Mockup with 8 tiles (10x10x5) mm³
#3

Mockup with 32 tiles (5x5x5) m³
#4
The surface view of Beryllium tiles (10x10) mm² after 1000 cycles at 10 MW/m² (no damages)

The surface view of Beryllium tiles (5x5) mm² after 2000 cycles at 12 MW/m² (the grid of micro-cracks)

Fig. 7. The surface of Beryllium after the thermocyclic tests.
Fig. 8. Small-scale beryllium mockups for First Wall application
Fig. 9. The transient Be/Cu brazed zones in two brazing techniques and after the test: top picture-brazing in resistive furnace, middle-brazing with e-beam heating, bottom-e-beam brazing after thermocyclic tests
Table 1.
The main conditions of tests with small-scale beryllium divertor mockups.

<table>
<thead>
<tr>
<th>Mockup type/ #</th>
<th>Test type</th>
<th>( Q_{\text{des}}, \text{MW/m}^2 )</th>
<th>( t_{\text{load/ pause per step}} )</th>
<th>( N_{\text{cycles}} )</th>
<th>( T_{\text{joints (measured)}}, \text{C} )</th>
<th>( T_{\text{surf (measured)}}, \text{C} )</th>
<th>Result</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mockup with two tiles (44x44x10 mm(^2)) #1</td>
<td>screening</td>
<td>0.5-2.0; 0.5 MW/m(^2) steps</td>
<td>15/15</td>
<td>10</td>
<td>120 (max)</td>
<td>280 (max)</td>
<td>OK</td>
</tr>
<tr>
<td></td>
<td>cyclic</td>
<td>2.0</td>
<td>15/15</td>
<td>2000</td>
<td>120</td>
<td>280</td>
<td>OK</td>
</tr>
<tr>
<td>Mockup with 20 tiles (10x10x7 mm(^3)), lock-type brazing #2</td>
<td>screening</td>
<td>1-7.5; 1 MW/m(^2) steps</td>
<td>15/15</td>
<td>10</td>
<td>235 (max)</td>
<td>750 (max)</td>
<td>OK</td>
</tr>
<tr>
<td></td>
<td>cyclic</td>
<td>7.5</td>
<td>10/10</td>
<td>1000</td>
<td>235</td>
<td>750</td>
<td>OK</td>
</tr>
<tr>
<td>Mockup with 8 tiles (10x10x5 mm(^3)) #3</td>
<td>screening</td>
<td>5.0-10; 1 MW/m(^2) steps</td>
<td>15/10</td>
<td>10</td>
<td>190 (max)</td>
<td>710 (max)</td>
<td>Overheating one of the tile, rest 7 tiles - OK</td>
</tr>
<tr>
<td></td>
<td>cyclic</td>
<td>10</td>
<td>15/10</td>
<td>1000</td>
<td>190</td>
<td>710</td>
<td>OK</td>
</tr>
<tr>
<td>Mockup with 32 tiles (5x5x5 mm(^3)) #4</td>
<td>screening</td>
<td>5.0-12; 1 MW/m(^2) steps</td>
<td>10/10</td>
<td>10</td>
<td>210 (max)</td>
<td>900 (max)</td>
<td>OK (the grid of micro-cracks on Be surface)</td>
</tr>
<tr>
<td></td>
<td>cyclic</td>
<td>12</td>
<td>10/10</td>
<td>2000</td>
<td>210</td>
<td>900</td>
<td>OK</td>
</tr>
</tbody>
</table>

5.2. Be-SS mockup

A small-scale mockup with beryllium brazed to stainless steel (SS) was fabricated and tested to check for First Wall application. The mockup is presented in figure 8 (bottom). Two beryllium (20x20x10) mm\(^3\) tiles were brazed to the (20x20x2) mm\(^3\) SS thin plate and to the CuCrZr block by CuInSnNi brazing alloy. The tiles were tested in the following sequence: 1-st tile- screening test in the range of 1-2.5 MW/m\(^2\), at a heat flux of 2.5 MW/m\(^2\) the tile detached in the Be-SS zone (brittle debonding); 2-nd tile- thermocycling test at \( Q=1 \text{ MW/m}^2 \), \( N=1000 - \text{OK} \), then screening and debonding at 2.5 MW/m\(^2\).

6. Metallographic analysis of beryllium-copper joints

The electron beam brazing with high heating rate was used for Be-CuCrZr joints. Brazing of Be/Cu joints in the resistive vacuum furnace with several hours of heating/cooling-down leads to formation of wide brittle intermetallic zones [3]. Figure 9 presents the Be/Cu zones after brazing in the resistive furnace and fast electron beam heating. The width of the brazing zone for the joint brazed in the resistive furnace is 420 m and only 20-30 m for the joint brazed by fast heating. Such difference is attributable to a substantial difference in heating/cooling time.

A thin (10 m) layer with pores in Be was detected near the Be/brazing alloy boundary for the first joint, but no pores for the second joint. It can be explained by a high rate of Be diffusion migration in brazing alloy resulting from an elevated temperature and long-term brazing. This assumption was verified by mechanical tests. Low shear strength for the first joint (22 MPa) with cracking through Be near the brazing boundary and higher strength (137 MPa) for the second joint were measured. The second joint was tested during \( 3 \times 10^3 \) cycles at a thermal load of \( 1.5 \text{ MW/m}^2 \). The joints brazed in resistive furnace have demonstrated poor reliability during the cyclic tests [3]. The Be/Cu joint after the test
had width of 40 m but no pores or cracks were found (figure 9).

The performed metallographic analysis of Be/Cu joints proved that brazing with high heating rate is more preferable for divertor components with beryllium armour.

7. Conclusions

The fabrication and testing of mockups with beryllium armour and analysis of the joints have demonstrated that the brazing bonding techniques with CulnSnNi brazing alloy can be used for the ITER divertor and first wall elements: brazing for Be/CuCrZr, Be/SS and Cu/CuCrZr joints. The beryllium mockup for divertor application with large armour tiles (44x44) mm$^2$ has demonstrated the cyclic reliability at 2 MW/m$^2$, with small Be tiles (5x5) mm$^2$ - the cyclic reliability at 12 MW/m$^2$. First wall mockups with PVD beryllium and Be/SS tiles have demonstrated their reliability in the heat flux range of 1-4 MW/m$^2$.

The described designs and fabrication processes of mockups can be used when designing and manufacturing large- and full-scale ITER divertor and first wall elements.

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5.5 Solid State Bonding of Beryllium-Copper for an ITER First Wall Application

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ABSTRACT
Several different joint assemblies were evaluated in support of a manufacturing technology for diffusion bonding a beryllium armor tile to a copper alloy heat sink for fusion reactor applications. Because beryllium reacts with all but a few elements to form intermetallic compounds, this study considered several different surface treatments as a means of both inhibiting these reactions and promoting a good diffusion bond between the two substrates. All diffusion bonded assemblies used aluminum or an aluminum-beryllium composite (AlBeMet-150) as the interfacial material in contact with beryllium. In most cases, explosive bonding was utilized as a technique for joining the copper alloy heat sink to an aluminum or AlBeMet-150 substrate, which was subsequently diffusion bonded to an aluminum coated beryllium tile. In this approach, a 250 μm thick titanium foil was used as a diffusion barrier between the copper and aluminum to prevent the formation of Cu-Al intermetallic phases. In all cases, a hot isostatic pressing (HIP) furnace was used in conjunction with canned assemblies in order to minimize oxidation and apply sufficient pressure on the assembly for excellent metal-to-metal contact and subsequent bonding. Several different processing schedules were evaluated during the course of this study; bonded assemblies were produced that failed outside the bond area indicating a 100% joint efficiency.

1. INTRODUCTION
The method of affecting a bond between two dissimilar or similar metals without the presence of a liquid phase is called diffusion bonding. For diffusion bonding to occur, the bare metal surfaces must come in contact at a distance close to that of the crystal lattice constant. In order for that to happen, the surfaces must be free of contamination and oxides. When these conditions have been met, temperature and pressure accomplish the desired bond. Reactive metals such as beryllium and aluminum form tenacious oxide films which are especially difficult to break up. However, these metal oxide films must be broken in order to obtain intimate contact and subsequent bonding between the metal surfaces. Much of the effort in developing a diffusion bonding process concerns minimizing or eliminating the effects of these oxide layers by adjusting the bonding parameters.

The application of pressure during the bonding process creates a shearing action at the bond interfaces which breaks up the oxide film and exposes bare metal surfaces. Also, through this shearing action, the diffusion process is accelerated through the generation of line and point defects. These bare metal-metal contact points mark the genesis of the diffusion bonding process. Relatively rough surfaces and high bonding pressures will enhance the diffusion process. The application of temperature serves to provide the thermal energy necessary for diffusion. As the bonding temperature is increased, those diffusion processes that create the bond (bulk diffusion, grain boundary diffusion, volume diffusion) become more active, resulting in void elimination and atomic exchange across the interface.

The application of a thin coating of metal which is compatible with the substrates provides a concentration gradient which further enhances diffusion. The other benefit gained by this coating is the replacement of the aluminum and beryllium oxides with a less tenacious oxide. Finally, a judicious material selection has to be made if the materials are to be compatible with both the beryllium armor tile and the copper heat sink material. That is, they do not form intermetallic compounds with either substrate. Those elements that do not form intermetallic compounds with beryllium include aluminum, germanium, silicon, and silver. Silver was precluded because of the transmutation products produced in the high neutron ITER environment [1]. The remaining possibilities were evaluated either in the brazing study conducted in joining these two materials [2] or in this study on diffusion bonding. Aluminum seemed the likely candidate for a compliant layer selection. It possesses good ductility at the expected interface service temperature (180°C) and excellent thermal conductivity but limited strength. However, the beryllium tiles would be sitting on a compliant layer which could not sustain residual stresses (derived from the thermal expansion differences between beryllium and copper) much greater than the flow stress of the aluminum (<100 MPa). Further, the compliant layer material would undergo annealing at the service temperatures which would further limit the retention of residual stresses. The material selection premise was to promote ductility over strength for this bond configuration.

In this study, the copper heat sink material (Hycon-3) was clad with a 1 mm layer of either 1100-Al alloy or AlBeMet-150 (50wt%Be-50wt%Al) using a thin titanium diffusion barrier (0.25 mm) as the interfacial material.
is isolating the copper from the aluminum. This layered composite was produced using an explosive bonding technique. Titanium as well as tantalum have been used in industry to separate these two materials. The chemical stability of these composite layers has been tested to the melting point of the aluminum [3]. Beryllium armor tiles were coated with a thin layer of aluminum. Thus, the diffusion bonding problem was reduced to bonding aluminum to itself if the material selections had been chosen correctly. The surfaces of the aluminum were coated with a thin coating of copper or silicon to eliminate the aluminum oxide and enhance the diffusion flow by producing a concentration gradient. If the titanium diffusion barrier could eliminate the reactions between beryllium-copper and copper-aluminum, the process should work. Much of the work in selecting materials for this study was made easier by an earlier study where brazing was used as the joining process [4].

2. EXPERIMENTAL PROCEDURE

The beryllium used in this study was Brush-Wellman grade S-65C, a product made by the vacuum hot pressing of impact ground powder. The precipitation hardenable copper alloys utilized were CuCrZr (Elbrodur G, Kablemetal) and CuNiBe (Hycon-3, Brush-Wellman). Both of these alloys were age hardened in the same temperature range [460-480°C] and soften quite rapidly at temperature above 500°C. The beryllium and copper alloy components were coated, with aluminum or aluminum alloys, by either explosive bonding, plasma spray (PS) or ion sputtering (PVD) techniques. Chemical composition of the various test materials are shown in Table 1.

Table 1

<table>
<thead>
<tr>
<th>Alloy Designation</th>
<th>Composition (wt. %)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CuCrZr (Elbrodur G)</td>
<td>Cu - 0.65%Cr - 0.10%Zr</td>
</tr>
<tr>
<td>CuBeNi (Hycon-3)</td>
<td>Cu - 1.4-2.2%Ni - 0.2-0.6%Be</td>
</tr>
<tr>
<td>S-65C</td>
<td>Be - &lt;1.0%Be - Fe/Al impurities</td>
</tr>
<tr>
<td>AlBeMet-150</td>
<td>50%Be-50%Al</td>
</tr>
<tr>
<td>1100-Al</td>
<td>99% Al (minimum)</td>
</tr>
</tbody>
</table>

Several different bonding sequences were evaluated in this study. All specimens consisted of essentially the same cylindrical geometry, shown schematically in Figure 1. A beryllium disk (S-65C) was sandwiched between two cylinders of coated copper. All of the components were 48 mm in diameter. Each specimen was assembled in a cylindrical stainless steel can and vacuum baked for 4 h at 100°C to remove the majority of surface moisture. A cap was then electron beam welded to the end of the can completing the encapsulation process and ensuring an inert environment during subsequent processing. The canned assemblies were then hot isostatically pressed (HIP).

Table 2 lists the specimen identification, materials, and HIP processing parameters for each specimen. The 1100-Al and AlBeMet-150 layers were explosively bonded to the copper alloys using an intermediate layer of 250 μm thick titanium designed to act as a diffusion barrier. Note that silicon and copper films were used to provide a concentration gradient and eliminate the aluminum oxide film at the bond surface. After first sputter etching the bonding surface to remove native oxide films, these thin coatings were applied by ion sputtering. Where these thin coatings were not employed, bond surfaces were: (1) chemically etched in an aqueous solution containing 45%HNO₃ and 10% HF, (2) rinsed in de-ionized water and (3) forced air dried immediately prior to assembly.

After HIP processing, the candidate assemblies were de-canned from the stainless steel and specimens were removed for metallographic examination and mechanical testing using wire electron discharge machined (WEDM). The specimen orientation is shown in Figure 1. The characteristics of the bond interface were examined by both optical and electron microscopy. The fracture strength was determined using a transverse tensile test specimen geometry with a reduced cross-section. These specimens were inexpensive to both machine and test, although they did not duplicate the in-service loading stresses found in real components. However, this test was used to compare various bonding sequences and not to determine absolute bonding strengths. Tests were conducted primarily at room temperature; selected specimens were also evaluated at 300°C.

3. TEST RESULTS

The room temperature tensile test results are shown in Table 3. Despite etching to remove surface oxides immediately prior to assembly, DB-1 and DB-2 exhibited problems associated with the presence of oxides at the bond interface. These oxides were visible, using optical metallography, as a thin black line at the interface. Tensile bar fracture surfaces appear as mixed mode consisting of dimple ductile rupture and regions of poor bonding where the surface had an oxide film.

DB-3 and DB-5 exhibited problems associated with the presence of a silicon-rich film at the beryllium interface. There is very little solubility of silicon in either beryllium or aluminum at room temperature. For reasons that are not completely understood, silicon in this system tends to segregate at the beryllium interface, particularly if the aluminum coating thickness (on beryllium) is kept low. In DB-3, this film was nearly continuous and failure occurred at a very low stress while machining. In DB-5, the silicon-rich film was much less prominent and consisted of a discontinuous phase. DB-4 exhibited no detectable bond line which indicates that sufficient diffusion had occurred to eliminate the presence of oxides and porosity at the bond interface. The copper promotes diffusion by providing a concentration gradient and eliminating the presence of an aluminum oxide. However, it is not of sufficient concentration to promote the formation of beryllium intermetallics at the beryllium surface. The fracture stress is essentially that of the tensile strength of pure aluminum.
Table 2
Identification, coatings and HIP parameters for diffusion bonded specimens

<table>
<thead>
<tr>
<th>Specimen ID</th>
<th>Be Coating</th>
<th>Cu Coating*</th>
<th>HIP Parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td>DB-1</td>
<td>none</td>
<td>AlBeMet-150</td>
<td>600°C / 1 h / 105 MPa</td>
</tr>
<tr>
<td>DB-2</td>
<td>25 μm PVD Al</td>
<td>AlBeMet-150</td>
<td>625°C / 1 h / 105 MPa</td>
</tr>
<tr>
<td>DB-3</td>
<td>25 μm PVD Al + 1 μm PVD Si</td>
<td>AlBeMet-150</td>
<td>625°C / 1 h / 105 MPa</td>
</tr>
<tr>
<td>DB-4</td>
<td>300 μm PS Al + 100 nm PVD Cu</td>
<td>1100-Al</td>
<td>625°C / 1 h / 105 MPa</td>
</tr>
<tr>
<td>DB-5</td>
<td>25 μm PVD Al + 1 μm PVD Si</td>
<td>1100-Al</td>
<td>625°C / 1 h / 105 MPa</td>
</tr>
<tr>
<td>DB-6</td>
<td>100 μm PVD Al + 300 nm PVD Cu</td>
<td>AlBeMet-150</td>
<td>625°C / 1 h / 140 MPa</td>
</tr>
</tbody>
</table>

*The explosive bonding of copper/titanium/aluminum plates was performed at Northwest Technical Industries, Inc. in Sequim WA, USA.

Necking was extensive in the bond regions which indicates excellent defect tolerance. The fracture surface exhibited dimple ductile rupture.

Based upon the results obtained for DB-4, another specimen (DB-6) was fabricated using PVD techniques to coat both the beryllium and AlBeMet-150 bonding surfaces. This specimen exhibited joint microstructures similar to DB-4 but fractured at a higher stress, presumably due to the improved strength of AlBeMet-150 compared to 1100-Al.

4. DISCUSSION

Etching with a dilute solution of HF/HNO₃ to remove oxides prior to bonding as was the case for the first two bonding configurations was not practical on a full-scale unit and at best was very difficult with lab-scale units. The etchant reacted very quickly with the beryllium. Cooling the etchant in an ice-bath to reduce the reactivity to a controllable rate resulted in a 10s immersion before a visible film began to form on the beryllium surface. Aluminum was etched with a NaOH solution. The transfer time to a vacuum chamber was critical if re-oxidation was to be kept at a minimum. Earlier studies by investigators had attempted to bond beryllium to Lockalloy (62%Be-38%Al) using a similar process [5]. They found that the beryllium removal rates using the HF/HNO₃ were as high as 1mm/m. In that study, an etching technique was developed in conjunction with a subsequent ion sputtered copper or silver coating on all surfaces to eliminate the oxidation problems and improve the likelihood of a good bond. In the current study, etching alone was insufficient surface preparation to obtain consistently good bonding (DB-1, DB-2), leading to the use of sputtering to both remove surface oxides and provide diffusion enhancers. Germanium was tried in the brazing experiments and excluded from this series of tests. The germanium was found to segregate to the beryllium interface and significantly reduce the bond strength. Silicon, if used in small volumes appeared to work, although, silicon-rich precipitates were observed at the beryllium interface. Copper worked the best and resulted in good bond strengths with no interfacial precipitates when the copper film thickness was kept low and aluminum volumes were sufficient to dilute the copper.

5. CONCLUSIONS

1. Diffusion bonding of aluminum surfaces to themselves is difficult because of the oxides formed by this very reactive material. Attempts to remove the oxide by using a HF/HNO₃ dilute solution (DB-1, DB-2) just prior to canning were only moderately successful.

2. Diffusion bonding of aluminum surfaces using a silicon film to eliminate aluminum oxide were unsuccessful (DB-3, DB-5). The silicon migrates to the beryllium interface and segregates as a silicon-rich phase. The dilution of this segregate can be improved by using thicker aluminum coatings (DB-5).

3. The use of a copper film on aluminum surfaces to eliminate aluminum oxide and promote diffusion by
Table 3
Metallographic and Tensile Test Results

<table>
<thead>
<tr>
<th>Specimen ID</th>
<th>Metallographic and SEM results and remarks</th>
<th>Fracture Stress at Room Temperature</th>
</tr>
</thead>
<tbody>
<tr>
<td>DB-1</td>
<td>oxide film still visible at the bond interface</td>
<td>86 MPa</td>
</tr>
<tr>
<td>DB-2</td>
<td>oxide film still visible at the bond interface</td>
<td>99 MPa</td>
</tr>
<tr>
<td>DB-3</td>
<td>semi-continuous, silicon-rich phase at the beryllium interface</td>
<td>60 MPa</td>
</tr>
<tr>
<td>DB-4</td>
<td>no apparent oxides, bond line was not visible</td>
<td>19 MPa</td>
</tr>
<tr>
<td>DB-5</td>
<td>semi-continuous, silicon-rich phase at the beryllium interface</td>
<td>113 MPa</td>
</tr>
<tr>
<td>DB-6</td>
<td>no apparent oxides, bond line was not visible</td>
<td>116 MPa</td>
</tr>
</tbody>
</table>

presenting a concentration gradient was successful. Bond fracture strengths at room temperature were at 100% efficiency based on the tensile strength of pure aluminum (105 MPa). The fracture morphology was dimple rupture. Extensive necking occurred in the bond region which indicates good defect tolerance.

4. The diffusion bonding process developed using the aluminum-beryllium compliant layer can produce beryllium-copper bonds that will survive at the 5-10 MW/m² level without failure.

REFERENCES


ACKNOWLEDGEMENTS

The authors would like to thank Mssrs S. Sastri (Surmet Corp), W. Bonivert (SNL/CA) and D. Butler (Northwest Technical Industries, Inc) for providing the coated beryllium and copper samples and Mssrs. K. Slattery and R. D. Watson for their valuable contributions in consulting, manufacturing, and testing throughout this study. A special thanks to C. Rood, A. Gardia, and N.Y.C. Yang for their assistance in the evaluation of the surfaces using optical and electron microscopy. Finally, we would like to acknowledge the work done by S. Goods and M. Tootle in the set-up, testing, and evaluation of the tensile specimens.

This work was supported by the United States Department of Energy under contract DE-AC04-94AL85000.
Figure 1. Bond assembly configuration showing orientation of specimen for transverse tensile testing.

Figure 2. Photograph showing tensile bars taken from the diffusion bonded sample (BD-5). Note the extensive necking in the sample on the left.

Figure 3. Micrograph showing the beryllium-aluminum and aluminum-AlBeMet-150 diffusion bond in specimen DB-6.

Figure 4. Micrograph showing the AlBeMet-150 to copper alloy explosive bond. Note regions of the AlBeMet-150 have intruded into the titanium.
Armor and Heat Sink Diagram:

16±0.25mm

66.46±0.125mm

outside surface of round portion - 32RHR roughness

10mm thick tiles

5mm thick tiles

14±0.08mm 59.2±0.5mm 43.51±0.125mm

10±0.25mm hole bored through center

Figure 5.
EBTS Test Specimen geometry
5.6 Bonding Be to Cu Alloys Using a Proprietary Brush Wellman Process and New Results for Bonding Be to AlBeMet

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A novel process has been investigated for bonding of beryllium to copper alloys. The process is compatible with current manufacturing capabilities and can be readily scaled up from laboratory to production scale. The effect of process variations on the bond are shown. Also shown are several new approaches for direct bonding of beryllium to an AlBeMet® interlayer. Bond strengths of Be/AlBeMet® 162/ Cu alloy couples are presented.

1. INTRODUCTION

Bonding beryllium to copper without the use of silver is important for the fabrication of several components in the ITER tokamak reactor. Preliminary results for a proprietary bonding process developed by Brush Wellman were presented previously [1]. Bond strengths were acceptable for ITER applications. Additional work showing the effect of several process variables on this bonding process are presented here. It was convenient in initial experiments to make finished bond interlayer thicknesses on the order of 6.4 mm. An experiment was conducted to obtain bond strength for a bond interlayer with a nominal 1.6 mm bond thickness.

A variety of bonding processes have been used to bond Hycon 3 HP™ (UNS C17510) to beryllium, but all compromise the mechanical properties of Hycon. Hycon is a high strength, high conductivity beryllium-copper alloy favored for its high strength compared to Glidcop Al-15 and CuCrZr. A second set of experiments was conducted to investigate variations in the thermal cycle of the Be to Hycon 3 HP bonding process which were designed to preserve the high strength of Hycon 3 HP. Slattery [2] has investigated Hycon to Hycon bonding using the Hycon solution heat treatment and aging cycle as the bonding process thermal cycle. This approach was used here also.

Bonding of beryllium to AlBeMet® was also investigated. Odegard and Cadden [3] have developed a multiple interlayer approach to bonding beryllium to copper. One of their interlayer components is AlBeMet® 150, an aluminum- 50 wt% beryllium alloy made by inert gas atomization and conventional powder metallurgy processing. The ITER community has been interested in the AlBeMet® family of alloys for its thixotropic properties. However, bonding AlBeMet® directly to beryllium has not been successful to this point. Several novel and successful approaches are described here, and bond strength data is presented.

2. BACKGROUND

The bonding of beryllium to an aluminum-beryllium alloy is governed by the tenacious oxide on both materials. Silver is known to promote bonding between beryllium surfaces [4], but is not permitted in ITER plasma facing components for neutron activation reasons. If the bonding problem is looked at primarily as penetration or breakdown of the beryllium oxide and aluminum oxide surface layers followed by diffusion bonding, the problem can be thought of as similar to sintering. Extensive work on activated sintering of beryllium powder in the 1960s [5-8], showed that the two most active sintering aids are elemental silicon and elemental germanium. The upper concentration limit to usefulness of activated sintering aids is at a low absolute concentration. Butcher is recommended 500 ppm [7]. Previous ITER related bonding research tended to use much greater quantities than 500 ppm and those exploratory tests failed. Note that high sintering temperatures (1100-1200°C) were used in the 1960s experiments but that ITER related bonding research is limited to much lower temperatures (500-800°C).

Butcher and Lowe [8] bonded two heavily oxidized beryllium disks together by sprinkling elemental silicon grains at a concentration of 0.005 g Si/cm2. Although this is more than 500 ppm based on the oxide thicknesses of two interfaces, it is an easily measurable quantity for small laboratory samples. Putting the sintering aid at the bonding surfaces eliminates the uncertainty of sintering aid distribution in a mixed powder. This approach was used in the present work.
Several silicon containing materials were investigated in the present work for use in bonding Be to AlBeMet. Optimum activated sintering or activated bonding behavior should be provided by the most uniform distribution of sintering/bonding aid on the bonding interface. Silicon powder can only be distributed in a macroscopically uniform manner. A liquid carrier would provide more uniform distribution. To that end, polydimethylsiloxane and sodium silicate were investigated as bonding agents.

Germanium powder was used for the present bonding experiments.

3. PROCEDURE

3.1. Proprietary Process for Bonding S-65C Grade Be to Cu: Interlayer Thickness

The details of this process are proprietary. No elements are used which would produce unsafe levels of radiation hazard or daughter products after neutron irradiation.

The first experiment (Trial #1) investigated the effect of decreasing bond interlayer thickness on bond strength. S-65C grade beryllium was bonded to Hycon 3 HP™, a high strength, high conductivity beryllium-copper alloy. The bond was manufactured to have a nominal 1.6 mm bond thickness. Straight sided and reduced section tensile samples were machined from the bonded material using electrical discharge machining (EDM). Figure 1 shows the geometry of the reduced section tensile specimen. The straight sided specimen had the same dimensions except for the reduced section. Standard tensile testing was conducted at room temperature and 400°C.

3.2. Proprietary Process for Bonding S-65C Grade Be to Cu: Thermal Cycle to Maintain Hycon Properties During Bonding

The next experiments investigated variations in the thermal cycle of the Be to Hycon 3 HP bonding process which were designed to preserve the high strength of Hycon 3 HP. The second experiment (Trial #2) explored the effect of the normal bonding cycle followed by a heat treatment to recover Hycon's mechanical properties. The optimum bonding cycle for the proprietary bonding process was used. The subsequent heat treatment is shown in Table 1. It consists of solutionizing (step A) and aging the Hycon (step B). In the third experiment (Trial #3), the proprietary bonding process was performed using the conditions in Step A of the heat treatment. i.e. a 927°C bonding temperature was used. A post-bonding heat treatment was done using the conditions of Step B. These tests are summarized in Table 2.

<table>
<thead>
<tr>
<th>Step</th>
<th>Soak Temp. (°C)</th>
<th>Soak Time (hr)</th>
<th>Cooling</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>927</td>
<td>1</td>
<td>Quench to RT</td>
</tr>
<tr>
<td>B</td>
<td>510</td>
<td>3</td>
<td>Cool in Ambient Temp. Gas</td>
</tr>
</tbody>
</table>

Table 2 Experiments to bond S-65C to Hycon 3 HP using proprietary process and maintain Hycon mechanical properties.

<table>
<thead>
<tr>
<th>Trial</th>
<th>Bond Temp. (°C)</th>
<th>Post Bond Heat Treatment</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>Proprietary</td>
<td>927°C for 1 hr and quench to RT and 510°C for 3 hr and Cool in Ambient Temp. Gas</td>
</tr>
<tr>
<td>3</td>
<td>927°C for 1 hr and quench to RT</td>
<td>510°C for 3 hr and Cool in Ambient Temp. Gas</td>
</tr>
</tbody>
</table>

3.3 Bonding AlBeMet to Be

Cylinders of S-65C beryllium were joined to cylinders of copper alloy (Hycon or C11000) using an AlBeMet® 162 cold pressed powder interlayer. AlBeMet® 162 was used as a matter of convenience instead of AlBeMet® 150; AlBeMet® 150 is expected to behave in a similar manner. One of four bonding aids was placed at the Be/AlBeMet® 162 interface: elemental silicon powder, polydimethylsiloxane (PDMS), sodium silicate, or elemental germanium powder. Silicon powder was introduced to provide 0.005 g Si/cm². The silicon powder was mixed with acetone and the resulting slurry was distributed...
uniformly over the AlBeMet® surface closest to the beryllium by pipette. The stack of cylinders were assembled in a HIP can as shown in Figures 2, degassed, and HIP'd at one of the temperatures shown in Table 3. A HIP pressure of 172.3 MPa was used.

Table 3 Bonding Conditions used for Bonding of Be to Cu using an AlBeMet 162 interlayer and a bonding aid at the Be/AlBeMet interface (Be/Bonding Aid/AlBeMet 162/Cu Alloy)

<table>
<thead>
<tr>
<th>Trial No.</th>
<th>Temp. (C)</th>
<th>Time (hr)</th>
<th>Bonding Aid</th>
<th>Cu Alloy</th>
</tr>
</thead>
<tbody>
<tr>
<td>4</td>
<td>570</td>
<td>1</td>
<td>Si</td>
<td>Hycon</td>
</tr>
<tr>
<td>5</td>
<td>700</td>
<td>1</td>
<td>Si</td>
<td>Hycon</td>
</tr>
<tr>
<td>6</td>
<td>570</td>
<td>1</td>
<td>Sodium Silicate</td>
<td>C11000</td>
</tr>
<tr>
<td>7</td>
<td>570</td>
<td>1</td>
<td>PDMS</td>
<td>C11000</td>
</tr>
<tr>
<td>8</td>
<td>500</td>
<td>1</td>
<td>Ge</td>
<td>C11000</td>
</tr>
<tr>
<td>9</td>
<td>700</td>
<td>1</td>
<td>Ge</td>
<td>C11000</td>
</tr>
</tbody>
</table>

The amount of polydimethylsiloxane (PDMS) used (0.0132 g/cm²) was calculated to have the equivalent amount of silicon as the powder used above. The PDMS (which is a liquid) was uniformly distributed over a cold pressed AlBeMet® disc in a manner similar to the silicon powder slurry. A HIP can was assembled and processed as described above. A similar procedure was used for sodium silicate, which is also a liquid; 0.04 g sodium silicate/cm² was used.

The amount of germanium powder used was calculated to be the molar equivalent of the silicon powder level. This was calculated to be 0.013 g Ge/cm². The powder was distributed on the bonding interface as was done for the silicon powder. All processing conditions except HIP temperature were identical to the silicon powder experiments. Tensile testing was conducted as described in section 3.1.

4.0 RESULTS AND DISCUSSION

4.1. Proprietary Process for Bonding S-65C Grade Be to Cu: Interlayer Thickness

The results of reducing the bond interlayer thickness are shown in Tables 4 and 5. Table 4 shows the results of testing on straight sided tensile specimens. Although these specimens are not ideal for tensile testing, they can be used in a semi-quantitative way to quickly rank bonding processes [1]. This data

Table 4 Tensile strength of bond for S-65C Be bonded to Hycon 3 HP (Cu-Be-Ni). Straight sided tensile specimens.

<table>
<thead>
<tr>
<th>Interlayer Thickness (mm)</th>
<th>Room Temp. Tensile Strength of Bond (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.4</td>
<td>117&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>104&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td>1.6</td>
<td>144&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>110&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>122&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>150&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
</tbody>
</table>

<sup>a</sup> EDM used to make entire specimen
<sup>b</sup> Ground to final thickness

Table 5 Tensile strength of bond for S-65C Be bonded to Hycon 3 HP (Cu-Be-Ni). Reduced section tensile specimens.

<table>
<thead>
<tr>
<th>Bond Thickness (mm)</th>
<th>Room Temp. Tensile Strength of Bond (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.4</td>
<td>161.4</td>
</tr>
<tr>
<td></td>
<td>148.8</td>
</tr>
<tr>
<td>1.6</td>
<td>71.7</td>
</tr>
<tr>
<td></td>
<td>31.7</td>
</tr>
<tr>
<td></td>
<td>80.9</td>
</tr>
<tr>
<td></td>
<td>94.3</td>
</tr>
<tr>
<td></td>
<td>133</td>
</tr>
<tr>
<td></td>
<td>143</td>
</tr>
</tbody>
</table>

Table 6 Tensile Failure Location for S-65C Bonded to Hycon 3 HP (Cu-Be-Ni). Reduced section tensile specimens.

<table>
<thead>
<tr>
<th>Bond Thickness (mm)</th>
<th>Tensile Failure Location</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.4</td>
<td>Broke in Bond</td>
</tr>
<tr>
<td>1.6</td>
<td>Be/Bond Interface</td>
</tr>
</tbody>
</table>

indicates that although there is more scatter in the 1.6 mm bond interlayer thickness data, the bond strength is in the neighborhood of the strength of the 6.4 mm
bond interlayer. This data also compares data from samples produced completely EDM and samples produced by EDM followed by grinding to final thickness. There appears to be some slight difference in the results, but the values are comparable on a semi-quantitative basis. The 1.6 mm bond interlayer appears to be robust enough to tolerate machining processes which are less gentle than EDM.

Table 5 shows the bond strength data obtained using conventional reduced section tensile samples. The 1.6 mm bond interlayer samples show a very low room temperature strength. A defect in the bond is suspected particularly because the bond layer process could not be controlled as well at this thickness. One indication of this is the location of failure in the tensile sample. As shown in Table 6, the location of failure for the 1.6 mm bond interlayer room temperature sample was at the Be/bond interface instead of in the bond layer as was seen for the 6.4 mm bond interlayer tensile sample. For this process, failure in the bond layer is an indication of a robust Be/interlayer bond. The 400°C results for the 1.6 mm bond interlayer are quite high compared to the 6.4 mm bond interlayer data. They will have to be confirmed. As shown in Table 6, the location of failure for the 1.6 mm bond interlayer was partially in the bond which is a an indication of good overall bond strength in this process.

At this point it can be concluded that making a bond thinner than 6.4 mm with the proprietary process is possible, but that some minor process development work is required to reduce the variability of the thinner bond. Preliminary indications from the straight sided tensile data are that the Be/interlayer bond is robust and should have similar bond strength to the 6.4 mm bond interlayer.

4.2. Proprietary Process for Bonding S-65C Grade Be to Cu: Thermal Cycle to Maintain Hycon Properties During Bonding

The results of testing designed to maintain Hycon properties while providing a good bond between S-65C and Hycon are shown in Table 7. Neither of the procedures provided a good bond. The bonding layer in both trials had large cracks.

4.3 Bonding AlBeMet® to Be

The results of the Be/AlBeMet bonding tests are shown in Table 8. The Be/Si Powder/AlBeMet 162/Cu Alloy bonded structure made by hot isostatic pressing (HIPing) at 570°C (Trial #4) produced good strength at room temperature and 200°C, and low strength at 400°C. However, the full significance of this result cannot be understood until the location of the failure is known. As shown in Table 9 and Figure 3, the failure occurred at the AlBeMet® /Cu interface. In all previous attempts to bond beryllium to AlBeMet, failure has occurred at the Be/AlBeMet interface. These results clearly show that the strength of the Be/AlBeMet bond for this process exceeds 125 MPa at room temperature, 112 MPa at 200°C and 49 MPa at 400°C.

Table 7 Results of bonding tests designed to maintain Hycon mechanical properties.

<table>
<thead>
<tr>
<th>Trial</th>
<th>Bond Temp. (°C)</th>
<th>Post Bond Heat Treatment</th>
<th>Result</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>Prop.</td>
<td>927°C for 1 hr and quench to RT, and 510°C for 3 hr and Cool in Ambient Temp. Gas</td>
<td>Large cracks in bond interlayer</td>
</tr>
<tr>
<td>3</td>
<td>927°C for 1 hr and quench to RT</td>
<td>510°C for 3 hr and Cool in Ambient Temp. Gas</td>
<td>Large cracks in bond interlayer</td>
</tr>
</tbody>
</table>

Table 8 Results for Bonding AlBeMet® to S-65C beryllium using various bonding aids.

<table>
<thead>
<tr>
<th>Trial No.</th>
<th>Temp (°C)</th>
<th>Bond Aid</th>
<th>Bond Strength (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>RT</td>
</tr>
<tr>
<td>4</td>
<td>570</td>
<td>Si</td>
<td>115</td>
</tr>
<tr>
<td>5</td>
<td>700</td>
<td>Si</td>
<td>125</td>
</tr>
<tr>
<td>6</td>
<td>570</td>
<td>Sodium Silicate</td>
<td>Failed in Machining</td>
</tr>
<tr>
<td>7</td>
<td>570</td>
<td>PDMS</td>
<td>50</td>
</tr>
<tr>
<td>8</td>
<td>500</td>
<td>Ge</td>
<td>Failed in Machining</td>
</tr>
</tbody>
</table>

- 132 -
This is a very promising result. It may remove the need for several intermediate layers between the beryllium and AlBeMet and thereby reduce the complexity of the bond.

This results also confirms the importance of using low concentrations of silicon as a beryllium bonding agent. Previous work using Si bonding aids at Brush Wellman and other institutions failed, and the reason for failure may not have been the intrinsic utility of silicon, but rather the fact that the silicon concentrations were too high. The silicon form and concentration used here are probably not the optimum. Other silicon carriers may perform better. Extremely thin plasma sprayed coatings or evaporated films which can more uniformly cover the surface may perform better still.

Table 9 Summary of Tensile Failure Location for Be/AlBeMet/Cu alloy Bonds

<table>
<thead>
<tr>
<th>Temp (°C)</th>
<th>Bond Aid</th>
<th>Tensile Failure Location</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>RT</td>
</tr>
<tr>
<td>570</td>
<td>Si</td>
<td>Cu/AlBe Interface</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Cu/AlBe Interface</td>
</tr>
<tr>
<td>570</td>
<td>PDMS</td>
<td>Cu/AlBe Interface</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Cu/AlBe Interface</td>
</tr>
</tbody>
</table>

The Be/Si Bonding Aid/AlBeMet 162/Cu Alloy bonded structure made by hot isostatic pressing (HIPing) at 700°C (Trial #5) did not have good bond strength. The tensile sample blanks fell apart as they were cut from the bonded structure by EDM. The Be/PDMS/AlBeMet 162/Cu Alloy bonded structure made by hot isostatic pressing (HIPing) at 570°C (Trial #7) was robust enough to survive machining by EDM. Moderate tensile strengths for the structure are obtained, but again as shown in Table 9 and Figure 4, the limiting bond was at the Cu/AlBeMet interface. This particular test shows that PDMS may be a useful bonding aid for bonding Be to AlBeMet. The Be/PDMS/AlBeMet bond did not fail at 50 MPa as did the Hycon/AlBeMet bond in Trial #4. A different copper alloy was used in this test - essentially pure Cu (C11000). These results indicate that the Hycon/AlBeMet bond is stronger than the pure Cu/AlBeMet bond.

Both bonding trials using germanium as a bonding aid produced bonded structures which did not have good bond strength. The tensile sample blanks fell apart as they were cut from the bonded structure by EDM.

5. CONCLUSIONS

5.1. Proprietary Process for Bonding S-65C Grade Be to Cu: Layer Thickness

1. Feasibility of decreasing the bond layer thickness from 6.4 mm to 1.6 mm was demonstrated. The bond was robust during machining.

2. Optimization of bond layer manufacture is needed to eliminate variability in bond strength results.

5.2. Proprietary Process for Bonding S-65C Grade Be to Cu: Thermal Cycle to Maintain Hycon Properties During Bonding

1. Bond cycles designed to maintain Hycon properties while providing a good bond between S-65C and Hycon did not provide a good bond.

5.3 Bonding AlBeMet® to Be

1. Low concentrations of Si are a very effective bonding aid for bonding Be to AlBeMet at HIP temperatures below 700°C.

2. Two combinations of bonding aid and HIP temperature produced Be/AlBeMet bonds which...
withstood machining: Si powder HIP'd at 570°C and polydimethylsiloxane HIP'd at 570°C.

3. **Si Powder Bonding Aid** Be/AlBeMet/Hycon bonded structure had a room temperature strength of 115-125 MPa. However, the failure was at the AlBeMet/Hycon interface, so the strength of the Be/AlBeMet bond is higher than this.

4. **Si Powder Bonding Aid** Be/AlBeMet/Hycon bonded structure had only mild decrease in bond strength from RT to 200°C.

5. AlBeMet stronger than AlBeMet/Hycon bond

6. Polydimethylsiloxane is not as effective a bonding aid as Si powder under the conditions tested.

7. AlBeMet/Hycon bond is stronger than AlBeMet/C10100 Cu bond.

REFERENCES


2. K. Slattery, Boeing Co., private communication, 1997


Figure 1 Drawing of Reduced Section Tensile Specimen for Samples of Be Bonded to Copper

Figure 2 Schematic of HIP Can with Position of Si powder and Cold Pressed AlBeMet® shown.
Figure 3 Reduced Section Tensile Specimens After Testing of S-65C Bonded to AlBemet and Hycon using a Si Powder Bonding Aid (Be/Si powder/AlBemet/Hycon): RT (a,b), 200°C (c,d), 400°C (e,f). Note that failure is at AlBemet/Hycon Cu interface.

Figure 4 Reduced Section Tensile Specimens After Testing of S-65C Bonded to AlBemet and Hycon using a PDMS Bonding Aid (Be/PDMS/AlBemet/Hycon): RT (a,b), 400°C (c). Note that failure is at AlBemet/ Cu interface.
ITER first wall beryllium mockups, which were fabricated by vacuum plasma spraying the beryllium armor, have survived 3000 thermal fatigue cycles at 1 MW/m$^2$ without damage during testing at the Plasma Materials Test Facility at Sandia National Laboratory in New Mexico. The thermal and mechanical properties of the plasma sprayed beryllium armor have been characterized. Results are reported on the chemical composition of the beryllium armor in the as-deposited condition, the through thickness and normal to the through thickness thermal conductivity and thermal expansion, the four-point bend flexure strength and edge-notch fracture toughness of the beryllium armor, the bond strength between the beryllium armor and the underlying heat sink material, and ultrasonic C-scans of the Be/heat sink interface.

1. INTRODUCTION

Investigations are being conducted within the International Thermonuclear Experimental Reactor (ITER) community to qualify a process for joining beryllium armor directly to an actively cooled copper heat sink for the primary first wall. A number of joining techniques are currently being evaluated by the international ITER community which include silverless brazing, diffusion bonding, hot isostatic press (HIPing) bonding, explosive bonding and plasma spraying [1]. In all cases joining beryllium directly to copper presents a challenging problem due to the formation of brittle intermetallic compounds (e.g. BeCu, Be$_2$Cu) at the interface. Plasma spraying is being investigated due to its flexibility of providing thick coatings of beryllium directly on large flat and curved copper surfaces on the primary first wall of ITER. In an initial study, Be/Cu mockups produced by plasma spraying survived 3000 thermal fatigue cycles at 1 MW/m$^2$ without damage during testing at the Plasma Materials Test Facility at Sandia National Laboratories [2]. This heat flux was twice the expected design heat flux for the first wall modules in ITER. Results of this test demonstrated the viability of plasma spraying as a method for fabricating the beryllium first wall armor for ITER.

In this investigation the thermal and mechanical properties of the plasma sprayed beryllium armor on the Be/Cu mockups were evaluated. Specific properties which were characterized included the through thickness and normal to the through thickness thermal conductivity and thermal expansion, the four-point bend flexure strength and edge-notch fracture toughness, and interfacial bond strength between the beryllium armor and the heat sink material. Chemical analysis of the beryllium armor after plasma spraying was also evaluated and compared to the initial powder feedstock material composition.

2. EXPERIMENTAL

Two Be/Cu mockups were vacuum plasma sprayed by translating the plasma sprayed torch in an X and Y motion over the surface of the copper heat sink and stainless steel fixturing while simultaneously depositing beryllium, Fig 1. Thick beryllium coatings (> 15mm) were deposited on the surface of two different types of copper heat sinks: 1) a CuNiBe heat sink and 2) a CuCrZr heat sink which had aluminum explosively bonded to the surface with a thin titanium diffusion barrier interlayer between copper and aluminum. Subsequent machining and profiling of the beryllium armor on the heat sinks was done in order to provide 5 and 10 mm thick individual beryllium tiles for the high heat flux testing. Details on the fabrication, machining and high heat flux testing of the plasma sprayed Be/Cu mockups can be found in reference [2].
Fig. 1. Schematic of the translation of the plasma spray torch to produce the Be/Cu plasma sprayed mockups.

To characterize the thermal and mechanical properties of the plasma sprayed beryllium, test samples were machined directly from the thick beryllium armor.

2.1 Mechanical Properties

2.1.1. Flexure strength and fracture toughness

To evaluate the mechanical properties of the beryllium plasma sprayed armor, four-point bend flexure strength and single-edge notched fracture toughness testing (in four-point bending) were performed, Fig. 2.

The four-point bend samples were electro-discharge machined (EDM) from the beryllium armor. The sample dimensions were 25 mm long x 2 mm wide x 2 mm thick. Each sample was polished to a 600 grit finish on all four sides before testing. The samples were tested in an Instron machine using a cross-head speed of 0.1 mm/min. An outer support span of 19 mm coupled with an inner loading span of 9.5 mm was used on all of the bend tests. Fracture toughness measurements were carried out using a single-edge notched beam specimen. Notches in the four-point bend samples were cut to one-third of the sample thickness (0.67 mm) using a slow speed diamond saw. A total of five samples were used to obtain the average value for both the flexure strength and the single-edge notched fracture toughness. The four-point bend flexure strength tests were carried out in accordance with ASTM standard C-203/85. The fracture toughness tests were carried out in accordance with ASTM standard STP 678.

2.1.2. Interfacial strength and adhesion

To determine the interfacial strength of the plasma sprayed beryllium armor on the CuNiBe heat sink and the CuCrZr heat sink with the explosive bonded aluminum layer, four-point bend testing was also performed. The samples tested were 25 mm long x 2 mm wide x 2 mm thick. The interface was located at the center of the sample (12.5 mm from the edge) within the inner loading span, Fig. 2. The four-point bend interfacial strength tests were performed under the same loading conditions used for the flexure strength and fracture toughness tests. The samples were all taken to failure during testing. Characterization of the interface between the beryllium plasma sprayed armor and the heat sink material was performed using scanning electron microscopy. A non-destructive evaluation using C-scan ultrasonics was also performed on the Be/Cu mockups to qualitatively determine the integrity of the bond between the beryllium plasma sprayed armor and the underlying heat sink. In performing the ultrasonic test, the energy level of the ultrasonic transmitter, the attenuation of the ultrasonic receiver and the relative location of the probe to the specimen were adjusted to give a good contrast between a relatively good and a bad bond at the interface. The NDE tests were performed at KARTA Technology using a Krauthramer HIS-1 ultrasonic analyzer with an attenuation of 39dB. A 5 and 10 Mhz probe with a .25 inch diameter...
piezoelectric crystal (Panametrics V312) was used on all scans.

2.2 Thermal Properties

2.2.1. Thermal conductivity and expansion

Thermal diffusivity samples (3 mm thick x 12.7 mm in diameter) and thermal expansion samples (5 mm in diameter x 8 mm long) were machined from scrap material after the beryllium armor had been machined to the required dimensions. To evaluate the microstructural effects on the thermal properties of the plasma sprayed armor, samples were machined from two different orientations; 1) through the thickness of the beryllium armor coating (i.e., in the sprayed direction) and 2) in the plane of the coating (i.e., normal to the spray direction), Fig 3.

Thermal diffusivity measurements were performed at Virginia Polytechnic University using a laser flash diffusivity technique [3]. The thermal conductivity of the beryllium armor was determined from the following relationship

\[ \kappa = \rho C_p D \]  

(1)

where \( \kappa \) is the thermal conductivity, \( \rho \) is the density and \( C_p \) is the specific heat and \( D \) is the thermal diffusivity. A density of 92% of theoretical (1.702 g/cm\(^3\)) was used to calculate the thermal conductivity. The specific heat as function of temperature (T) was determined using the following relationship [4]:

\[ C_p = 1741.8 + 3.3358 \times T - 3.1125 \times 10^{-3} \times T^2 + 1.2748 \times 10^{-5} \times T^3 \]  

(2)

The calculated values for thermal conductivity of the beryllium plasma sprayed armor were then compared directly to the thermal conductivity values for S-65B beryllium [4]. Microstructural characterization of the thermal diffusivity samples was done using polarized and light microscopy after the thermal diffusivity measurements.

The coefficient of thermal expansion (CTE) of the beryllium armor was measured on a quenching and deformation dilatometer manufactured by Materials Measuring Corporation. The beryllium samples were heated in vacuum to 1000 °C and furnace cooled to room temperature. The heating and cooling cycle took approximately 30 minutes to complete. The measured CTE for the beryllium armor was also compared to S-65B CTE data [4].

3. RESULTS AND DISCUSSION

3.1. Chemical Analysis

A chemical analysis of the inert gas atomized beryllium powder used for plasma spraying and the resulting beryllium armor on the CuNiBe and CuCrZr heat sink is given in Table 1. Substantial differences in the chemical analysis between the beryllium powder and the beryllium armor was observed for N, C, BeO, Al and Cu. Increases in the N, C and BeO were attributed to the commercial purity of the processing gases used during the plasma spraying process. The observed increases in the Al and Cu was attributed to erosion of the copper anode inside of the plasma spray torch and erosion of Al parts inside of the powder feeders.

3.2. Four-point bend flexure strength of armor

The flexure strength of the beryllium plasma sprayed armor on the CuNiBe and CuCrZr heat sinks is given in Fig. 4. The stress/displacement curves show a linear strain to failure for the beryllium armor in both cases. The flexure strength of the beryllium armor deposited on the CuNiBe was significantly stronger (~285 MPa) then the beryllium which was deposited on the CuCrZr with the aluminum explosive bonded layer (~155 MPa).
Table 1. Chemical analysis of beryllium powder and Be armor material

<table>
<thead>
<tr>
<th>Element</th>
<th>Be Powder</th>
<th>Be on CuNiBe</th>
<th>Be on CuCrZr</th>
</tr>
</thead>
<tbody>
<tr>
<td>*BeO</td>
<td>.34</td>
<td>.84</td>
<td>.98</td>
</tr>
<tr>
<td>*C</td>
<td>.074</td>
<td>.130</td>
<td>.104</td>
</tr>
<tr>
<td>Fe (ppm)</td>
<td>920</td>
<td>965</td>
<td>980</td>
</tr>
<tr>
<td>Al (ppm)</td>
<td>440</td>
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<td>625</td>
</tr>
<tr>
<td>Si (ppm)</td>
<td>310</td>
<td>405</td>
<td>385</td>
</tr>
<tr>
<td>Ni (ppm)</td>
<td>110</td>
<td>105</td>
<td>105</td>
</tr>
<tr>
<td>Cu (ppm)</td>
<td>35</td>
<td>115</td>
<td>90</td>
</tr>
<tr>
<td>N (ppm)</td>
<td>80</td>
<td>405</td>
<td>395</td>
</tr>
</tbody>
</table>

* wt.%

Fig. 4. Results of the 4-point bend flexure strength of the beryllium armor on the CuNiBe and CuCrZr heat sinks.

Strength of plasma sprayed armor is predominately determined by the cohesive strength between the individual splat layers which make up the bulk of the armor. Improvements in the cohesive strength can be accomplished by enhancing the bond between individual splat layers. Microstructural analysis using optical and polarized light microscopy of the beryllium armor on both heat sinks revealed similar microstructural features including porosity striations and unmelted beryllium particles, Fig. 5. Immersion densities were also similar and were approximately 92% of theoretical density (1.702 g/cm³). Transmission electron microscopy studies are currently in progress to determine if their are any significant differences between the inter-splat bonding. The differences in the flexure strength of the beryllium armor is not well understood at this time but may be influenced by differences in the residual stresses that develop during the deposition of the thick beryllium coatings on the two different copper heat sink materials (CuNiBe vs. CuCrZr) and depositing on a copper surface versus depositing on an aluminum surface. In addition, BeO can influence the mechanical strength of beryllium at room and elevated temperatures [5]. Further investigations will need to be performed in order to determine these effects.

Fig 5. Bright field image of the plasma sprayed beryllium armor a) on the CuNiBe heat sink and b) on the CuCrZr heat sink with an explosive bonded aluminum surface layer. Note porosity striations.
3.3. Edge-notched fracture toughness of armor

The stress-displacement curves for the four-point bend edge-notched samples is given in Fig. 6. The maximum stress in bending was approximately 160 MPa for the beryllium armor on the CuNiBe and CuCrZr heat sinks. The two samples exhibited similar stress-displacement behaviors during four-point loading. There was an initial load increase followed by load leveling. Beyond a critical displacement the load increased until sample failure had occurred.

A constant load with an increase in displacement is typically observed in materials which exhibit R-Curve behavior [6]. R-curves characterize the resistance to fracture of a material during crack extension. The constant load and increase crack extension represents a “pop-in” effect which arises from a sudden unstable, rapid crack propagation before the crack slows down to a more stable crack advancement. In the plasma sprayed beryllium armor, porosity striations were observed throughout the coating thickness and can potentially influence the crack advancement through the armor. The fracture toughness, $K_{IC}$, for the beryllium armor on the CuNiBe was approximately 9.26 MPa m$^{1/2}$ and 8.56 MPa m$^{1/2}$ for the beryllium armor on the CuCrZr heat sink with an explosive bonded aluminum surface layer. The presence of porosity in beryllium has been shown to improve the fracture toughness of beryllium [6].

3.4. Interfacial strength of Be armor/heat sink

The stress-displacement results from a four point bend test to determine the interfacial strength between the plasma sprayed beryllium armor on the CuNiBe heat sink and the CuCrZr heat sink with aluminum explosively bonded to the surface are shown in Fig. 7.

A constant load with an increase in displacement is typically observed in materials which exhibit R-Curve behavior [6]. R-curves characterize the resistance to fracture of a material during crack extension. The constant load and increase crack extension represents a “pop-in” effect which arises from a sudden unstable, rapid crack propagation before the crack slows down to a more stable crack advancement. In the plasma sprayed beryllium armor, porosity striations were observed throughout the coating thickness and can potentially influence the crack advancement through the armor. The fracture toughness, $K_{IC}$, for the beryllium armor on the CuNiBe was approximately 9.26 MPa m$^{1/2}$ and 8.56 MPa m$^{1/2}$ for the beryllium armor on the CuCrZr heat sink with an explosive bonded aluminum surface layer. The presence of porosity in beryllium has been shown to improve the fracture toughness of beryllium [6].

![Fig. 6. Stress-displacement plots for edge-notched four-point bend testing of the beryllium armor.](image1)

Fig. 6. Stress-displacement plots for edge-notched four-point bend testing of the beryllium armor.

![Fig. 7. Four-point bend strength of the interface between the beryllium armor on the CuNiBe heat sink and on the CuCrZr heat sink.](image2)

Fig. 7. Four-point bend strength of the interface between the beryllium armor on the CuNiBe heat sink and on the CuCrZr heat sink.

![Fig. 8. Location of the failure in the beryllium armor during the four-point bend interfacial strength test. A) beryllium armor on CuNiBe heat sink and b) beryllium armor on the CuCrZr heat sink with an explosive bonded aluminum surface layer.](image3)

Fig. 8. Location of the failure in the beryllium armor during the four-point bend interfacial strength test. A) beryllium armor on CuNiBe heat sink and b) beryllium armor on the CuCrZr heat sink with an explosive bonded aluminum surface layer.
In both cases, failure occurred in the beryllium armor and not at the beryllium/heat sink interface during the four-point bend test, Fig 8. The strength of the beryllium armor on the CuNiBe heat sink (~70 MPa) was significantly lower than the strength of the beryllium armor on the CuCrZr heat sink (~110 MPa). Failure typically initiated in the beryllium armor and propagated along a path parallel to the heat sink interfaces. Cracking parallel to the interface is often seen in joints of dissimilar materials like ceramic/metal systems [7-9]. The thermal expansion mismatch between dissimilar materials can result in compressive stresses at the interface causing a crack to be deflected away from the interface, but still travel parallel to the interface. The plasticity and relative toughness of the interface along with residual tensile stresses in the plasma sprayed beryllium armor can promote cracking in the beryllium armor at coating defects. The interfacial strength between the beryllium armor/heat sink was stronger, in both cases, than the beryllium armor itself.

3.5 Ultrasonic C-scan results of beryllium armor/heat sink interface

Baseline ultrasonic C-scans of the interface between the 5 and 10 mm beryllium armor tiles on the CuNiBe and the CuCrZr heat sinks are given in Figs 9 and 10. These preliminary ultrasonic C-scans were done to determine the relative bond quality of the beryllium armor to the heat sink materials before high heat flux testing. The squares in the ultrasonic C-scans represent the individual beryllium tiles on the Be/Cu mockups. A dark region in the ultrasonic C-scan indicates a high intensity spot where a potential defect/discontinuity at the interface existed. The low intensity ultrasonic C-scans for the beryllium armor on the CuNiBe and CuCrZr heat sinks was an indication of no appreciable defects at the armor/heat sink interface.

Fig. 9. Benchmark ultrasonic C-scan of interface of Be on CuNiBe heat sink. The sample contains two 5-mm tiles (a) and two 10-mm tiles (b). The squares represent the boundaries of the armor tiles.

Fig 10. Benchmark ultrasonic C-scan of interface of the Be armor on a CuCrZr heat sink with an explosive bonded aluminum surface layer. The sample contains two 5 mm tiles (a) and two 10 mm tiles (b). The squares represent the boundaries of the armor tiles.
In the case of the beryllium armor on the CuCrZr heat sink, Fig. 10, the ultrasonic C-scan did indicate a region of high intensity on the lower portion of each of the 5 mm tiles. Although the spots indicated localized areas of poor bond quality, we suspected the damages were caused by drilling of thermocouple holes in the copper heat sink and the beryllium tiles.

### 3.6. Thermal expansion of beryllium armor

Thermal expansion through the thickness of the beryllium armor (i.e., in the spray direction) and in the plane of the armor (i.e., normal to the spray direction) is given in Fig. 11. The CTE for both orientations had a behavior similar to S65-B from room temperature to \(400^\circ C\). A slight anisotropy in CTE was observed for the two orientations. Above \(400^\circ C\) the plasma sprayed beryllium armor showed a significant contraction for both orientations. This behavior has been previously observed in plasma sprayed beryllium and has been attributed to microstructural effects in the plasma sprayed beryllium coating [10].

Fig. 11. Thermal expansion through the thickness of the beryllium armor (i.e., in the spray direction) and in the plane of the armor (i.e., normal to the spray direction)

Under an applied load at elevated temperatures the layered microstructure, which is typically seen in plasma sprayed coatings, can slide against each other producing the observed contraction. The amount of contraction can vary depending upon the bond strength between individual splat layers. At higher temperatures and applied loads the contraction due to sliding can potentially lead to densification of the beryllium plasma sprayed coating. The differences in the CTE between the armor tested through the thickness of the coating and along the plane of the coating can be attributed to the difference in orientation of individual splat layers and the effective resistance to sliding.

### 3.7. Thermal conductivity of beryllium armor

A comparison of the through thickness thermal conductivity from room temperature to \(600^\circ C\) for beryllium plasma sprayed armor on the CuNiBe and the CuCrZr heat sink is shown in Fig. 12, and compared to S65-B beryllium.

The thermal conductivity of the beryllium deposited on the CuNiBe heat sink was approximately 40-50% of S65-B over the room temperature to \(600^\circ C\) temperature range whereas the thermal conductivity of the beryllium deposited on the CuCrZr heat sink, which had an explosive bonded aluminum surface layer, was approximately 60-70% of S65-B. Both beryllium armor samples showed a similar trend in thermal conductivity to S65-B from room temperature to \(600^\circ C\). The microstructure, Fig 5, and immersion density (approximately 92% of theoretical) of the beryllium armor samples were similar and showed no obvious differences. In both cases, porosity striations were observed throughout the thickness of the beryllium coating. Although the difference between the thermal conductivity of the beryllium armor is not fully understood, a number of factors such as pore size and pore distribution [11], splat-to-splat cohesion within the coating [12] and the amount and distribution of BeO in beryllium [10] may contribute to the observed differences.

Fig. 12. A comparison of the through thickness thermal conductivity from room temperature to \(600^\circ C\) for the beryllium plasma sprayed armor on the CuNiBe and the CuCrZr heat sink. Results are compared to S65-B.
The residual stresses during the deposition and build-up of the thick beryllium armor may also influence both the mechanical and thermal properties of the final beryllium coating. Further investigations are needed in order to evaluate the contribution of these various factors.

An anisotropy in thermal conductivity was observed in the plasma sprayed beryllium armor, Fig. 13. The thermal conductivity in the spray direction was lower than the thermal conductivity normal to the spray direction over the room temperature to 600°C temperature range.

![Graph showing thermal conductivity comparison](image)

Fig. 13. A comparison of the thermal conductivity through the thickness of the beryllium armor (i.e., in the spray direction) versus in the plane (i.e. normal to the spray direction) of the beryllium armor.

This difference in conductivity can also be attributed to microstructural effects of the plasma sprayed beryllium armor which consists of individual molten particles which impact and spread into individual splat layers building-up the thickness of the coating. Layer separation between individual splats can result in a thermal resistance across the splat interfaces which can reduce the through thickness thermal conductivity \[12\]. The presence of the porosity striations which are normal to the through thickness of the coating and in the heat conduction path, can also provide a thermal resistance at each striation. Porosity striations along the length of the heat conduction path, which were present in the samples tested in the plane of the coating, would not provide as much resistance to thermal conduction.

4. CONCLUSION

Be/Cu mockups for high heat flux testing were produced by plasma spraying beryllium on heat sink materials of CuNiBe and CuCrZr with an explosive bonded layer of aluminum. Characterization of the thermal and mechanical properties of the plasma sprayed beryllium armor and the interface between the armor and the heat sink material was performed prior to the high heat flux testing. In general, both the thermal and mechanical properties where effected by the microstructural development of the plasma sprayed beryllium armor. The layered structure which is produced from the individual splats which make up the bulk of the beryllium armor contributed to differences in the thermal expansion, thermal conductivity, flexure strength and fracture toughness of the beryllium armor. Other factors such as the oxide content, residual stress and splat-to-splat cohesion may also have contributed to the observed differences in the thermal and mechanical properties. Further investigations will need to be performed in order to evaluate the contributions of each of these factors.

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6. Plasma / Tritium Interactions

6.1 Erosion of Be and Deposition of C and O Due to Bombardment with C\(^+\) and CO\(^+\)

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The bombardment of Be with 3 and 5 keV C\(^+\) and CO\(^+\) at normal incidence is investigated experimentally and by computer simulation with the program TRIDYN. The deposited amount of C and O is determined experimentally and found in good agreement with calculated data for C bombardment. Chemical erosion dominates at higher fluences for CO\(^+\) bombardment. Calculations are then used to determine the sputter yield of Be at steady state conditions as a function of the plasma edge electron temperature for two C impurity concentrations in the incident D flux, typical for fusion plasmas. The fluence to reach steady state conditions is also investigated.

1 Introduction

In the design of future fusion plasma devices like ITER several elements are regarded as possible first wall materials. Both, low-Z elements as Be and C, and heavy elements as Mo and W are under discussion. If these elements are eroded from walls or divertor plates, they will eventually lead to impurity contamination of the discharge. In this case plasma facing components will be bombarded simultaneously with hydrogen isotopes and these impurity species. Of the low-Z elements Be has been used in JET [1] and C in most fusion machines. Earlier investigations have shown that experimental data are well reproduced by simulations for the bombardment of W with C only [2]. Some information on Be is given in [3, 4]. Therefore, in this paper the effects of a simultaneous bombardment of Be with C and O from CO\(^+\) are investigated experimentally and compared to computer simulations to validate the modelling of this more complex situation. Furthermore, co-implantation of D and C is studied in computer simulations for fusion plasma relevant conditions.

2 Experiment

A polycrystalline polished Be target was bombarded at normal incidence with 3 and 5 keV C\(^+\) and CO\(^+\) ions in the apparatus BOMBARDON [5]. The ions are produced by an electron impact source from CO gas (Linde 3.7) and subsequently mass-separated by a 80° magnetic deflection. The base pressure in the target chamber was in the low 10\(^{-11}\) hPa range, during implantation and analysis in the 4-5 \(\times\) 10\(^{-11}\) hPa range. The Be target was cleaned by periodic 5 keV Ar\(^+\) bombardment and annealing to 400 °C. Analysis of the implanted C and O was performed by Rutherford backscattering (RBS) using 0.6 MeV \(^4\)He\(^+\) at normal incidence. The detector was situated at a scattering angle of 163°.

Fig. 1 shows RBS spectra of the sample after cleaning (a) and after CO\(^+\) implantation (b). The spectrum (a) of the clean sample exhibits the Be edge and peaks originating from Ar caused by the cleaning procedure, as well as a small signal from tantalum. The Ta surface contamination is caused by sputter deposition of Ta from an aperture used in early steps of the cleaning procedure. The Ta intensity corresponds to an amount of 6.0 \(\times\) 10\(^{13}\) cm\(^{-2}\) (about 0.04 monolayers). Fig. 1b shows the RBS spectrum of the Be target after bombard-
3 Simulation

The calculations were performed with the Monte Carlo program TRIDYN (version 40.3) [6, 7]. This program takes into account all collisional effects as implantation, reflection, and sputtering. Target composition changes due to the bombardment are regarded as well, so that effects like sputter yields, reflection coefficients, and composition profiles can be determined as a function of the bombarding fluence. The program allows also simultaneous bombardment with several species of fixed energy or with a Maxwellian incident distribution. Surface binding energies, important for sputtering, are based on the elemental heats of sublimation and are interpolated due to the surface composition of the target [8]. Chemical erosion, diffusion and segregation are neglected.

4 Results and Discussion

4.1 C\(^+\) implantation

The Be target was bombarded with C\(^+\) ions up to a fluence of 55 \(\times 10^{16}\) cm\(^{-2}\). The maximum fluence was determined by the lifetime of the ion source filament. Fig. 2 shows the calculated depth distribution of 5 keV C\(^+\) implanted in beryllium for several fluences which correspond to the experimentally applied values. Beginning with 5 \(\times 10^{16}\) cm\(^{-2}\), carbon is deposited in a depth of roughly 200 Å with almost no carbon at the sample surface. For higher fluences, the carbon concentration on the target surface increases up to almost unity. This is due to the simultaneous carbon deposition and sputtering by C\(^+\) ions. Since the self sputtering coefficient of C\(^+\) on carbon is below 1 [3], a carbon layer is expected to build up under these conditions.

The experimental results confirm the predictions from the TRIDYN simulation. Fig. 3 shows the deposited amount of carbon on the target as a function of the C\(^+\) fluence. The lines represent...
Figure 2: Depth distributions of C in Be. Be is bombarded with 5 keV C\(^+\) at normal incidence (TRIDYN). The calculated fluences correspond to the values of the experiments.

the TRIDYN calculations for a primary energy of the C\(^+\) ions of 3 and 5 keV. The data points are the measured carbon concentrations in the Be target after the C\(^+\) bombardment. The lines represent simulations with KrC \([9]\) and ZBL \([10]\) interaction potentials. The difference between them is negligible. The amount of deposited carbon after ion bombardment is calculated from the carbon peak area in the RBS spectra after background subtraction. For 5 keV C\(^+\) the agreement between simulation and experiment is very good, for 3 keV a deviation can be recognized. The experimental error is either in the measured fluence or in the deposited amount. For a final assessment for this primary energy, further experimental data points at higher C\(^+\) fluences are desirable.

As predicted by the simulation, the experiments show the buildup of a carbon layer on the sample surface. In the studied fluence range no saturation in the deposited amount of carbon is found. The carbon layer thickness at the maximum experimental C\(^+\) fluence of 54.8 \(10^{16}\) cm\(^{-2}\) is 433 Å, calculated for a graphite density of 2.26 g cm\(^{-3}\). This is in good agreement with the result from the calculation (Fig. 2), where the carbon concentration starts to decrease at a depth of about 400 Å. However, the maximum carbon concentration in the calculation is only 90%, but the additional carbon in deeper layers leads to a total amount of carbon comparable to that detected in the RBS experiments. The carbon layer at the maximum fluence after 3 keV C\(^+\) bombardment (20 \(10^{16}\) cm\(^{-2}\)) reaches a thickness of 93 Å.

4.2 CO\(^+\) implantation

Since the CO\(^+\) currents from the ion source are much larger than for C\(^+\), implantation fluences up to 200 \(10^{16}\) cm\(^{-2}\) are accessible in the experiment. Compared to the C\(^+\) beams at 3 and 5 keV, the carbon and oxygen atoms impinging on the surface for 3 and 5 keV CO\(^+\) beams have lower energies. The kinetic energy is split between the atoms in the CO\(^+\) molecule according to the mass ratio of C (3/7) and O (4/7). Therefo-
re, carbon possesses 1.286 keV and 2.143 keV for the 3 and 5 keV primary beams, whereas oxygen has energies of 1.714 and 2.857 keV, respectively.

At low fluences, the behaviour is similar to that for C\textsuperscript{+} implantation, see Fig. 4. The experimentally measured amounts of deposited carbon and oxygen in the sample increase according to the calculations by the TRIDYN program. However, beginning with a CO\textsuperscript{+} fluence of about 4 \cdot 10\textsuperscript{16} cm\textsuperscript{-2}, the C and O concentrations start to saturate. This effect cannot be explained by mere atomic collisions and is therefore not predicted by the calculations. One reason is the chemical erosion of carbon in the formed layer by oxygen, which is about unity [11, 12, 3]. The formation of CO and CO\textsubscript{2} by chemical erosion of graphite and beryllium was observed under O\textsuperscript{+} irradiation [13, 14]. As observed in the O\textsuperscript{+} experiments [14], this process will also be effective here only after an appreciable C concentration has been accumulated on the surface and after this layer is saturated in oxygen. Therefore, the initial increase in deposited carbon and oxygen following the calculation results is explained by the formation of graphite and anticipated oxide layers. The subsequent deviation from the TRIDYN results is due to the erosion of the previously formed layers.

The measurements suggest that the amounts of deposited C and O pass through a maximum between 10 and 40 \cdot 10\textsuperscript{16} cm\textsuperscript{-2}. This effect is more pronounced for a primary energy of 3 keV. After 40 \cdot 10\textsuperscript{16} cm\textsuperscript{-2}, the deposition/erosion ratio is not completely at equilibrium. However, the total amounts of C and O decrease only slightly. The erosion process dominates the deposition process. After 40 \cdot 10\textsuperscript{16} cm\textsuperscript{-2}, the C/O ratio for the 3 keV implantation scatters around 0.75, whereas for the 5 keV irradiation this value is 0.85. This means that in the case of the lower primary energy, the O fraction in the formed layer is larger than in the 5 keV case. It is also noticeable that the total amount of deposited carbon and oxygen is higher in the case of higher primary energy, as shown in Fig. 4. For an erosion process which takes place via the formation and desorption of molecules, the material deposited in greater depths is less eroded than material close to the surface. For the desorption of molecules it is necessary that they travel from their point of origin to the surface of the layer, where the desorption can take place. This is more likely for molecules formed close to the surface, as in the case of lower deposition energies, than for molecules formed deeper in the sample. Therefore, in quasi-equilibrium, the layer deposited by CO\textsuperscript{+} ions at 3 keV should be thinner than from 5 keV CO\textsuperscript{+}, as observed.

The thickness of the deposited adlayer on the beryllium target can also be deduced from the RBS spectra by the shift of the Be edge compared to the edge position in the clean target. In both CO\textsuperscript{+} ion energy cases, the thickness calculated from the edge shift under the assumption of a graphite layer is larger than calculated from the C integral. The RBS spectra also show oxygen after irradiation, which both lead to the conclusion that BeO is additionally formed in the target. However, to clarify the composition of the layer,
4.3 D, C co-implantation

Assuming that the calculations give a reasonable approach to the problem of simultaneous bombardment, calculations for a Maxwellian distribution of simultaneous bombardment of Be with D\(^{+}\) and C\(^{4+}\) have been performed. The results for the sputter yield of Be at steady state are given in Fig. 5 for three fusion plasma relevant C concentrations in the bombarding flux versus the plasma edge electron temperature. Steady state is reached when the composition stays constant with further bombardment; the thickness of a deposited C layer may still increase. Whereas at higher temperatures (above 200 eV) the C impurity level is not important in the range considered, the Be sputter yield remains higher at lower impurity levels. The reason is a higher remaining Be concentration at the surface. Due to the much lower incident energies and the smaller C fraction compared to the keV bombardment discussed above the sputter yield is strongly reduced and leads to a C deposition below about 10 eV depending on the C impurity flux, an effect also discussed earlier [15].

The fluence to reach steady state conditions for the Maxwellian bombardment with D and C\(^{4+}\) is shown in Fig. 6 for two C impurity levels in the incident flux. Below 10 eV electron temperature steady state means an increasing C film on Be with zero sputter yield of Be. At higher electron temperatures steady state (here the Be sputter yield is not zero, see Fig. 5) is reached at the lowest fluences around 30 eV; the steady state fluence is increasing to lower and higher electron temperatures. Which conditions are reached in fusion devices depends on the incident wall flux and the discharge time.
5 Conclusions

It is shown that the experimental data are in good agreement with results calculated by TRIDYN for 3 and 5 keV C+ bombardment. C+ bombardment at 3 and 5 keV leads to the formation of a carbon layer on Be. CO+ bombardment leads to the formation of a layer containing both carbon and oxygen. In this case, saturation is reached after initial layer formation due to chemical interactions. The observed deviation from the simulation is consistent with O+ erosion models and implantation depths of the incident particles. These processes are not included in the simulation. Calculations are applied to a simultaneous bombardment of Be with D+ and C4+ to model more fusion plasma relevant conditions. At steady state conditions the sputter yield of Be decreases strongly below about 20 eV electron temperature leading to C deposition below about 10 eV electron temperature. Maximum Be sputter yields are reached at slightly higher electron temperatures than for the pure D bombardment. Minimum fluences to reach steady state conditions are found at about 30 eV electron temperature.

6 Acknowledgement

The authors thank R. Bastasz for the polished Be target.

References

6.2 Thermal Desorption of Deuterium from Be, and Be with Helium Bubbles

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Deuterium desorption measurements carried out on a single-crystalline beryllium sample are presented. Deuterium ions were implanted at room temperature at the energy of 0.7 and 1.2 keV up to doses ranging from $10^{19}$ to $3.6\times10^{21}$ m$^{-2}$. In order to eliminate the influence of the beryllium-oxide surface layer, before the implantation the surface of the sample was cleaned by argon sputtering. After the implantation the sample was annealed up to 1200 K at a constant rate of 10 K/s. Deuterium released from the sample was monitored by a calibrated quadrupole mass-spectrometer. The desorption spectra revealed two different contributions. One is a well-defined and very narrow peak centered around 450 K. This peak is observed only at high implantation doses $> 7.8\times10^{20}$ m$^{-2}$, which is close to the deuterium saturation limit of 0.3 D/Be and is related to deuterium release from blisters or interconnected bubbles. The activation energy of 1.1 eV and the threshold implantation dose are consistent with the values reported in literature.

The second contribution in the release spectra is found in the temperature range from 600 to 900 K and is present throughout the whole range of the implantation doses. The activation energies corresponding to this release lie in the range between 1.8 and 2.5 eV and are ascribed to the release from deuterium-vacancy type of defects.

In a number of experiments the deuterium implantation was preceded by helium implantation followed by partial annealing to create helium bubbles. The resulting deuterium desorption spectra indicate that deuterium detrapping from helium bubbles is characterized by an activation energy of 2.7 eV.

1. INTRODUCTION

In the present concept of ITER program Be is considered as a prime candidate for the plasma facing material [1,2]. Therefore during the last years a considerable amount of studies were devoted to deuterium retention in beryllium. A number of experimental results on deuterium retention in beryllium are summarized in table 1. The table presents the following information:
deuterium ion implantation energy and dose, deuterium concentration in beryllium matrix as D/Be ratio, temperatures and the corresponding activation energies for deuterium detrapping and the deuterium traps responsible for the release. Values in brackets are not explicitly quoted in the original papers and are estimated by the present authors. Despite different experimental conditions, e.g. implantation parameters, annealing rates and sample preparation, some features appear to be common and are reproduced in most experiments. Deuterium

Table 1. Summary of the experimental study on deuterium retention in beryllium.

<table>
<thead>
<tr>
<th>Energy (keV)</th>
<th>Dose (m^2)</th>
<th>D/Be</th>
<th>T release (K)</th>
<th>dT/dt (K/s)</th>
<th>Ea (eV)</th>
<th>detrapping mechanism</th>
<th>reference / exp. technique</th>
</tr>
</thead>
<tbody>
<tr>
<td>60</td>
<td>2x10^19</td>
<td></td>
<td>723-773</td>
<td>0.03</td>
<td>2.3</td>
<td>D-vac. clusters</td>
<td>Wampler [3] / NRA</td>
</tr>
<tr>
<td>4x10^20</td>
<td></td>
<td></td>
<td>843</td>
<td>(2.7)</td>
<td></td>
<td>D2 - bubbles</td>
<td></td>
</tr>
<tr>
<td>2x10^19+</td>
<td></td>
<td></td>
<td>873</td>
<td>(2.8)</td>
<td></td>
<td>D2 from helium bubbles</td>
<td></td>
</tr>
<tr>
<td>15 keV</td>
<td>He 4x10^20</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.5-1.5</td>
<td>&gt; 10^21</td>
<td>0.31</td>
<td>398</td>
<td>0.08</td>
<td>1</td>
<td>blisters</td>
<td>Wampler [6] / NRA</td>
</tr>
<tr>
<td></td>
<td>2x10^20</td>
<td>(0.1)</td>
<td>573-773</td>
<td>1.8</td>
<td>(D-vac. clusters)</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>and &gt; 10^21</td>
<td>0.31</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>10^20-10^21 (&lt; 0.2)</td>
<td>3.2</td>
<td>850-1050</td>
<td>4</td>
<td>2.2-2.8</td>
<td>D-vac. clusters</td>
<td>Markin et al</td>
</tr>
<tr>
<td></td>
<td>&gt; 2x10^21 (&gt;0.3)</td>
<td></td>
<td>470, 490</td>
<td>1.3</td>
<td>(1.3)</td>
<td>blisters, channels</td>
<td>[4] / TDS</td>
</tr>
<tr>
<td>about 50</td>
<td>0</td>
<td>0.5</td>
<td>320</td>
<td>0.6</td>
<td>1</td>
<td>from surface:</td>
<td>Lossev and Kuppers [5] / TDS</td>
</tr>
<tr>
<td>2.5</td>
<td>0.5x10^22</td>
<td>0.36</td>
<td>423</td>
<td>0.5</td>
<td></td>
<td>O covered Be</td>
<td>Kawamura et al [7] / ERD</td>
</tr>
<tr>
<td>3.3x10^22</td>
<td></td>
<td></td>
<td>673</td>
<td></td>
<td></td>
<td>clean Be</td>
<td>TDS</td>
</tr>
<tr>
<td>0.7</td>
<td>1.3x10^20</td>
<td>0.1</td>
<td>450</td>
<td>10</td>
<td>1</td>
<td>D-vac. clusters</td>
<td>present work / TDS</td>
</tr>
<tr>
<td>3.7x10^21</td>
<td>&gt; 0.3</td>
<td></td>
<td>700-1000</td>
<td>1.8-2.7</td>
<td></td>
<td>blisters</td>
<td>TDS</td>
</tr>
<tr>
<td>1.2</td>
<td>10^18-10^19 &lt; 0.01</td>
<td></td>
<td>900-1000</td>
<td>2.5-2.7</td>
<td></td>
<td>D2 - bubbles</td>
<td></td>
</tr>
<tr>
<td>3keV He</td>
<td>3.2x10^14</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>D2 in helium bubbles</td>
<td></td>
</tr>
</tbody>
</table>

detrapping from deuterium-vacancy clusters is observed in the temperature region 600-800 K corresponding to an activation energy of 1.8-2.3 eV. When the implantation dose increases the release temperature shifts up to 1000 K and is ascribed to deuterium release from small D2 - bubbles. Deuterium release from helium bubbles studied by Wampler [3] takes place at the
temperature of 843 K. Further increase of the implantation dose ($>10^{17}$) according to Markin et al [4] results in two sharp release peaks located at 470 and 490 K. A TEM study on the beryllium samples implanted with deuterium up to the saturation dose of about 0.3 D/Be indicated blisters or bubbles interconnected with channels [4]. In a different study, where beryllium was exposed to a hydrogen ion beam, Lossev and Kuppers [5] have shown that unlike molecular hydrogen, H atoms get absorbed at the beryllium surface. The activation energies for desorption were reported to be 0.6 eV for the oxygen covered and 0.87 eV for clean beryllium surfaces. According to the latter observation deuterium release from the blisters, probably already opened after implantation, is governed by desorption from the surface. Therefore the activation energy of about 1 eV related to this desorption peak is much lower then the values characteristic for deuterium detrapping from vacancy clusters and bubbles in the bulk and are close to the values reported by Lossev and Kuppers.

The present study is aimed to investigate deuterium detrapping from implanted beryllium and from beryllium pre-implanted with helium by means of thermal desorption.

2. EXPERIMENTAL

Thermal desorption experiments were performed on a single crystal beryllium in a UHV-chamber equipped with an ion gun and a calibrated quadrupole mass-spectrometer. The implantation energies used for deuterium were 0.7 and 1.2 keV. The implantation doses varied from $10^{19}$ till $7 \times 10^{21}$ m$^{-2}$. The implantation energy for $^3$He and $^4$He ions was 3.0 keV. The ramp annealing was carried out at a constant rate of 10 K/s up to 1200 K. During the annealing deuterium release was simultaneously monitored for two masses: HD and D$_2$.

3. RESULTS AND DISCUSSION

3.1. Deuterium trapping in Be

The desorption spectra for HD and D$_2$ after 2.5 keV D$_2^+$ implantation in Be for the doses $10^{19}$, $3.2 \times 10^{19}$, $5.6 \times 10^{19}$ and $10^{20}$ m$^{-2}$ are shown in Fig. 1. The broad desorption peak in the temperature region 600-900 K is ascribed to the deuterium escape from small vacancy clusters created by implantation. A small peak in the beginning of the spectra at 450 K is observed for HD and D$_2$ and is ascribed to desorption from the surface. The activation energy of 1.1 eV which corresponds to this release is close to the values reported by Lossev and Kuppers [5] for hydrogen desorption from beryllium covered with oxygen and clean beryllium of 0.6 and 0.78 eV, respectively. A number of sharp peaks at 1000-1100 K can be ascribed to deuterium release from D$_2$-bubbles or from helium bubbles not fully released from the previous experiments.
Thermal desorption spectra for deuterium ions implanted with 0.7 keV in Be with high doses are shown in Fig. 2. A sharp peak at 430 K is observed only for the implantation doses higher than $10^{21}$ m$^{-2}$. The activation energy for this release of 1.1 eV is in reasonable agreement with the values reported earlier [3,4]. In these references, the release is ascribed to detrapping from the blisters or bubbles interconnected with channels which were observed by TEM [4]. Note that the activation energy attributed to this release is close to the value observed for the surface detrapping for low dose deuterium ion implantation.

3.2. Helium trapping in Be

Helium desorption spectra implanted with the energy of 3.0 keV and doses varied from $10^{18}$ till $10^{19}$ m$^{-2}$ are shown in Fig. 3. Helium release peaks are located at 490, 700, 950 and 1050 K and are in agreement with the helium desorption study of Eleveld et al [8]. Assuming a first order release model, these temperatures correspond to the activation energies of 1.2, 1.8, 2.5 and 2.8 eV, respectively. In the work of Eleveld et al [8] on helium irradiated Be, it was
also shown that after 600-800 K ramp annealing the helium-vacancy clusters evolve into bubbles which appear to be stable even at 1000 K.

3.3. Deuterium in Be pre-implanted with helium

In the following experiment the influence of the helium bubbles on the deuterium trapping in beryllium was investigated. \(^3\)He ions were implanted in beryllium with 3 keV energy and doses \(3.2 \times 10^{18}\) and \(10^{19}\) m\(^2\). After that the sample was ramp annealed till 800 K to grow helium bubbles. Then 1.2 keV deuterium ions were implanted up to a dose of \(10^{18}\) m\(^2\). The resulting spectra are shown in Fig. 4 and the

<table>
<thead>
<tr>
<th>Spectrum</th>
<th>(^3)He pre-implantation</th>
<th>ramp annealing</th>
<th>D implantation</th>
<th>ramp annealing</th>
</tr>
</thead>
<tbody>
<tr>
<td>a)</td>
<td>-</td>
<td>-</td>
<td>1.2 keV, (10^{19}) m(^2)</td>
<td>-</td>
</tr>
<tr>
<td>b)</td>
<td>3.0 keV, (3.2 \times 10^{18}) m(^2)</td>
<td>-</td>
<td>1.2 keV, (10^{19}) m(^2)</td>
<td>-</td>
</tr>
<tr>
<td>c)</td>
<td>3.0 keV, (3.2 \times 10^{18}) m(^2) to 800 K</td>
<td>1.2 keV, (10^{19}) m(^2)</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>d)</td>
<td>3.0 keV, (10^{19}) m(^2) to 800 K</td>
<td>1.2 keV, (10^{19}) m(^2) to 800 K</td>
<td>-</td>
<td></td>
</tr>
</tbody>
</table>
actual treatment of the sample before the measurements corresponding to the spectra a), b), c) and d) are presented in Table 2. In all the spectra obtained with $^3$He pre-implantation a new peak at 950 K is observed. This contribution in the release is ascribed to deuterium escape from the helium clusters/bubbles. The annealing of the sample at 800 K in-between the helium and deuterium implantation cause the 950 K peak to become broader. In the last experiment (spectrum d) in Fig. 4) the annealing after the deuterium implantation was done in two steps. First, the sample was ramp annealed to 800 K to desorb the deuterium trapped in vacancy clusters, and then the sample was annealed to 1200 K. Evidently, the desorption peak moved to higher temperatures (above 1100 K). A possible explanation for this effect is that during the second annealing the deuterium/helium bubbles became larger and therefore gave rise to higher desorption temperature.

4. CONCLUSIONS

Thermal desorption studies on deuterium trapping in a beryllium single crystal have revealed two desorption stages:

1) deuterium desorption from the surface characterized by the activation energy of 1 eV.
2) deuterium desorption from vacancy clusters and small deuterium bubbles with an activation energy of 1.8-2.5 eV

Helium pre-implantation in beryllium with the subsequent annealing at 800 K creates strong traps for deuterium. The activation energy for this release is estimated as 2.7 eV.

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6.3 Experimental Studies and Modeling of Processes of Hydrogen Isotopes Interaction with Beryllium


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The objective of this work was to clarify the surface beryllium oxide influence on hydrogen-beryllium interaction characteristics. Analysis of experimental data and modeling of processes of hydrogen isotopes accumulation, diffusion and release from neutron irradiated beryllium was used to achieve this purpose as well as the investigations of the changes of beryllium surface element composition being treated by H\textsuperscript{+} and Ar\textsuperscript{+} plasma glowing discharge.

1. INTRODUCTION

The analysis of literary data shows that parameters of hydrogen interaction with beryllium strongly depend on surface conditions of this material, which always has an oxide film on the surface. In many cases the influence of this thin film is the most important and consequently to know its behaviour under various conditions is necessary for correct understanding of processes of hydrogen isotopes interaction with beryllium.

2. MODELING OF EXPERIMENTAL DATA ON HYDROGEN ISOTOPES RELEASE FROM BERYLLIUM SAMPLES

All the experimental data, brought in given work, are presented for the EHP-56 beryllium grade, manufactured by Ulba metallurgical plant, Kazakhstan. The chemical composition of beryllium samples, as specified by manufacturer, is shown in Table 1. The density of EHP-56 beryllium is 1.854 g/cm\textsuperscript{3} (100% of theoretical).

To understand the processes, connected with the influence of fission reactor irradiation on the parameters of hydrogen isotopes retention and release from beryllium, the series of experiments on EHP-56 beryllium in-pile loading has been carried out [1]. It was obtained, that hydrogen release from the samples loaded with hydrogen at the temperature 1150K in IVG.1M reactor (irradiation time - 6 hours, the pressure of hydrogen 10\textsuperscript{5} Pa) is much more in comparison with control samples loaded under the same conditions without irradiation and with that loaded after irradiation in nitrogen atmosphere, Fig. 1.

Table 1. Chemical composition of beryllium samples, as specified by manufacture, weight %

<table>
<thead>
<tr>
<th>Element</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Beryllium</td>
<td>98.0</td>
</tr>
<tr>
<td>Oxygen (beryllium oxide)</td>
<td>1.0 (1.60)</td>
</tr>
<tr>
<td>Aluminium</td>
<td>0.04</td>
</tr>
<tr>
<td>Carbon</td>
<td>0.10</td>
</tr>
<tr>
<td>Iron</td>
<td>0.20</td>
</tr>
<tr>
<td>Manganese</td>
<td>0.01</td>
</tr>
<tr>
<td>Nickel</td>
<td>0.01</td>
</tr>
<tr>
<td>Silicon</td>
<td>0.04</td>
</tr>
</tbody>
</table>

Analysis of experimental data on hydrogen diffusion coefficients in polycrystalline beryllium (Fig. 2) and in beryllium oxide [2-6] and carried out numerical modeling of hydrogen isotopes release from a cylindrical beryllium sample (23 mm with the thickness of surface oxide film \(l=5\) nm), using the method of experimental curve fitting, have shown, that only the high-temperature peak (\(T_{\text{peak}}=1100\) K) on TDS spectrum can be described within the framework of diffusion mechanism from
Fig. 1. Typical experimental TDS curves for beryllium samples, subjected to different treatment at the temperature 1150K, $\beta=0.33$K/sec.

Fig. 2. Results of hydrogen release modeling from the beryllium sample $\varnothing3$mm, using the data of different authors.

the bulk of beryllium sample.

The temperature dependences of hydrogen diffusion coefficients in Be and BeO, obtained as the fitting results, nevertheless are a little bit outside the range of experimental data of the various authors being available for us. Fig. 3 shows the ranges of hydrogen isotopes diffusion coefficients in Be and BeO, including our data, received by the modeling of high-temperature peak (Fig.4, $T_{\text{peak}} \approx 1100$K), using the values:

$D_{\text{Be}}=3\times10^7 \exp(-28k\text{J/mole}), \text{[m}^2/\text{sec}]$,

$D_{\text{BeO}}=8\times10^7 \exp(-192k\text{J/mole}), \text{[m}^2/\text{sec}]$.

Hydrogen amount, released from beryllium sample only according to the given mechanism,

exceeds the equilibrium solubility of hydrogen in beryllium under our loading conditions, if to use the data on hydrogen solubility in Be from the work of V.Shapovalov [7]. It assumes initially high concentration of hydrogen in beryllium, obtained during the process of its manufacturing. All the other peaks at lower temperatures cannot be simulated according to diffusion mechanism, using the reasonable values for diffusion coefficients in Be and BeO, and consequently, for their correct description it is necessary to use other mechanisms.

One can try to describe these peaks within the framework of second order desorption model, thus the flux of desorbing particles, is usually written down as:

$$J = N^2\nu^2 \exp\left(-\frac{2E_{\text{des}}}{kT}\right) = kN^2$$ (1)

where $k$ – recombination coefficient, $N$ – concentration of particles on a surface, atoms/cm$^2$, $\nu$ – oscillation frequency of atom on a surface, $10^{13}$ sec$^{-1}$, $\alpha$ – lattice parameter, $\approx 2.7\times10^{-8}$ cm, $E_{\text{des}}$ – activation energy of desorption, as a rule it equals to a difference of activation energy of adsorption and the heat of chemosorption.

When calculating all the frequency factors in the recombination coefficient have been taken equal to $7.3\times10^3$ cm$^2$/sec, as in expression for the flux (1) that are written down as $\nu^2$.

The results of such modeling are shown in
Fig. 4, where high temperature sixth peak is described by the diffusion mechanism. The activation energies of the other gas release peaks, calculated earlier in the work of I. Tazhibaeva [8] using the model of the second order desorption, have been corrected and are represented in Table 2.

Table 2. Activation energies for TDS curve of Fig. 4.

<table>
<thead>
<tr>
<th>Peak number</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
</tr>
</thead>
<tbody>
<tr>
<td>E. eV/atom</td>
<td>0.65</td>
<td>0.78</td>
<td>0.99</td>
<td>1.17</td>
<td>1.33</td>
</tr>
</tbody>
</table>

While it is possible to find in the literature reasonable explanations for the values of binding energies between atom and surface having been calculated for the first two peaks and connected with the energy of decay of Be(H) and Be(OH) compositions, for peaks 3, 4, 5 the existence of traps with such binding energies on BeO surface seems hardly probable. Therefore we assume, that for the first two peaks the recombination of two hydrogen atoms located on a surface takes place, and for peaks 3, 4, 5 the recombination occurs between the atom, located on a surface (in adsorbed state) and the atom, located in near surface region (in absorbed state), and the atom can be located in the sites having different energy levels. For such process the flux of desorbing atoms is commonly written down as:

\[ J = NC^* n^3 \exp\left(-\frac{E_{\text{des}} + E_{\text{b-s}}}{kT}\right) \]  

(2)

where \( C \) - concentration of atoms in near surface layer, atoms/cm\(^3\), \( E_{\text{b-s}} \) - activation energy of bulk-surface transition and, assuming that there is the same concentration of absorbed hydrogen atoms in the near surface layer (recalculated to atoms/cm\(^3\)), as on a surface, it is possible to carry out the modeling of gas release flux according to this mechanism using expression for flux (1).

However, in this case the meaning of activation energy of recombination process changes. If for the first two peaks this energy represents double energy of desorption, for the other three peaks this energy is a sum of activation energy of desorption and the activation energy of hydrogen atom transfer from the bulk onto a surface. Here we make an important assumption, that during the growth of oxide film in the atmosphere of hydrogen under the influence of reactor irradiation there occurs an active oxide formation with the concentration of hydrogen inside significantly exceeding the equilibrium one [2]. The data, received in the experiments on IVG.1M reactor, where the increase of hydrogen retention was observed, confirm the high probability of such mechanism existence.

To confirm that surface region of the sample plays a key role in hydrogen retention the loading in RA reactor was carried out in the deuterium atmosphere, at the same pressure and temperature, as it was done in IVG.1M reactor (T=1150K, loading time 50 hours, deuterium pressure \( 10^5 \) Pa). Gas release spectra for these experiments are submitted on fig. 5 and 6. It is necessary to note, that the effect, having been observed for the samples loaded in IVG.1M reactor, was not observed for the samples loaded in RA reactor, the reactor with less intensity of irradiation. In that case gas release was practically identical for irradiated and non-irradiated samples. It is clearly seen, that gas release in a high-temperature region, responsible for bulk diffusion, has low intensity.

As it is seen from the data of Tables 2-4, the activation energies of gas release remain practically constant, and only the quantity of atoms, released according to various mechanism, are changed. It is also seen, that the 6-th peaks, which was attributed to the diffusion mechanism of gas release, was not displaced with the decrease of released gas quantity.
Fig. 5. Results of modeling of deuterium release from non-irradiated beryllium samples, loaded in deuterium at \( T=1150 \) K for 50 hours, \( P(D_2)=10^5 \) Pa.

Table 3. Activation energies for TDS curve of Fig. 5.

<table>
<thead>
<tr>
<th>Peak number</th>
<th>E, eV/atom</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.72</td>
</tr>
<tr>
<td>1</td>
<td>0.9</td>
</tr>
<tr>
<td>2</td>
<td>0.99</td>
</tr>
<tr>
<td>3</td>
<td>1.14</td>
</tr>
<tr>
<td>4</td>
<td>1.27</td>
</tr>
</tbody>
</table>

Fig. 6. Results of modeling of deuterium release from beryllium samples, irradiated in deuterium in RA reactor at \( T=1150 \) K for 50 hours, \( P(D_2)=10^5 \) Pa.

Table 4. Activation energies for TDS curve of Fig. 6.

<table>
<thead>
<tr>
<th>Peak number</th>
<th>E, eV/atom</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.79</td>
</tr>
<tr>
<td>1</td>
<td>0.95</td>
</tr>
<tr>
<td>2</td>
<td>1.08</td>
</tr>
<tr>
<td>3</td>
<td>1.18</td>
</tr>
<tr>
<td>4</td>
<td>1.3</td>
</tr>
</tbody>
</table>

While the other peaks are shifted to higher temperatures. One can see from the Fig. 4 and Fig. 5.6 that the gas release from the samples, irradiated in RA reactor, is one order of magnitude less than gas release from the samples, irradiated in IVG.1M reactor, even taking into account the greater time of irradiation, greater fluence and the same temperature of irradiation.

As far as the total quantity of desorbed molecules is about \( 10^{13} \) molec./cm\(^2\) and \( 10^{16} \) molec./cm\(^2\), for the samples, loaded in IVG.1M and RA reactors, respectively, an assumption was made about the different degree of surface fracturing for these samples. The sample surface fractures, probably, during the temperature increasing or decreasing, as a result of BeO film cracking on the surface. If to use a widely used simplified expression, having received in complete form in [9] for the estimation of stress, appearing in oxide film on the sample surface, it is possible to write down:

\[
\sigma = \frac{(\alpha_{BeO} - \alpha_{Be}) \times \Delta T \times E_{BeO} \times E_{Be}}{(1 - \mu_{BeO})E_{Be} - \frac{h_{BeO}}{h_{Be}}(1 - \mu_{Be})E_{BeO}}
\]

where \( \sigma \) – stress, arising in BeO when the temperature changes on \( \Delta T \), \( \mu_{BeO}, \mu_{Be} \) and \( \alpha_{BeO}, \alpha_{Be} \) – Poisson’s ratios and thermal expansion coefficients for BeO and Be, respectively, \( E_{BeO}, E_{Be} \) – their Young’s modulus and \( h_{BeO}, h_{Be} \) – the thickness of oxide and the radius of beryllium sample, respectively.

Though our samples have the cylindrical form, but, taking into account that \( h_{BeO}h_{Be} << 0.1 \), it is possible to neglect by radial component of stress, i.e. not to take into account the bend of the surface and to use the expression (3).

After carrying out estimations with the values: \( \alpha_{Be}=1.6 \times 10^{-3} \) 1/K, \( \alpha_{BeO}=9.4 \times 10^{-6} \) 1/K, \( \mu_{BeO}=0.18 \), \( \mu_{Be}=0.07 \), \( E_{BeO}=375 \) GPa, \( E_{Be}=243 \) GPa; \( h_{BeO}=5 \) nm, \( h_{Be}=1.5 \times 10^{-3} \) m, it turns out, that already after the sample heating up to 313-393 K the compressing stress arises in the oxide film, \( \sigma=60-300 \) MPa, exceeding the strength of beryllium oxide, 40-190 MPa [10] at these temperatures. Hence, it is reasonable to assume, that during the sample heating up to our temperature of loading and during the decreasing of temperature, the surface oxide film formed under these conditions, was cracked. Thus, depending on the conditions, in which the oxide film was formed, its structure, mechanical properties and, respectively, the character of its destruction changes. It results in a
Table 5. Irradiation parameters of the reactors used for experiments

<table>
<thead>
<tr>
<th>Irradiation parameters</th>
<th>IVG.1M</th>
<th>RA</th>
</tr>
</thead>
<tbody>
<tr>
<td>Heat power, MW</td>
<td>6.0</td>
<td>0.5</td>
</tr>
<tr>
<td>Neutron flux, n/cm²/sec</td>
<td></td>
<td></td>
</tr>
<tr>
<td>fast (0.1 MeV &lt; E &lt; 15 MeV)</td>
<td>2.2x10¹³</td>
<td>2.8x10¹³</td>
</tr>
<tr>
<td>thermal (E &lt; 0.67 eV)</td>
<td>1.0x10¹⁴</td>
<td>2.0x10¹²</td>
</tr>
<tr>
<td>γ-flux, photons/cm²/sec</td>
<td>7.3x10¹³</td>
<td>6.1x10¹²</td>
</tr>
<tr>
<td>Experiment duration, hours</td>
<td>6</td>
<td>50</td>
</tr>
<tr>
<td>Neutron fluence per experiment, n/cm²</td>
<td></td>
<td></td>
</tr>
<tr>
<td>fast (0.1 MeV &lt; E &lt; 15 MeV)</td>
<td>4.8x10¹⁷</td>
<td>5.0x10¹⁸</td>
</tr>
<tr>
<td>thermal (E &lt; 0.67 eV)</td>
<td>2.2x10¹⁸</td>
<td>3.6x10¹⁷</td>
</tr>
<tr>
<td>γ-fluence per experiment, photons/cm²</td>
<td>1.6x10¹⁸</td>
<td>1.1x10¹⁹</td>
</tr>
</tbody>
</table>

Various degree of the surface fracturing for the samples, loaded in fission reactors with various spectra of radiation. This assumption, initially based on the data of TDS experiments, then was confirmed by the results of electron microscopy and X-ray structural microanalysis, which have shown the active growth of oxide layer and the formation of cracks on the surfaces of beryllium samples, irradiated in hydrogen in IVG.1M reactor.

From the data obtained one can make a conclusion, that during the loading of beryllium samples with hydrogen in IVG.1M reactor the oxide film grows with the gas concentration exceeding equilibrium one that does not occur during the loading in RA reactor or without irradiation. The mechanisms of this phenomenon are not quite clear, as since the number of defects, produced per unit of time in unit of volume is approximately identical in both reactors (1.8x10⁸ dpa/sec and 1.6x10⁸ dpa/sec), fast neutrons fluence and, respectively, total number of dpa is greater for RA reactor, the only one difference of irradiation characteristics for these reactors - the difference in the fluxes of γ-photons and thermal neutrons, see Table 5. This part of radiation spectrum, according to a literary data, is responsible for formation of so called freely-migrating defects [11], which, obviously, are of great importance for the formation of oxide film with high concentration of hydrogen. During the irradiation of solids a large part of vacancies and interstitial atoms, which are generated by particles with high energy, either annihilate, via recombinations with near-by defects of an opposite type, or become immobilized upon absorption into a cluster of similar defects. As a result only a fraction of the total number of defects, that are generated by irradiation, become free and is able to migrate over substantial distances, i.e. distances, exceeding the size of the primary cascade. Several important changes of properties of materials, occurring in reactor irradiation conditions, are connected with this fraction of total defect population. For example they are responsible for void swelling, radiation induced segregation, radiation enhanced diffusion, and radiation embrittlement.

Carried out experiments have shown, that the influence of reactor radiation on the process of molecular hydrogen (deuterium) interaction with beryllium results in creation of anomalous high concentration of hydrogen in a surface film of beryllium oxide for researched samples. To answer the question about the significance of this effect in respect to ITER conditions, it is necessary to know the behaviour of beryllium oxide film on the surface of the first wall, which, besides γ and neutron loads, is exposed to the fluxes of DT neutrals and is actively sputtered. Therefore it seems pertinent, within the framework of present work, to investigate the changes of beryllium surface element composition as a result of its interaction with hydrogen ions with fluxes, comparable to fluxes in ITER.

3. INTERACTION OF GLOWING DISCHARGE WITH BERYLLIUM SURFACE

As researched sample a disk of EHP-56 beryllium with diameter 20 mm and thickness 1 mm was used.
For creation of hydrogen ions flux of necessary intensity the glowing discharge was used. The changes of element composition of sample surface were monitored by the Auger electron spectroscopy technique using cylindrical mirror type of Auger electrons energy analyzer, without exposing sample to atmosphere.

Residual gases pressure in the chamber, where the sample was located, was about $5 \times 10^{-7}$ Pa before admittance of gases, being necessary for the glowing discharge ignition. Hydrogen, refined by passing through palladium-silver filter, and purified argon were used as working gases.

Fig. 7. Auger-spectrum of the sample surface, obtained after 24 hours of a sample exposure to the $H_2$ plasma of glowing discharge. Concentration of carbon on the surface is 100%.

The Auger-spectra were decoded using the atlas of Auger-spectra [12], the concentration of carbon on the sample surface was calculated by the method of the pure standards with the matrix amendments according to technique, suggested at the same atlas. Auger-spectrum of beryllium surface sample, exposed to plasma of hydrogen glowing discharge for 24 hours at $I=50$ mA/cm$^2$, $U=300$V, $P(H_2)=400$ Pa, $T=300$K is presented on fig. 7. For this time a layer about 20 microns was sputtered. As the exposition of the sample in $H_2$ glowing discharge has not resulted in removing oxide film from the sample surface, but resulted in covering it with carbon, the sample was exposed to glowing discharge with Ar as a working gas.

Fig 8 shows an Auger-spectrum of the sample surface after its exposure to Ar glowing discharge plasma at the following parameters of discharge - $I=50$ mA, sample temperature 300K, $U=200$V, $P(Ar)=400$ Pa. The exposition of the sample to the glowing discharge was performed for 24 hours. For this time a layer about 40 microns was sputtered.

It is clearly seen, that the peak of oxygen on a Auger-spectrum is not observed, there is small peak of carbon (272 eV), which concentration on a surface is equal 5.5% and the peak of beryllium (105 eV), not bound in BeO.

After sample heating the element composition of sample surface changes, the peak of oxygen (502 eV) appears at the temperature 873K, the intensity of carbon peak decreases (concentration 4%). The kinetics of process can be observed on the Auger-spectra of sample surface, obtained at the temperatures 300, 473, 673 and 973K (Fig. 9). It is seen, that already at the temperature of a sample 473K beryllium oxidation occurs, the shape of Be Auger peak changes and the shift of the bottom maximum down to 95 eV is observed. The weak reduction of C concentration on the surface is observed only at the temperature 973K. Partial pressure of oxygen-containing gases in the chamber during the experiment did not exceed $5 \times 10^{-7}$ Pa.

To confirm the results of the previous experiment, one more experiment on the sample exposure to plasma of hydrogen glowing discharge...
was carried out at the sample temperature 300 K and the same discharge parameters \((P=400 \text{ Pa}, U=300 \text{ V}, I=50 \text{ mA}, t=24 \text{ hour})\). Auger-spectrum of the surface after such treatment is shown on Fig. 10.

The enrichment of a sample surface with carbon occurs in a much less degree (concentration 14.8%), in comparison with the previous experiments. It is seen, that beryllium on the surface is bound in the oxide BeO (Auger-peak at 95 eV) and there is the peak of oxygen (502 eV). The sample heating up to 873K results in reduction of carbon peak intensity on the sample surface.

One of the basic results obtained is the fact, that in the conditions of beryllium sample exposure to hydrogen plasma of glowing discharge it was not possible to obtain the sample surface free of oxide film BeO despite observed sample sputtering. Partial pressure of oxygen-containing gases did not exceed \(5 \times 10^{-7} \text{ Pa}\) before the admittance of hydrogen inside the chamber of experimental installation. Besides, the conditions of sample exposition to \(H_2\) plasma were identical to the conditions of Ar glowing discharge. However in the first case the oxide is removed, in the second it is not removed. For explanation of such effect it is necessary to allow the existence of preferential beryllium atoms sputtering in comparison with atoms of oxygen in case of interaction with light hydrogen ions. This effect should not be observed in case of interaction BeO with heavy Ar ions.

The second essential result obtained in the carried out experiments is observable enrichment of the sample surface with carbon after its exposure to hydrogen glowing discharge. This is not observed while realizing the experiments in argon glowing discharge plasma. In the last case the initial vacuum conditions before the gas admittance into the chamber were the same.

For interaction of nonequilibrium hydrogen with beryllium at the fluences, considerably smaller, than for these experiments, different authors observed significant development and cracking of beryllium samples surfaces. Under the conditions of our experiments realization, the fracturing and cracking of the surface should occur rather quickly and already at the first experiments with hydrogen plasma it is significant and increases with the subsequent experiments. Thus uncovered atoms of carbon in cracks are binding with hydrogen atoms and then, as a result of surface diffusion release on "visible" surface, available for control by means of Auger analysis. One can confirm this assumption of rather fast fracturing of beryllium sample surface by specific estimations.

It is known, that when implantation fluence reaches a level \(10^{15}-10^{23} \text{ atom/m}^2\) (in our experiments it occurs in a few minutes) then comes
saturation of retained gas in some part of implanted layer and almost in all metals drastic change of surface condition is observed. Blisters and pores arise on a surface. In the works [13, 14] the authors observed cracks in beryllium after its implantation with H⁺ with energy 33 keV, at room temperature up to the fluences, specified above. In our experiments the energy of ions is much lower, ≈300 eV, however, it is possible to show, that blistering is possible in our case. To have blistering, cracking and etc., two main conditions should be provided [15]:
1. In solids critical concentration $C_{cr}$ of implanted gas should be achieved: $0.3 < C_{cr} < 1$ atom of gas per atom of target. As the maximum concentration is limited by reflection of particles and surface approaching as a result of sputtering, the phenomenon of blistering type are observed under the condition:

$$\frac{1 - R_N}{S} \geq C_{cr}$$

(4)

Where $R_N$ – factor of reflection, and $S$ – sputtering factor. The ratio $(1-R_N)/S$ is the maximum concentration of implanted atoms, which can be achieved in the absence of saturation effects.

2. The specified critical concentration should be at first achieved at the distance not less than several tens of nm from surface. Blistering and cracking are suppressed, if $C_{cr}$ is reached at first on a surface and then is distributed in the bulk of a sample.

Owing to sputtering the maximum of implantation profiles can move to surface yet before the achievement of $C_{cr}$. In case of free paths profile having the form of gaussian, neglecting by diffusion and considering factor $S$ to be not dependent on fluence, such displacement will take place under the following condition [16]

$$C_{cr} \geq \frac{1}{S} \text{erf} \left( \frac{R_P}{\sqrt{2} \sigma_R} \right)$$

(5)

At high implantation energies we have $R_P/\sigma_R > 3$ and the conditions (4) and (5) are equivalent, while at low energies the condition (5) appears to be more strict. For ions of hydrogen the right part of an inequality (5) is more than $C_{cr}$ at all ion energies $E_p > 300-500$ eV. For the ions of heavy gases, for example, Ar, it is close to 1 at $E \leq 100$ keV and exceeds 1 at higher energies of ions. Therefore the blistering at the bombardment by heavy ions (Ar, N, Ne) was observed by the researchers only at energy $E_p > 100$ keV. It is obvious, that during the realization of our experiments at the bombardment by Ar ions neither blistering no cracking are observed, so carbon atoms in pores and cracks are not able to be uncovered and, hence, they cannot achieve, in the result of enhanced surface diffusion, the surface, available for Auger analysis control, for reasonable times of experiments carrying out. Besides this the complexes are sputtered faster on the visible surface of a sample by heavy ions Ar⁺ in comparison with light H⁺.

CONCLUSIONS

Analysis of earlier obtained TDS spectra for samples, loaded in H₂ and D₂ at $T=1150$K, $P=10^5$ Pa in two different fission reactors, with different spectra of radiation has been carried out. Activation energies of gas release were calculated, assuming, that the first five peaks can be described by second order desorption, and the sixth peak - by bulk diffusion.

Assumption was made, that in the conditions of irradiation in IVG.1M reactor oxide surface film grows in the process of irradiation with hydrogen concentrations, considerably exceeding equilibrium ones.

It is shown, that the observed development of sample surface can be explained by cracking of oxide film because of internal stresses arising in it at temperature changes of few tens degrees. Thus the conditions of oxide film growth can determine the concentration of hydrogen inside it and, accordingly, the character of its destruction.

Assumption was made, that γ-radiation and thermal neutrons effects were responsible for observed effects.

Experiments on beryllium sample exposition to hydrogen and argon glowing discharge plasma were carried out.

It is shown, that at the flux density of H⁺ about $10^{30}$ atoms/m²sec and ion energy ≈300 eV the situation was realized, when the sputtering of beryllium sample surface occurred with
conservation of oxide film on the surface at vacuum conditions, being typical for glowing discharge functioning. At the same vacuum conditions and the same fluxes of Ar ions oxide film can be easily removed at the sample temperature 300 K.

Acknowledgements
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A possibility of the correct determination of deuterium solubility and diffusivity in Be on the basis of degassing experiments is demonstrated. It has been found that the main fraction (above 90 %) of deuterium retained under D₂ exposure is removed under slight electropolishing (descaling of ~ 2-5 μm) of the samples before TDS measurement. This deuterium seems to be located in the near surface oxide layers formed during the exposure as a result of interaction of beryllium with oxygen containing molecules of residual gas. In all degassing runs the diffusion of deuterium in the bulk of beryllium samples was not a limited-stage of gas release.

1. INTRODUCTION

The interaction of beryllium with hydrogen is of great interest with regard to safety requirements and fuel balance. Some codes are available for estimations of tritium inventory in fusion reactor components, permeation into the coolant and ejection in case of accidents [1,2]. Nevertheless, there is a lack of consistent data on hydrogen solubility, diffusion and interaction with defects which gives no grounds for reliable prognosis.

Analysis of degassing kinetics of specimens saturated with hydrogen isotope is a well-established procedure which allows to derive solubility and diffusion constants provided that diffusion is a rate-determined factor of gas release [3]. In the initial phase of the annealing, degassing may be diffusion-limited if ζ > > 1, where

\[ \zeta = K_r c_o / D, \]

where \( K_r \) is recombination coefficient for second order surface reaction and \( D \) is diffusion coefficient. The sample is assumed to be a slab of thickness \( l \) with an initial uniform concentration of \( c_o \). In diffusion-limited regime for times \( t > 0.04 l^2 / D \) degassing rate \( R \) of a sample is governed with sufficient accuracy by the equation:

\[ R = \frac{8 c_o D}{l} \exp\left( -\frac{t}{\tau_d} \right), \]

where \( \tau_d \) is a degassing constant

\[ \tau_d = \frac{l^2}{\pi^2 D}. \]

Hence \( D \) and \( c_o \) can be derived by fitting Eq. (2) to the exponential portion of the measured degassing curve. Obviously, firm conclusion about a rate-determined factor of degassing process is available only from accurate measurements of desorption rate during prolonged degassing run [3]. This is not possible in all cases. A further procedure is that Eq. (2) is fitted to the degassing curves measured on the specimens of different sizes. In the diffusion-limited regime the parameter \( \tau_d \) varies quadratically with the sample thickness. Surface processes must be taken into account when the thickness dependence of \( \tau_d \) appears to be weaker than quadratic one.

In present study this procedure has been applied to investigate deuterium behavior in beryllium loaded with deuterium by heating in D₂ atmosphere.
2. EXPERIMENTAL

Beryllium grades S-65B (Brush Wellman) and beryllium refined by zone melting (ZM) produced by Kharkov Institute of Physics and Technology, Ukraine were used. According to supplier’s information ZM material is characterized by an extremely low oxygen content, and namely, less than 0.005 wt%.

The samples of different thickness were used to clarify either diffusion or surface processes are rate-determined factors of gas release. The samples from S-65B were disks 10 mm in diameter. For the making plate-like specimens from ZM beryllium a bar with dimensions 6.0x7.5x45 mm$^3$ was cut perpendicular to its long axis. Prior deuterium loading the samples were mechanically and electrochemically polished. The examination of the specimens by means of EPMA revealed that oxygen content in the near-surface layer was about $5 \times 10^{20}$ O/m$^2$.

D$_2$ exposure of the samples was performed as follows. Aluminum tube heated by radiant furnace with the sample inside was set in vacuum chamber equipped with liquid nitrogen trap to collect water vapor. Before deuterium admission the chamber was evacuated to pressure of $-10^{-4}$ Pa. Air leakage into vacuum chamber lead to increase in oxygen pressure to ~ 0.1 Pa for 2 hours. Eight samples were exposed to D$_2$ pressure of 10 kPa at temperature of 920 K for 2 hours. Oxygen content in the near-surface layer of samples was proved to be increased during this procedure up to ~ $6 \times 10^{21}$ O/m$^2$. One-half of the samples loaded were then electropolished to remove 5 - 10 μm thick surface layer.

TDS measurements were conducted in the special UHV system with a background pressure of $10^{-6}$ Pa. The samples were heated by electron bombardment at a constant input power. Temperature of the samples was measured with an optical pyrometer. Deuterium release in the form of D$_2$ and HD molecules was monitored by quadrupole mass spectrometer (QMS). The sensitivity of QMS for D$_2$ and HD signals was evaluated from calibrated $^4$He and $^3$He leaks, respectively.

3. RESULTS AND DISCUSSION

Deuterium release from as-loaded ZM samples are shown in Fig. 1 a. As seen the degassing (as a total deuterium release in the form of D$_2$ and HD molecules) constants of the samples of different thickness agree very closely, suggesting that the degassing rate is limited by an evolution of deuterium retained in a near surface layer, not in the sample bulk.

![Figure 1](image-url)  
*Figure 1. Thermal desorption curves of D$_2$ (filled symbols) and HD (open symbols) molecules and degassing curves (crosses) of ZM samples of different thicknesses exposed to D$_2$ gas at pressure of 10 kPa at 920 K for 2 hours: (a) as-loaded samples, (b) followed by electropolishing to remove 5 - 10 μm thick surface layer. The value at the beginning of fitting line is the temperature of isothermal annealing.*
Table 1
Average deuterium concentration in the bulk of beryllium exposed to D₂ atmosphere.

<table>
<thead>
<tr>
<th>Material</th>
<th>Sample thickness mm</th>
<th>Degassing constant s</th>
<th>Deuterium concentration appm</th>
</tr>
</thead>
<tbody>
<tr>
<td>S-65B</td>
<td>0.90</td>
<td>36</td>
<td>1.0 ± 0.2</td>
</tr>
<tr>
<td></td>
<td>0.15</td>
<td>11</td>
<td>5.0 ± 1.0</td>
</tr>
<tr>
<td></td>
<td>0.88</td>
<td>26</td>
<td>0.70 ± 0.15</td>
</tr>
<tr>
<td>ZM</td>
<td>0.45</td>
<td>18</td>
<td>0.90 ± 0.20</td>
</tr>
</tbody>
</table>

Figure 2. Thermal desorption curves of D₂ (filled symbols) and HD (open symbols) molecules and degassing curves (crosses) of S-65B samples of different thicknesses exposed to D₂ gas at pressure of 10 kPa at 920 K for 2 hours followed by electropolishing to remove 5-10 μm thick surface layer. The value at the beginning of fitting line is the temperature of isothermal annealing.

Degassing curves of the samples polished after loading are presented in Figs. 1 b. The lines show the results of fitting of Eq. (2) to the exponential portion of the degassing curves. Two points are noteworthy. First, desorption rate of D₂ and HD molecules from the samples is one order of magnitude less than from as-loaded ones. That is to say that the majority of deuterium retained in the samples under loading is located in the near-surface layers of 5 - 10 μm in thickness or lesser.

Second, the degassing constants depend markedly on the sample thickness. This indicates that the degassing rate is limited by an evolution of deuterium from the sample bulk. Qualitatively similar results has been obtained for S-65B beryllium. Corresponding degassing curves of polished samples are shown in Fig. 2. Average deuterium concentrations in the polished samples calculated by integrating D₂ and HD spectra are collected in Table 1. The error in the value of concentration was mainly due to variations in QMS sensitivity in the course of degassing run. For ZM beryllium, concentrations in the samples of both thicknesses agree within experimental uncertainties suggesting that deuterium-beryllium equilibrium has been reached during 2 hours exposure. This result is in good agreement with deuterium permeation transients measured by Abramov et al. [4]. As to S-65B beryllium, corresponding concentrations differ considerably indicating that deuterium is distributed nonuniformly throughout the specimen thickness. One possible reason of difference between ZM and S-65B beryllium is that BeO inclusions presented in the later contribute significantly to the deuterium accumulation.

As seen from the Table, thickness dependence of \( r_2 \) appears to be weaker than quadratic one. Therefore the diffusion of deuterium in the bulk of
beryllium samples is not a limited-stage of gas release at all.

4. CONCLUSIONS AND FUTURE PLANS

It has been found that the majority of deuterium retained in the samples during D₂ exposure is located in the near-surface oxide layers. In all degassing runs the diffusion of deuterium in the bulk of beryllium samples is not a limited-stage of gas release.

Of some interest is the fact that the main fraction of deuterium retained under D₂ exposure is removed under slight electropolishing. Having in mind extremely low hydrogen solubility in beryllium this deuterium seems to be located in the near surface oxide layers formed during the exposure as a result of interaction of beryllium with oxygen containing molecules of residual gas. This can lead to an apparent increase of deuterium solubility in beryllium. Hence, a more detailed study of hydrogen retention in the near-surface oxide layers is required. Also the data on thermal stability of hydrogen loaded beryllium oxide are of great importance for the assessments of tritium permeation/inventory in PFC and mobilization of tritium retained in codeposited layers. Corresponding study is in progress [5].

In relation to hydrogen diffusivity in beryllium, a diffusion-limited regime of the degassing should be firstly obtained. This can be done using large samples and/or providing for a clean surface in the course of degassing run. We plan to remove the surface oxide by in-situ Ar ions sputtering.

ACKNOWLEDGMENTS

The authors would like to thank Dr. R.Kh. Zalavutdinov for providing EPMA measurements. This work was supported by the International Atomic Energy Agency under Contract RUS 8889.

REFERENCES

6.5 Effect of Deposited Tungsten on Deuterium Accumulation in Beryllium in Contact with Atomic Deuterium

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*Moscow State University, Moscow

Usually ion or plasma beams are used in experiments with beryllium which simulates the interaction of plasma with first wall in fusion devices. However, the using of thermal or subthermal atoms of hydrogen isotopes seems to be useful for that because the flux of the neutral charge-exchange atoms composes a significant part of particle flux which interacts with different elements of discharge chamber in real fusion devices. Moreover, the application of deuterium thermal atoms proved to be more fruitful when study the influence of surface state on the deuterium retention and release processes in comparison with energetical ions which pass through the surface layers and do not "feel" the defects or impurities at surface.

Recently, we have studied the deuterium accumulation in beryllium in contact with atomic deuterium. A three-electrode ion-plasma source with heated cathode on the base of dc glow discharge was used to produce deuterium atoms. In Figure 1 the scheme of experimental setup is given. The tungsten cathode is located inside the ionization chamber and it is not in direct view of other parts of source. Cathode is ohmically heated up to high temperature to increase the efficiency of source work at deuterium pressure of 5-10^{-1} Pa. When the potential is applied to the anode and cathode the plasma column is formed in the space inside the plasma chamber. Normally \( V_{CA} = 50-100 \text{ V}, I_{CA} = 0.1-4 \text{ Amps} \). Be sample is assembled on heating holder in front of the plasma column. In experiments with atomic deuterium the positive potential of +80 V was applied to the holder to screen the sample from positive D ions.

Therefore, the sample was mainly exposed to atomic deuterium and electron fluxes. Besides, molecular deuterium at pressure 5-10^{-1} Pa (deuterium pressure in the chamber during the source operation) interacted also with sample. The thermal atoms flux was approximately \( (1-2) \times 10^{16} \text{ cm}^{-2}\text{s}^{-1} \) which was estimated using so-called "Langmuir effect" [1]. The base vacuum in chamber was 1.33 \times 10^{-4} \text{ Pa}. Beryllium TIP-30 manufactured in Bochvar Institute by hot isostatic pressing was used.

![Figure 1. Scheme of experimental setup.](image-url)
The deuterium distribution depth and concentration depend on the exposure time and after exposure time of 240 min they reach the values of 400 nm and \((8-9) \times 10^{21} \text{ D/cm}^2\), respectively (Figure 2).

![Figure 2. Depth profiles of deuterium concentration in TIP-30 exposed to atoms with flux of \(10^{16} \text{ D/cm}^2\) for different exposure times.](image)

The deuterium content \(N_0\) growth in dependence on the exposure time \(\tau_{\text{exp}}\) occurs as \(N_0 \sim \sqrt{\tau_{\text{exp}}}\) that points out the diffusion character of deuterium accumulation process.

During the atomic deuterium exposure the oxide film grows on the plasma-facing side of sample at growth rate of 0.4-0.5 nm/min and after exposure time of 4 hours the thickness reaches value of 125 nm. On the back side of the sample, which was not affected by the atomic deuterium, the thickness of the initial oxide film was changed insignificantly.

To understand the mechanism of deuterium trapping the experiments with D atoms were performed using Secondary Ion Mass Spectrometry (SIMS) and Residual Gas Analysis (RGA) methods [3]. This work was done in collaboration with Dr. V. Alimov. SIMS and RGA were used for measurements of \(H^+\) and \(D^+\) secondary ion yields and partial pressures of \(D_2\) and HD molecules in the course of sputtering of the surface with 4 keV \(\text{Ar}^+\) ions. It was found after comparison of depth profiles of deuterium and oxygen that deuterium is mainly accumulated in oxide layer and is retained in two states - in atomic and in molecular forms. Deuterium content in beryllium oxide proved to decrease if sample was kept in vacuum or air at room temperature. This decreasing (dependent on the storage time) is shown in Figure 3 for the sample which was exposed to D atoms at temperature of 340K.

![Figure 3. Evolution of D atoms concentration (on the top) and \(D_2\) molecules concentration (on the bottom) in Be sample exposed to D atoms for 1 hour at 340 K in dependence on storage time in vacuum in room temperature.](image)

It is seen that this decreasing takes place for atomic part of trapped deuterium mainly. The molecular part of trapped deuterium was constant independent on the storage time within the experimental error.
The same results were obtained for the samples which were exposed to D atoms at 500 and 740K.

Analysis of these results gave the possibility to suggest that atomic part of trapped deuterium may be explained by deuterium retention in hydroxide which is formed in result of chemical reactions in oxide layer:

$$2\text{BeO} + 2\text{D} \rightarrow \text{Be(OD)}_2 + \text{Be} + 0.704 \text{ eV/D}$$

The possibility of this reaction was discussed earlier in [4]. This reaction may explain the fact of oxide layer growth on the sample side which was exposed to atomic deuterium. Really, according to this reaction the formation of hydroxide is accompanied by the release of free beryllium atoms. Then these beryllium atoms react with residual oxygen forming the growing oxide layer. These processes take place not only on the surface but in oxide beryllium bulk as well. That means that deuterium atoms diffuse into the sample bulk and free beryllium atoms (formed in bulk) migrate to surface. Thus, deuterium accumulation in beryllium oxide is determined by deuterium diffusion (the diffusion character of deuterium accumulation was mentioned earlier) and oxide film growth - by beryllium atoms diffusion through the film to surface.

The decreasing of retained deuterium content during Be sample storage in vacuum or air at room temperature is probably occurred in result of hydroxide decay. We calculated the kinetic constant of deuterium content decreasing in Be at room temperature. This value proved to be close to that which was extrapolated to room temperature from experimental data on the isothermal hydroxide decay at high temperatures [5]. This fact proves also the formation/decay of hydroxide during/after exposition to D atoms.

Molecular part of retained deuterium is stable and is not changed during sample storage at room temperature. Apparently, it is connected with trapping of deuterium atoms in bubbles which are formed on the base of oxide structure imperfections.

Thus, under D atoms exposure deuterium is mainly retained in surface oxide layer because thermal deuterium atoms penetrate into very short distance from the surface. Therefore, the surface conditions (impurities, defects and so on) may strongly influence on the deuterium retention and release.

It was mentioned earlier that ion-plasma source with heated tungsten cathode was used in our experiments. During the source operation tungsten atoms are evaporated and sputtered out of cathode surface and then deposited on different elements of plasma chamber. The Be sample is located far from cathode and it is not in direct visibility. Nevertheless, the part of evaporated and sputtered tungsten atoms can reach the sample and they are deposited on its surface. It was decided to investigate the influence of such tungsten deposits on deuterium accumulation in Be in contact with atomic deuterium.

Deuterium inventory and distribution in sample were measured by means of ERD and impurities on the sample surface were detected by Rutherford Back Scattering (RBS) techniques. ERD analysis were made using helium beams at 1.9 MeV energy. The depth resolution was about 20 nm. The minimal deuterium concentration which can be detected is approximately $5 \times 10^{-6}$ D/cm$^3$ or ~0.4 at%. RBS analysis was carried out using helium beam at the same energy.

RBS spectra of Be samples after different D atoms exposure times at 740K are given in Figures 4, 5 and 6. RBS spectrum of back side surface after 720 min exposure is shown in Figure 7. This side was not exposed to D atoms or plasma and therefore it may be considered as initial one. It is seen that tungsten content at exposed side increases when the exposure time grows. These data are given in Table.
Table

<table>
<thead>
<tr>
<th>$t_{\text{exp.}}$ min</th>
<th>90</th>
<th>240</th>
<th>720</th>
</tr>
</thead>
<tbody>
<tr>
<td>W content, at/cm$^2$</td>
<td>$5.6 \times 10^{14}$</td>
<td>$3.1 \times 10^{15}$</td>
<td>$1.2 \times 10^{16}$</td>
</tr>
</tbody>
</table>

Figure 4. RBS spectrum of Be surface after D atoms exposure for 90 min.

Figure 5. RBS spectrum of Be surface after D atoms exposure for 240 min.

Figure 6. RBS spectrum of Be surface after D atoms exposure for 720 min.

Figure 7. RBS spectrum of Be back side surface after D atoms exposure for 720 min.
It is possible to evaluate the tungsten atoms flux if one suggests that sticking coefficient of tungsten atoms to BeO surface is equal to 1. The flux calculated proved to be changed from $1 \times 10^{11}$ at/cm$^2$·s for exposure time 90 min to $3 \times 10^{11}$ at/cm$^2$·s for exposure time 720 min, i.e. five orders of magnitude less than flux of D atoms. Therefore for small exposure times it is possible to ignore tungsten contamination.

The lines of other elements are presented in the spectra as well. The amounts of carbon and phosphorus are small. These elements as well as chromium are probably appeared at the Be surface in result of preliminary electropolishing procedure. Carbon and phosphorus quantities don’t practically change during D atoms exposure. The content of chromium (and may be iron) increases insignificantly during exposition. The reason is probably the ion sputtering of plasma chamber wall which is made of stainless steel.

The deuterium content in these samples was measured by means of ERD. The deuterium profiles for exposure times of 90, 240 and 720 min are shown in Figure 8. When exposure time increased from 90 to 240 min deuterium content in Be grew from $6 \times 10^{16}$ at/cm$^2$ to $1.9 \times 10^{17}$ at/cm$^2$.

The depth of deuterium distribution in bulk increased as well. These features are similar to those described earlier [2]. But after exposure time of 720 min both deuterium content ($7 \times 10^{16}$ at/cm$^2$) and depth of deuterium distribution proved to be less than those after exposure time of 240 min. As it is seen from Table, the tungsten content in Be surface layer after exposure time of 720 min is higher by a factor of 4 in comparison with that after exposure time of 240 min.

We consider that tungsten deposit at Be surface is the reason of deuterium content decreasing in Be oxide. Taking into account that atom density at the Be surface is about $2 \times 10^{15}$ at/cm$^2$ the integral tungsten coverage is equivalent to near one monolayer in the sample exposed during 240 min and some monolayers after 720 min exposure. Therefore, it is suggested that tungsten changes the adsorption - desorption parameters of Be oxide surface. Hydrogen recombination rate for beryllium oxide is much less than that for tungsten. In [6] the recombination rate for beryllium oxide is described by following expression:

$$k_{r,BeO} \text{ (m}^4/\text{s}) = 1.58 \times 10^{-32} \exp(-57.5(kJ/mol)/RT)$$

Using this expression the recombination coefficient for 740K was calculated. It is equal to $2 \times 10^{-36}$ m$^4$/s. For tungsten the surface recombination coefficient for 735K is equal to $k_r = 1.0 \times 10^{-22}$ m$^4$/s [7]. So, deuterium recombination coefficient for tungsten surface is much higher than that for Be oxide and therefore the tungsten coverage decreases the deuterium solution in Be oxide. It is happened when tungsten coverage is equivalent to one monolayer or more. After that the deuterium supply into Be oxide is decreased. Deuterium content in oxide layer is determined by the equilibrium between deuterium supply from atomic phase and deuterium release out of sample in result of hydroxide decay. The decreasing of deuterium supply leads to reduction of deuterium content in oxide layer and therefore the deuterium content in the

![Figure 8. Depth profiles in Be exposed to D atoms at 740 K for different lengths of time.](image-url)
sample exposed during 720 min is less than that after 240 min exposure.

It is necessary to note one more fact. The oxygen content in Be sample exposed during 720 min is less than that after 240 min exposure by a factor of 2. It means that W coverage suppresses not only deuterium accumulation but oxide film growth as well. Probably, tungsten coverage detaines the exit of free beryllium atoms out of bulk at surface. During exposure to D atoms oxide film grows and therefore tungsten atoms don't cover the surface continuously but distribute in the growing oxide film. The important question is: how such amount of tungsten atoms can influence on the oxide film growth?

Thus, deposited at Be surface tungsten changes the quantitative parameters of deuterium accumulation in beryllium. It seems to us that this result is of great interest for fusion applications as both metals are considered as the advanced plasma-facing materials. During fusion devices operation the simultaneous sputtering of PFM will lead to formation of mixed layers and this may influence on the deuterium retention and release and change the deuterium inventory in plasma-facing materials and hydrogen isotopes balance in discharge chamber.

REFERENCES

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6.6 Modeling of Hydrogen Interactions with Beryllium

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In this paper, improved mathematical models are developed for hydrogen interactions with beryllium. This includes the saturation effect observed for high-flux implantation of ions from plasmas and retention of tritium produced from neutronic transmutations in beryllium. Use of the models developed is justified by showing how they can replicated experimental data using the TMAP4 tritium transport code.

1.0 INTRODUCTION

Not long after it was discovered that beryllium surfaces in a tokamak gave improved performance because of the low atomic number and good oxygen gettering capabilities, project management of the International Thermonuclear Experimental Reactor (ITER) decided that the plasma-facing surfaces should feature beryllium. Even though areas within the ITER divertor are now planned to be faced with tungsten or carbon fiber composite, plans are still to use beryllium for the plasma-facing surfaces of the first wall in the main plasma chamber.

A significant technical question regarding beryllium used in this way relates to its hydrogen recycling properties and in particular to the retention and release of tritium from beryllium surfaces in the event of an accident or during routine operations interruptions. Deuterium and tritium will be implanted from the plasma, and tritium will be bred by neutron transmutations in the beryllium. Efforts to measure hydrogen transport properties such as solubility, diffusivity, and the recombination coefficient resulted in widely differing values. This proved problematic for those trying to make reasonable estimates of tritium inventory and its vulnerability to support safety analyses for ITER. It became apparent that the usual models of tritium interaction with materials, especially the recombination model for return of hydrogen isotopes to the plasma, could not be counted on for beryllium surfaces under high plasma fluxes. Further, trap concentrations and energies associated with neutron transmutations were not well understood.

In response to the need for additional information on hydrogen interactions with beryllium, several experimental programs were begun to gain insight into the problem. One was in the Russian Federation where work at the Kurchatov Institute and at the Institute of Physical Chemistry of the Russian Academy of Sciences provided detailed information on structural changes that take place in beryllium as a result of hydrogen ion implantation. [1,2] Another experiment series was conducted using the Tritium Plasma Experiment (TPE) originally located at Sandia National Laboratory, Livermore but now housed at the Tritium Systems Test Assembly at Los Alamos National Laboratory.[3] There, samples of beryllium were exposed to intense fluxes of deuterium and tritium ions at elevated temperatures, representative of their exposure in ITER.

Other work on hydrogen interaction with beryllium was performed at McMaster University,[4,5] at the University of Toronto Institute for Aerospace Studies,[6] and at the Idaho National Engineering and Environmental Laboratory (INEEL).[7,8] These efforts combine to reveal an interesting and somewhat complicated combination of processes that must be considered when attempting to predict the response of beryllium to intense hydrogen plasma. Few tests have been conducted on hydrogen retention in neutron-irradiated beryllium, but experiments have been performed at Battelle Pacific Northwest Laboratory [9] and at the INEEL[7] on beryllium irradiated in the Advance Test Reactor in Idaho. Information on trap concentrations and energies can be extracted from those data. In this report, I explore observations made in these experiments and what they imply regarding hydrogen interactions with beryllium.

2.0 SURFACE INTERACTIONS

It has been known for several years that beryllium saturates with hydrogen under ion bombardment.[10,11] The work of Chernikov et al.[1] and of Guseva et al.[2] showed saturation of the surface by the implanting ions. Chernikov et al. showed graphic evidence of the formation and growth of bubbles during implantation of Russian TIP-30 beryllium with 3- and 10-keV deuterons at fluences ranging from 3 x 10^{20} to 8 x 10^{21} D/m^2. Sample temperatures were 300, 500, and 700 K. Guseva et al. exposed Russian TShP beryllium (98.7 wt.% Be, 0.9 wt.% O, 0.2 wt.% Fe) to 5-keV H ions with flux density 6.2 x 10^{21} H/m^2 s in the SAPPHIRE facility. They reported that elastic recoil detection (ERD) and secondary ion mass spectroscopy (SIMS) measurements of the hydrogen profile showed an unexpected reduction in the inventory of implanted hydrogen and a shift of the spatial distribution of that hydrogen toward the surface with increasing ion fluence. After a fluence of 2.3 x 10^{21} H/m^2, surface pits were observed to develop, mostly along grain boundaries. After exposure to 1.2 x 10^{24} H/m^2, erosion cones were beginning to form, and by 1.5 x 10^{25} H/m^2, the cones had grown to an advanced stage with the surface behind the cones taking on an amorphous appearance.

Additional insight was gained from the experiments on the TPE.[3] Five-cm diameter disks of hot-pressed and sintered S-65 beryllium, 2-mm thick were exposed to high fluxes of deuterium ions with a 3% tritium tracer. Plasma currents of 0.8 - 9.2 A gave fluxes from 2.5 x 10^{21} to 2.8 x 10^{22} (D+T)/m^2 s at energies of about 100 eV. Exposures lasted one hour with samples maintained at constant temperatures from 100 to 700°C. Following the implantation, the samples were cooled, transported through air to a separate outgasing furnace, and subjected to a thermal desorption tests in which the samples were thermally ramped in a flowing mixture of helium and 1% hydrogen. Gases passed through an ionization chamber and an oxidizing catalyst bed into ethylene glycol bubblers. Temperatures were increased from room temperature to 800°C at a constant rate of 20°C/min. Tritium retention was measured by counting the tritium activity in the bubblers, and overall hydrogen content was estimated by extrapolation using the known D/T ratio. Post-experiment observation of the samples under a scanning electron microscope revealed that the surface was “hairy” from formation of may slender and closely spaced sputter-erosion cones.

INEEL experiments[8] conducted in the early 1990s were characterized by implantation fluxes of 5 - 6 x 10^{19} D/m^2 s at 1 keV/D and sample temperature of the order of 450°C. The samples were 25 - 71 μm thick foils. The upstream (plasma-side) surfaces of the targets were found to have severe damage and pitting to a depth of 1 μm following the implantation. Permeation was observed to take much longer and to be of correspondingly lower rate than was expected.

Experiments by Haasz and Davis[6] implanted 0.13-mm thick beryllium targets at room temperature with 1-keV deuterons at fluences ranging from 10^{21} to 10^{25} D/m^2 and...
measured deuterium retention by thermal desorption spectroscopy. They saw retained deuterium increase with fluence up to about $10^{22}$ D/m$^2$ after which there was little or no further increase in retention with increasing fluence.

2.1 General Characteristics

Observations from these and other similar experiments lead to the following general characteristics for hydrogen retention of implanted hydrogen in beryllium.

1. Hydrogen isotopes exist in both molecular and atomic form in ion-implanted beryllium and in neutron-irradiated beryllium.[1] Much if not most hydrogen is molecular, appearing to reside in bubbles. Some exists as single atoms, probably attached to free beryllium surfaces or to BeO or other impurities.

2. Hydrogen saturates in beryllium surfaces reaching a maximum concentration that is temperature dependent but essentially independent of ion flux or further fluence.[1,3] Concentration of both molecular and atomic forms grows from the implantation zone to the surface, possibly a consequence of erosion of the surface. Recombination or re-emission occurs very quickly once the surface reaches saturation concentration.

3. Gas pressure in the bubbles that form is very close to equilibrium values.[1,12]

4. There is a damage region much deeper than the implantation zone where traps are more numerous than they are in the remaining bulk of the beryllium.[1,8] The extent of the region may correlate with implanting ion energy. The process may also be linked to hydrogen precipitation at bubble nucleation sites.

5. Films of plasma contaminants, even at very low concentrations in the plasma, can form on the plasma-facing surfaces at all operating pressures. In many experiments involving hydrogen transport in beryllium, the rate limiting processes are associated with surface films.[4,13]

6. Diffusivities for mobile atoms in Brush Wellman S-65 and similar grades of beryllium appear to fit those given by Abramov for high-grade beryllium.[14] Disparate values in measurements may be ascribed to the effects of surface films or trapping.[15] In the implantation zone, the effective diffusivity is higher because of the damage.

7. At the high ion fluxes of the TPE experiments and in ITER divertors, surface erosion must be taken into account in modeling.

2.2 Modeling Transport Processes

To model tritium inventory, permeation, and release, it is usual to use one of the tritium transport codes available. TMAP4,[16] DIFFUSE,[17] and PIDAT[18] are representative transport codes that model processes of implantation, retention and re-emission of tritium from materials. They are all one-dimensional with respect to tritium transport through solids and use the same equations for most transport processes. For implantation of hydrogen isotopes into plasma-facing materials under ITER-like conditions it is appropriate to consider three zones in series: the implantation zone with a thickness of a little more than the implantation depth (typically a few nanometers), the damage zone where the majority of the bubble traps reside, and the bulk or the rest of the material layer. Some useful techniques have been found that allow good fits to be obtained to experiments where saturation is evident.

2.2.1 Diffusion With Erosion

The equation for diffusive flux in the codes mentioned is

$$J_i = -D \left( \nabla n_i + \frac{n_i Q^*}{kT^2} \nabla T \right)$$  \(1)$$

where $D$ is the diffusivity, $n_i$ is the concentration of atoms of species $i$, $k$ is Boltzmann’s constant, $T$ is temperature, and the second term in parentheses accounts for the Ludwig-Soret effect (mass transport due to a temperature gradient) characterized by the heat of transport $Q^*$. When erosion is taking place, the face of the eroding surface is receding at a rate $u$ (m/s) away from the plasma-exposed face, and it is
convenient to assume that all near-surface transport processes are taking place in a coordinate system moving with velocity \( u \) through the medium. In that case the diffusion equation becomes

\[
J_i = -D \left( \nabla n_i + \frac{n_i Q^*}{kT^2} \right) - un_i \tag{2}
\]

assuming that the positive direction for diffusion is away from the plasma-exposed face. For convenience in avoiding recoding of the computer models, one can let

\[
\frac{Q^*}{k} = \frac{Q''}{k} + \frac{uT^2}{DVT} \tag{3}
\]

where \( Q'' \) is the ordinary heat of transport (often neglected) and where the second term uses values of \( u, D, T, \) and \( VT \) from separate analyses or input equations, depending on the code used. For example, in TMAP4, running the code without concern for hydrogen transport will allow the local values of \( VT \) to be established, \( D \) is available as a temperature-dependent input equation, and \( T \) is an available state variable from the code.

### 2.2.2 Recombination

Embedded hydrogen atoms near the surface are being removed along with the eroded matrix atoms. For a solution-law diffusive boundary condition, the flux of atoms from the surface is calculated using Eq.(2). If the surface is recombination limited, the flux of hydrogen atoms to the surface is

\[
J_i = -(2K_r n_i^2) \tag{4}
\]

where \( K_r \) is the recombination coefficient and \( n_i \) is the concentration of mobile hydrogen atoms of species \( i \) at the surface. The negative sign denotes that the recombination flux is in the direction toward the plasma for positive \( u \) away from the plasma. As the surface material becomes saturated with hydrogen isotope atoms, a path must be opened for their rapid return to the plasma, effectively bypassing the rate-limiting processes of diffusion from the implantation depth to the surface and recombination at the surface. A change can be made in \( K_r \) to accommodate both erosion and saturation effects. This is accomplished by allowing the recombination coefficient to grow exponentially when the surface concentration reaches the saturation level. The resulting equation for \( K_r \) is

\[
K_r = \left[ 3.4 \times 10^{-29} \exp \left( \frac{-0.28 \text{ eV}}{kT} \right) \right] \times \frac{1 + \exp \left( \frac{10n_i}{n_{sat} - 10} \right)}{1 + n_i} + \frac{u}{n_{sat}} \tag{5}
\]

Here the first term in brackets is the Hsu-Andrew-Causey[19] value for the recombination coefficient obtained at low fluxes. It is important that this functional dependence be retained for short-term responses under ITER-like conditions or for low-flux operation. The term in the second set of brackets accomplishes the exponential growth at saturation, thought the abruptness factor, 10, is somewhat arbitrary. The last term accommodates loss of hydrogen from the surface by surface erosion.

The saturation concentration is a key in this analysis. It is obtained by assuming that the mobile atom concentration in the matrix will never be greater than that corresponding through Sieverts' law to gas pressure in the bubbles and that the gas pressure will never exceed the yield strength of the material. Hence, \( n_{sat} \) is the product of the square root of the material yield stress and the solubility (Sieverts' law with yield stress substituted for gas pressure). Using the Shapovalov and Dukel'ski solubility,[20] and yield stress fit from data of Henshall et al.[12] that becomes

\[
n_{sat} = \frac{4.079 \times 10^{26}}{1 + 0.0012 + \left( \frac{T}{950} \right)^{16/2}} \times \exp \left( \frac{-0.17 \text{ eV}}{kT} \right) \left( \frac{\text{atom}}{\text{m}^3} \right) \tag{6}
\]
With these changes or adaptations, the conventional tritium transport codes can be used successfully in situations where saturation in beryllium becomes noticeable.

2.2.3 Simplified Model

For inventory purposes, the quantity of hydrogen isotopes stored in the thin implantation zone can be neglected in comparison with what will be found in the damage zone and in the bulk of the beryllium. Under ITER-like conditions, saturation will be achieved in only a few seconds of operation, so there is a motivation to look for a simpler way of performing the calculation that does not rely on the recombination coefficient. Following the notion that the mobile atom concentration near the surface will reach some saturation value, \(n_{\text{sat}}\), almost immediately, one may model the plasma-facing surface as a constant concentration boundary if the appropriate concentration can be found. Ordinarily one would consider the value

\[
n_i = \frac{\delta \phi}{D} \tag{7}
\]

where \(\delta\) is the implantation flux and \(D\) is the implantation depth. This is the conventional zero-surface-concentration boundary condition. This gives only moderately good results because of the much more rapid diffusivity in the implantation zone caused by the lattice disturbances there. It overpredicts retained inventories by roughly factors of 3-4. An empirically derived expression that gives excellent agreement with experiments at reasonable temperatures (below 700°C) is

\[
n_i = \frac{S_y}{3} (\delta \phi)^{2/3} \tag{8}
\]

The motivation for this expression is the observation that the temperature sensitivity of the retained inventory in several experiments is essentially controlled by the material yield strength, but implantation flux and implantation depth do play some role in the process.

2.2.4 Comparison of Theory with Experiments

2.2.4.1 Tritium Plasma Experiment

The code adaptations described above were used with the TMAP4 code to model experiments conducted using the TPE at Los Alamos National Laboratory described previously above.[3] Figure 1 shows the distribution of experimental results of hydrogen inventory released during thermal desorption and the corresponding calculations using TMAP4 simulating the three connected zones of implantation, damage, and bulk with the erosion and recombination modifications given above.

These results were obtained by assuming a triangular implantation distribution 3 nm deep that peaked at 2 nm, consistent with results from TRIM calculations for these implantation conditions. In this layer were assumed a trap concentration of 0.15 atom fraction and 2.3-eV trap energy. This energy, observed by Wampler,[11] is the same as that associated with dissociation of hydrogen molecules and is assumed to represent the trap energy of molecules in bubbles formed in the implantation zone. Behind that zone were assumed a 1-μm thick damaged zone with traps of 0.0025 atom fraction and 0.8-eV trap energy. The 0.8 eV was an adjusted parameter, sized to fit experimental results. It is similar to the 1-eV trap seen by Wampler[10] and essentially the same as that seen in thermal desorption experiments by Baldwin[9,21] and
inferred to be associated with uptake in BeO by Macaulay-Newcombe.[22] It is also interesting that Krimmel and Fahnle[23] have recently concluded from purely theoretical considerations that hydrogen in beryllium is most favored at hexahedral sites with a heat of solution of 0.8 eV. In the bulk of the samples, 500 appm traps with the same 0.8 eV trap energy were assumed, again the result of adjusting parameters. The recombination model of Eq.(5) was used, and diffusivity in the bulk beryllium was taken as the Abramov high-grade value.[14]

\[ D = 8.0 \times 10^{-9} \exp\left(\frac{-0.36 \text{ eV}}{kT} \right) \left( \frac{m^2}{s} \right) \] (9)

The diffusivity in the implantation zone was adjusted to 1.5 times the value in Eq.(9) to account for the greater transport expected there. It was also multiplied by the same exponential multiplier as in Eq.(5) to accommodate the very rapid transport to the surface once the saturation concentration had been achieved. Measured temperatures for the sample faces with an assumed 10°C drop from front to rear were used. With those modifications, the only changes in input parameters among all the tests were the temperatures and the ion currents to the surface.

The fit between the model and the experimental values is quite good. However, it was found that the results are rather strongly influenced by the spatial distribution of the implanted ions and by the diffusivity in the implantation region. Inclusion of the erosion term in the recombination coefficient equation and in the diffusion equation (through the \( Q^*/k \) expression) in the bulk of the sample seems to make little difference, but failing to include it in the damage zone resulted in errors of about a factor of three in inventory.

Figure 2 shows the comparison between experiment and calculation for the simplified model with surface concentration given by Eq.(8).

Agreement with experimental data is very good, especially over the mid range of temperatures. It should be noted that a fraction of the inventory (3 \( \times 10^{19} \) D+T/m² for the high-temperature case, nearly the same at lower temperatures) was calculated to be trapped in the implantation layer. That inventory is not included in the calculation values shown in Figure 2.

2.2.4.2 Early INEEL Results

Permation experiments on beryllium conducted at the INEEL in 1990-1992 were reported in Ref.[8] To reiterate, those experiments were characterized by implantation fluxes of 5 - 6 \( \times 10^{19} \) D/m² s at energies of 1 keV/D and sample temperatures near 450°C. For one such test, the sample was 33 \( \mu \)m thick. Conventional analysis using then accepted recombination theory was unsuccessful in replicating the experimental data.[24] However, using the same model used for the fits of Figure 2 with appropriate changes in sample thickness, temperature, plasma loading conditions, and minor changes in trap concentrations, the results of Figure 3 were obtained.

2.2.4.3 Sandia Livermore Experiment

The basic experiments conducted by Hsu [25] in which he sought to obtain the recombination coefficient for deuterium on beryllium were
also modeled. (Hsu’s data were later interpreted by Andrew et al.[26] and cited by Causey and Wilson[19]) In those experiments, a deuterium glow discharge was initiated between two 5.1-cm diameter, 1.6-mm thick beryllium disk electrodes. Ion fluxes to the surface were 7.4 x 1020 D/m2s for three minutes. The electrodes became implanted with deuterium, and when the discharge was stopped, residual gas analysis of the experimental chamber revealed the rate at which the deuterium came out of the electrodes. Again, the same model with minor changes for ion implantation rate and energy, trap concentration, and temperature was used. Figure 4 shows the comparison of the experimental data of Hsu and the TMAP4 calculation. Again agreement is excellent. Similar agreement is obtained when modeling the experiments of Saibene et al.[27] (not shown). Saturation was not an issue in the experiments of Hsu and of Saibene et al.

The success of the saturation model in simulating experiments of diverse kinds conducted under rather different conditions is a reasonable indication that it could be counted on for predicting tritium inventory and permeation rates for ITER or other applications.

3.0 NEUTRON RADIATION EFFECTS

When analyzing for tritium retention and transport in beryllium used in fusion machines such as ITER where there will be significant neutron flux, two effects due to neutrons must be considered. One is the production of tritium by the transmutation of beryllium. The other is the creation of traps by neutron induced vacancies and by helium and hydrogen isotopes produced by transmutations. These will affect transient times as well as inventories.

3.1 Tritium Production

Tritium production has been estimated by Forty et al.[28] for various locations in a representative fusion reactor design using the FISPACT code. Fits to the calculations of Forty et al. give the tritium breeding inventory in fully dense beryllium as

\[ I = 280 \phi t - 2350 \left[ 1 - \exp(-0.1\phi t) \right] \quad \text{(appm)} \quad (10) \]

where the \( \phi t \) product is in MWa/m2. Differentiating in time and making unit conversions leads to a volumetric source rate for tritium production in beryllium as
\[ \frac{dI}{dt} = 1.092 \times 10^{18} \phi - 
9.166 \times 10^{17} \exp\left(-\frac{\phi t}{3.156 \times 10^8}\right) \left(\text{atom m}^{-3} \text{s}^{-1}\right) \]  
(11)

where \( \phi \) is in MW/m\(^2\) and \( t \) is in seconds.

3.2 Trapping

Determining trap energies and concentrations due to neutrons is somewhat problematic because of the lack of data on interactions of neutron irradiated beryllium with hydrogen isotopes. Experience with porous beryllium irradiated in the ATR\[29\] showed that two samples, one of 80.9% and one of 99.6% of theoretical density, irradiated to the same fluence (2.6 \times 10^{21} \text{n/cm}^2, E > 1 \text{ MeV}) under the same conditions, had substantially different amounts of tritium retained after the irradiation was complete. Following the irradiation, the 80.9% dense material had only 55.3 appm tritium (atom fraction) compared with 71.8 appm that was left in the 99.6% dense material.

The thermal release characteristics of the remaining tritium were also different. Corresponding fractional release of tritium from the 99.6% dense sample occurred at about 200°C higher temperatures during stepped thermal anneals as corresponding releases from the 80.9% dense sample. This is illustrated in Figure 5. The activation energy for the 80.9% dense material was 0.64 eV while that for the 99.6% dense material was 0.67 eV. The offset at 300°C was a factor of 42 in release fraction.

Subsequent surface area analysis[30] of beryllium from the same lots showed that the porosity of the low-density material was mostly interconnected with the ratio of BET-detected surface area to apparent geometric area of about 790. The same ratio for the 99.6% dense material was about 4.9. Earlier work[31] on fully dense beryllium of similar composition irradiated in ATR to about ten times the fluence (3 \times 10^{22} \text{n/cm}^2, E > 1 \text{ MeV}) revealed that at ambient temperature the tritium and the much more copious quantities of helium generated were finely distributed in the beryllium grains. No bubbles of gas were evident under scanning electron microscopy. When specimens from that group of samples were heated to 300, 400, and 500°C for one hour (single step), bubbles were observed at progressively larger sizes of 3, 6, and 9 nm in diameter, respectively.

The implications of those observations for the present problem are that tritium forming in the low-density material will be deposited in a dispersed form. Tritium atoms will migrate and can find their way more easily to a free surface and leave during the irradiation if the material is already porous. That process will go faster at higher temperatures. Hence, the amount of tritium retained in material irradiated at elevated temperatures would be expected to be less than that retained in material irradiated at lower temperatures, and less would be retained in porous material than in dense.

Regrettably, experimental data on beryllium irradiated at elevated temperatures to quantify those generalities have yet to be produced. This difference in retention and release may be related to trapping characteristics, as will be shown in subsequent sections here.

3.3 Diffusion

Even though Baldwin and Billone[29] found effective diffusivities for the release of tritium from irradiated beryllium in their
stepped-anneal experiments, they did not consider the effects of surface oxide films on their specimens in their analysis. These films have been found very important in almost all experiments on movement of hydrogen isotopes in beryllium. Oxide film effects were considered by Cho et al.[32] who gave an effective value for diffusivity of the high-density beryllium using the data of Baldwin and Billone as

\[ D_{Be} = 4.5 \times 10^{-7} \exp \left(- \frac{1.08 \text{ eV}}{kT} \right) \left( \frac{m^2}{s} \right) \]  

(12)

They used fitting techniques to estimate a diffusivity for BeO as

\[ D_{BeO} = 6.28 \times 10^{-16} \exp \left(- \frac{0.553 \text{ eV}}{kT} \right) \left( \frac{m^2}{s} \right) \]  

(13)

and they estimated the BeO thickness on the samples as effectively 18.6 nm.

The Cho et al. value of diffusivity in dense beryllium is apparently a consequence of trapping. It can be reconciled with that of Abramov et al.[14], Eq.(9), as follows.

It may be shown[33] that beryllium under the conditions of this problem falls into a regime in which

\[ \frac{D_{eff}}{D} = \frac{\lambda^2 v}{\rho D_0} \exp \left( \frac{E_d - E_t}{kT} \right) \]  

(14)

where

- \( \lambda \) = lattice constant (m)
- \( v \) = Debye frequency (10\(^{13}\) s\(^{-1}\))
- \( \rho \) = trap concentration (atom fraction)
- \( D_0 \) = diffusivity pre-exponential (m\(^2\)/s)
- \( E_d \) = diffusion activation energy (eV)
- \( E_t \) = trap binding energy (eV)
- \( k \) = Boltzmann's constant (8.615 \times 10^{-5} eV/atom K)
- \( T \) = temperature

Taking the diffusivity of Cho et al. as \( D_{Be} \) and the Abramov value as \( D \), it is straightforward to show that \( \rho = 1.08 \) atom fraction and \( E_t = 1.44 \text{ eV} \). These values are reasonable for the traps that would be associated with the small bubbles generated by the production of helium and tritium in the beryllium and are generally consistent with other observations.[24] They should not be considered as definitive, however, because of uncertainties in the values for \( D \) and \( D_{eff} \).

With regard to the effects of oxide film coatings on beryllium, Cho et al. gave a value for the recombination coefficient for tritium on BeO though they apparently did not account in their analysis for the strong surface hydrogen concentration present in the experiments, which would make surface release first order rather than second as they supposed. Baldwin and Billone used 0.1% H\(_2\) in the sweep gas over the samples during the desorption. To perform successful TMAP4 calculations for oxide coated surfaces, it is necessary to know the hydrogen solubilities in the base material and in the oxide or at least their ratio. For beryllium, the solubility of Shapovalov and Dukel'ski,[20]

\[ S_{Be} = 2.165 \times 10^{22} \exp \left(- \frac{0.17 \text{ eV}}{kT} \right) \left( \frac{\text{atom}}{m^3 \text{Pa}^{1/2}} \right) \]  

(15)

is preferred. For solubility in BeO, the value of Macaulay-Newcombe and Thompson[22] was used:

\[ S_{BeO} = 10^{18} \exp \left(- \frac{0.8 \text{ eV}}{kT} \right) \left( \frac{\text{atom}}{m^3 \text{Pa}^{1/2}} \right) \]  

(16)

Both of these are for fully dense material.

There have been no adequate measurements of diffusivity for low-density beryllium. However, in the data of Baldwin and Billone, the time constants for release of tritium from the two samples were very similar for corresponding temperatures. As pointed out elsewhere,[34] the transient data are consistent with expected effective diffusivity values.

To assist in resolving that issue, the 80.9%-dense and 99.6% dense sample stepped-anneal
experiments of Baldwin and Billone were simulated using TMAP4 with an assumed oxide film thickness of 40 nm. The diffusivity used for beryllium was Abramov's,[14] the solubility was that of Shapovalov and Dukel'ski,[20] the BeO diffusivity was Cho's,[32] and the solubility of BeO was that of Macaulay-Newcombe and Thompson.[22]. Trap energy and concentration were adjusted to give optimal fits to the experimental data. For the 80.9% dense case, traps at 0.3% atom fraction concentration and 1.06 eV trap energy gave a good fit. The transients calculated are shown in Figure 6 together with selected data points from Baldwin and Billone.

![Figure 6](image)

Figure 6. Approximation of the diffusion transients of tritium from 80.9% dense beryllium in experiments of Baldwin and Billone.[29] Simulations used standard diffusivity and solubility for Be and BeO and traps at 0.3% atom fraction and 1.06 eV trap energy. The 573-K transient as measured apparently had a second, low-energy trap that was not occupied at higher temperatures.

The fit is quite good except at the 300°C temperature. That difference at 300°C could be explained by the presence of a second trap (not modeled) at a fractional eV trap energy that would retain tritium at 300°C but be invisible at higher temperatures. That trap may correspond to the binding of tritium in the oxide film, possibly as hydroxyl radicals. It was previously observed[21] that a more or less instantaneous release process (for example, from the surface oxide) combined with an effective diffusivity similar to that of Jones and Gibson[34] would give a very satisfactory explanation for the Baldwin and Billone observations for the higher-density material, supporting the notion of a second, low-energy trap.

The traps that gave a good fit were not so numerous nor at quite as high a trap energy as was estimated using the effective diffusivity theory for the fully dense material. Both of those points are consistent with the notion that the more porous material provides much less retention of transmuted tritium because of the ready pathways to the surface that exist in the interconnected porosity.

The same process was undertaken for the Baldwin and Billone 99.6% dense data. Here the trap concentration and trap energy that gave the best fits were 2% atom fraction and 1.4 eV trap energy. The comparison with the experimental measurements appears in Figure 7.

![Figure 7](image)

Figure 7. Approximation of the tritium diffusion transients from 99.6% dense beryllium measured by Baldwin and Billone.[29] Simulations used standard diffusivity and solubility for Be and BeO with traps at 2% atom fraction and 1.4 eV trap energy.

For want of a better technique, it may be appropriate to use linear interpolation on trap concentrations and logarithmic interpolation on trap energies for grades of beryllium intermediate between the two densities considered here. However, this remains to be confirmed by experiments.
4.0 SUMMARY AND CONCLUSIONS

Modeling of the interaction of hydrogen isotopes with beryllium has proven to be less than straightforward. It has been found in experimental data that there is a saturation phenomenon in beryllium that is only apparent when implantation fluxes are high, of order $10^{20}$ ion/m$^2$s or more. The mechanism for this effect appears to be the opening of channels from the implantation depth to the surface by the action of implanting ions precipitating at rates too great to be carried away by diffusion. It has been successfully modeled in TMAP4, which gives calculational results that are in good agreement with a wide range of experiments.

Diffusion, retention, and trapping in neutron irradiated material are also interesting in that neutron displacements and gaseous transmutation products generate traps for hydrogen isotopes in beryllium as well as producing tritium. Values for trap energies and concentrations have been chosen that allow relatively straightforward modeling of neutron effects using standard values for diffusivity, solubility, and other processes.

These models should make possible much more accurate estimates of tritium inventory and permeation in beryllium plasma-facing components in future fusion reactors.

6.0 REFERENCES

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7. First Wall Applications

7.1 Response of Beryllium to Severe Thermal Shocks - simulation of Disruption and Vertical Displacement Events in Future Thermonuclear Devices

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Beryllium will play an important role for plasma facing components in next step thermonuclear fusion devices such as ITER. In particular for the first wall beryllium will be used with an armor thickness of several millimeters. During normal plasma operation these components are subjected to quasi-stationary, cyclic heat loads; however, during plasma instabilities they also will experience severe thermal shocks. Here plasma disruptions with deposited energy densities of several ten MJm\textsuperscript{2} are the most essential damaging mechanism. However, a significant fraction of the incident energy will be absorbed by a dense cloud of ablation vapor, hence reducing the effective energy density at the beryllium surface to values in the order of 10 MJm\textsuperscript{2}. Even under these conditions which occur on a millisecond time scale severe melting of the plasma facing material to a depth of several hundreds of microns occurs. Splashing of liquid Be-droplets and the complete removal of the melt layer due to electromagnetic forces during the plasma disruption will result in an enhanced erosion. Beside material losses, the formation of cracks in the recrystallized zone and in the base material will have strong impact on the integrity of the beryllium tiles, in particular during the cool down phase. Plasma instabilities on a much longer time scale, i.e. with a duration of 0.3 to 1.0 s have to be anticipated. These so-called vertical displacement events (VDEs) result in deposited energy densities of approx. 60 MJm\textsuperscript{2}. In contrast to the rather short disruption events beside surface effects also material damage to the joining zone has to be considered.

To investigate the material response to all these plasma instabilities thermal shock tests on small scale test coupons (disruption effects) and on actively cooled divertor modules (VDEs) have been performed in the electron beam test facility JUDITH at ITER relevant surface heat loads. These tests have been performed on different bulk beryllium grades and on plasma sprayed coatings; the influence of pulse duration, power density, and temperature effects has been investigated experimentally. Detailed in-situ diagnostics (for beam characterization, optical pyrometry etc.) and post mortem analyses (profilometry, metallography, optical and electron microscopy) have been applied to quantify the resulting material damage. 1D- and 2D models have developed to verify the experimental results obtained in the electron beam simulation experiments.

1. INTRODUCTION

Disruptions and vertical displacement events result in substantial surface modifications due to evaporation, melting and crack formation. In general these effects will be lifetime limiting for plasma facing components in future fusion reactors [1, 2]; additional material damage due to neutron irradiation will result in an even accelerated material degradation. The type of material damage induced by the deposition of large energy densities within short periods depends on the type of beryllium, its microstructure, the oxygen content, and the thermomechanical properties (in particular thermal conductivity and melting temperature) [3]. The thickness of the heat affected zone strongly depends on the time scale of the thermal shock loading: milliseconds for disruptions, several hundred milliseconds for VDEs. The former event is independent from the active cooling system; in the
latter case heat removal by the metallic heat sink has to be considered.

2. EXPERIMENT

To simulate the material loading in future tokamak facilities such as ITER, thermal shock experiments on a wide spectrum of candidate PFMs have been performed in the electron beam device JUDITH. Beside beryllium these tests also include a variety of different graphites and carbon fiber composite materials [4], ceramics based on SiC or B$_4$C [5], and various tungsten alloys [4, 6]. The test facility supplies a focused 60 kW electron beam; homogenous heat load distribution to the sample is done by fast scanning; typical loaded areas are 5 x 5 mm$^2$ for disruptions and 10 x 10 mm$^2$ for VDEs.

The maximum deposited energy densities were 8 MJm$^{-2}$ (disruptions) and 60 MJm$^{-2}$ (VDEs). For the short disruption pulses the electrical power (U = 120 kV) has been supplied from a capacitor; the typical pulse duration was 5 ms (beam rise time 130 μs). For VDE-tests the gun power was extracted from the high voltage transformer which implies a much slower rise time: the applied loading scenario included a 0.5 s beam ramp-up, 0.5 s at full power and a 0.5 s ramp-down.

The electron beam test facility JUDITH (Figure 1) has been equipped with a set of powerful diagnostics to quantify temperatures / temperature distributions in test samples and to calibrate the incident heat fluxes to the components' surface. Among these are [7]:

- single color pyrometer for the temperature range from 200 to 1100°C (emissivity dependent signal).
- a fast single color pyrometer for $T \geq 1100°C$ with a time resolution in the order of 1 μs (for disruption simulation experiments).
- Two color pyrometer for a temperature range from 1000 to 3500°C (independent from the emissivity ε).
- diagnostics for the quantification of the absorbed current in the test sample to determine the fraction of backscattered electrons.

Figure 2: Beryllium test specimens for the simulation of material damage during plasma disruptions.
2.1 Disruption simulation

Test coupons for disruption simulation (cf. Figure 2) experiments consist of polished specimens for multiple spot loading (25 x 25 x 10 mm$^3$) and single spot loading (12 x 12 x 5 mm$^3$). The latter type of test sample has been designed for neutron irradiation experiments. On these test specimens material erosion during simulated disruption events have been quantified by two independent methods: weight loss measurements and 2D/3D surface profiles determined by a laser micro-profilometer.

The latter method supplies additional information on the melt motion during electron beam loading. Figure 3 shows surface profiles measured at different energy densities. Even at rather low deposited energies (i.e. below 1 MJm$^{-2}$) the surface shows a distinct swelling of 1 to 2 μm; however, SEM micrographs from this surface area do not indicate any visible surface modifications. This swelling obviously occurs during electron beam loading which is associated with a plastic deformation of the thermally expanding material. With increasing energy densities (e.g. 1.4 MJm$^{-2}$) material swelling becomes more essential. Up to a threshold value of approx. 2 to 3 MJm$^{-2}$ no net erosion (i.e. crater formation below the original surface) has been detected. At higher energy densities (3.4 MJm$^{-2}$ and above) the material response is controlled by different effects: swelling, net erosion and melt ejection.

The effect of material erosion for different grades of beryllium is shown in Figure 4. In the graph the maximum crater depth determined from 3D-profiles has been plotted vs. absorbed energy density. 4 % of the incident electrical power density has been reflected. This value which has been determined by measuring the absorbed current is rather small for beryllium compared to high-Z materials (e.g. 52 % for tungsten). The investigated materials include the grade S65 C by Brush Wellman and three Russian grades (DShG200, DShG56, and TR-30). All these materials have been produced by power
metallurgical methods. DShG200 is a coarse grain material. TR-30 has a fine grain structure and a relative high oxygen content; this material is expected to show good neutron irradiation resistance. DShG56 covers an intermediate grain size. According to the electron beam tests in JUDITH the grades S65 C and DShG56 show the best thermal shock performance. The onset of a clear crater formation is at approx. 3 MJm$^2$; beyond this level the crater depth grows rapidly with increasing energy density. Less thermal shock resistant materials such as DShG200 and TR-30 show a more rapid crater formation.

![Figure 4](image)

Figure 4: Erosion of different beryllium grades. Negative values indicate material swelling.

Beyond 5 MJm$^2$ crater depths in the order of 200 μm are realistic for all Be-grades. To extrapolate from these data to the conditions in a thermonuclear fusion reactor vapor shielding effects [8] have to be taken into account; hence, the energy densities plotted in Figure 4 should be compared with the fraction of incident energy to the first wall, the baffle, or divertor which penetrates through the optically dense cloud of ablated material. In addition it should be noted that convection in the melt layer and the fraction of melt loss will not necessarily be identical under electron beam load tests (with vertical beam incidence) and the realistic situation in a large tokamak reactor.

![Figure 5](image)

Figure 5: Crack formation in beryllium TR-30 (energy density: 5.2 MJm$^2$, pulse duration: 5 ms) top: polarized light, bottom: etched metallographic section.
Figure 6: Crack formation in beryllium S65 C (energy density: 5.2 MJm\(^{-2}\), pulse duration: 5 ms)
top: polarized light, bottom: etched metallographic section.

The formation of cracks in recrystallized beryllium and in the underlying base material is of essential importance for the components' integrity and life time. Figure 5 shows an example for the formation of deep cracks in a less thermal shock resistant beryllium grade. At incident energy densities of 5.2 MJm\(^{-2}\) the resulting cracks penetrate deep (approx. 1800 µm) into the base material; the thickness of the melt layer is in the order of 250 µm. The trace of the crack is mainly intergranular, with some transgranular damage. Below the recrystallized melt there is almost no heat effected zone; grain size and shape are almost unaffected.

A different thermal shock resistance has been established for the beryllium grade S65 C, c.f. Figure 6. This material grade shows a similar thickness of the melt layer under identical heat load conditions (5.2 MJm\(^{-2}\)). However, thermally induced cracks only extend to the bottom of the melt layer and not into the base material.

To compare different Be-materials the maximum thickness of the melt layer and the maximum crack length has been plotted in Figure 7 for S65 C and the three Russian grades. The former material shows similar values for melt layer thickness and crack length (approx. 100 - 150 µm at 2.2 MJm\(^{-2}\), and 250 µm at 5.2 MJm\(^{-2}\), resp.). This implies that disabling cracks only occur in the recrystallized zone. Any new disruption event will remelt this layer and new cracks will be generated during the recrystallization phase (which lasts for approx. 20 ms) or the subsequent cool-down period. The other three material grades reveal additional crack formation in the base material below the melt layer, in particular at high energy densities. This effect is worse for TR-30.

Figure 7: Thickness of recrystallized zone and crack length in different beryllium grades after thermal shock loading at different energy densities.

All thermal shock tests described so far have been performed on cold (i.e. not preheated) test specimens. Plasma facing components in future thermonuclear fusion reactors will experience severe thermal shocks primarily during plasma operation, i.e. at elevated temperatures. Plasma disruptions on
the first wall are expected to occur at surface temperatures above 400°C. Under these conditions beryllium behaves more ductile compared to room temperature. Hence, part of the Be-test coupons have been loaded under experimental conditions which take care of this effect. The samples (12 x 12 x 5 mm³) have been mounted on a copper holder which was thermally isolated from the movable support structure. A defocused electron beam with low power density was used to heat up the copper holder including the test coupons to temperatures slightly above the experimental conditions. Thermal shock loading by means of a focused beam (cf. Chapter 2) has been performed in the cool down phase after having achieved the envisaged temperature range.

A typical result from these tests is shown in Figure 8. Two test specimens made from the Be grade TR 30 have been loaded at 5 MJ m⁻² at room temperature and at 450°C, respectively. In agreement with Figure 5 intense crack formation occurred on the cold sample within the area of direct beam incidence; some of these cracks extend to an area outside the melt crater.

![Figure 8: Beryllium TR30 loaded at 5.0 MJm², 5 ms pulse duration, 5 shots](image1)

Figure 8: Beryllium TR30 loaded at 5.0 MJm², 5 ms pulse duration, 5 shots
Top: loading at room temperature, bottom: sample heated to 450°C prior to electron beam loading.

A similar thermal shock behavior has also been established for tungsten in electron beam and plasma accelerator simulation experiments [6, 11], here the formation of cracks can be suppressed almost completely at elevated temperatures.

![Figure 9: Crack formation and weight loss of different beryllium grades at room temperature and 450°C.](image2)

Figure 9: Crack formation and weight loss of different beryllium grades at room temperature and 450°C.
Figure 10: Actively cooled divertor module FT38-2 with S65 C armor on CuCrZr (CuMnSnCe braze) after VDE-simulation at 60 MJm$^{-2}$. Bottom: metallographic section from the 6 mm thick Be armor.

Figure 11: Actively cooled divertor module FT29-4 with S65 C armor on CuCrZr (InCuSil braze) after VDE-simulation at 60 MJm$^{-2}$. Bottom: metallographic section from Be armor.
In other beryllium grades (e.g. S65C or plasma sprayed beryllium) this effect of sample temperature on the formation of surface cracks is less significant or does not exist at all. In Figure 9 the total crack length measured by optical image analysis has been compared for the above mentioned beryllium grades at room temperature and 450°C up to absorbed energy density values of 8 MJ m⁻².

Figure 9 also gives results from weight loss measurements on the same set of test specimens at identical test conditions. All samples loaded at elevated temperature show an increased material erosion compared with the RT conditions.

Other non-beryllium material candidates for plasma facing components in ITER show a more favorable behavior under intense energy deposition [4]. This is mainly due to the significantly higher melting point (W-alloys) or in the case of carbon fiber composites due to the absence of a liquid phase.

2.2 Simulation of Vertical Displacement Events

Compared with the fast thermal shock loading during disruptions, vertical displacement events occur on a much longer time scale (typically in the order of 0.3 to 1 s). Under these conditions thermal conduction into the base material is essential; in addition - depending on the thickness of the PFM - the bond or braze layer will be affected. Part of the incident energy will be removed by the coolant water.

The heat load experiments in the electron beam facility JUDITH have been performed on miniaturized flat tile mock-ups with beryllium (S65 C) armor. The Be-tile thickness was 3 and 6 mm, respectively. The armor was brazed to a CuCrZr heat sink by an induction brazing process with a CuMnSnCe braze (FT38-2), Figure 10. Similar tests have been performed on mock-ups using an InCuSil braze (FT29-4), Figure 11, and on a HIPed module (DS-Cu substrate). Results from tests with monoblock CFC mock-ups with identical armor thicknesses and loading parameters are reported elsewhere [4, 12].

Electron beam loading was performed with a beam spot size of 10 x 10 mm². Since the energy supply system (transformer) does not allow fast transients at high beam currents, a loading cycle of 1.5 s was applied (0.5 s each for the ramp-up, for the full power and for the ramp-down phase). The applied heat flux at full power was 60 MWm⁻² (this corresponds to a deposited energy density of 60 MJm⁻³).

In Figure 12 the resulting surface temperatures are plotted for the two different armor thicknesses. The emissivity of beryllium depends strongly on temperature, surface morphology and BeO-content; thus temperature measurements using integrated flux pyrometers are rather inaccurate. To overcome this deficiency, a two-color pyrometer (with a sensitivity range from 1000 - 3500°C) has been applied additionally.

Figure 12: Surface temperature during VDE simulation by intense electron beams for a brazed (FT38-2) and a HIPed module (CEA1); deposited energy density: 60 MJm⁻³.

The surface temperature increases continuously during the ramp-up phase and reaches a maximum value of about 2100°C (independent from the armor thickness). During the full power phase the surface temperature seems to decrease. This is explained by the formation of a vapor cloud over the melted beryllium surface. This cloud is transparent to the incident electrons but not for the emitted IR radiation. During the power ramp down phase mock-ups with a thin beryllium armor (3mm) show a faster temperature drop.
Metallographic sectioning of the mock-ups has been performed after electron beam loading. Figures 10 and 11 show micrographs from the S65 C beryllium brazed to the CuCrZr heat sink. For both types of modules the surface layer shows a rather homogeneous melt zone; castellations are bridged with melt; in the cool down phase a deep crack (which penetrates the melt layer completely) follows the former castellation. The resulting melt layer has a thickness of approx. 900 \mu m for a Be thickness of 3 mm and up to 1500 \mu m for the thicker armor. The recrystallized layer consists of long columnar grains which extend from the surface down to the interface with the unaffected material. There are almost no cracks detectable, neither in the recrystallized zone, nor in the undamaged S65 C. Metallographic examinations from the joints do not indicate any defects for the two brazed modules. The HIPed joint however showed intense delamination at the interface, particularly at the corner of the mock-up.

Monoblock mock-ups with CFC armor do not show any visible damage at identical heat load scenarios. Beside some minor discoloration at the point of beam incidence the carbon fiber composite material has not been eroded; the braze joint remains intact [4].

3. MODELLING

3.1 Disruption simulation

Finite element calculations using the ABAQUS code have been performed to determine the temperature distribution during short term thermal shock loading on beryllium surfaces. Figure 13 shows the temperature evolution for a 5 ms electron beam pulse resulting in an absorbed energy density of 5 MJm\(^{-2}\). Volumetric heating of the surface layer due to the 120 keV electrons was implemented in the model. In addition the effect of intense convection in the melt layer could be implemented; both calculations are shown in Figure 13. In a shallow surface region down to the penetration depth of the electrons (approx. 150 \mu m) the resulting temperature remains rather constant. At a depth of 200 \mu m melting occurs after approx. 4 ms if no convection is permitted; for the convection case melting occurs at approx. 3 ms of beam incidence. Below a melt layer of 200 to 250 \mu m (cf. Figure 7) the material will not melt.

The braze layer or bonding zone which is several mm below the loaded surface remains (thermally) almost unaffected by the disruption heat pulses. However, a complicated stress distribution will be generated due to the intense heating of the outer surface layer. Calculations performed without or with the convection term show a similar tendency, however, under the latter conditions higher surface temperatures and a shorter recrystallization time of 10 ms have been detected (with convection: 15 ms).

![Figure 13: Modelling of the temperature evolution during thermal electron beam shock loading of beryllium with and without convection in the meltlayer, \(E_{abs} = 5 \text{ MJm}^{-2}\), pulse duration = 5 ms.](image-url)
3.2 VDE simulation

2D thermal analyses of the vertical displacement events were carried out by means of the ANSYS finite element code [13]. The heat transfer coefficient vs. wall temperature was computed by means of the EUPITER code [14]. The thermal input parameters were identical to those in the electron beam tests in JUDITH, i.e. 0.5 s ramp-up to 60 MW m\(^{-2}\), 0.5 s of full power, and 0.5 s ramp-down. The effect of evaporation cooling was taken into account implementing a previously developed model in the ANSYS code [15].

The results of the numerical analyses are shown in Figure 14 for different geometries and beryllium armor thicknesses of 3 and 6 mm, respectively. Compared with the experimental results in Figure 12, similar maximum surface temperature are obtained from the experimental and the calculated data. In this respect one should consider, that the calculations assume evaporation cooling only during the VDE tests, i.e. up to 1.5 s [12]. After that time a more rapid decrease of the experimentally measured surface temperature can be noted because of the contribution of the evaporation cooling.

![Figure 14: Modelling of VDEs with a deposited energy density of \(E_{\text{abs}} = 60\) MJm\(^{-2}\).](image)

- \(\text{GEC - 6 mm}\)
- \(\text{ACCEL - 3 mm}\)
- \(\text{GEC - 3 mm}\)

In the 6 mm thick beryllium a constant surface temperature of \(T = 1280^\circ\text{C}\) was computed at the time 1.7-1.9 s because of the resolidification of the Be armor. This effect is less pronounced in the experimental curve. This may be explained by the occurrence of convective motion in the melt layer which results in a more effective cooling [16].

As far as the depth of the melt layer is concerned, a value of 1.0 and 1.25 mm was computed for the 3 and 6 mm thick armor, respectively. An experimental depth higher than that computed is the consequence of the above mentioned convective motion.

4. CONCLUSIONS

Different beryllium grades have been investigated under severe thermal shocks to simulate disruptions and VDEs. The experimental results indicate that components with Be armor can withstand a limited number of these events without disabling failure of the entire component.

To optimize the performance under short energetic thermal shocks which are typical for plasma disruptions, a careful selection among different Be grades is indispensable. Alternative PFMs such as W or CFC show a superior performance if compared with beryllium; this is mainly due to the higher melting point or the absence of a liquid phase.

Vertical displacement events on actively cooled modules with beryllium armor with deposited energy densities of 60 MJm\(^{-2}\) result in a severe melting of the plasma facing material (the thickness of the melt layer is in the order of 1 mm). In electron beam tests the bond layers with brazed joints (InCuSil or CuMnSnCe) remained intact; HIPed joints failed under identical thermal loads.

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7.2 Thermal Cycling Tests of Actively Cooled Beryllium Copper Joints

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Screening tests (steady state heating) and thermal fatigue tests with several kinds of beryllium – copper joints have been performed in an electron beam facility. Joining techniques under investigation were brazing with silver containing and silver-free braze materials, hot isostatic pressing (HIP) and diffusion bonding (hot pressing). Best thermal fatigue performance was found for the brazed samples.

1. Introduction

Beside carbon materials and tungsten, beryllium is one of the candidates for plasma facing materials in next generation tokamaks as ITER. For the construction of components, beryllium tiles have to be attached to heat sinks made from copper alloys. Different joining techniques were developed during the last years, including brazing, diffusion bonding and HIPing. Testing of such beryllium copper joints have been performed in the electron beam facility JUDITH in Forschungszentrum Jülich, Germany. In these tests, small-scale mock-ups have been loaded in screening tests (steady state heating) and fatigue tests (thermal cycling) up to 8 MW/m² and 1000 cycles.

During the operation of ITER, high heat flux components will be exposed to 14 MeV neutrons. The degradation of various materials properties due to neutron irradiation has been studied since several years, but up to now no heat flux simulations have been performed with actively cooled samples after neutron-irradiation. During the last year, JUDITH has been upgraded for this kind of experiments. First tests with samples irradiated up to 0.4 dpa at the High Flux Reactor in Petten (The Netherlands) have been started.

2. Experimental Details

Hot Cell Electron Beam Facility JUDITH

The electron beam facility JUDITH consists of a 60 kW electron gun with an acceleration voltage of 120 kV, a vacuum chamber measuring 800 x 600 x 900 mm³, and a number of diagnostic devices. The focused electron beam has a diameter of 1 mm approx. and can be swept in two directions at frequencies up to 100 kHz. The technical data for the machine are listed in [1], a more detailed description is given in [2]. Diagnostics installed with the testing of actively cooled beryllium monoblock are described below.

For the investigation of the influence of neutrons on the properties of high heat flux components, some modification to former testing procedures were required. These modifications concerned on one hand the samples (cf. ch. 3.1) and on the other hand the testing facility.

In order to save volume in the neutron irradiation rig, the irradiation mock-ups were designed without tube connectors for cooling water supply (cf. fig. 1). Therefore a clamping mechanism for the water supply was developed (cf. fig. 2). For installing, the radioactive samples are placed on a small tray which is transported to the clamping mechanism by manipulator. When the sample is in the correct position, the water connectors are clamped to the sample. This is performed by a motor while the force is controlled by a load cell. Sealing is achieved by special sealing adapters machined from soft copper in combination with O-ring sealings and springs. The whole clamping system is attached to the door of the vacuum chamber of JUDITH. Once the sam-
Fig. 1: Miniaturized Be/Cu mock-up for neutron-irradiation

Fig. 2: Clamping system for water supply to miniaturized Be/Cu mock-ups

- A quadrupole mass spectrometer is mainly used to detect the evaporation of beryllium, but it also indicates water leaks in the cooling water supply of the mock-ups.
- Thermo-couples (number and position are dependent on mock-up design and experimental parameters).
- Instrumented cooling loop: During the experiment pressure, flow rate, inlet- and outlet temperature of cooling water are controlled (s. below).

Measurement of Surface Temperatures

For the assessment of heat removal efficiency of the different mock-ups, it is essential to measure the surface temperature. As thermo-couples can measure the temperature only in a certain distance from the surface, IR measurements are required (IR scanner, pyrometer). These measurements are subjected to uncertainties in emissivity. The emissivity of beryllium is strongly dependent on surface condition (roughness, oxidation grade etc.) and temperature. In order to minimize the errors in the IR scanner measurements, each individual mock-up is calibrated. In these calibration experiments the cooling water is removed from the module while it is heated uniformly. After the equilibrium temperature is reached (thermo-couple reading) the corresponding emissivity is obtained from the IR image of the sample surface. This method considers the surface condition of the mock-up at the beginning of the experiment. However, in long-term fatigue experiments the surface condition undergoes changes...
caused by oxidation. Therefore a re-calibration is required at the end of each experiment.

Absorbed Heat Flux

The power absorbed by the mock-ups during the test is determined by water calorimetry. It can be calculated from:

\[ P_{\text{abs}} = C_p \Delta T \phi \]

where \( C_p \) is the specific heat, \( \Delta T \) the change of temperature increase, and \( \phi \) the mass flow of cooling water.

If the calculated values \( P_{\text{abs}} \) are compared with the incident powers \( P_{\text{inc}} \), absorption coefficients of 77% are found for beryllium.

During the heat loading, the area covered by the electron beam is a little smaller than the total surface. This heated area is not completely reproducible, but after the experiment it can be recognised as a discoloration on the sample surface and can be measured. Typically the electron beam stays away 1 mm from all sample edges, and the sweeping amplitudes in x- and y-direction are 2 mm smaller than the length and width of the mock-up surface.

If the power density is calculated, the result depends strongly on the reference area. The differences are between 5 and 25%, depending on the size of mock-ups. For the assessment of the joints, a power density which refers to the whole surface area \( D_i \) is thought to be more suitable and thus the corresponding numbers are used in the following.

3. Testing of Un-Irradiated Mock-Ups

In the frame of the European ITER tasks beryllium - copper mock-ups have been produced by several European associations and companies [3]. Different material combinations, designs and joining techniques have been developed. In order to assess the heat removal efficiency and the thermal fatigue behaviour some of these variants have been tested in JUDITH.

3.1 Brazed flat tile mock-ups

These small-scale mock-ups (cf. fig. 1) were developed mainly for neutron irradiation purposes. Due to the limited volume in the irradiation rig the surface area of these samples was limited to 15 x 25 mm. The beryllium thickness is 3 or 8 mm. For better comparison these samples were also used for the un-irradiated reference tests.

The production process followed the guidelines for induction brazing given by JET for InCuSil braze material [4, 5]. In this process the samples are heated slowly to 450 °C; after a hold time of two minutes, they are heated rapidly (8 °C / s) up to 5 – 10 °C above liquidus. After a dwell time of 10 to 300 s they are rapidly cooled (4 °C / s) down to 450 °C.

Under the operational condition of ITER, silver is strongly activated by neutrons and transmutes rapidly to cadmium. Therefore silver-free braze materials are preferred. In addition to the "classical" InCuSil braze, mock-ups with a new silver-free CuMnSnCe braze were produced by GEC-Marconi in co-operation with JET.

A mock-up with an InCuSil braze was tested before and showed an excellent behaviour during a loading procedure of 500 heat cycles at a power density of 4.8 MW/m². The results of this test were reported elsewhere [6]. A different mock-up brazed with the CuMnSnCe braze and a Be armour thickness of 8 mm was loaded up to 1000 heating cycles at a power density of \( D_i = 5.4 \) MW/m² (referred to the total surface area of 25 x 15 mm²). During each load cycle the mock-up was heated for 10 seconds; then it was allowed to cool down for another 10 seconds. Ramp-up and ramp-down times were 0.5 seconds each. The flow rate of cooling water during the test was 15 m/s at a pressure of 40 bars.

During the thermal fatigue experiment, the surface temperature was measured by pyrometer. In addition the braze layer temperature was controlled by means of a thermo-couple located 1 mm below
Fig. 4: Graph of temperatures during thermal cycling, above: temperatures on surface and in braze layer, below: power absorbed by cooling water (from water calorimetry)

the joint within the copper body. This thermo-couple was mounted through a small hole drilled from the bottom of the mock-up. Fig. 3 shows the temperature on the surface and at the thermo-couple during the first two heat cycles. After ten seconds the surface temperature had reached approximately 95% of the equilibrium temperature.

During the 1000 heating cycles, both temperature curves stay stable. This becomes also clear from fig. 4, where the peak value of the temperatures in each load cycle is plotted versus the number of cycles. If for the pyrometer a constant emissivity is assumed, the surface temperature during the 1000 cycles increases steadily from 430 to 490 °C although the power absorbed by the coolant water stays constant (P_{abs} = 2.0 kW, cf. fig. 4b). But this temperature measurement does not take into account a change in emissivity. In fact, the sample surface is progressively oxidised by the residual oxygen in the vacuum chamber and the emissivity is increased. By calibration experiments before and after the fatigue experiment, an increase of emissivity from ε = 0.13 to 0.38 was observed. Thus, if the results from the pyrometer are corrected with respect to this changed emissivity, the surface temperature at the end of the 1000th cycle has not increased.

Although former shear test results for the CuMnSnCe braze were not so promising [7], the behaviour of this mock-up in the thermal fatigue test was excellent. It survived 1000 cycles at 5.4 MW/m² without any indication of failure. After the high heat flux experiment, metallographic sectionings were made. In these sectionings, an intermetallic phase with some crack formation was observed in the braze material. Evidence of these features was also seen prior to high heat flux testing. No cracking was observed at the root of the castellation slots.

3.2 Beryllium monoblock mock-up

A beryllium monoblock design has been developed by JET. In this design twenty tiles of beryllium S65C were brazed to a CuCrZr tube. The brazing process was as described above, braze material was InCuSil. A beryllium monoblock loaded several times up to 20 MW/m² in the JET NBI testbed showed no visible degradation, but some cracking was observed on the re-solidified Be surface [8]. In order to study the thermal fatigue behaviour, the same mock-up has been submitted to 1000 cycles in the JUDITH facility at a power density of D_{0} = 4.5 MW/m² (heated area: 40 x 25 mm, corresponding to 4 tiles of 10 mm width). No catastrophic failure was observed during these 1000 load cycles. The surface temperature measured by the two-colour pyrometer was above 700°C but it was not stable during the test. For the first 500 cycles it increased by more than 30 °C, but came back to the initial temperature during the last 500 cycles. This fluctuation of the surface temperature was confirmed by several independent measuring methods (two-color pyrometer, IR camera, thermo-couple inside the tiles). The absorbed power during the test was constant according to water calorimetry. It is assumed that the observed temperature fluctuations are caused by crack opening and crack closure processes in the braze material. In order to clarify this situation, metallographic sectioning are carried out.
3.3 Hot isostatic pressed (HIPed) mock-ups

Two European associations produced solid HIPed BE/CU mock-ups (CEA and Studsvik Material). The HIPing process in both cases was somewhat different.

CEA produced two flat tile mock-ups with a heat sink of Glidcop A125 and a stainless steel back-plate. One was developed for the first wall, and the second one for baffle applications. In both cases the modules were HIPed for two hours at 800 °C and 120 MPa. A titanium interlayer was used to avoid the migration of beryllium to copper [9, 10]. The two mock-up were designed as follows:

- first wall mock-up: total surface area 96 x 86 mm$^2$, four cooling tubes, steel back-plate, 9 mm armour thickness, no castellations,
- baffle mock-up: total surface area 72 x 70 mm$^2$, three cooling tubes, steel back-plate, 4 mm armour thickness, castellations 24 x 10 mm$^2$ approx.

A somewhat different production process was used by Studsvik Material. Two samples were produced by HIPing of beryllium S65C to a CuCrZr heat sink at 850°C and 140 MPa for 2 hours, no interlayer was used [11]. The total surface area of the modules was 100 x 24 mm$^2$. Originally the beryllium thickness was 8 mm, but for half of the mock-up it was reduced to 4 mm by electric discharge methods.

The first wall mock of CEA was loaded 1000 load cycles at 2.5 MW/m$^2$. For the first 100 cycles heating and cooling times were 10 seconds, then these times were increased to 30 seconds each. Several thermo-couples were installed in the copper, near the braze and in the beryllium. Fig. 5 shows the reading of the pyrometer and of a thermo-couple located in the Be approx. 5 mm below the sample surface) during the first six load cycles for heating and cooling periods of 10 and 30 seconds respectively. It is obvious that in the former case the mock-up was still far from equilibrium conditions and the latter load case is more relevant. During the 1000 heating cycles neither the thermo-couples nor the IR images of the beryllium sample did show any indication of failure for the module.

The baffle mock-up developed by CEA was loaded with 100 cycles at heat loads of 3.7 MW/m$^2$. No detachment of tiles was observed at this heat load. But after increasing the power density to values of 6.5 MW/m$^2$, five tiles failed in the second load cycle. Fig. 6 shows the IR image of the mock-up immediately before failure of these tiles. Four of them showed only over-heating, while the last one jumped off. Fig. 7 shows this mock-up after failure.

The behaviour of the Be/Cu mock-ups produced by Studsvik Material was different for those parts with 4 mm and those with 8 mm beryllium thickness. The IR camera indicated a significant overheating on 8 mm Be armour areas even at rather low power densities below 4 MW/m$^2$. Such an overheating due to reduced cooling is an indication for a faulty braze layer. On the thin beryllium part (4 mm Be) both samples sustained 100 heating cycles at an absorbed power density of $D_i = 3.5$ MW/m$^2$ without any indication of failure; after an increase of power density to $D_i = 7.0$ MW/m$^2$ they failed by sudden detachment of the beryllium tiles.

It can be summarized that HIPed Be/Cu mock-ups have not yet reached the standards for application in high heat flux components but it seems to be a promising technique for the production of first wall components. At present the limits of this method are reached for heat loads around 5 MW/m$^2$. 

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Fig. 5: Thermal response of a HIPed first wall mock-up 10 s heating/ 10 s cooling (above), and 30 s heating/ 30 s cooling (below)
Diffusion Bonded (hot pressed) mock-ups

Five flat tile mock-ups were produced by diffusion bonding (hot pressing) at ENEA [12]. The production process was as follows: After removal of oxide layers from the beryllium by anodic etching, an interlayer of electrolytic copper was deposited on the activated beryllium. This interlayer which shall minimize the beryllium diffusion was ≈ 10 μm in thickness. The diffusion bonding was performed at 420 °C and 98 MPa for 30 minutes. The temperature limit was determined by the fact that CuCrZr cannot sustain heat treatments above 450 °C. The time was limited to 30 minutes in order to avoid the formation of intermetallic phases.

Heat sink materials were CuCrZr and Glidcop Al25. Some of the mock-ups were castellated to reduce the thermal stresses between Be and Cu. The beryllium thickness for most samples was 4 mm; for one mock-up it was 8 mm. The five samples showed a wide variety of failure limits depending on heat sink material (CuCrZr and DS-Cu) and the existence of castellations. Most samples showed a non-uniform temperature distribution from the beginning i.e. at rather low power densities. These samples failed at power densities below 5 MW/m². An example for
such a sample is given in fig. 8 (heat sink: CuCrZr, no castellations). At a power density of 3.3 MW/m$^2$ a strong over-heating is observed. Although after the test no detachment was visible, it could be detected in an ultra sonic inspection.

Best behaviour of all hot pressed samples was found for the variant with a heat sink made from Glidcop and with castellation in the beryllium which failed at $D_e = 6.8$ MW/m$^2$. At present, diffusion bonding (hot pressing) seems not be a promising method for solutions of high heat flux components. But optimized joining techniques may be appropriate for first wall applications.

4. Summary

Several variants of beryllium - copper mock-ups have been tested in screening tests (steady state heating) and thermal fatigue tests. At present brazing seems to be superior to diffusion bonding or HIPing methods. The "classical" InCuSil braze and the new silver-free CuMnSnCe braze showed a good behaviour during cycling tests. The first test with a beryllium monoblock brazed to a CuCrZr tube shows promising results.

HIPing and diffusion bonding have not yet reached the standards for high heat flux components, but they may be a solution for the first wall.

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The surface chemical states of beryllium and carbon binary systems at elevated temperature were investigated by means of x-ray photoelectron spectroscopy (XPS) and secondary ion mass spectroscopy (SIMS). The XPS measurements revealed that the mixed subsurface layers containing Be and C readily yield Be$_2$C layers by vacuum heating and ion bombardment. The SIMS measurements showed that hydrogen isotope atoms are trapped by three distinct sites; namely Be, C, and O-sites on the sample surface. The SIMS measurements also showed that carbon atoms lose its ability to bind with hydrogen isotope atoms on forming Be$_2$C. It would be a key to control hydrogen inventory when Be and C are used together as PFM.
deposited with arc discharge between two graphite rods (4 mm in diameter) attached to the stainless steel electrodes. The applied voltage and current were 70 V and 40 A, respectively. The thickness of the deposited carbon film was determined as 10 nm with a calibrated quartz oscillator.

The [C(H)-Be] sample was prepared by implantation of C₂H₄ ions into Be plate. The Be plate was cleaned by Ar ion sputtering before use. As for the C₂H₄ ion implantation, the applied acceleration voltage and ion current were 5 kV and 20 μA cm⁻², respectively. The total ion dose was 2 x 10¹⁸ ions cm⁻².

2.2. XPS and SIMS measurements

An UHV system equipped with XPS and SIMS optics was used to measure changes in the surface state of the samples with elevated temperature. The system was evacuated with a sputter ion pump and a turbomolecular pump backed with an oil-sealed rotary pump. The residual pressure of the system was routinely below 7 x 10⁻⁸ Pa.

The sample was mounted on a Ta-heater plate attached to the sample holder made of stainless steel. It could be heated up to 900°C, and its temperature was measured using a Pt/Pt-Rh(13%) thermocouple spot-welded to the backside of the Ta-heater. The sample was heated at a given temperature for 10 min, and was cooled to room temperature to measure XPS spectra. For XPS measurements, MgKα radiation (1253.6 eV) with 400 W power was used as probe. Energy of photoelectrons was measured with use of a cylindrical mirror analyzer (PHI 12-255). The binding energy readout was calibrated by referring the Au4f7/2 peak position to be 83.7 eV.

A conventional ion gun (PHI 04-191) was used for SIMS measurements and also for in-situ ethylene ion implantation. (Ar + D₂) mixed gas was used as source of probe ions for SIMS measurements. Argon and deuterium gases were purchased from Nippon Sanso Co. (guaranteed as 99.999%) and Showa Denko Co. (guaranteed as 98%), respectively. They were used without further purification. Negative and positive secondary ions were measured using quadrupole mass spectrometer (Extranuclear Lab., 19 mm rod, 1.5 MHz).

3. RESULTS

3.1. [C/Be] sample

Fig. 1 shows the temperature dependence of the Be Is XPS spectrum for the [C/Be] sample with vacuum heating. As seen in this figure, no Be Is peak was observed below 500°C, indicating that the Be surface was covered with deposited C layer. A Be Is peak appeared above 600°C at 112 eV with a shoulder at higher binding energy side, indicating that surface Be atoms exist in two or more different chemical states on the sample surface. This point will be discussed later.

![Fig. 1. Change in the Be Is spectrum of [C/Be] sample with elevated temperatures.](image-url)
Fig. 2 shows the temperature dependence of the Cls XPS spectrum for the [C/Be] sample with vacuum heating. A single Cls peak was observed irrespective of vacuum heating conditions. However, it shifted toward the lower binding energy side, in a range from 286 to 282 eV, with increasing temperature to 600°C, and remained at 282 eV above 600°C.

Fig. 3 shows the temperature dependence of the O1s XPS spectrum for the [C/Be] sample with vacuum heating. At room temperature, the O1s spectrum consist of two peaks: namely, one peak was centered at 533 and the other at 531.5 eV. After elevating temperature from 300 to 500°C, those peaks disappeared and a peak at 530 eV, Zn-O, became dominant. This is due to contamination from the overheated electrode partly made of brass. It disappeared above 600°C. On the contrary, new O1s peak appeared at 531.5 eV above 600°C.

Fig. 4 (a) and (b) show examples of positive and negative SIMS spectra for the [C/Be] sample measured at room temperature after annealing at 800°C for 10 min. These spectra were measured at room temperature using (Ar + D2) mixed gas as source of probe ions. Most of the observed peaks in the positive SIMS spectrum could be attributed to the secondary ions relating to Be and probe ions; namely, M/e = 2(D+), 4(D2+), 9(Be+, 10(BeH+), 12(C+ and/or BeD+), 18(Be2+), 20(Ar+), 21(BeC+), 24(C+), and 25(BeO). The species relating to deuterium, such as M/e = 2(D), 14(CD+), 18(OD), 26(CD2+ and/or BeOH+), and 27(BeOD+) were observed as well.
3.2. [C(H)-Be] sample

Fig. 5 shows the temperature dependence of the Be 1s XPS spectrum of the [C(H)-Be] sample with vacuum heating. Broad peaks were observed at various heating temperatures; a feature of shoulders and/or double peaks were apparent. It indicates that Be atoms exist in two or more different chemical states on the surface. This point will be discussed in the following section.

Fig. 6 shows the temperature dependence of the C 1s XPS spectrum for the [C(H)-Be] sample. The C 1s peak showed a shoulder at higher binding energy side below 500°C. The results indicate that carbon atoms on the surface also exist in different chemical states and certain amount of hydrogen atoms are implanted as hydrocarbons. The C 1s spectrum, however, changed to a single peak centered at 282 eV above 500°C.

Fig. 7 shows the temperature dependence of the O 1s XPS spectrum. The spectrum consisted of the single peak centered at 532 eV irrespective of the annealing temperature range, indicating that oxygen on the surface exist as Be-O.

4. DISCUSSION

4.1. Carbon deposited sample, [C/Be]

The C 1s peak shift toward the lower binding energy side with vacuum annealing should be ascribed to the change in the chemical states of deposited carbon atoms. The binding energy of C 1s peak of the as-prepared [C/Be] sample (286.5 eV) was higher than that of graphite. It suggests that the carbon layer contains impurities such as hydrogen, oxygen and/or hydroxide. They reduce the electron density of carbon atoms to increase the apparent binding energy of C 1s electrons. These impurities might be
included by the carbon layer during the deposition from residual gases due to the poor vacuum condition of the sample preparation chamber (1.3 × 10⁻⁴ Pa or higher). The observations for O1s shown in fig. 3 support this interpretation. Namely, the as prepared sample showed two peaks at 533 and 531.5 eV; the former is attributed to adsorbed -COH and the latter to adsorbed -CO₁₀₁¹.

The C1s peak of 285 eV observed after 300°C heating could be attributed to hydrogen containing carbon, and peak of 284 eV to the contaminant type carbon. These observations suggest that most of the impurities included in the deposited carbon layer desorb as volatile gases such as CO, CO₂, H₂O and hydrocarbons during the annealing. The C1s peak of 282 eV appeared above 600°C is identified as metal carbide, indicating that most part of carbon atoms changed to Be-carbide.
Similarly, atomic ratio in the Be-oxide is determined to be [Be] : [O] = 9% : 10% \approx 1 : 1, corresponding to the chemical formula of BeO, which is well known as commonly observed impurity on/in Be.

Concerning the chemical states of Be above 600°C, most of the surface Be was found to exist as Be-carbide for [C/Be] sample. The noticeable changes in XPS spectra above 600°C (Figs.1, 2 and 3) is due to the formation of Be-carbide. This indicates that Be migrates fast to the surface from inner layers in higher temperature region above 500°C and reacts with surface carbon atoms as

\[ C + 2\text{Be} \rightarrow \text{Be}_2\text{C}. \]  

Fig.9 shows a schematic drawing of the formation of surface Be$_2$C layer at elevated temperature: namely, Be migration to the surface becomes much easier with temperature

The observed BeLs spectra could be deconvoluted into three component peaks by the least square spectral fitting calculations. Fig.8 shows the deconvolution of the BeLs spectrum for the [C/Be] sample heated at 600°C for 10 min. In this figure, the solid line shows the observed spectrum and dotted lines calculated component peaks. With respect to the component peaks, the most intense peak centered at 113 eV could be ascribed to Be-carbide. One of the two weak peaks centered at 115 eV could be ascribed to Be-oxide, and the other at 112 eV to pure metallic state. The ratio of integrated peak intensities of these component peaks was [carbide] : [oxide] : [metal] = 5 : 1 : 1. On the other hand, from the ratio of the corresponding photoelectron peak areas, surface atomic composition of Be, C and O after annealing at 600°C was determined as 65%, 23% and 10%, respectively. Taking account of the deconvolution, the atomic ratio of the surface Be-carbide is determined to be [Be] : [C] = 46% : 23% = 1. Therefore, the chemical formula of the Be-carbide should be written by Be$_2$C.
and hence the chemical reaction to form $\text{Be}_2\text{C}$ is enhanced.

4.2. Carbon implanted sample, $[\text{C(H)}-\text{Be}]$

Doubly peaked structure of the Cls spectra below 500°C (Fig. 6) indicates that carbon atoms existed both as carbide and amorphous hydrocarbons. Since Be and carbon atoms did not react easily to form $\text{Be}_2\text{C}$ at around room temperature for the $[\text{C}/\text{Be}]$ sample, the existence of $\text{Be}_2\text{C}$ in the as-prepared $[\text{C(H)}-\text{Be}]$ sample is due to local mixing and reaction of implanted carbon (ethylene ions) with surface Be atoms caused by the energy deposition and/or transfer during ion bombardment. Fig. 10 shows a schematic drawing of the formation of surface mixed layer of H, C, $\text{Be}_2\text{C}$ and BeO owing to the C$_2$H$_4$ ion implantation. The disappearance of the shoulder and the growth of the peak at 283.5 eV indicates that amorphous hydrocarbons on the sample surface decompose to form $\text{Be}_2\text{C}$ according to the reaction (1).

The complicated Be Is spectra as shown in fig. 5 could be deconvoluted into three peaks by the least square spectral fitting calculations. Fig. 11-(a) and -(b) show examples of the deconvolution for the $[\text{C(H)}-\text{Be}]$ sample heated at 300 and 600°C for 10 min, respectively, where solid line shows observed Be Is spectrum and dotted lines calculated component peaks. The ratio of integrated peak intensities of the component peaks was [carbide] : [oxide] : [metal] = 3 : 2 : 0 and 6 : 6 : 1 for the sample heated at 300 and 600°C, respectively.

The surface atomic composition was determined from the ratio of the corresponding photoelectron peak areas as $[\text{Be}] : [\text{O}] : [\text{C}] = 54\% : 28\% : 18\%$ for the sample annealed at 300°C. Oxygen and a part of beryllium form BeO as mentioned above. Excluding this oxide contribution, the oxygen free beryllium to carbon ratio should be $[\text{Be}] : [\text{C}] = 11 : 9$. The deviation from Be$_2$C stoichiometry suggests the existence of excess carbon atoms on the sample surface. This consideration is in accordance with that of the observed Cls spectra below 500°C that indicates the existence of amorphous hydrocarbons. The ratio of $[\text{Be}] : [\text{O}]$ is evaluated to be 32 : 28, being almost equal to unity. Therefore the chemical formula of Be-oxide should be written by BeO.

On the other hand, surface atomic composition is $[\text{Be}] : [\text{O}] : [\text{C}] = 61\% : 26\% : 13\%$ for the sample annealed at 600°C. On account of the spectrum deconvolution that beryllium to carbon ratio is $[\text{Be}] : [\text{C}] = 28\% : 13\% \approx 2 : 1$, the chemical formula of Be-carbide should be written by Be$_2$C. Atomic ratio of Be-oxide is also concluded to be $[\text{Be}] : [\text{O}] = 28\% : 26\% \approx 1 : 1$. The chemical formula of Be-oxide should be written by BeO. The presence of Be-oxide agrees well with the fact that oxygen atoms on the surface exist as Be-O for all the experimental temperatures as shown in fig. 7.
4.3. SIMS measurements

4.3.1. Carbon related species

The negative secondary ions (Fig.4-(b)) of the M/e = 12 and 24 are typical species from carbon and/or graphite, and they should be assigned to C- and C2-, respectively. Accordingly, the peaks at 13 and 14 should be due to CH and CD-, respectively. Although the later (M/e=14) can also be ascribed to CH2, the above conclusion is supported by the absence of the CHD (M/e = 15). Similarly, the peaks at 25 and 26 were also assigned to C3H+ and C3D+, respectively. Then, it is concluded that hydrogen isotope atoms are captured by carbon atoms on the sample surface as well as by Be atoms as shown in Fig.4-(a).

Emission of the negatively charged secondary ions of M/e = 21 (BeC) appears due to Be2C, whose formation was confirmed by the XPS measurements for both the [C/Be] and [C(H)-Be] samples. On the other hand, the absence of the secondary ions such as M/e = 22 and 23 strongly suggests that the Be-carbide can not capture/trap hydrogen isotope atoms.

4.3.2. Oxygen related species

The peak at M/e = 16 could be assigned to O2; the possibility of CD2+ is eliminated by the absence of CHD-. The peaks at 17 and 18 are assigned to OH and OD, respectively. From these observations, it is concluded that hydrogen isotope atoms were captured by oxygen atoms as -OX (X = H and D) on the sample surface. The peak at 19 is assigned to F captured in Be during its refining processes.

5. CONCLUSIONS

It was found by means of XPS and SIMS that the mixed subsurface layers containing Be and C readily yield Be2C layers by vacuum heating and ion bombardment.

The XPS measurements revealed that the Be-carbide formation takes place above 600°C for both the samples. The chemical formula of the Be-carbide is written by Be2C.
As for the carbon evaporated sample, [C/Be], Be atoms diffuse to the sample surface to form carbide. On the other hand, Be$_2$C was observed even for the as-prepared sample of [C(H)-Be], indicating that Be$_2$C would be formed at low temperatures due to the local energy transfer and/or deposition by energetic carbon ion incidence. In addition, Be-oxide inevitably exists on the sample surface because of the strong affinity of Be to oxygen. It could not be completely removed from the surface only by vacuum heating. The stoichiometry of the Be-oxide is Be:O = 1:1, corresponding to the chemical formula of BeO for both samples.

The SIMS measurements showed that hydrogen isotope atoms are trapped by carbon atoms on the sample surface. These observations are very similar to those for graphite. Carbon atoms, however, lose its ability to bind with hydrogen isotope atoms on forming Be$_2$C. It would be a key to control hydrogen recycling and inventory when Be and C are used together as PFM. SIMS measurements also revealed that hydrogen isotope atoms were captured by oxygen atoms as -OX (X = H and D) as well as Be atoms. Therefore, it is concluded that hydrogen isotope atoms are trapped by three distinct sites; namely Be, C, and O-sites on the sample surface.

REFERENCES

7.4 Energy Dependence of Sputtering Yields of Be, Be-C and Be-W Films by Be$^+$- Ions

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The energy dependence measurements of Be, Be-C and Be-W deposited layer sputtering yields by Be$^+$-ions were performed. The ion energy was varied in the range (0.3-5.0) keV. The temperature in the process of irradiation was sustained at the level of 670 K. The mixed layers were prepared by simultaneous sputtering of pair targets, Be and C, Be and W, and Be-targets with Ar$^+$- and Be$^+$ - ions and codeposition of the sputtered atoms on silicon collectors. The codeposited layer thickness was changed in the range of (500-1000) nm. The content of oxygen in the Be, Be-C, Be-W deposited layers did not exceed 20 at. %.

The mixed layer sputtering yields were compared with the experimental and calculated data, obtained for the self-sputtering yields of beryllium and carbon. It was found that the sputtering yields of the Be-C and Be deposited layers by Be$^+$-ions in the energy range (0.3-5.0) keV are within the range between the corresponding self-sputtering yields for Be and C. The sputtering yields for the mixture Be-W are close to the corresponding self-sputtering yields of beryllium.

1. INTRODUCTION

As shown in calculations and experiments, the redeposition process essentially affects the erosion rate of the divertor plate and the first wall materials. The study in detail shows that the errors in determination of the redeposition coefficient result in greater errors in determination of the lifetime for ITER plasma facing components. Until recently, the Be self-sputtering yields were estimated by the computer simulation only. By the end of 1996, the studies of energy, temperature and angular dependences for the Be self-sputtering yield were completed [1-4]. The lifetime of the divertor plates made of beryllium has been calculated, and it has been shown that this material cannot be used under expected thermal loadings onto the ITER divertor[5]. Tungsten and CFC are expected to be used as materials for the divertor and beryllium - as material for the first wall [6]. In this connection, the production of the mixed layers, Be-C, Be-W, will take place upon the first wall. Therefore, a particular attention is paid to the study of mixing layers at present [7]. Recently, study results on sputtering of the mixing layers made from the plasma facing materials by the D$^+$-ions have been reported [8].

In this paper the energy dependence of sputtering yields for Be, Be-C and Be-W deposited layers by Be$^+$-ions in the energy range (0.3-5.0) keV at 670 K is investigated.

2. EXPERIMENTAL TECHNIQUE

The targets of beryllium, TShP-56, graphite, MPG-8, and those of electrolytically-pure tungsten were used in the experiments.

The experiments were carried out using a 180°- and 60°- accelerators. A special ion receiver for the production of the deposited Be, Be-C and Be-W layers by the simultaneous ion sputtering of pair targets, Be+C, Be+W, and Be-tartgets was made.

* This work was supported by the International Atomic Energy Agency.
incident ions
Ar or Be
A scheme of ion receiver is shown in Fig. 1. The targets of Be, graphite and W were fastened at the angle of 60° towards the ion beam direction. The targets of different pair materials were simultaneously sputtered by Be⁺-ions with the energy of 5 keV or by Ar⁺-ions with the energy of 20 keV. The Be⁺-ions were generated within the high temperature-ion source-plasma, where metallic beryllium was used as a working substance. The beryllium and argon ion beam areas were 4.5x10⁻³ m² and 2x10⁻² m², respectively. The plates of monocrystalline silicon, as well as beryllium, TShP-56 trade mark, were used as collector for sputtered target material (See, Fig. 1). A silicon/beryllium collector with 17 mm in diameter was placed at a distance of 25 mm from the target at a scattering angle of about 150°. Atoms sputtered from the target are deposited on the collector.

Figure 1. Scheme of ion receiver for the production of the deposited Be, Be-C, Be-W layers.

The thickness and the roughness of the mixed layers were measured with a profilometer, Sloan Instruments Dektax. The chemical composition of these layers, as well as their thicknesses, were found by the Rutherford back scattering (RBS) technique and by the Auger electron spectroscopy (AES). The RBS-measurements were done in the Van-de-Graaf accelerator, where the channeling He⁺-ions with the energy of 1.5 MeV were scattered at the angle 160° from the targets under study.

The energy dependence studies of the sputtering yields by Be⁺-ions for the deposited layers, Be, Be-C and Be-W, were done with the 180° accelerator. The experimental set-up for irradiation by Be⁺-ions is schematically shown in Fig. 2. The Be⁺-ion flux was equal 4x10⁻¹⁰ m²·s⁻¹. The ion energy was varied in the range 0.3-5.0 keV. The temperature in the process of irradiation was sustained at the level of 670 K.

Figure 2. Schematic drawing of the experimental set-up for irradiation by Be⁺-ions.
The sputtering yields were determined by the weight technique. The sensitivity of a microbalance was equal to 10^{-6} kg. The amount of Be^{+}-ions arriving at the target was taken into account in the calculation of sputtering yields by Be^{+}-ions. The surface microstructures of the deposited Be-films, mixed beryllium-carbon and beryllium-tungsten layers were investigated by means of JEOL-scanning electron microscope before and after irradiation by Be^{+}-ions.

3. EXPERIMENTAL RESULTS


The depth distribution of elements measured by the RBS technique in the Be-C mixed layer deposited on silicon collector is given in Fig. 3. As seen in Fig. 3, a layer including the mixture of Be and C atoms is about 1000 nm thick. In the mixed layer the atoms of beryllium and carbon are uniformly distributed. The concentration of Be atoms is 60 at.%, the amount of carbon is approximately twice lower (26 at.%). The content of oxygen in the mixed layer is varied in depth from 10 at.% to 15 at.%, it being seen that the oxygen enrichment up to 15 at.% is in the surface layer only.

3.2. Microstructures of the mixed layers.

Some typical microphotographs of the mixed Be-C layer surfaces before (a,b) and after (c) Be^{+}-ion bombardment at the energy of 0.5 keV are given in Fig. 5. The films have the fine-grained structure which is clearly manifested after ion etching (Fig. 5c). The film fragment
Table 1
Element composition of the mixed layer surface on silicon after sputtering by Be\(^{+}\)-ions with the energy of 0.5 keV

<table>
<thead>
<tr>
<th>No.</th>
<th>Mixture</th>
<th>Be</th>
<th>C</th>
<th>O</th>
<th>W</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Be</td>
<td>54.8</td>
<td>9.5</td>
<td>35.7</td>
<td>-</td>
</tr>
<tr>
<td>2</td>
<td>Be-C</td>
<td>44.0</td>
<td>29.7</td>
<td>26.4</td>
<td>-</td>
</tr>
<tr>
<td>3</td>
<td>Be-W</td>
<td>39.9</td>
<td>7.3</td>
<td>34.3</td>
<td>18.6</td>
</tr>
<tr>
<td>4</td>
<td>Be-W*</td>
<td>44.3</td>
<td>7.5</td>
<td>19.0</td>
<td></td>
</tr>
</tbody>
</table>

shown in Fig. 5b is about 1 \(\mu\)m thick. The similar structure is observed in the films of Be and Be-W. According to the data measured with profilometer, the roughness of the mixed layer surfaces under study does not exceed (15-20) nm.

### 3.3. Energy dependences of the sputtering yields for the mixed layers.

The determined sputtering yields for the mixed layers, Be-C, Be-W, and for deposited Be films are compared with experimental energy dependences of the self-sputtering yields for beryllium [1,3] and for carbon [9] in Fig. 6. For the Be\(^{+}\)- and C\(^{+}\)-ion energies lower than 0.1 keV, the curves are approximated in accordance with the calculations by Eckstein [10]. One can see that the sputtering yields of the deposited layers Be-C and Be by Be\(^{+}\)-ions are within the area between two curves for the self-sputtering yields, Be-Be\(^{+}\) and C-C\(^{+}\). The sputtering yields for the mixture Be-W by Be\(^{+}\)-ions are close to the beryllium self-sputtering yields. Probably, it is a result of two factors: preferential sputtering of Be-atoms and simultaneous growing of Be layer during the sputtering of the Be-W mixture by Be\(^{+}\)-ions.

Thus, under sputtering of the deposited mixed layers Be-C, Be-W and Be by Be\(^{+}\)-ions, in difference from the results obtained in the experiments on sputtering of the mixed Be-C layers produced under interaction of a deuterium plasma with beryllium [8],
the sputtering yield values are close to the yields of self-sputtering for their components.

4. CONCLUSION

1. In the deposited Be and mixed Be-C, Be-W layers, ~1000 nm thick, produced by simultaneous sputtering of pair targets, Be+C, Be+W, Be+Be, by an Ar+ and Be+- ion beam with a large area, the content of oxygen does not exceed 20 at.%. 
2. It has been found out that the sputtering yields of the deposited Be and mixed Be-C layers by Be+-ions with energy from 0.3 keV to 5.0 keV are within the range between the corresponding self-sputtering yields for their components, Be and C. The sputtering yields for the mixture Be-W are close to the corresponding self-sputtering yields of beryllium.

REFERENCES

The main damaging factors which impact the ITER divertor components are neutron irradiation, cyclic surface heat loads and hydrogen environment. One of the important questions in divertor mockups development is the reliability of beryllium/copper joints and the beryllium resistance under neutron irradiation and thermal cycling. This work presents the experiment, where neutron irradiation and thermocyclic heat loads were applied simultaneously for two beryllium/copper divertor mockups in a nuclear reactor channel to simulate divertor operational conditions.

Two mockups with different beryllium grades were mounted facing each other with the tantalum heater placed between them. This device was installed in the active zone of the nuclear reactor SM-2 (Dimitrovgrad, Russia) and the tantalum block was heated by neutron irradiation up to a high temperature. The main part of the heat flux from the tantalum surface was transported to the beryllium surface through hydrogen, as a result the heat flux loaded two mockups simultaneously. The mockups were cooled by reactor water. The device was lowered to the active zone so as to obtain the heating regime and to provide cooling lifted. This experiment was performed under the following conditions: tantalum heater temperature - 1950°C; hydrogen environment -1000 Pa; surface heat flux density -3.2 MW/m²; number of thermal cycles (lowering and lifting) -101; load time in each cycle - 200-5000 s; dwell time (no heat flux, no neutrons) - 300-2000 s; cooling water parameters: v~ 1 m/s, Tin - 50°C, Pin - 5 MPa; neutron fluence - 2.5x10²⁰ cm⁻² (~8 years of ITER divertor operation from the start up). The metallographic analysis was performed after experiment to investigate the beryllium and beryllium/copper joint structures, the results are presented in the paper.

1. Introduction

The beryllium/copper joints of the divertor components that have active water cooling operate under several damaging factors: neutron irradiation, cyclic surface heat loads and hydrogen environment [1]. For successful development of high heat flux components with beryllium armoring the simulation experiments reflecting the combined damaging factors are required. The approach of in-pile experimental method is described, where neutron irradiation and thermocyclic heat loads were applied simultaneously with hydrogen environment for two beryllium/copper divertor mockups in a nuclear reactor channel to simulate divertor operational conditions.

2. Description of tested mockups

The design of the mockups was selected in view of the main requirements for actively cooled high heat flux components, i.e. armour has to be bonded to the actively cooled heat.

The following materials were used for small-scale mockups:
1) Beryllium as an armour material, four Russian beryllium grades were used - TR-30, TShG-56, DSbG-200, TGP-56;
2) CuCrZr copper alloy for cooling tube;
3) Pure copper as the interlayer between armour material and cooling tube;
4) Copper-based brazing alloy.

Five (20x10x10) mm³ beryllium tiles were brazed with a 1.5-mm-thick saddle copper interlayer and a 16x1.5 mm CuCrZr tube as a set in the vacuum furnace. The general view of the mockup is presented in figure 1 (top - cross-sectional view, bottom - picture).
3. Description of the experimental method

The method of cyclic heat loading under neutron irradiation and hydrogen environment was developed to investigate the influence of combined damaging factors. Two mockups were mounted facing each other in one testing block (figure 2). The (8x40x100) mm$^3$ tantalum heater was placed between them. Gaps between the mockup and tantalum surfaces were provided by slots in copper discs brazed to the mockups tubes. This device was placed in the active zone of the nuclear reactor and the tantalum block was heated by neutron irradiation up to a high temperature. The main part of the heat flux from the tantalum surface was transported to the beryllium surface through dissociated hydrogen (provided by high temperature of tantalum) and the rest part of heat flux was transported via heat radiation, so the heat flux loaded two mockups simultaneously. The mockups were cooled by reactor water. To obtain the thermocycling regime the device was lowered in the active zone and lifted. The scheme of the experiment is presented in figure 3.

The principle possibility of heat transfer via hydrogen was examined by out-pile experiment using Mo and Nb heaters in the hydrogen atmosphere. It was shown that at various hydrogen pressures and heater temperatures the heat flux on the cooled samples was in the range of 50-610 W/cm$^2$ [2]. To estimate the total heating of the heater in the active zone of nuclear reactor the experimental scheme was calculated using Monte-Carlo method.

Measurement of absorbed heat flux was provided by gradient thermocouples in the copper block between beryllium tiles. The temperature of Ta-heater and Be tiles was also measured by thermocouples. The mockups were lifted from the middle to the outside of the active zone with a speed of 1 mm/s (lifting time-350 s).

In the channel of the nuclear 101 thermal cycles were performed - 50 cycles with 200 s loading per each cycle and 51 cycles with 5000 s loading. The experimental conditions and main measured data are presented in table 1.

After testing the mockups were cut from the device. The first inspection did not reveal any damages of materials and joints. The view of the mockup after the test and cutting is presented in figure 4. Top view do not show any cracks of beryllium surface. The oval-shape iridescence-trace proves the axial symmetry and transverse uniformity at heat load. Thin film on the surface is a result of Ta-deposition from hot Ta-heater. Side view demonstrates un-damaged Be and Be/Cu boundary, the side heat loading trace proves the presence of temperature gradient and longitudinal uniformity of heat load.

Several microsections from irradiated and non-irradiated mockups were prepared for further metallographic analysis.

4. Results of metallographic analysis

To investigate the effect of neutron irradiation under cyclic heat loading and hydrogen environment on the behavior of armour and heat sink materials and their joints the metallographic analyses were performed on tested and non-tested samples fabricated under the same conditions and from the same materials.

The beryllium/copper joints were inspected before and after the test by the optical microscopes with a magnification of 100. Figures 5 and 6 show no cracks, voids and changes in the Be/Cu joints and no changes were detected in the width of the joints. The same result was observed in beryllium and copper before and after the test. The irradiated beryllium microsections were chemically etched for more detailed examination. No damages were detected as seen in figure 7.

Figures 5, 6 and 7 show the metallographic analysis results for TShG-56 beryllium grade. The same results were obtained for another beryllium grades (tiles). More detailed analysis of mockup materials and joints is under way.

Fig.2. Two beryllium mockup mounted together for in-pile experiment.
Fig. 3. The scheme of in-pile experiment
Fig. 4. The view of mockup after in-pile experiment (top picture-surface view, bottom picture-side view)
Fig. 5. Beryllium/copper joint before in-pile test.

Fig. 6. Beryllium/copper joint after in-pile test.
Fig. 7. The structure of the Beryllium after the in-pile test.
Table 1.
The results of in-pile experiment with beryllium mockup.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Heat power density, W/cm²</td>
<td>350</td>
</tr>
<tr>
<td>Number of thermal cycles</td>
<td>101</td>
</tr>
<tr>
<td>Time of load at each cycle, s</td>
<td>200 (50 cycles)</td>
</tr>
<tr>
<td>Dwell time (no heat flux, no neutrons), s</td>
<td>5000 (51 cycles)</td>
</tr>
<tr>
<td>Time of reaching of Tsurf max., s</td>
<td>~40</td>
</tr>
<tr>
<td>Cooling water parameters</td>
<td>v~1m/s, Tin=50°C, P=5 Mpa</td>
</tr>
<tr>
<td>Neutron fluence, n/cm²</td>
<td>~2.5x10²⁰</td>
</tr>
<tr>
<td>Hydrogen environment (dissociated hydrogen), Pa</td>
<td>1000</td>
</tr>
<tr>
<td>Temperature of Be surface, C</td>
<td>550</td>
</tr>
<tr>
<td>Temperature of Ta-heater, C</td>
<td>1950</td>
</tr>
</tbody>
</table>

5. Conclusions

This work has demonstrated the principle possibility of the in-pile experiment with actively cooled mockups. It was shown that combined damaging factors such as neutron irradiation, cyclic heat loads and hydrogen environment can be simulated at a time. Two small-scale mockups that simulated the materials, geometry and technique of bonding of divertor components were successfully tested by the developed in-pile method. Several Russian beryllium grades were used in the experiment and all of them successfully survived the test.

This experimental approach can be used to develop high heat flux components with different armour materials (graphite, tungsten, beryllium) for fusion application.

References:

2. Report of ITER RF Home Team #G18TT15/RF 1 F35 24/04/95E.
8. Neutron Irradiation Effects

8.1 Influence of Neutron Irradiation on the Tritium Retention in Beryllium

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1. INTRODUCTION

Carbon-based materials and beryllium are candidates for protective layers on plasma-facing components of fusion reactors. In contact with the D-T-plasma these materials absorb tritium and it is anticipated that tritium retention increases with neutron damage due to neutron-induced traps, leading eventually to tritium inventories which represent a safety problem. Previous investigations indeed show that for carbon-based materials tritium retention increases with neutron damage in the range \(< 0.1\) dpa by one to three orders of magnitude [1, 2]. For beryllium only one data set exists, indicating a gradual increase of tritium retention with neutron damage in the range \(< 40\) dpa by about a factor ten, which is assumed to be due to irreversible changes of the microstructure of the samples [2].

Because of the poor data base for beryllium, the work concentrated on this material. The samples of this study were only irradiated to a fast neutron fluence of \(\sim 1.6 \times 10^{21}/\text{cm}^2 = 1.6\) dpa (previous work \(\leq 40\) dpa [2]). Tritium is loaded into the samples from a stagnant \(\text{T}_2/\text{H}_2\) atmosphere (simulating the D-T-plasma) and afterwards the amount of loaded tritium is determined by purged thermal annealing. Irradiated beryllium contains a huge amount of neutron-produced tritium. To determine the amount of loaded tritium the two types of tritium must be separated, which is possible due to the different release kinetics.

2. SAMPLES

The samples studied were manufactured by Brush-Wellman, material type S200F HIP. Specifications by the manufacturer are [3]:

<table>
<thead>
<tr>
<th>Component</th>
<th>Specification</th>
</tr>
</thead>
<tbody>
<tr>
<td>BeO</td>
<td>0.9 wt%</td>
</tr>
<tr>
<td>Other metallic impurities</td>
<td>0.31 wt%</td>
</tr>
<tr>
<td>Average grain size</td>
<td>7.1 (\mu)m</td>
</tr>
<tr>
<td>Percentage theor. density</td>
<td>99.9 % (measured 99.5 %)</td>
</tr>
</tbody>
</table>

The samples were irradiated in Mol (irradiation Mol F-BSBE1). Irradiations conditions were (samples Z0 were not irradiated) [4]:

<table>
<thead>
<tr>
<th>Samples</th>
<th>U9</th>
<th>U3</th>
<th>U5</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fast fluence ((10^{21}/\text{cm}^2))</td>
<td>1.66</td>
<td>1.51</td>
<td>1.63</td>
</tr>
<tr>
<td>Dpa</td>
<td>(-1.7)</td>
<td>(-1.5)</td>
<td>(-1.6)</td>
</tr>
<tr>
<td>Temperature (\text{°C})</td>
<td>235</td>
<td>485</td>
<td>600</td>
</tr>
</tbody>
</table>

The samples have been analyzed by SEM before and after irradiation [3]. The BeO is observed as small \((\leq 1 \mu\)m) precipitates. About 600 appm He is produced during irradiation, which is at irradiation temperatures \(\sim 485 \text{ °C}\) partly accumulated in small \((\sim 0.1 \mu\)m) bubbles and causes a swelling of \(\sim 0.5\) \%.

3. EXPERIMENTS

The loading and the annealing equipment are contained in two different glove boxes with inert \(\text{N}_2\) atmosphere. During handling and transfer (between the two boxes) the samples were under inert atmosphere.
3.1. Loading

After insertion of the samples (normally an irradiated together with an unirradiated comparison sample) into a $\text{Al}_2\text{O}_3$-capsule, the capsule is evacuated (membrane pump, final pressure ~ 2 mbar) and heated to the loading temperature (normally 850 °C). Then the capsule is filled with the loading gas (normally to a pressure of 2 bar) and closed. After 6 - 7 h loading under stagnant atmosphere, first the heating is stopped, then the capsule is evacuated and the samples are transferred into the annealing capsule.

The loading gas normally was $\text{H}_2 + 5 \text{ ppm T}_2$ (specified value, April 1995). Correcting for tritium decay, the specified activity at the time of this investigation (June 1996) was 4.7 ppm which agrees satisfactorily with a measured value of 4.3 ppm [5].

For the usual loading conditions (850 °C, 2 bar) $1.6 \times 10^7$ Bq tritium are contained in the loading capsule (64 cm$^3$). The amount loaded into the samples was always < $5 \times 10^6$ Bq and $2 \times 10^6$ Bq in the average causing a reduction of the effective tritium activity by ~ 10% in the average. Normally, irradiated and unirradiated comparison samples were loaded together in one run. During loading (under stagnant atmosphere) some neutron-produced tritium may be released causing an increase of the tritium activity in the gas phase. This was checked by a loading run with pure $\text{H}_2$ (2 bar) (table 1). With pure $\text{H}_2$ the loaded tritium in the unirradiated sample ($3.6 \times 10^5 \text{ Bq}$) was at least a factor three less than with $\text{H}_2 + 5 \text{ ppm T}_2$ loading, indicating that the tritium released from the irradiated sample causes an increase of the gas activity of ~ 1 ppm at most.

In addition, essentially no "loaded" tritium was observed in the irradiated sample after pure $\text{H}_2$ loading (factor 20 less than for loading with 5 ppm T$_2$). This proves that a direct transfer of "neutron-produced" into "loaded" tritium within the irradiated samples does not occur.

To summarize:

1. both, the loss (due to sample loading) and the gain (due to release of neutron-produced tritium from the irradiated sample) are small (~ 1 ppm) compared to the initial tritium content of the loading gas. In addition, they compensate each other, therefore the effective tritium of the loading gas is estimated to be $4.5 \pm 1 \text{ ppm}$.

2. during loading essentially no tritium is transferred within the irradiated sample from the "neutron-produced" into the "loaded" state.

3.2 Annealing

Tritium is annealed by purging with He+0.1 % H$_2$ (50 SCCM) and heating the samples slowly with 5°C/min or fastly with 26°C/min to a maximum temperature (mostly 850 °C) and keeping this temperature for a certain time. From the tritium activity measured with an ionization chamber, the release rate and by integrating over the time the total release are determined. In fig. 1 the release curves from the three samples (irradiated, loaded; irradiated, not loaded; not irradiated, loaded) of one test run are shown.

3.3 Determination of loaded tritium in irradiated samples (separation of neutron-produced and loaded tritium)

From the fast neutron fluence ($\sim 1.6 \times 10^{21} \text{ /cm}^2$) the neutron-produced tritium in the irradiated samples is estimated to be $\sim 4 \times 10^9$ Bq/g, which is much higher than the loaded tritium ($\leq 5 \times 10^7$ Bq/g). To allow separation of the two components, therefore first a larger fraction of the neutron-produced tritium is released by at least one annealing (mostly at 850 °C).

Fortunately it turns out that the release kinetics of the residual neutron-produced tritium is quite different from that loaded from the gas phase at temperatures $\leq 950 \text{ °C}$: whereas the release of the residual neutron-produced tritium starts only near 850 °C and decreases very slowly exponential (with $\sim 10 \%$/h) at 850 °C, release of loaded tritium starts already at $\sim 500 \text{ °C}$ and at 850 °C it is essentially completely released within 0.5 h (i.e. at the end of annealing) (fig. 2).
The separation of the two tritium components (neutron-produced and loaded) is based on the different release kinetics. By annealing a loaded, irradiated sample the release of both components is determined. The release characteristic of the the residual neutron-produced tritium is determined by annealing an unloaded irradiated sample with a similar annealing history. The release curve of the unloaded sample is normalized to that of the loaded at the end of annealing (but still at 850 °C), assuming that at this time essentially all loaded tritium has been released (fig. 2). The normalized curve therefore represents the release of residual neutron-produced and the difference of the two curves represents the release of loaded tritium from the loaded, irradiated sample (fig. 3).
Table 1  Summary of experimental results

<table>
<thead>
<tr>
<th>Samples</th>
<th>Temp. (°C)</th>
<th>Neutron-gen.</th>
<th>Neutron-gen.</th>
<th>Irrad. (Bq/g)</th>
<th>Unirrad. (Bq/g)</th>
<th>Ratio irrad/unirrad.</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Irrad.</td>
<td>Unirrad.</td>
<td>Load</td>
<td>Anneal</td>
<td>+loaded</td>
<td>(estimated)</td>
<td></td>
</tr>
<tr>
<td>U.3.2</td>
<td>Z0.1</td>
<td>1</td>
<td>500</td>
<td>---</td>
<td>---</td>
<td>---</td>
<td></td>
</tr>
<tr>
<td>0.1021 g</td>
<td>0.0761 g</td>
<td>2</td>
<td>500</td>
<td>700</td>
<td>1.51E7</td>
<td>1.51E7</td>
<td>-0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3</td>
<td>700</td>
<td>850</td>
<td>---</td>
<td>---</td>
<td>--</td>
</tr>
<tr>
<td></td>
<td></td>
<td>4</td>
<td>850</td>
<td>850</td>
<td>6.78E6</td>
<td>2.87E6</td>
<td>3.83E7</td>
</tr>
<tr>
<td></td>
<td></td>
<td>5</td>
<td>850</td>
<td>850</td>
<td>4.37E6</td>
<td>2.18E6</td>
<td>2.14E7</td>
</tr>
<tr>
<td></td>
<td></td>
<td>6</td>
<td>850</td>
<td>850</td>
<td>3.42E6</td>
<td>2.07E6</td>
<td>1.33E7</td>
</tr>
<tr>
<td></td>
<td></td>
<td>7</td>
<td>850</td>
<td>850</td>
<td>8.38E6</td>
<td>3.58E6</td>
<td>4.70E7</td>
</tr>
<tr>
<td></td>
<td></td>
<td>8</td>
<td>850</td>
<td>850</td>
<td>1.72E6</td>
<td>1.61E6</td>
<td>1.07E6</td>
</tr>
<tr>
<td>U.3.3</td>
<td>Z0.1</td>
<td>1</td>
<td>500</td>
<td>---</td>
<td>---</td>
<td>---</td>
<td>--</td>
</tr>
<tr>
<td></td>
<td>Z0.2</td>
<td>2</td>
<td>500</td>
<td>700</td>
<td>1.53E7</td>
<td>1.34E7</td>
<td>3.70E7</td>
</tr>
<tr>
<td></td>
<td>Z0.3</td>
<td>3</td>
<td>700</td>
<td>850</td>
<td>2.49E7</td>
<td>2.43E7</td>
<td>1.16E7</td>
</tr>
<tr>
<td>U.9.4</td>
<td>Z0.2</td>
<td>4</td>
<td>850</td>
<td>850</td>
<td>3.08E6</td>
<td>1.64E6</td>
<td>2.84E7</td>
</tr>
<tr>
<td></td>
<td>Z0.3</td>
<td>2</td>
<td>500</td>
<td>850</td>
<td>7.24E6</td>
<td>6.60E6</td>
<td>6.79E6</td>
</tr>
<tr>
<td></td>
<td>Z0.4</td>
<td>3</td>
<td>700</td>
<td>850</td>
<td>2.96E6</td>
<td>2.70E6</td>
<td>2.78E6</td>
</tr>
<tr>
<td>U.9.5</td>
<td>Z0.3</td>
<td>4</td>
<td>850</td>
<td>850</td>
<td>6.78E6</td>
<td>2.71E6</td>
<td>4.08E7</td>
</tr>
<tr>
<td>U.9.5</td>
<td>Z0.4</td>
<td>1</td>
<td>500</td>
<td>850</td>
<td>1.31E6</td>
<td>1.15E7</td>
<td>1.38E7</td>
</tr>
<tr>
<td>0.0119 g</td>
<td>0.0598 g</td>
<td>2</td>
<td>500</td>
<td>850</td>
<td>1.66E6</td>
<td>8.04E5</td>
<td>7.16E7</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3</td>
<td>850</td>
<td>850</td>
<td>8.30E6</td>
<td>7.13E5</td>
<td>9.84E6</td>
</tr>
<tr>
<td></td>
<td></td>
<td>4</td>
<td>850</td>
<td>850</td>
<td>8.30E6</td>
<td>7.13E5</td>
<td>9.84E6</td>
</tr>
</tbody>
</table>

Estimated error: ±1E6 ±4E5

Loading: H₂ + 5 ppm T₂, 2 bar, 6-7 h; if not otherwise specified
3.4 Determination of loaded tritium in unirradiated sample

The unirradiated comparison sample is loaded together with the irradiated sample. Because the unirradiated sample contains no neutron-produced tritium, the determination of the loaded tritium is straightforward (consisting essentially in subtracting the system background). But it should be mentioned that the amount of tritium loaded into the unirradiated samples is pretty low (<3 x 10^6 Bq/g) and correspondingly the error (due to uncertainties in the background correction) is relatively high (~ 4 x 10^5 Bq/g).

4. RESULTS AND DISCUSSION

In fig. 3 the the specific release (per mass unit) of loaded tritium from an irradiated and an unirradiated sample is shown. It is evident that the irradiated sample contains much more loaded tritium than the not irradiated sample.

In table 1 the main results of this investigation are summarized.

Before discussing the other data, the results of the loading in a pure H₂ atmosphere (table 1) shall be considered. Compared to the normal loading (H₂ + 5 ppm T₂), the loaded inventory in the unirradiated sample (~ 4 x 10^5 Bq/g, this corresponds to the experimental uncertainty) is about a factor three and that of the irradiated sample about a factor twenty lower. From this we conclude that during loading

1. no neutron-produced tritium is transferred over the gas phase into the unirradiated sample and
2. no tritium is transferred within the irradiated sample from the "neutron-produced" into the "loaded" state.

This proves that the loaded tritium in both, the irradiated and the unirradiated samples, originates from the tritium content (5 ppm) of the loading gas.

From the data of table 1 it can be seen that

1. after 6 - 7 h at 850 °C not all traps are saturated (loading at 950 °C is higher than at 850 °C)
2. for both, the irradiated and the unirradiated sample, the loaded tritium decreases from run to run, indicating partial annealing of the traps at 850 °C and especially at 950 °C.

The most important result of this investigation is that for beryllium, similar as for carbon-based materials, the loaded tritium in the irradiated samples (~ 1.6 dpa) is definitely higher than in the unirradiated samples, indicating that tritium traps are generated by fast neutrons. The ratio of loaded tritium in the irradiated to that in the unirradiated samples varies between 3 and 43 and amounts to about 20 in the average (table 1 and fig. 3).

In table 2 average values of loaded tritium have been transferred into tritium (T)/beryllium (Be) atomic ratios (1 Bq/g = 0.84 x 10^14 T/Be) and have been corrected for the isotopic H/T ratio of the loading gas (2 x 10^5 in this work).

Tab. 2 Tritium retention in unirradiated and irradiated beryllium and graphite

<table>
<thead>
<tr>
<th>Material</th>
<th>Loading atmosphere</th>
<th>Loaded Tritium</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Unirrad.</td>
</tr>
<tr>
<td>Be</td>
<td>H₂ + 5 ppm T₂, 2 bar, 850 °C, 6-7 h</td>
<td>(Bq/g)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1 x 10⁴</td>
</tr>
<tr>
<td></td>
<td></td>
<td>~3 x 10⁻⁷</td>
</tr>
<tr>
<td>T₂, 0.8 bar, 850 °C, 10 h</td>
<td>(T/C)</td>
<td>~1 x 10⁻⁴</td>
</tr>
</tbody>
</table>

*Corrected for isotopic ratio

For both, the unirradiated and the irradiated beryllium samples, the estimated loaded tritium in a pure T₂ atmosphere is very high and certainly much higher than in graphite [2].

5. REFERENCES


8.2 Mechanical Performance of Irradiated Beryllium Pebbles

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For the Helium Cooled Pebble Bed (HCPB) Blanket, which is one of the two reference concepts studied within the European Fusion Technology Programme, the neutron multiplier consists of a mixed bed of about 2 and 0.1-0.2 mm diameter beryllium pebbles. Beryllium has no structural function in the blanket, however microstructural and mechanical properties are important, as they might influence the material behavior under neutron irradiation. The EXOTIC-7 as well as the "Beryllium" experiments carried out in the HFR reactor in Petten are considered as the most detailed and significant tests for investigating it.

This paper reviews the present status of beryllium post-irradiation examinations performed at the Forschungszentrum Karlsruhe with samples from these irradiation experiments, emphasizing the effects of irradiation on essential material properties and trying to elucidate the processes controlling the property changes.

The microstructure, the porosity distribution, the impurity content, the behavior under compression loads and the compatibility of the beryllium pebbles with lithium orthosilicate (Li$_4$SiO$_4$) during the in-pile irradiation are presented and critically discussed. Qualitative information on ductility and creep obtained by hardness-type measurements are also supplied.

1. Introduction

For the Helium Cooled Pebble Bed (HCPB) Blanket the neutron multiplier consists of a mixed bed of about 2 and 0.1-0.2 mm diameter beryllium pebbles. The main structure of the pebble bed is given by the larger pebbles with a packing factor of 63.3%. In the space between them are placed the smaller beryllium pebbles with a packing factor of 17.5%.

Both kind of pebbles are fabricated by melting, however for the larger ones a relatively inexpensive intermediate product of the beryllium fabrication route (Brush-Wellmann Company) has been chosen [1]. Because of their poorly defined production conditions, the 2 mm Be pebbles are usually characterized by a large scatter in the microstructural and mechanical data. Therefore, a relatively large number of both unirradiated and irradiated pebbles were analyzed in order to get representative data necessary for characterizing the material as far as possible before its use in a nuclear fusion environment.

The EXOTIC-7 [2] as well as the "Beryllium" [3] experiments carried out in the HFR reactor in Petten are considered as the most detailed and significant tests for investigating the beryllium pebble response under neutron irradiation. The beryllium irradiated in both the EXOTIC-7 and the "Beryllium" experiments consists of a mix of about 2 mm and 0.08-0.18 mm diameter beryllium pebbles. However, while in EXOTIC-7 the beryllium pebbles were - during irradiation - in contact with Li$_4$SiO$_4$-pebbles, in the "Beryllium" experiment mainly pure beryllium pebble beds were irradiated.

For some post-irradiation examinations (PIE) the components of the EXOTIC-7 mixed beds had to be separated. The 2 mm diameter Be pebbles were separated from the small (0.1-0.2 mm) Li$_4$SiO$_4$ and Be pebbles by sieving. The small Li$_4$SiO$_4$ and Be pebbles were separated by a wet procedure using an organic liquid with a density in-between that of the Li$_4$SiO$_4$ and Be pebbles. The visual inspection of the Li$_4$SiO$_4$ pebbles showed that only a small number of pebbles were fractured, whilst the beryllium pebbles look almost all intact.
2. Microstructural Analysis

The bigger unirradiated beryllium pebbles usually show a relatively large number of indentations on their external surface. This is probably due to the fact that during the fabrication process very hot (or still partially molten) beryllium pebbles come in contact with cold and already solidified ones. With optical microscopy, a quite strong variation of coarse pores was observed. Some pebbles show big voids which seemed to be generated during the cooling phase of the fabrication process. Relatively often a coarse porosity with a pore size of 0.1-0.2 mm has been observed. On the other hand, most of the pebbles reveal a very small micro-porosity usually oriented along the crystal axis showing a very fine dendritic or cellular structure. Fig. 1 shows a typical porosity distribution in the 2 mm beryllium pebbles.

![Porosity distribution in 2 mm pebbles.](image)

In general, it has been observed that the porosity of small pebbles is always smaller than that of the bigger ones, which clearly confirms that coarse porosity is generated during the cooling phase of the fabrication process. Furthermore, a large number of pebbles presents, near the external surface, a dense region the depth of which usually reaches 0.2-0.3 mm as shown in Fig. 2.

![Cross-section of a 2 mm pebble.](image)

The metallographic structure of both smaller and bigger pebbles shows the presence of large grains, in some cases as large as the pebble diameter [4]. In Fig. 3 a typical grain size distribution for the 2 mm Be-pebbles is presented.

![Grain size distribution in the 2 mm pebbles.](image)
From the chemical point of view, insoluble impurities have been usually observed on the grain boundaries, while iron and chrome are almost exclusively present in solid solution in the beryllium-matrix. The external surface of the large unirradiated Be pebbles is usually covered by a 2 μm thick SiO₂ layer and/or a fluorine layer (probably BeF₂) the thickness of which is generally lower than 2 μm. Two extraneous phases are present in almost all analyzed pebbles.

Mostly a round bright phase looking like an eutectic stored in the beryllium matrix and occasionally a dark square phase which appears as a primary precipitated phase has been observed. The dominant precipitated phases are Be₁₃Mg, Be₁₃(Mg, Zr, U), Mg₂Si and Al₂O₃ [5]. The concentration of the main impurities is shown in Tabs. 1 and 2 for the 2 mm and the 0.1-0.2 mm diameter pebbles respectively.

| Table 1 Main impurity concentrations in the 2 mm pebbles (data in wt%). |
|------------------|-----|-----|-----|-----|-----|
| BeO   | F   | C   | Mg  | Fe  |
| 0.078 | 0.0585 | 0.05 | 0.16 | 0.073 |
| Al    | Si  | Zr  | U   | Cr  |
| 0.025 | 0.0205 | 0.0125 | 0.007 | 0.015 |

| Table 2 Main impurity concentrations in the 0.1-0.2 mm pebbles (data in wt%). |
|------------------|-----|-----|-----|-----|-----|
| BeO   | F   | C   | Mg  | Fe  |
| 0.34  |     | 0.07 | <0.01 | 0.09 |
| Al    | Si  | Zr  | U   | Cr  |
| 0.04  | 0.03 |     |     |     |
Because of the relatively low irradiation temperatures in both EXOTIC-7 and "Beryllium" experiments (T_{irr}=410-550 °C), no remarkable thermally induced changes in the beryllium structure (macroscopic and microscopic) have been observed after the neutron irradiation except the almost complete disappearance of the porosity fraction in the range 230-300 μm. However, although the fast neutron fluence was almost the same in both experiments (Φ≈1.3×10^{21} cm^{-2}; E_{n}>1 MeV), the beryllium pebbles from EXOTIC-7 showed strong irradiation induced structural changes. In particular a new type of porosity was observed in a boundary layer of the pebbles not deeper than 40-50 μm. The new pores are of an oblong shape, are oriented along the crystals and have diameters of up to 2 μm as shown in Fig. 4. This new porosity is essentially due to the contact between beryllium and Li₄SiC₄ during irradiation. According to previous studies [6] it is in fact expected that, if beryllium is in direct contact with ceramics during irradiation, a fraction of the tritium as well as helium ions produced in the Li₄SiC₄ during irradiation is implanted in a surface layer of beryllium the depth of which is about 40μm.

Etching the microsections reveals a polygonization pattern consisting of bundles of oblong lines, mostly aligned in the same direction as shown in Fig. 5. Polygonization is the first step in subgrain formation, and is initiated by an array of step dislocations towards small-angle grain boundaries. At higher temperatures, this gives rise to rounded subgrain shapes, which already existed in the structure of the unirradiated pebbles. Along with the line structures of polygonization, etching causes a dense network of etching pits. A striking phenomenon are narrow strips of approx. 2 μm width on both sides of the grain boundaries, which remain free from etching attack. This phenomenon is almost identical to that observed in the UO₂ fuel elements of fission reactors with the only difference that, in case of beryllium, instead of the fission product gases, helium precipitated from the Be (n, 2n) ²He reaction is present.

Fig. 4 New porosity due to the contact between Be and ceramics.

Fig. 5 Poligonization patterns in the pebbles.
3. Mechanical Behavior

The mechanical behavior of a significant number of unirradiated pebbles with the larger diameter has been investigated by submitting them to compressive loads of up to 1600 N at room temperature. The plastic deformations of the pebbles have been measured and correlated with the applied loads. Relatively large variations have been observed in the mechanical response of the pebbles. However, probably due to the very small amount of BeO impurities, all the pebbles showed a high ductility at room temperature. Pebbles loaded up to 400 N show diameter reductions up to 13% but, in spite of evident large plastic deformations, no fracturing or crack formation was registered [4]. On the other hand, pebbles loaded with 800 N (diameter deformation up to 25%) or more, reveal cracks on their “meridian” planes [4].

Figs. 6 to 8 show beryllium pebbles which were subjected to 400, 800 and 1600 N respectively.

Fig. 6 Pebble subjected to a 400 N load.

Fig. 7 Pebble subjected to a 800 N load.

Fig. 8 Pebble subjected to a 1600 N load.
Microhardness measurements were performed in the microsections prepared by metallographic techniques. On the basis of hardness levels, techniques were developed to record the tendencies of irradiation-induced changes in the mechanical properties, i.e., plasticity and elasticity. The hardness measurements were initially conducted with a holding time of 5 s. Afterwards, the same position was subjected to pressure once again, but with a holding time of 60 s. The increase in area, expressed in percent, produced by the second impression was used as a benchmark for the subsequent evaluation of the plastic deformation of the material. The permanent deformation of the hardness impression is in fact a consequence of both plastic and elastic deformation fractions. The concave dishing of the sides of the impression square is a measure of the elastic reset forces, comparable to the compression limit of the material. The unit of measurement used was the percentage difference in areas between the impression calculated from diagonals of the impression and the real impression as determined by microscopy analysis [7].

Although the applied load was always the same all over the sample, in some cases different microhardness impression shapes have been obtained. This was due to the different orientation of the beryllium grains, what causes an anisotropic behavior of the material itself. Because of this fact the pebbles were characterized by a quite large scatter in the mechanical data. Therefore, a very large number of pebbles was analyzed to be able to get statistically representative data.

The mechanical properties of the pebbles from the “Beryllium” experiment did not significantly change during irradiation: only an increase of less than 5% of their elastic behavior and a decrease of less than 10% of their plastic behavior has been observed after irradiation. On the contrary, pebbles from EXOTIC-7 show an increase up to about 30% of their elastic behavior range and a decrease up to about 50% of their plastic behavior range after irradiation. The decrease of the plasticity is much more evident in the smaller pebbles due to the fact that the implantation depth of tritium coming from ceramics (~40-50 µm) is of the same order of magnitude as the pebble radius (~100-200 µm), which results in a higher implantation efficiency than in the case of the larger Be pebbles.

4. Summary

The behavior of both unirradiated beryllium pebbles and beryllium pebbles irradiated in the EXOTIC-7 as well as in the “Beryllium” experiments has been investigated. The bigger unirradiated beryllium pebbles usually show a relatively large number of indentations on their external surface. Relatively often a coarse porosity with a pore size of 0.1-0.2 mm has been observed. On the other hand, most of the pebbles reveal a very small micro-porosity usually oriented along the crystal axis showing a very fine dendritic or cellular structure. The metallographic structure of both smaller and bigger pebbles shows the presence of large grains, in some of the smaller pebbles as large as the pebble diameter. Two extraneous phases are present in almost all the analyzed pebbles. Because of the relatively low irradiation temperatures in both EXOTIC-7 and “Beryllium” experiments no remarkable thermally induced changes in the beryllium structure (macroscopic and microscopic) have been observed after the neutron irradiation. However, although the fast neutron fluence was almost the same in both experiments, the beryllium pebbles from EXOTIC-7 showed strong irradiation induced structural changes due to the contact with ceramics during the irradiation. From the mechanical point of view it has been observed that, probably due to the very small amount of BeO impurities, all the pebbles showed a high ductility at room temperature. Pebbles loaded up to 400 N show diameter reductions up to 13% but, in spite of evident large plastic deformations, no fracturing or crack formation was registered. The mechanical properties of the pebbles from “Beryllium” experiment did not significantly change after irradiation. On the contrary, pebbles from EXOTIC-7 show an increase up to about 30% of their elastic behavior range and a decrease up to about 50% of their plastic behavior range. The decrease of the plasticity is much...
more evident in the smaller pebbles due to the fact that the implantation depth of tritium coming from ceramics is of the same order of magnitude as the pebble radius, which results in a higher implantation efficiency than in the case of the larger Be pebbles.

Acknowledgement

The authors would like to thank E. Kaiser and F. Weiser of the FZK-Hot Cells for performing the measurements. This work was performed in the framework of the Forschungszentrum Karlsruhe Fusion Project and is supported by the European Union within the European Fusion Technology Program.

References


8.3 Tensile and Fracture Toughness Test Results of Neutron Irradiated Beryllium

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Tensile and fracture toughness test results of four Beryllium grades are reported here. The flow and fracture properties are investigated by using small size tensile and round compact tension specimens. Irradiation was performed at the BR2 material testing reactor which allows various temperature and irradiation conditions. The fast neutron fluence (>1 MeV) ranges between 0.65 and 2.45 \(10^{21}\) n/cm\(^2\). In the meantime, un-irradiated specimens were aged at the irradiation temperatures to separate if any the effect of temperature from irradiation damage. Test results are analyzed and discussed, in particular in terms of the effects of material grade, test temperature, thermal ageing and neutron irradiation.

1. INTRODUCTION

Irradiation embrittlement of nowadays ductile beryllium grades is investigated here as part of the European Fusion Technology contribution for ITER. Four beryllium grades, namely the S65 and S200 obtained either by hot (HIP) or vacuum isostatic pressing (VHP) were investigated. Tensile tests allow to determine the effect of radiation on the flow properties, i.e., the yield stress, tensile strength and ductility. On the other hand, fracture toughness tests allow to characterize the fracture resistance of the above mentioned materials. Irradiation is known to induce a material hardening and a loss of ductility which result in a loss of fracture resistance. Therefore, small tensile and round compact specimens were irradiated in the BR2 material research reactor in various temperature and fluence conditions. Beside irradiation, some specimens were heat treated to separate if any the effect of temperature from irradiation damage.

2. MATERIALS

Starting from impact grinded powder, four beryllium grades are produced: axial vacuum hot pressed (VHP) S-200-F and S-65 and direct hot isostatic pressed (HIP) S-200-FH and S-65-H. All specimens were manufactured by Brush Wellman [1]. A series of samples was aged at various temperatures in order to simulate the thermal conditions prevailing in the reactor. The nominal chemical composition and tensile properties are reported in Table 1 and 2. The four materials differ mainly by their Be- and BeO-contents. The HIP process results in higher strength (yield and ultimate stress).

<table>
<thead>
<tr>
<th>Material</th>
<th>Be (wt.%)</th>
<th>BeO (wt.%)</th>
<th>Fe (wt.%)</th>
<th>C (wt.%)</th>
<th>Al (wt.%)</th>
<th>Mg (wt.%)</th>
<th>Si (wt.%)</th>
<th>Others</th>
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<td>0.10</td>
<td>0.08</td>
<td>0.04</td>
<td>0.02</td>
<td>0.03</td>
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<td>0.03</td>
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<td>0.02</td>
<td>&quot;</td>
</tr>
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<td>0.6</td>
<td>0.06</td>
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<td>0.02</td>
<td>&lt;0.01</td>
<td>0.03</td>
<td>&quot;</td>
</tr>
</tbody>
</table>

Table 1
Chemical analysis of the four beryllium grades (in wt. %) [1].
Table 2
Density, average grain size and tensile properties at ambient of the four beryllium grades [1].

<table>
<thead>
<tr>
<th>material</th>
<th>theoretical density (%)</th>
<th>average grain size (μm)</th>
<th>specimen orientation</th>
<th>yield stress (MPa)</th>
<th>UTS (MPa)</th>
<th>total elongation (%)</th>
</tr>
</thead>
<tbody>
<tr>
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<td>7.1</td>
<td>Z</td>
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</tr>
<tr>
<td>S200 VHP</td>
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<td>L</td>
<td>261.3</td>
<td>377.1</td>
<td>2.1</td>
</tr>
<tr>
<td>S65 HIP</td>
<td>99.9</td>
<td>8.4</td>
<td>X</td>
<td>342.0</td>
<td>509.5</td>
<td>4.8</td>
</tr>
<tr>
<td>S65 VHP</td>
<td>99.8</td>
<td>6.6</td>
<td>Z</td>
<td>339.9</td>
<td>492.3</td>
<td>3.6</td>
</tr>
</tbody>
</table>

3. EXPERIMENTAL

3.1. Specimens

Specimens were irradiated in the BR2 Materials Testing Reactor for a period of 2340 hours. Irradiation conditions are such that irradiation temperatures range from 185 to 610 °C while cumulated fast neutron fluence \( E_n > 1 \text{MeV} \) is between 0.75 to \(2.5 \times 10^{21} \text{n/cm}^2\). To improve the temperature uniformity, the specimens are submerged in a peritectic NaK liquid metal bath (60 at% Na, 40 at% K). The aged samples were also immersed in a perlite NaK bath to receive a temperature/time profile in a furnace similar to the one experienced into the reactor.

Specimen size is selected to accommodate the space available for irradiation in the BR2 reactor. Therefore, small tensile specimens of initial diameter \( D_0 = 3.2 \text{mm} \) and initial gauge length \( L_0 = 13.2 \text{mm} \) were selected. For fracture toughness determination, a small round compact tension specimen of 8 mm thickness was selected. This was supported by two preliminary tests which resulted in fracture toughness values within the ASTM validity requirements. Both geometries are shown in Figure 1.

The VHP compact tension specimens are machined in the weakest L-R orientation i.e., the crack front is parallel to the axial direction of pressing (L) and propagates in the radial (R) direction. For the HIP compact specimens, which are more isotropic, the L-T orientation was selected, T being the thickness or minor dimension of the HIP rectangular billets.

![Figure 1. Specimen geometries: round compact tension (W=16mm, B=8mm a_o=8mm) and tensile (D_0=3.2mm, L_0=13.2mm).](image)

It should be noted that the compact tension specimens were precracked prior irradiation.

3.2. Irradiation conditions

Three temperature zones of the irradiation capsule were initially aimed: 200, 400 and 600°C. The actual temperature distribution is shown in Figure 2. The specimen geometry and location, the irradiation temperature and the resulting neutron irradiation damage are depicted in Figure 3. The
irradiation damage is expressed in terms of displacement per atom (dpa), helium production (appm He) and fast neutron fluence. More details on neutron calculations can be found elsewhere [3].

As can be seen, the neutron induced damage can be equally expressed in dpa, appm He or neutron fluence.

Figure 2. Temperature distribution in the reactor. The three levels of irradiation temperatures nominally selected are 200, 400 and 600°C.

Figure 3. Neutron damage and temperature distribution of the tensile and compact tension specimens.
3.3. Tensile testing

Tensile tests on un-irradiated specimens were performed at CRM using a servohydraulic MTS 810 tensile machine. Irradiated samples were tested at SCK•CEN with an Instron servohydraulic tensile machine. In the unirradiated condition, the tests were first performed attaching an extensometer on the gauge length for elongation measurement (samples tested at 185°C). However, the results obtained by measuring the total displacement (including the loading system) are in very good agreement with data obtained with extensometer. Furthermore, the total elongation allowed by the extensometer is limited to 1 mm only. Therefore, the subsequent tests were performed without using the extensometer. In the irradiated condition, all tests were carried out without extensometer.

Prior to testing, all irradiated samples were measured to determine the initial dimensions, \(D_0\) and \(L_0\). In the unirradiated condition, only the specimen diameter \(D_0\) is measured, \(L_0\) is taken equal to its nominal value, 13.2 mm.

The specimens are first maintained at least 30 min. at the testing temperature and further tested under displacement control. The loading rate is 0.2mm/min, which corresponds to a strain rate of \(\dot{\varepsilon}=2.5 \times 10^{-4} \text{ s}^{-1}\). However, the unirradiated samples instrumented with extensometer and tested at 185°C were loaded at a strain rate of \(\dot{\varepsilon}=5.5 \times 10^{-4} \text{ s}^{-1}\) up to 1%-elongation and 12.5 \(10^{-4} \text{s}^{-1}\) beyond.

The yield stress is measured at 0.2% permanent deformation. In some situations, however, the specimen fails before the 0.2% elongation. In these cases, the yield equals the ultimate tensile strength. The various flow parameters are derived according to Figure 2:
- yield stress, \(\sigma_y\);
- ultimate tensile strength, UTS (also called tensile strength);
- uniform elongation, \(\varepsilon_u\);
- total elongation, \(\varepsilon_t\);
- reduction of area, RA.

The fracture stress was also determined.

3.4. Fracture toughness testing

All specimens were fatigue precracked according to ASTM E399 standard [3]. Because of the brittle character of Be-alloys, precracking is carried out in both tension and compression. Typical values of the applied loads are 0.89 and 2.2 kN in tension and compression, respectively. The maximum stress intensity factor during fatigue precracking was maintained below 9MPa\(\sqrt{\text{m}}\). With a loading frequency of 20 Hz, the number of cycles was 50000 to 300000. The crack length is measured optically after testing. It is found that the "crack length-to-width" ratio, \(a/W\), is close to 0.5. However, in the irradiated condition, the initial fatigue crack length is not yet measured. In the present evaluation, \(a_0\) is assumed equal to 8mm. This is supported by crack length measurements performed on 77 unirradiated samples showing that \(a_0=8.02\pm0.46\) mm. The derived fracture toughness values are thus accurate within \(\pm10\%\) uncertainty.

Originally, the fracture toughness tests were planned to be performed according the ASTM E399 standard [3]. However, while an extensometer is mounted to measure the crack mouth opening displacement of un-irradiated specimens, such measurement is not used for the irradiated samples. However, as irradiation induces hardening embrittlement, the load versus displacement (total displacement including the loading system) is linear and therefore only the load at fracture is used to
determine the fracture toughness. Furthermore, the contribution of the loading system is small compared to the crack mouth opening displacement.

In the un-irradiated condition, most of the tests performed at temperatures below 250°C exhibit a linear "load versus displacement" behaviour. The stress intensity factor, $K$, is determined using the relation:

$$K = \frac{F}{B \sqrt{W}} f \left( \frac{a}{W} \right)$$

where:

$$f \left( \frac{a}{W} \right) = \frac{\left( \frac{2 + a}{W} \right)}{\left( 1 - \frac{a}{W} \right)^{2}} \left( 0.76 + 4.8 \left( \frac{a}{W} \right) - 11.58 \left( \frac{a}{W} \right)^{2} + 11.43 \left( \frac{a}{W} \right)^{3} - 4.08 \left( \frac{a}{W} \right)^{4} \right)$$

$F$ is the load at fracture, $a$ is the crack length, $W$ in the specimen width and $B$ is the specimen thickness.

At higher test temperatures, the load-displacement trace becomes non linear. In this case, the $J$-integral concept is used. The determination of fracture toughness from the compact tension specimen is standardized [4]. From the load versus displacement record, the $J$-integral can be calculated using the relation:

$$J = J_{el} + J_{pl} = \frac{K^2 (1-v^2)}{E} + \frac{\eta U_{pl}}{B (W-a)}$$

where $E$ is the Young modulus, $v$ is the Poisson ratio, $\eta$ is a factor equal to $\eta=2+0.522 \left( \frac{1}{1-a/W} \right)$ and $U_{pl}$ is load-displacement area as shown on Figure 5.

The elastic constants, the Young modulus and Poisson ratio, are taken from literature [5]. Poisson ratio, $v$, is taken equal to 0.1, independent of testing temperature. The Young modulus can be represented by the fitting equation:

$$E=310-0.095833 T+3.75 \times 10^{-4} T^2 -7.29167 \times 10^{-7} T^3$$

where $T$ is the test temperature.

4. RESULTS AND DISCUSSION
4.1. Tensile data

Two interesting parameters are examined to investigate the effect of ageing and irradiation: the material strength (measured by the yield or ultimate stress) and the material ductility (reduction of area or total elongation). These data are summarized in Tables 3 and 4 for the as-received and aged samples, respectively.

Table 3
Tensile test results of the unirradiated as received materials. \( \sigma_y \) is the yield stress, UTS is the tensile strength, \( \varepsilon_u \) is the uniform elongation, \( \varepsilon \) the total elongation and RA the reduction of area.

<table>
<thead>
<tr>
<th>Material</th>
<th>test ( T ) (°C)</th>
<th>20</th>
<th>20</th>
<th>185</th>
<th>230</th>
<th>310</th>
<th>455</th>
<th>540</th>
<th>605</th>
</tr>
</thead>
<tbody>
<tr>
<td>S200 HIP</td>
<td>( \sigma_y ) (MPa)</td>
<td>353</td>
<td>356</td>
<td>333</td>
<td>320</td>
<td>251</td>
<td>221</td>
<td>174</td>
<td>137</td>
</tr>
<tr>
<td></td>
<td>UTS (MPa)</td>
<td>445</td>
<td>439</td>
<td>387</td>
<td>345</td>
<td>291</td>
<td>242</td>
<td>215</td>
<td>141</td>
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<tr>
<td></td>
<td>( \varepsilon_u ) (%)</td>
<td>3.7</td>
<td>4.8</td>
<td>10.8</td>
<td>11.4</td>
<td>8.6</td>
<td>6.4</td>
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<tr>
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<td>( \sigma_y ) (MPa)</td>
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<td>271</td>
<td>241</td>
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<td>206</td>
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<td>364</td>
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<td>305</td>
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<td>29.4</td>
<td>28.8</td>
<td>24.5</td>
<td>20.1</td>
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</table>

Within the experimental uncertainties, examination of these data indicates that:
1. All Be-grades exhibit a similar behaviour, except the S200 VHP.
2. There is no significant effect of ageing, particularly at test temperatures lower than 455°C.
3. As expected, the HIP grades results in higher strength as compared to the VHP materials.
4. The yield stress and UTS decrease with increasing test temperature while ductility increases up to 310°C and then, a loss of ductility is observed for higher temperatures.

At room temperature, no necking is observed, i.e., fracture occurs at the maximum load. When testing temperature increases, necking formation is promoted.

All the observations made here are in agreement with data published by Deknis et al. [5] on similar materials.
Table 4
Tensile test results of the unirradiated aged materials.

<table>
<thead>
<tr>
<th>Material</th>
<th>ageing T(°C)</th>
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<th>185</th>
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<td>40.8</td>
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<td>3.5</td>
<td>6.3</td>
<td>5.5</td>
<td>4.7</td>
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<td>3.0</td>
</tr>
<tr>
<td></td>
<td>ε (%)</td>
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<td>4.2</td>
<td>8.9</td>
<td>3.7</td>
<td>23.5</td>
<td>20.8</td>
<td>10.9</td>
</tr>
<tr>
<td></td>
<td>RA (%)</td>
<td>1.2</td>
<td>5.5</td>
<td>9.1</td>
<td>3.7</td>
<td>28.8</td>
<td>27.8</td>
<td>6.6</td>
</tr>
<tr>
<td>S65 HIP</td>
<td>σ₀ (MPa)</td>
<td>356</td>
<td>325</td>
<td>285</td>
<td>247</td>
<td>201</td>
<td>194</td>
<td>154</td>
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<tr>
<td></td>
<td>UTS (MPa)</td>
<td>423</td>
<td>458</td>
<td>414</td>
<td>358</td>
<td>255</td>
<td>196</td>
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<td></td>
<td>εₜ (%)</td>
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<td>8.0</td>
<td>9.1</td>
<td>6.7</td>
<td>6.7</td>
<td>3.7</td>
<td>23.9</td>
</tr>
<tr>
<td></td>
<td>ε (%)</td>
<td>0.9</td>
<td>8.0</td>
<td>15.2</td>
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<td>23.9</td>
<td>23.6</td>
<td>12.4</td>
</tr>
<tr>
<td></td>
<td>RA (%)</td>
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<td>5.5</td>
<td>13.8</td>
<td>45.9</td>
<td>47.9</td>
<td>34.3</td>
<td>14.3</td>
</tr>
<tr>
<td>S65 VHP</td>
<td>σ₀ (MPa)</td>
<td>240</td>
<td>215</td>
<td>151</td>
<td>180</td>
<td>161</td>
<td>117</td>
<td>90</td>
</tr>
<tr>
<td></td>
<td>UTS (MPa)</td>
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<td>114</td>
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<td>3.7</td>
<td>12.7</td>
<td>42.6</td>
<td>51.5</td>
<td>37.7</td>
<td>34.1</td>
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<tr>
<td></td>
<td>ε (%)</td>
<td>3.7</td>
<td>8.3</td>
<td>17.1</td>
<td>9.7</td>
<td>6.8</td>
<td>5.6</td>
<td>3.3</td>
</tr>
<tr>
<td></td>
<td>RA (%)</td>
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<td>13.2</td>
<td>44.5</td>
<td>64.8</td>
<td>60.7</td>
<td>42.3</td>
<td>24.1</td>
</tr>
</tbody>
</table>

In the testing program, a number of samples that were submitted to various thermal ageing treatments were dedicated to investigate their impact on the tensile properties. All these samples were tested at 250°C. The results, given in Table 5 and represented in Figure 6 clearly show the non-influence of the ageing treatment on the tensile properties.

It is known that upon irradiation, the material hardens and looses its ductility. Table 6 summarizes the tensile properties of the irradiated samples. It clearly shows an increase of both the yield stress and tensile strength and a drastic reduction of ductility. Figures 7 to 9 illustrate the radiation effects on the yield stress, tensile strength and reduction of area. As mentioned before, ageing of unirradiated samples does not affect the tensile properties. On the other hand, the increase of material strength (measured by the yield stress and UTS) is significant except at very high testing (and irradiation) temperature. In the mean time, the ductility, measured by the reduction of area drops drastically by a factor of 5 to 6. According to Beeston [6,7], this loss of ductility can be associated to the Helium production during irradiation. However, density measurements performed on these materials have not shown any significant density changes (less than 1%) [8].

Scanning electron microscopy (SEM) observations indicated that all samples fail by a transgranular cleavage fracture mechanism. In the irradiated samples, some regions of intergranular fracture were identified [9].
Table 5
Effect of ageing temperature on the flow properties. All specimen are tested at 250°C.

<table>
<thead>
<tr>
<th>Test T = 250°C</th>
<th>ageing T (°C)</th>
<th>310</th>
<th>455</th>
<th>540</th>
<th>605</th>
</tr>
</thead>
<tbody>
<tr>
<td>S200 HIP</td>
<td>σ_y (MPa)</td>
<td>315</td>
<td>279</td>
<td>251</td>
<td>269</td>
</tr>
<tr>
<td></td>
<td>UTS (MPa)</td>
<td>341</td>
<td>339</td>
<td>332</td>
<td>334</td>
</tr>
<tr>
<td></td>
<td>ε_y (%)</td>
<td>10.7</td>
<td>9.8</td>
<td>10.8</td>
<td>10.8</td>
</tr>
<tr>
<td></td>
<td>ε (%)</td>
<td>31.8</td>
<td>29.4</td>
<td>31.8</td>
<td>22.7</td>
</tr>
<tr>
<td></td>
<td>RA (%)</td>
<td>31.6</td>
<td>35.0</td>
<td>29.7</td>
<td>23.6</td>
</tr>
<tr>
<td>S200 VHP</td>
<td>σ_y (MPa)</td>
<td>230</td>
<td>178</td>
<td>173</td>
<td>199</td>
</tr>
<tr>
<td></td>
<td>UTS (MPa)</td>
<td>303</td>
<td>305</td>
<td>305</td>
<td>302</td>
</tr>
<tr>
<td></td>
<td>ε_y (%)</td>
<td>5.0</td>
<td>7.8</td>
<td>8.3</td>
<td>7.6</td>
</tr>
<tr>
<td></td>
<td>ε (%)</td>
<td>8.2</td>
<td>13.9</td>
<td>16.4</td>
<td>8.3</td>
</tr>
<tr>
<td></td>
<td>RA (%)</td>
<td>9.7</td>
<td>12.1</td>
<td>15.5</td>
<td>7.4</td>
</tr>
<tr>
<td>S65 HIP</td>
<td>σ_y (MPa)</td>
<td>274</td>
<td>298</td>
<td>294</td>
<td>279</td>
</tr>
<tr>
<td></td>
<td>UTS (MPa)</td>
<td>403</td>
<td>412</td>
<td>405</td>
<td>405</td>
</tr>
<tr>
<td></td>
<td>ε_y (%)</td>
<td>8.9</td>
<td>10.2</td>
<td>9.3</td>
<td>8.5</td>
</tr>
<tr>
<td></td>
<td>ε (%)</td>
<td>18.8</td>
<td>19.7</td>
<td>17.3</td>
<td>19.7</td>
</tr>
<tr>
<td></td>
<td>RA (%)</td>
<td>7.4</td>
<td>19.0</td>
<td>15.5</td>
<td>18.6</td>
</tr>
<tr>
<td>S65 VHP</td>
<td>σ_y (MPa)</td>
<td>169</td>
<td>217</td>
<td>168</td>
<td>205</td>
</tr>
<tr>
<td></td>
<td>ε_y (%)</td>
<td>282</td>
<td>301</td>
<td>292</td>
<td>295</td>
</tr>
<tr>
<td></td>
<td>ε (%)</td>
<td>12.1</td>
<td>11.2</td>
<td>11.7</td>
<td>10.5</td>
</tr>
<tr>
<td></td>
<td>UTS (MPa)</td>
<td>43.3</td>
<td>45.2</td>
<td>32.4</td>
<td>39.8</td>
</tr>
<tr>
<td></td>
<td>RA (%)</td>
<td>48.6</td>
<td>45.6</td>
<td>33.4</td>
<td>45.5</td>
</tr>
</tbody>
</table>

While ageing temperature does not affect the tensile properties of unirradiated materials, it plays an important role when associated with neutron irradiation. Indeed, some of the irradiation induced defects are annealed by increasing temperature. Unfortunately, the available data are not sufficient to better understand the effect of irradiation temperature on the damage mechanisms.

Figure 6. Ageing does not affect the tensile properties of the un-irradiated materials.
Table 6
Tensile test results of the irradiated materials.

<table>
<thead>
<tr>
<th>Material</th>
<th>test T (°C)</th>
<th>185</th>
<th>230</th>
<th>310</th>
<th>455</th>
<th>540</th>
<th>605</th>
</tr>
</thead>
<tbody>
<tr>
<td>Neutron fluence ((10^{21} \text{n/cm}^2))</td>
<td>0.85</td>
<td>2.35</td>
<td>0.65</td>
<td>2.0</td>
<td>2.45</td>
<td>2.2</td>
<td></td>
</tr>
<tr>
<td>appm He</td>
<td>300</td>
<td>715</td>
<td>240</td>
<td>610</td>
<td>750</td>
<td>680</td>
<td></td>
</tr>
<tr>
<td>dpa</td>
<td>1</td>
<td>2.45</td>
<td>0.8</td>
<td>2.1</td>
<td>2.5</td>
<td>2.3</td>
<td></td>
</tr>
<tr>
<td>S200 HIP</td>
<td>$\sigma_x$ (MPa)</td>
<td>405</td>
<td>458</td>
<td>524</td>
<td>327</td>
<td>155</td>
<td>106</td>
</tr>
<tr>
<td></td>
<td>UTS (MPa)</td>
<td>463</td>
<td>492</td>
<td>524</td>
<td>331</td>
<td>185</td>
<td>124</td>
</tr>
<tr>
<td></td>
<td>RA (%)</td>
<td>2.5</td>
<td>0.2</td>
<td>2.1</td>
<td>11.2</td>
<td>2.4</td>
<td>0.9</td>
</tr>
<tr>
<td></td>
<td>$\varepsilon$ (%)</td>
<td>0.2</td>
<td>0.1</td>
<td>-</td>
<td>2.6</td>
<td>2.1</td>
<td>1.7</td>
</tr>
<tr>
<td>S200 VHP</td>
<td>$\sigma_x$ (MPa)</td>
<td>457</td>
<td>458</td>
<td>444</td>
<td>238</td>
<td>99</td>
<td>94</td>
</tr>
<tr>
<td></td>
<td>UTS (MPa)</td>
<td>480</td>
<td>457</td>
<td>444</td>
<td>273</td>
<td>110</td>
<td>110</td>
</tr>
<tr>
<td></td>
<td>RA (%)</td>
<td>-</td>
<td>0.2</td>
<td>1.1</td>
<td>1.1</td>
<td>0.3</td>
<td>0.2</td>
</tr>
<tr>
<td></td>
<td>$\varepsilon$ (%)</td>
<td>0.1</td>
<td>-</td>
<td>-</td>
<td>0.9</td>
<td>0.4</td>
<td>1.1</td>
</tr>
<tr>
<td>S65 HIP</td>
<td>$\sigma_x$ (MPa)</td>
<td>391</td>
<td>489</td>
<td>556</td>
<td>266</td>
<td>101</td>
<td>87</td>
</tr>
<tr>
<td></td>
<td>UTS (MPa)</td>
<td>391</td>
<td>558</td>
<td>556</td>
<td>288</td>
<td>124</td>
<td>91</td>
</tr>
<tr>
<td></td>
<td>RA (%)</td>
<td>2.9</td>
<td>0.2</td>
<td>1.5</td>
<td>7.5</td>
<td>0.1</td>
<td>0.0</td>
</tr>
<tr>
<td></td>
<td>$\varepsilon$ (%)</td>
<td>-</td>
<td>0.1</td>
<td>-</td>
<td>5.7</td>
<td>0.3</td>
<td>0.9</td>
</tr>
<tr>
<td>S65 VHP</td>
<td>$\sigma_x$ (MPa)</td>
<td>374</td>
<td>412</td>
<td>449</td>
<td>282</td>
<td>203</td>
<td>82</td>
</tr>
<tr>
<td></td>
<td>UTS (MPa)</td>
<td>410</td>
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<td>449</td>
<td>293</td>
<td>248</td>
<td>102</td>
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<tr>
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<td>RA (%)</td>
<td>-</td>
<td>0.2</td>
<td>4.2</td>
<td>6.4</td>
<td>9.6</td>
<td>1.3</td>
</tr>
<tr>
<td></td>
<td>$\varepsilon$ (%)</td>
<td>0.1</td>
<td>0.4</td>
<td>-</td>
<td>3.1</td>
<td>2.7</td>
<td>2.5</td>
</tr>
</tbody>
</table>

Figure 7. Radiation effect on the yield stress.

Figure 8. Radiation effect on the ultimate tensile strength (UTS).
The following conclusions can be drawn:
1. Irradiation results in classical effects on the tensile properties, namely, loss of ductility and strengthening of the material.
2. No clear distinction can be made between the four Be-grades, except the S200 VHP which exhibits a different behaviour. In addition, the HIP materials results in higher strength than the VHP materials.
3. Thermal ageing does not affect much the tensile properties of unirradiated materials.
4. Upon irradiation, classical strengthening and reduction of ductility are observed. Beside, the irradiation temperature starts to play an important role.

Table 7. Fracture toughness data in the unirradiated condition.

<table>
<thead>
<tr>
<th>test T</th>
<th>ageing T</th>
<th>S65 VHP</th>
<th>S65 HIP</th>
<th>S200 VHP</th>
<th>S200 HIP</th>
<th>average</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>-</td>
<td>11.8</td>
<td>10.8</td>
<td>11.5</td>
<td>10.7</td>
<td></td>
</tr>
<tr>
<td>20</td>
<td>185</td>
<td>12.5</td>
<td>9.9</td>
<td>11.6</td>
<td>10.3</td>
<td></td>
</tr>
<tr>
<td>20</td>
<td>230</td>
<td>11.5</td>
<td>9.4</td>
<td>12.9</td>
<td>9.5</td>
<td>11.1</td>
</tr>
<tr>
<td>185</td>
<td>-</td>
<td>16.1</td>
<td>13.0</td>
<td>14.4</td>
<td>11.3</td>
<td></td>
</tr>
<tr>
<td>185</td>
<td>185</td>
<td>15.9</td>
<td>13.6</td>
<td>13.0</td>
<td>13.9</td>
<td>13.9</td>
</tr>
<tr>
<td>230</td>
<td>-</td>
<td>22.4</td>
<td>15.1</td>
<td>15.5</td>
<td>14.7</td>
<td></td>
</tr>
<tr>
<td>230</td>
<td>230</td>
<td>-</td>
<td>14.1</td>
<td>12.0</td>
<td>15.1</td>
<td>15.6</td>
</tr>
<tr>
<td>250</td>
<td>310</td>
<td>17.8</td>
<td>14.8</td>
<td>17.8</td>
<td>14.8</td>
<td></td>
</tr>
<tr>
<td>250</td>
<td>455</td>
<td>-</td>
<td>-</td>
<td>11.3</td>
<td>15.3</td>
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<tr>
<td>250</td>
<td>605</td>
<td>23.6</td>
<td>15.0</td>
<td>28.8</td>
<td>17.9</td>
<td></td>
</tr>
<tr>
<td>250</td>
<td>605</td>
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<td>18.0</td>
<td>14.4</td>
<td>16.7</td>
<td>18.0</td>
</tr>
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<td>-</td>
<td>32.0</td>
<td>14.9</td>
<td>22.9</td>
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<td>40.5</td>
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<td>455</td>
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<td>37.4</td>
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<tr>
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<td>-</td>
<td>33.8</td>
<td>25.3</td>
<td>14.1</td>
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<td>605</td>
<td>33.1</td>
<td>24.8</td>
<td>26.1</td>
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</tr>
<tr>
<td>605</td>
<td>605</td>
<td>-</td>
<td>25.1</td>
<td>24.2</td>
<td>23.0</td>
<td>25.1</td>
</tr>
</tbody>
</table>

4.2. Fracture toughness data
Fracture toughness test results are summarized in Table 7 and 8 for unirradiated and irradiated materials, respectively. In the test temperature range of 20 to 230°C, most of the unirradiated samples fails in the linear region, allowing determination of a $K_{IC}$-value. Some of these values can be considered as $K_{IC}$ values. Beyond 230°C, the load-displacement becomes non linear, requiring the use of the J-integral concept. Note that for the specimens exhibiting a stable crack growth process, the fracture toughness is determined at the maximum load.
Table 8. Fracture toughness data for the various testing and irradiation conditions.

<table>
<thead>
<tr>
<th>test T</th>
<th>ageing T</th>
<th>fluence</th>
<th>appm He</th>
<th>dpa</th>
<th>S65 VHP</th>
<th>S65 HIP</th>
<th>S200 VHP</th>
<th>S200 HIP</th>
<th>average</th>
</tr>
</thead>
<tbody>
<tr>
<td>25</td>
<td>200</td>
<td>1.4</td>
<td>420</td>
<td>1.6</td>
<td>9.1</td>
<td>6.9</td>
<td>7.4</td>
<td>9.8</td>
<td>7.9</td>
</tr>
<tr>
<td>25</td>
<td>230</td>
<td>2.0</td>
<td>600</td>
<td>2.3</td>
<td>8.1</td>
<td>7.7</td>
<td>6.1</td>
<td>9.1</td>
<td>8.3</td>
</tr>
<tr>
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<td>200</td>
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<td>420</td>
<td>1.6</td>
<td>8.7</td>
<td>7.4</td>
<td>7.7</td>
<td>9.1</td>
<td>9.3</td>
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<td>600</td>
<td>2.3</td>
<td>7.8</td>
<td>11.5</td>
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<td>9.0</td>
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</tr>
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<td></td>
</tr>
<tr>
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<td>600</td>
<td>2.25</td>
<td>700</td>
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<td>12.8</td>
<td>14.8</td>
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<tr>
<td>250</td>
<td>610</td>
<td>2.2</td>
<td>680</td>
<td>2.5</td>
<td>11.7</td>
<td>13.4</td>
<td>15.6</td>
<td>12.8</td>
<td>11.7</td>
</tr>
<tr>
<td>350</td>
<td>350</td>
<td>1.1</td>
<td>330</td>
<td>1.25</td>
<td>8.5</td>
<td>10.9</td>
<td>-</td>
<td>13.0</td>
<td>10.8</td>
</tr>
<tr>
<td>435</td>
<td>435</td>
<td>1.65</td>
<td>500</td>
<td>1.85</td>
<td>-</td>
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<td>25.2</td>
<td>15.9</td>
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<td>-</td>
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<td>13.2</td>
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<td></td>
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<tr>
<td>610</td>
<td>610</td>
<td>2.2</td>
<td>680</td>
<td>2.5</td>
<td>14.9</td>
<td>-</td>
<td>17.2</td>
<td>-</td>
<td>16.6</td>
</tr>
</tbody>
</table>

The average $K_c$ value for the S200 VHP grade tested at 20°C is 11.8 MPa.m$^{-1}$. This value lies about 10% higher than fracture toughness data performed by Shabbits and Longdon [10] on large compact tension specimen of a similar material. This may be due to either the higher stress intensity factor during fatigue or to a size effect.

Figure 10 shows the fracture toughness results as a function of test temperature. It should be emphasized here that the irradiated samples were irradiated and tested at the same temperature.

Fracture toughness behaviour of the unirradiated is very similar to the evolution ductility with test temperature. This was unexpected as the fracture mechanism was found as transgranular cleavage type. Perra and Finnie [11] suggested also that fracture toughness is not related to reduction of area in the tension test when cleavage fracture dominates.

Irradiation drastically reduces the fracture toughness, in particular in the 400 to 450°C temperature range. It should be reminded that in this range, unirradiated samples fail by stable crack growth while unstable fracture occurs when irradiated.

The effect of irradiation fluence can be better seen on Figure 11. Fracture toughness first decreases and then increases for higher fluences. However, the irradiation temperature is not identical. There is a benefit effect of temperature as it anneals some of the defects induced by irradiation.
Figure 10. Radiation effect of the fracture toughness. Unirradiated data result in higher fracture toughness values. No distinction can be made between the beryllium grades before and after irradiation.

Fig. 11 - Fracture toughness data tested at 250°C show that fracture toughness decreases with increasing neutron fluence but re-increases with higher fluences. This is due to the benefit effect of irradiation temperature: the higher the fluence, higher the irradiation temperature.
5. CONCLUSIONS

The main conclusions that can be drawn from this work can be summarized as follow:
1. All beryllium grades exhibit a quite similar behaviour from the tensile properties as well as fracture toughness point of view.
2. In the un-irradiated condition, the ageing treatment does not significantly affect the tensile properties as well as the fracture toughness. However, the irradiation damage is much affected by the irradiation temperature: at a higher temperature, a benefit effect on fracture toughness is observed.

Further work is however required to determine if crack initiates at maximum load when tested at higher temperatures. Scanning electron microscopy examination of the fractured compact tension specimens is also needed to better understand the fracture mechanisms and the relations between fracture toughness and flow properties.

ACKNOWLEDGMENTS

This work was supported by the European Commission Fusion Program. This work would not have been achieved without the technical assistance of Teo Noels, F. Verstrepen, R. Mertens, W. Hendrix from VITO, J. Defourny and M. Delooz from CRM.

REFERENCES

8.4 Tritium Release from Neutron Irradiated Beryllium Pebbles

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One of the most important open issues related to beryllium for fusion applications refers to the kinetics of the tritium release as a function of neutron fluence and temperature. The EXOTIC-7 as well as the "Beryllium" experiments carried out in the HFR reactor in Petten are considered as the most detailed and significant tests for investigating the beryllium response under neutron irradiation. This paper reviews the present status of beryllium post-irradiation examinations performed at the Forschungszentrum Karlsruhe with samples from the above mentioned irradiation experiments, trying to elucidate the tritium release controlling processes. In agreement with previous studies it has been found that release starts at about 500-550 °C and achieves a maximum at about 700-750 °C. The observed release at about 500-550 °C is probably due to tritium escaping from chemical traps, while the maximum release at about 700 -750°C is due to tritium escaping from physical traps.

The consequences of a direct contact between beryllium and ceramics during irradiation, causing tritium implanting in a surface layer of beryllium up to a depth of about 40 mm and leading to an additional inventory which is usually several times larger than the neutron-produced one, are also presented and the effects on the tritium release are discussed.

1. Introduction

For the Helium Cooled Pebble Bed (HCPB) Blanket, which is one of the two reference concepts studied within the European Fusion Technology Programme, the neutron multiplier consists of a mixed bed of about 2 and 0.1-0.2 mm diameter beryllium pebbles. The main structure of the pebble bed is given by the larger pebbles with a packing factor of 63.3%. In the space between them are placed the smaller beryllium pebbles with a packing factor of 17.5%. In the fast neutron field of the blanket, helium and tritium are produced in beryllium. Therefore, besides compatibility with other blanket materials, helium-induced swelling and tritium retention are of concern.

Although the tritium production rate in beryllium is two to three order of magnitude smaller than that in the breeder ceramic, because of the slow release at typical blanket temperatures [1,2], the inventory in beryllium might become large during the HCPB DEMO blanket lifetime (≈3 years) and is therefore a safety concern.

Once tritium is produced/implanted in beryllium it diffuses at a significant rate to sites of lower free energy (i.e. He bubbles) and/or it chemically reacts with impurities for which it has a particular affinity (i.e. BeO) [2]. Release kinetics is, therefore expected to be dependent on the particular trapping mechanisms and, in particular, to be hindered both by structural sinks (physical trapping) and by beryllium oxide impurities (chemical trapping).

If tritium is trapped in a helium-filled bubble, it follows the fate of the bubble and will be released only if the bubble is vented into an open porosity network, through which the gas can escape with effectively no activation energy. On the other hand, because beryllium of commercial purity always contains some oxide inclusions (which tend to collect at grain boundaries where they form globular phases), beryllium oxide can react with tritium to form beryllium hydroxide which is energetically stable with respect to single tritium atoms. Tritium chemically bound in form of beryllium hydroxide at oxide inclusions requires a sufficiently high thermal energy to be released. In fact, under the action of heating, beryllium hydroxide may dissociate and tritium in atomic form can be re-injected into the lattice. Tritium in excess of what the lattice can dissolve either
diffuses through the lattice and escape from the specimen or is captured by gas bubbles, depending on the present global bubble density and size. Due to the two different tritium trapping mechanisms in beryllium, the release kinetics will be thus different depending on gas bubble density and size, and on irradiation or annealing temperature.

The EXOTIC-7 [3] as well as the "Beryllium" [4] experiments carried out in the HFR reactor in Petten are considered as the most detailed and significant tests for investigating the beryllium pebble response under neutron irradiation.

2. Samples and Irradiation Conditions

The beryllium irradiated in both EXOTIC-7 and "Beryllium" experiments consists of a mix of about 2 mm and 0.08-0.18 mm diameter beryllium pebbles.

The bigger pebbles are are fabricated by melting and are a relatively inexpensive intermediate product of the beryllium fabrication route (Brush-Wellmann Company) [1]. On the contrary the smaller beryllium pebbles are produced by Rotating Electrode Process (REP).

While in EXOTIC-7 the beryllium pebbles were - during irradiation - in contact with Li4SiO4 pebbles, in the "Beryllium" experiment mainly pure beryllium pebble beds were irradiated. For some post-irradiation examinations (PIE) the components of the EXOTIC-7 mixed beds had to be separated. The 2 mm diameter Be pebbles were separated from the small (0.1-0.2 mm) Li4SiO4 and Be pebbles by sieving. The small Li4SiO4 and Be pebbles were separated by a wet procedure using an organic liquid with a density in-between that of the Li4SiO4 and Be pebbles.

Type, mass and drying procedure of the pebbles delivered to ECN-Petten for irradiation in EXOTIC-7 are given in Table 1.

The capsules were filled at ECN/JRC November 1993 under air. The mixed beds were prepared by filling first the 2 mm diameter Be pebbles and afterwards into the residual space a mixture of about equal volume fractions of 0.1-0.2 mm Li4SiO4 and 0.1-0.2 mm Be pebbles. The irradiation started February 1994 and has been completed in February 1995 (11 reactor cycles, 261 full power days). During ir-
radiation the pebbles were purged with He+0.1 vol% H₂. The peak neutron fluences near the capsule walls were: fast fluence \((54^{\text{Mn}}) 1-2 \times 10^{21} \text{n/cm}^2\), thermal fluence \((60^{\text{Co}}) 1-1.6 \times 10^{21} \text{n/cm}^2\). The depression of the thermal fluence at the capsule centers was a factor 2 to 4. Other irradiation conditions are given in Table 3.

### Table 1 Pebbles delivered to ECN-Petten for irradiation in EXOTIC-7

<table>
<thead>
<tr>
<th>Material type</th>
<th>Mass (g)</th>
<th>Drying</th>
<th>%TD</th>
<th>Grain size (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\text{Li}_4\text{SiO}_4) (+ 1.4 wt% SiO₂), 50 % (6^\text{Li}) not tempered, pebble diameter 0.1-0.2 mm type Schott B-4153, 42/93</td>
<td>~ 20</td>
<td>300 °C, 3 h stagnant air</td>
<td>97</td>
<td>20 - 40</td>
</tr>
<tr>
<td>Be, pebble diameter ~ 2 mm</td>
<td>~ 28</td>
<td>650 °C, 3 h</td>
<td>100</td>
<td>___</td>
</tr>
<tr>
<td>type REP, pebble diameter 0.1-0.2 mm</td>
<td>~ 5.5</td>
<td>purging He</td>
<td>100</td>
<td>___</td>
</tr>
</tbody>
</table>

### Table 2 Loading characteristics of EXOTIC-7 capsules

<table>
<thead>
<tr>
<th>Capsule number</th>
<th>(V) (cm³)</th>
<th>Material</th>
<th>M (g)</th>
<th>(\rho_b = M/V) (g/cm³)</th>
<th>(\rho_b/\rho_{th}) (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>26.2</td>
<td>3.43</td>
<td>(\text{Li}_4\text{SiO}_4) Be 0.1 - 0.2 mm</td>
<td>1.40</td>
<td>0.408</td>
<td>17.1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.94</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>2.59</td>
<td>3.53</td>
<td>1.029</td>
<td>55.9</td>
</tr>
<tr>
<td>28.1</td>
<td>3.38</td>
<td>(\text{Li}_4\text{SiO}_4) Be 0.1 - 0.2 mm</td>
<td>4.99</td>
<td>1.476</td>
<td>61.8</td>
</tr>
<tr>
<td>28.2</td>
<td>8.06</td>
<td>(\text{Li}_4\text{SiO}_4) Be 0.1 - 0.2 mm</td>
<td>1.88</td>
<td>0.233</td>
<td>9.8</td>
</tr>
<tr>
<td></td>
<td></td>
<td>144</td>
<td>9.81</td>
<td>1.217</td>
<td>66.2</td>
</tr>
</tbody>
</table>

### Table 3 EXOTIC-7 Irradiation conditions

<table>
<thead>
<tr>
<th></th>
<th>26.2</th>
<th>28.1</th>
<th>28.2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Central temperature (°C)</td>
<td>410 - 545</td>
<td>465 - 730</td>
<td>410 - 480</td>
</tr>
<tr>
<td>Total lithium burn-up (%)</td>
<td>14 ± 2</td>
<td>7 ± 2</td>
<td>19 ± 2</td>
</tr>
</tbody>
</table>
With reference to the "Beryllium" Experiment, mixtures of large and small pebbles were dried by purging them with helium at 650 °C for 3 hours and filled into steel capsules. The capsules were evacuated, then filled with 1.1 bar helium and closed by welding. The capsule cladding was made from stainless steel AISI 316L and each capsule was provided with a gas plenum of about 0.5 cm$^3$ to accommodate released helium and tritium. The capsules were irradiated from April to August 1994 over four reactor cycles in the HFR reactor at a temperature of about 420 °C (capsules 2 and 3) and 510 °C (capsules 5 to 8) to a fast neutron fluence (E>1 MeV) of about 1.0$\times$10$^{21}$ cm$^{-2}$ (capsules 2 and 3) and 1.2$\times$10$^{21}$ cm$^{-2}$ (capsules 5 to 8) [4]. After irradiation the samples were handled under inert atmosphere before annealing.

Results and Discussion

A series of tests showed that the release characteristics of each type of pebbles is reproducible. The representative release curves for large pebbles, pieces ($\Phi < 0.7$ mm) from crushed large pebbles and for small pebbles of the "Beryllium" experiment are shown in Fig. 2. Useful parameters to characterize tritium release kinetics are the fractional release (release R/production P), the temperature $T_{\text{max}}$ corresponding to the maximum release rate (during the 5 °C/min temperature ramp) and the factor DF by which the release rate decreases (at 850 °C within 3 hours). For the pebbles investigated in the frame of the present work these parameters are given in Tab. 4.

### 3. Facilities and Procedures

One of the main characteristics of the tritium release facilities is that the sample chamber is connected by a short, heated line ($\approx$300 °C) to a Zn-reductor ($\approx$390 °C). The reductor transforms any tritium water to tritium gas. This avoids problems with tritium water absorption and allows quantitative tritium measurements. The tritium activity of the purge gas is measured with an ionization chamber and/or a proportional counter. The released tritium inventory is determined by integrating the measured release rate over the time. Release kinetics and total amount of released tritium were determined by annealing the pebbles by temperature ramps of 5 °C/min up to 850 °C, keeping constant this temperature for several hours and purging them with 50 SCCM He+0.1 vol% H$_2$.

![Fig.2 Tritium release for large pebbles, pieces from crushed large pebbles and small pebbles.](image-url)

Table 4 Tritium release characteristics of beryllium samples from the "Beryllium" experiment.

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Material</th>
<th>$\Phi$ [cm$^2$] (E&gt;1 MeV)</th>
<th>$P$ [Bq/g]</th>
<th>$R$ [Bq/g]</th>
<th>$R/P$ [%]</th>
<th>$T_{\text{max}}$ [°C]</th>
<th>DF</th>
</tr>
</thead>
<tbody>
<tr>
<td>&quot;Beryllium&quot; Caps. 2</td>
<td>Large pebbles</td>
<td>1.0$\times$10$^3$</td>
<td>2.1$\times$10$^9$</td>
<td>8.0$\times$10$^8$</td>
<td>40</td>
<td>&gt;850</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td>Large pebbles</td>
<td>1.0$\times$10$^3$</td>
<td>2.1$\times$10$^9$</td>
<td>1.4$\times$10$^8$</td>
<td>70</td>
<td>800</td>
<td>50</td>
</tr>
<tr>
<td></td>
<td>(pieces)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Small pebbles</td>
<td>1.0$\times$10$^3$</td>
<td>2.1$\times$10$^9$</td>
<td>2.0$\times$10$^8$</td>
<td>100</td>
<td>400, 800</td>
<td>5$\times$10$^7$</td>
</tr>
</tbody>
</table>
As seen in the table, the fractional release, R/P, is relatively small in case of the 2 mm pebbles and increases by decreasing the pebble dimensions. The $T_{\text{max}}$ for the three cases (large pebbles, pieces ($\Phi < 0.7$ mm) from crushed large pebbles and small pebbles) shows a slight decrease with decreasing the specimen dimensions, from 850 °C to 800 °C, as expected. The factor DF is relatively low for the 2 mm pebbles, even at the high hold temperature of 850 °C, indicating only gradual continuing release at the hold temperature. On the contrary the DF is considerably higher in case of both pieces ($\Phi < 0.7$ mm) from crushed large pebbles and small pebbles.

In general the beryllium specimens irradiated in the "Beryllium" experiments show that the release from 0.7 mm diameter pieces from the large pebbles is faster than that from the whole pebbles ("particle size" effect), but a further decrease of the particle size to ≤0.5 mm diameter indicated no further release improvement. However, release from the small pebbles was faster than that from 0.7 mm diameter pieces from the large pebbles.

Furthermore, it has been found that the release of previously investigated Brush-Wellman samples (B-26, S200 HIP) [5-7] is comparable with that from the large pebbles. Therefore, it can be assumed that the experimentally determined tritium residence times of beryllium B-26 [5] apply also to the large pebbles from the "Beryllium" experiment.

With reference to EXOTIC-7, release of all investigated Be samples (Be 2 mm, Be 2 mm broken, Be 0.1-0.2 mm) from the mixed beds (Be + Li$_4$SiO$_4$) of capsules 28.2 and 26.2-1 is very similar. In agreement with previous studies [5] [8] the release starts at about 500-550 °C and achieves a maximum at about 700 -750°C as shown in Fig. 3. The total release of the large Be pebbles (Be 2 mm) is slightly larger ($\approx 6\times10^{10}$ Bq/g) than that from Li$_4$SiO$_4$ from the same capsules, while that of the small Be pebbles (Be 0.1-0.2 mm) is about a factor 30 larger ($\approx 1\times10^{12}$ Bq/g). Once tritium is produced/implanted in Be it diffuses at a significant rate to sites of lower free energy (i.e. He bubbles) and/or it chemically reacts with impurities for which it has a particular affinity (i.e. BeO) [2]. Release kinetics is, therefore expected to be dependent on the particular trapping mechanisms and, in particular, to be hindered both by structural sinks (physical trapping) and by beryllium oxide impurities (chemical trapping). The observed release at about 500-550 °C is probably due to tritium escaping from chemical traps, while the maximum release at about 700-750°C is due to tritium escaping from physical traps.

![Fig. 3 Tritium release from EXOTIC-7 beryllium pebbles](image-url)
cific tritium production in Be of $1.3 \times 10^9$ Bq/g, in reasonable agreement with a value of $2 \times 10^9$ Bq/g based on tritium production vs. fast fluence graph [10]. In any case, the neutron-generated tritium in Be is about a factor 50 lower than the released tritium from the larger Be pebbles and about a factor 700 lower than that from the smaller Be pebbles, assuming no release during the irradiation. This high tritium inventory in both the large and small Be pebbles from EXOTIC-7 capsules 26.2-1 and 28.2 can be only due to implantation from ceramics. Furthermore, the total release from the small Be pebbles is about a factor 15 larger than from the bigger ones. This is probably due to the fact that in the small Be pebbles the implantation depth of tritium coming from ceramics ($\approx 40 \mu m$) is of the same order of magnitude as the pebbles radius ($\approx 50 - 100 \mu m$), which results in a higher implantation efficiency than in the case of larger Be pebbles.

4. Summary

The behavior of beryllium pebbles irradiated in the EXOTIC-7 as well as in the “Beryllium” experiments has been investigated. Concerning the observed tritium release kinetics of the pebbles from the “Beryllium” experiment it can be seen that the release from 0.7 mm diameter pieces from the large pebbles is faster than that from the whole pebbles (“particle size” effect). However, a further decrease of the particle size to $\leq 0.5$ mm diameter indicated no further release improvement. With reference to EXOTIC-7, the tritium release of all investigated Be samples from the mixed beds is very similar and, in agreement with previous studies, it starts at about $500-550 ^\circ C$ and achieves a maximum at about $700-750 ^\circ C$. In any case, the neutron-generated tritium in Be is about a factor 50 lower than the released tritium from the larger Be pebbles and about a factor 700 lower than that from the smaller Be pebbles, assuming no release during the irradiation. This high tritium inventory in both the large and small Be pebbles from EXOTIC-7 is due to implantation from ceramics.

Acknowledgement

The authors would like to E. Damm and H. Ziegler of the FZK-Hot Cells for performing the measurements. This work was performed in the framework of the Forschungszentrum Karlsruhe Fusion Project and is supported by the European Union within the European Fusion Technology Program.

References


[6] W. Dienst et al., Tritium Release of Li$_4$SiO$_4$, Li$_2$O and Beryllium and chemical compatibility of Beryllium with Li$_4$SiO$_4$, Li$_2$O and Steel (SIBELIUS Irradiation), KfK Bericht 5109, Karlsruhe, December 1992.


Microstructure and mechanical properties of neutron irradiated beryllium with total fast neutron fluences of \(1.3 - 4.3 \times 10^{21} \text{n/cm}^2 (E > 1 \text{MeV})\) at 327-616°C were studied. Swelling increased by high irradiation temperature, high fluence, and by the small grain size and high impurity. Obvious decreasing of the fracture stress was observed in the bending test and in small grain specimens which had many helium bubbles on the grain boundary. Decreasing of the fracture stress for small grain specimens was presumably caused by crack propagation on the grain boundaries which weekend by helium bubbles.

1. Introduction

Beryllium are expected to be used as a neutron multiplier and plasma facing material in the International Thermonuclear Experimental Reactor (ITER). For a neutron multiplier of blanket, beryllium pebble and/or low density beryllium are candidate materials because these material are expected to give low swelling. As a plasma facing material, hot-pressed beryllium (S-65C) is a candidate material because of its high thermal shock resistance [1]. Recently, some studies for neutron irradiated beryllium were reported [2-4]. However, neutron irradiation data are poor in these materials, and discussion about grain size and/or impurity effects is insufficient. Therefore, microstructure and mechanical properties of neutron irradiated beryllium which produced by different method, and which had different grain size and impurity level were studied to get the engineering data for the fusion reactor design.

2. Samples

Specimens were bending specimens produced by the hot-press or vacuum cast method, tensile specimens produced by hot-press or HIP (hot isostatic press) method and beryllium pebbles produced by the rotating electrode method. Each specimen had different grain size and impurity level. Information of each specimen is shown in Table 1 and 2. Four sets of these specimens were irradiated with four inner capsules in the Japan Materials Testing Reactor (JMTR). Irradiation conditions are shown in Table 3. Helium and tritium production rate calculated by HEINBE code [5].

### Table 1

<table>
<thead>
<tr>
<th>Beryllium specimens</th>
<th>Fabrication method</th>
<th>Grain size (mm)</th>
<th>Configuration</th>
</tr>
</thead>
<tbody>
<tr>
<td>BHP (S-65C)</td>
<td>HP</td>
<td>0.01</td>
<td>Bending</td>
</tr>
<tr>
<td>BVC (B-26D)</td>
<td>VC</td>
<td>0.56</td>
<td>Tensile</td>
</tr>
<tr>
<td>THP (S-200F)</td>
<td>HP</td>
<td>0.02</td>
<td></td>
</tr>
<tr>
<td>THP</td>
<td>HIP</td>
<td>0.13</td>
<td>Pebble (6x6mm)</td>
</tr>
</tbody>
</table>


### Table 2

<table>
<thead>
<tr>
<th>Element (wt.%)</th>
<th>Be</th>
<th>BeO</th>
<th>Fe</th>
<th>C</th>
<th>Al</th>
<th>Mg</th>
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<tr>
<td>BHP</td>
<td>99.1</td>
<td>0.9</td>
<td>0.10</td>
<td>0.14</td>
<td>0.04</td>
<td>&lt;0.01</td>
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<tr>
<td>BVC</td>
<td>99.8</td>
<td>0.02</td>
<td>0.03</td>
<td>0.08</td>
<td>0.04</td>
<td>0.04</td>
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<tr>
<td>THP</td>
<td>98.9</td>
<td>1.2</td>
<td>1.0</td>
<td>0.14</td>
<td>0.05</td>
<td>0.02</td>
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<tr>
<td>THP</td>
<td>99.72</td>
<td>0.18</td>
<td>0.047</td>
<td>0.093</td>
<td>0.026</td>
<td>0.0115</td>
</tr>
<tr>
<td>P</td>
<td>98.2</td>
<td>1.51</td>
<td>0.11</td>
<td>0.023</td>
<td>0.078</td>
<td>0.017</td>
</tr>
</tbody>
</table>

### Table 3

<table>
<thead>
<tr>
<th>Neutron irradiation condition</th>
<th>Inner capsule No.</th>
<th>(T_m) (°C)</th>
<th>Fluence (E &gt; 1 \text{MeV}) (\times 10^{21} \text{n/cm}^2)</th>
<th>Helium (\times 10^5 \text{appm})</th>
<th>Tritium (\times 10^1 \text{GBq/cm}^3)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>327</td>
<td>1.3</td>
<td>0.3</td>
<td>0.5</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>445</td>
<td>3.0</td>
<td>0.7</td>
<td>1.8</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>616</td>
<td>4.3</td>
<td>1.0</td>
<td>3.0</td>
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<tr>
<td>4</td>
<td>524</td>
<td>4.1</td>
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</tbody>
</table>

* Fast neutron energy : \(E > 1 \text{MeV}\)
* Production rate were calculated by HEINBE code
3. Experimental

Swelling was measured with a density meter (Mettler SGM-3) using the echilarchale for pebble, or water for other specimens.

Fracture strength of two bending specimens, one tensile specimen and 5-10 compression specimens were tested at room temperature [6] for each of the inner capsules. Testing speed was 0.2 mm/min for each specimen. The fracture stress of the three points bending test was calculated by using the following equation [7].

\[ \sigma_b = \frac{3PL}{2wt^2} \]  

(1)

where \( \sigma_b \) is fracture stress in bending test, \( P \) is fracture load, \( L \) is the span width of lower support (30 mm), \( w \) is specimen width, \( t \) is specimen thickness. For the evaluation of fracture strength about pebble, we used the following equation.

\[ \sigma_c = \frac{L}{r^2} \]  

(2)

where \( \sigma_c \) is the evaluation value of compression test, \( L_c \) is fracture load, \( r \) is the radius of pebble.

Scanning electron micrograph (SEM) observation of the fracture surface for each specimen was carried out by using an equipment of JMTR hot-laboratory. Metallographical observation was also carried out after chemical etching [dipping in mixture of ethyl alcohol (45cm³) and hydrogen fluoride (5cm³) for 15-30 s]. Then, micro vickers hardness was measured under the conditions of 200 g and 30 s.

4. Results and discussion

4.1 Swelling

Measured swelling is shown in Fig. 1. Swelling increased with the increasing of irradiation temperature and fluence, and with the decreasing of the grain size and the increasing of impurity level. These results are reasonable because the single crystal specimen (with large grains and low impurity level) showed low swelling [8]. Swelling of the pebble specimen showed negative values. This reason was presumed to be caused by oxidation because the pebble specimen had a larger specific surface area than that of the other specimens. The formula of swelling evaluation by Beeston and Semyaev [8] is also shown in the same figure. The formula by Beeston showed a rather low value. This difference was caused by the difference in neutron spectrum. The formula by Sernyaev gave good agreement with the results using different structure-sensitive factors [8] ("Max" and "Min" mean the maximum and the minimum of structure-sensitive factor). However, further studies will be needed to understand the swelling mechanism, for example, structure-sensitive factor and separation of effects of the grain size and impurities.

![Swelling as a function of irradiation temperature.](image1)

![Results of the bending test.](image2)
4.2 Microstructure and mechanical properties

Results of the bending and tensile test are shown in Fig. 2 and Fig. 3. By increasing irradiation temperature, the fracture stress of small-grained specimens (BHP, THP) rapidly decreased. These results were due to helium bubbles because many helium bubbles were observed on the grain boundaries (Fig. 4). Observed maximum diameter of the helium bubbles is shown in Fig. 5. Rapid decreasing of fracture stress for the lager-grained specimens was not observed though lager grain kept larger helium bubbles.

Fig. 4 Helium bubble on the grain boundary (Inner capsule No.3, BHP specimen).

Fig. 5 Observed maximum diameter of the helium bubbles for the each specimens.

Fig. 6 Ratio of the intergranular fracture for the each specimens.

Generally, small-grained specimen shows high fracture stress (Hall-Petch relation). However, we
should pay attention to the selection of grain size in case neutron irradiation, and should find the most suitable grain size for each neutron irradiation condition.

Fig. 7 Occupied area ratio of the helium bubbles on the grain boundaries.

Fig. 8 Results of compression test.

Results of compression test are shown in Fig. 8. Vertical axis is normalized by the average of evaluation value [see equation (2)] for the unirradiated sample. Mechanical properties of the beryllium pebble specimen shows the feature of larger-grained specimens, and rapid decreasing was not observed. The dispersion at low temperature was presumed to be caused by the anisotropy in strength [9], and anisotropy became small with the growth of helium bubbles. Fractured surface of BVC and pebble is shown in Fig. 9. Helium bubbles on the grain boundaries of the pebble specimen were smaller than those of BVC specimen although both specimens had a similar grain size. This reason was presumed to be caused by the size effect of specimen (Most of helium was released because of the short diffusion length in the pebbles) and/or the initial defects introduced in production process; pebble specimen had many initial defects because of rapid cooling, and helium will be trapped by these defects. Anyway, beryllium pebbles have some advantage compare with small grained specimens.

As precious photograph, hexagonal pore were observed in the grains of BVC specimen (Fig. 10).

Fig. 9 Fracture surface of BVC and pebble specimen.

From the results of the metallographical observations, it is concluded that helium bubbles concentrated on the grain boundaries at first, and were grown up with the increase of irradiation temperature and neutron fluence.

Measured vickers hardness is shown in Fig. 11. Decreasing of hardness was observed on the smaller-grained specimens, while, obvious tendency of the decreasing was not observed on the larger-grained specimens. This tendency is similar to the case of fracture stress.
4.3 Impurity effects on mechanical properties

Reevaluation of the existing data were attempted because the discussion on the impurity effects was insufficient by using only the data obtained in these experiments.

Fig. 10 Hexagonal pore in the grain of BVC specimen.

Fig. 11 Measurement results of vickers hardness.

Fracture stress was measured as the JMTR surveillance test more than ten years ago [10]. Two kinds of beryllium specimens [NGK and KBI (the present name is NGK Metals Corp.)] were used in this test, and it became clear by the reevaluation that these specimens had different levels of Fe impurity caused by different production processes (other impurities were in the same level). Replotted figure for the specimens with different Fe impurity is shown in Fig. 12. Fracture stress for the specimen with higher Fe impurity decreased in the high neutron fluence range. This results shows that Fe impurity affects the fracture stress in the high fluence range. As an explanation of this results, Fe impurity like as Be$_2$Fe traps helium bubbles [11], and helium bubbles weaken the mechanical properties.

Fig. 12 Effects of Fe impurity on fracture stress.

5. Conclusion

Microstructure and mechanical properties of the neutron irradiated beryllium which was produced by different methods were discussed, and the following results were obtained:

- Swelling increased with the increasing of irradiation temperature, and fluence, and the decreasing of grain size and the increasing of impurity level.
- Fracture stress of the small-grained specimens rapidly decreased at high irradiation temperature. Rapidly decreasing was presumed to be caused by the propagation of cracks on the grain boundaries. This result means that we should pay attention for the selection of grain size in case of neutron irradiation.
- Beryllium pebble has some advantages due to larger grain, smaller size and initial defects in comparison with the other small grained specimens.
- Fe impurity affects the fracture stress in the high fluence range.

Some results were obtained from this study. However, these data are insufficient to discuss the grain size and impurities effects in detail. Further studies are required to use beryllium for the fusion application.
Acknowledgments

The authors greatly appreciate the helpful comments on this paper by Dr. O. Baba (Director, Department of JMTR, JAERI).

References

8.6 Neutron Irradiation Behavior of ITER Candidate Beryllium Grades

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\(^b\) Scientific Research Institute of Atomic Reactors, Dimitrovgrad, Russia

Beryllium is one of the main candidate materials both for the neutron multiplier in a solid breeding blanket and for the plasma facing components. That is why its behaviour under the typical for fusion reactor loading, in particular, under the neutron irradiation is of a great importance. This paper presents mechanical properties, swelling and microstructure of six beryllium grades (DshG-200, TR-30, TshG-56, TRR, TE-30, TIP-30) fabricated by VNIINM, Russia and also one - (S-65) fabricated by Brush Wellman, USA. The average grain size of the beryllium grades varied from 8 to 25 \(\mu\)m, beryllium oxide content was 0.8 - 3.2 wt. %, initial tensile strength was 250 - 680 MPa. All the samples were irradiated in active zone of SM-3 reactor up to the fast neutron fluence \((5.5 - 6.2) \cdot 10^{21} \text{ cm}^{-2}\) \((2.7 - 3.0 \text{ dpa}, \text{helium content up to } 1150 \text{ appm, } E > 0.1 \text{ MeV at two temperature ranges: } T_1 = 130-180^\circ\text{C} \text{ and } T_2 = 650-700^\circ\text{C}. \text{ After irradiation at } 130-180^\circ\text{C} \text{ no changes in samples dimensions were revealed. After irradiation at } 650-700^\circ\text{C} \text{ swelling of the materials was found to be in the range } 0.1-2.1\% \text{. Beryllium grades TR-30 and TRR, having the smallest grain size and highest beryllium oxide content, demonstrated minimal swelling, which was no more than } 0.1\% \text{ at } 650-700^\circ\text{C} \text{ and fluence } 5.5 \cdot 10^{21} \text{ cm}^{-2}. \text{ Tensile and compression test results and microstructure parameters measured before and after irradiation are also presented.}

1. INTRODUCTION

Beryllium now is considered as a main candidate material for a fusion application. However, the efficiency of beryllium as a plasma-facing material and a neutron multiplier for a breeding blanket strongly depends on its behaviour under neutron irradiation. In terms of a fusion application the most important consequences of neutron irradiation on beryllium are helium induced swelling, embrittlement and tritium release. In spite of a rich variety of data on effect of irradiation on beryllium, which available in literature [1-4], not much is known about of the irradiation behaviour of main fusion-candidate beryllium grades [5].

This paper presents the results of investigation on effect of high-temperature and low-temperature irradiation on swelling, mechanical properties and microstructure of beryllium. The very first results of this work have been reported earlier [6].

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Table 1

Chemical composition of beryllium grades, wt %

<table>
<thead>
<tr>
<th>Material</th>
<th>Be</th>
<th>BeO</th>
<th>Fe</th>
<th>Al</th>
<th>Si</th>
<th>C</th>
<th>Mg</th>
</tr>
</thead>
<tbody>
<tr>
<td>DshG-200</td>
<td>99.34</td>
<td>0.79</td>
<td>0.024</td>
<td>0.0045</td>
<td>0.013</td>
<td>0.077</td>
<td>0.003</td>
</tr>
<tr>
<td>TshG-56</td>
<td>99.10</td>
<td>0.95</td>
<td>0.10</td>
<td>0.013</td>
<td>0.014</td>
<td>0.077</td>
<td></td>
</tr>
<tr>
<td>TR-30</td>
<td>97.64</td>
<td>3.2</td>
<td>0.12</td>
<td>0.015</td>
<td>0.013</td>
<td>0.05</td>
<td>0.006</td>
</tr>
<tr>
<td>TRR</td>
<td>97.64</td>
<td>3.2</td>
<td>0.12</td>
<td>0.015</td>
<td>0.013</td>
<td>0.05</td>
<td>0.006</td>
</tr>
<tr>
<td>S-65B</td>
<td>≥99.0</td>
<td>0.98</td>
<td>0.09</td>
<td>0.018</td>
<td>0.026</td>
<td>≤0.1</td>
<td>≤0.003</td>
</tr>
<tr>
<td>TE-30</td>
<td>98.13</td>
<td>2.5</td>
<td>0.11</td>
<td>0.015</td>
<td>0.013</td>
<td>0.088</td>
<td></td>
</tr>
<tr>
<td>TIP-30</td>
<td>98.27</td>
<td>2.1</td>
<td>0.11</td>
<td>0.014</td>
<td>0.013</td>
<td>0.075</td>
<td>0.006</td>
</tr>
</tbody>
</table>

2. MATERIALS AND EXPERIMENTAL PROCEDURE

Seven grades of beryllium were investigated with the different characteristics of the initial powder and properties of the billets made of them. Among them six grades (DshG-200, TR-30, TshG-56, TRR, TE-30, TIP-30) were manufactured by VNIINM (Russia) and other one (S-65B) - by Brush Wellman (USA). Chemical composition and initial characteristics of the materials are presented in table 1 and 2.

All the materials were irradiated in active zone of CM-3 reactor in leaktight capsules up to the fast neutron fluence \((5.5 - 6.2) \times 10^{21} \text{ cm}^{-2}(E > 0.1 \text{ MeV})\) at two irradiation temperatures \(T_1 = 130 - 180^\circ\text{C}\) and \(T_2 = 650 - 700^\circ\text{C}\). Irradiation parameters are presented in table 3. Two types of the specimens have been tested. First type, which was used for swelling and compressive characteristics measurements, represents the solid cylinders machined to close tolerances with 6 mm in diameter and 9 mm in length. The other type of the specimens with 3 mm in diameter and 28 mm in length was used for tensile testing and microstructure investigations. The testing was performed in air environment at room temperature (RT) and in vacuum at higher temperatures ( >RT). The rate of straining was 1 mm/min both for tensile and compression testing.

3. EXPERIMENTAL RESULTS AND DISCUSSION

3.1. Swelling

The results of swelling measurement are presented in table 4. After the irradiation at 130-180°C swelling was not revealed for all the samples investigated. After the irradiation at 650-700°C swelling for all the samples was found to be in the range 0.1 - 2.1 %. All the materials can be divided into two groups by the swelling values. The swelling value of the first group of materials (TR-30, TRR, TE-30, TIP-30) was rather small and did not exceed 0.4 %, while that for the second group (S-65, DshG-200, TshG-56) was significantly higher > 1.3 %. Data presented in tables 2 and 4 confirm the conclusions of previous works [1-4], that fine-grained beryllium grades with increased beryllium oxide content are less susceptible to swelling at high temperature irradiation. In TR-30 and TRR grades, having the smallest grain size and the highest beryllium oxide content (3.2 %), swelling did not...
Table 2
Characteristics of beryllium

<table>
<thead>
<tr>
<th>Material</th>
<th>Average Grain Size, µm</th>
<th>Direction</th>
<th>Ultimate Tensile Strength, MPa</th>
<th>Yield Stress, MPa</th>
<th>Total Elongation, %</th>
<th>Ultimate Compressive Strength, MPa</th>
<th>Yield Compressive Stress, MPa</th>
<th>Failure deformation method</th>
</tr>
</thead>
<tbody>
<tr>
<td>DshG-200</td>
<td>22</td>
<td>trans.</td>
<td>314</td>
<td>288</td>
<td>0.8</td>
<td>1606</td>
<td>497</td>
<td>31.0</td>
</tr>
<tr>
<td></td>
<td>25</td>
<td>long.</td>
<td>240</td>
<td>-</td>
<td>0</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>TshG-56</td>
<td>22</td>
<td>trans.</td>
<td>458</td>
<td>388</td>
<td>1.6</td>
<td>1653</td>
<td>561</td>
<td>29.5</td>
</tr>
<tr>
<td></td>
<td>25</td>
<td>long.</td>
<td>263</td>
<td>256</td>
<td>0.2</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>TR-30</td>
<td>9</td>
<td>trans.</td>
<td>691</td>
<td>579</td>
<td>1.8</td>
<td>1927</td>
<td>609</td>
<td>30.1</td>
</tr>
<tr>
<td></td>
<td>9</td>
<td>long.</td>
<td>589</td>
<td>580</td>
<td>0.2</td>
<td>2074</td>
<td>674</td>
<td>31.4</td>
</tr>
<tr>
<td>TRR</td>
<td>9-8</td>
<td>trans.</td>
<td>688</td>
<td>-</td>
<td>0</td>
<td>2100</td>
<td>688</td>
<td>33.4</td>
</tr>
<tr>
<td></td>
<td>9-8</td>
<td>long.</td>
<td>690</td>
<td>-</td>
<td>0</td>
<td>1986</td>
<td>694</td>
<td>27.5</td>
</tr>
<tr>
<td>S-65B</td>
<td>18-20</td>
<td>long.</td>
<td>441</td>
<td>312</td>
<td>3.5</td>
<td>1752</td>
<td>563</td>
<td>37.8</td>
</tr>
<tr>
<td>TE-30</td>
<td>16</td>
<td>trans.</td>
<td>376</td>
<td>351</td>
<td>0.7</td>
<td>1296</td>
<td>504</td>
<td>23.4</td>
</tr>
<tr>
<td></td>
<td>23</td>
<td>long.</td>
<td>669</td>
<td>472</td>
<td>14</td>
<td>1807</td>
<td>398</td>
<td>35.8</td>
</tr>
<tr>
<td>TIP-30</td>
<td>18</td>
<td>trans.</td>
<td>487</td>
<td>416</td>
<td>1.8</td>
<td>1692</td>
<td>453</td>
<td>31.3</td>
</tr>
<tr>
<td></td>
<td>18</td>
<td>long.</td>
<td>482</td>
<td>463</td>
<td>0.8</td>
<td>1677</td>
<td>463</td>
<td>30.5</td>
</tr>
</tbody>
</table>

<sup>a</sup> Hot isostatic pressing, <sup>b</sup> vacuum hot pressing

exceed 0.1 % after irradiation with a fluence 5.5 \(10^{21}\) cm<sup>-2</sup> (E >0.1 MeV) at 650-700°C.

3.2. Tensile test results

The data on mechanical properties of beryllium under tensile testing before and under irradiation are presented in table 5. Testing was performed both at room temperature (RT) and at 650°C. Irradiation at both temperatures results in a significant change of beryllium properties.

After irradiation at 650-700°C ultimate tensile strength (\(\sigma_b\)), yield stress (\(\sigma_0.2\)) and total elongation (\(\delta\)) of beryllium decrease on 21-49 %, 7-44 % and 86-100%, accordingly, when testing was performed at RT. Under testing at 650°C, \(\sigma_b\), \(\sigma_0.2\) and \(\delta\) change on +10 % - 47 %, +42 % - 40 %, - (94 +100) %, accordingly. Under testing both at RT and 650°C \(\sigma_b\) and \(\sigma_0.2\) for the first group of materials (TR-30, TRR, TE-30, TIP-30) were significantly higher than that for the second group (S-65, DshG-200, TshG-56). Elongation for the first group of materials is identical to that for the second group or higher.

After the irradiation at 130-180°C, \(\sigma_b\) and \(\sigma_0.2\) reduce on 26-55 %, when testing at room temperature, or change on + 70 % - 27 % under the testing at 650°C. Elongation falls on 22-93 % at RT and on 92±100 % at 650°C. Under the testing at RT, \(\sigma_b\) and \(\sigma_0.2\) of the first group of materials exceed significantly that for the second group. When testing at 650°C, \(\sigma_b\) and \(\sigma_0.2\) for the first group of materials are generally higher than that for the materials of the second group in unirradiated condition. Elongation
Table 3
Irradiation parameters of beryllium

<table>
<thead>
<tr>
<th>Parameter</th>
<th>DshG-200</th>
<th>TshG-56</th>
<th>TR-30</th>
<th>TRR</th>
<th>TE-30</th>
<th>S-65</th>
<th>TIP-30</th>
</tr>
</thead>
<tbody>
<tr>
<td>Neutron flux (E &gt; 0.1 MeV), cm⁻²·c⁻¹</td>
<td>(5.0-7.5)·10¹³</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Neutron flux (E &lt; 0.68 eV), cm⁻²·c⁻¹</td>
<td>(1.0-1.5)·10¹⁵</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>( F_s (E &gt; 0.1 \text{ MeV}) ), cm⁻²</td>
<td>(4.2-6.2)·10²¹</td>
<td>(4.2-6.2)·10²¹(T₁)</td>
<td>(3.9-5.5)·10²¹(T₁)</td>
<td>(3.9-5.5)·10²¹</td>
<td>(3.9-5.5)·10²¹</td>
<td>(4.2-6.2)·10²¹</td>
<td>(3.9-5.5)·10²¹</td>
</tr>
<tr>
<td>( F_{th} (E &lt; 0.68 \text{ eV}) ), cm⁻²</td>
<td>(2.1-3.1)·10²⁰</td>
<td>(2.1-3.1)·10²⁰(T₁)</td>
<td>(1.8-2.8)·10²⁰(T₁)</td>
<td>(1.8-2.7)·10²⁰</td>
<td>(1.8-2.7)·10²⁰</td>
<td>(2.1-3.1)·10²⁰</td>
<td>(1.8-2.7)·10²⁰</td>
</tr>
<tr>
<td>Radiation damage, dpa</td>
<td>2.0-3.0</td>
<td>1.8-3.0</td>
<td>1.8-2.8</td>
<td>1.8-2.7</td>
<td>1.8-2.7</td>
<td>2.0-3.0</td>
<td>1.8-2.7</td>
</tr>
<tr>
<td>Irradiation temperature, °C</td>
<td>650-700(T₁) 650-700;</td>
<td>(T₁) 650-700;</td>
<td>(T₂) 130-170;</td>
<td>650-700(T₁) 650-700;</td>
<td>650-700(T₁) 650-700;</td>
<td>650-700(T₁) 650-700;</td>
<td>(T₂) 130-170;</td>
</tr>
</tbody>
</table>

3.3. Compression test results

Table 6 presents the data on effect of irradiation on mechanical properties of beryllium under compression testing. Testing was performed at RT and in the range of 450-650°C.

Testing at RT shows, that after irradiation at 650-700°C compressive strength \( (\sigma_{0.2}) \) and failure deformation \( (\varepsilon) \) decrease on 30±49% and 56±97%, accordingly, while compressive yield stress \( (\sigma_{0.2}) \) increases on 10±78%, that is a characteristic of the irradiated beryllium. Failure deformation for the materials of the first group was in the range of 3.5±14%, while that for the second group was 1.0±6.2%.

Irradiation at 130-180°C results in a more significant embrittlement of beryllium, than that at 650-700°C. The \( \sigma_{0.2} \) and \( \varepsilon \) values decrease on 14±54% and 74±100%, accordingly, while \( \sigma_{0.2} \) increases on 17±172%. By testing at 450-650°C it was shown that \( \sigma_{0.2} \) increase on 10±78% after irradiation at
Table 4
Swelling of beryllium samples

<table>
<thead>
<tr>
<th>$T_{in}$, °C</th>
<th>Material</th>
<th>DshG-200</th>
<th>TshG-56</th>
<th>TR-30</th>
<th>TRR</th>
<th>TE-30</th>
<th>S-65B</th>
<th>TIP-30</th>
</tr>
</thead>
<tbody>
<tr>
<td>130-180</td>
<td>Swelling, %</td>
<td>-0.04±0.3</td>
<td>-0.1±0.5</td>
<td>-0.06±0.2</td>
<td>-0.08±0.1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>650-700</td>
<td>Swelling, %</td>
<td>1.36±1.1</td>
<td>2.1±1.0</td>
<td>0.1±0.05</td>
<td>0.1±0.1</td>
<td>0.36±0.3</td>
<td>1.77±1.0</td>
<td>0.38±0.1</td>
</tr>
<tr>
<td></td>
<td>Helium content, at. %</td>
<td>1060-1140</td>
<td>1060-1140</td>
<td>940-1010</td>
<td>940-1010</td>
<td>940-1010</td>
<td>1060-1140</td>
<td>940-1010</td>
</tr>
</tbody>
</table>

Table 5
Tensile test results of beryllium samples before and after irradiation with fluence up to 5.1-5.8 \times 10^{21} \text{ cm}^2 \text{ (up to 1.8- 2.8 dpa) The rate of straining - 1 mm/min.}

<table>
<thead>
<tr>
<th>Material</th>
<th>Condition</th>
<th>$T_{in}$, °C</th>
<th>Test temperature, °C</th>
<th>20</th>
<th>650</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>$\sigma_b$, MPa</td>
<td>$\sigma_{0.2}$, MPa</td>
<td>$\delta$, %</td>
</tr>
<tr>
<td>DshG-200</td>
<td>unirr</td>
<td>-</td>
<td>314</td>
<td>288</td>
<td>0.8</td>
</tr>
<tr>
<td></td>
<td>irr</td>
<td>650-700</td>
<td>61</td>
<td>161</td>
<td>0</td>
</tr>
<tr>
<td>TshG-56</td>
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Table 6
Compression test results of beryllium samples before and after irradiation with fluence up to $5.5-6.2 \cdot 10^{21}$ cm$^{-2}$ (up to 2.7-3.0 dpa) The rate of straining - 1 mm/min

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<th>Test temperature, °C</th>
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<td>$\sigma_0^0$, MPa</td>
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<td>$&gt;793$</td>
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650-700°C and on 17-172 % after irradiation at 130-180°C. Unfortunately, attempts to carry a test to failure at 450-650°C have not met with success due to a good plasticity of the initial beryllium at higher temperatures (> 50-60 %) and also because of the restrict force of the testing machine, used for testing at higher temperatures. At the maximal stress of 900 MPa, the deformation of beryllium irradiated at 650-700°C was 10-44%, and that of the samples irradiated at 130-180°C either was lower than $\sigma_0^t$ ($\sigma_{max} < \sigma_0^t$) or varied in the range of 0-11 %.

These results show that the materials of the first group, having higher mechanical properties in the initial condition, demonstrate less degradation of the mechanical properties under irradiation. The advantage in elongation of the second group of materials (S-65) disappears under irradiation.
Fig. 1 Microstructure of beryllium irradiated at 650-700°C
a,b - TE-30 grade; c,d - TR-30 grade; e,f - S-65 grade; (a-e) - x100 000; f - x50 000.
The tendency for a degradation of mechanical properties revealed at this work, allows to anticipate, that under the neutron irradiation a resistance of the second group of materials against a thermal fatigue will reduce faster than that of the first group. It can lead to a loss of the advantage in the resistance against thermal shock/fatigue, which were reported in [7,8] for S-65 and DshG-200 grades in unirradiated condition. Therefore, it seems improper to choose beryllium grade taking into account either thermal fatigue or neutron irradiation behaviour separately.

To validate the choice for fusion application there is a need to summarise the data on beryllium behaviour under combined exposure to thermal loading and neutron irradiation. In addition, it seems reasonable to say that the different approaches should be used, when choosing beryllium for the application in the different fusion elements.

3.4. Microstructure of irradiated beryllium

Microstructures of the beryllium irradiated at different temperatures are shown on Fig. 1 and Fig. 2. It was found that the bubble formation in the samples investigated takes place on the grain boundaries, inside the grains of a matrix and on the interface of matrix-second phase. The bubbles have a specific crystallographic form. In addition, the linear aggregates of pores have been brought out in matrix, whose formation took place on dislocations occurred in material before the irradiation annealing.

The main features of the helium bubbles formation in the samples are as follows:

- the most large-sized bubbles are found at matrix-second phase interface. Bubble dimension is comparable with the dimension of the inclusion;
- the most large-sized bubbles in matrix decorate the initial dislocations dissociated at irradiation by creep across;
- among the matrix bubbles the disk-like bubbles are generally revealed with a thickness of $2\pm2.5$ nm and a diameter of $15\pm20$ nm. Gabirus of the bubbles lies in the basis plane (002) of hexagonal close-packed lattice of beryllium;
- the size of grain boundary bubbles depends on the grains disorientation. They have a lens-like form with

Fig.2 Microstructure of beryllium irradiated at 130-180°C
a - TR-30 grade ($\times100\ 000$), b - TshG-56 ($\times25\ 000$); c - TIP-30 ($\times50\ 000$).
axis ratio 2:1. Grain boundary bubbles formation as well as the bubbles formation inside the grains result in the distortion of crystal lattice.

The most average size of the bubbles was found in S-65 beryllium grade. It is 20.6 nm in matrix, although the bubbles formed earlier at dislocations are by the factor 2 larger. The average size of the bubbles, which formed on the boundaries, varies from 17.5 to 20 nm, depending on the disorientation of the grains. The sizes of matrix and boundary bubbles in DshG-200 and TshG-56 grades were 15 nm and 7-12 nm (depending on the grains disorientation), accordingly. In TR-30 grade the size of grain-boundary pores was 3-10 nm. In the samples irradiated at 130-180°C the presence of bubbles was not revealed.

CONCLUSION

The study on swelling, mechanical properties and microstructure of seven grades of beryllium has been carried out. It has been confirmed, that fine-grained beryllium grades with high beryllium oxide content are less susceptible to swelling at high temperature irradiation. Beryllium grades TR-30 and TRR having the smallest grain size and the highest beryllium oxide content (3.2%), demonstrate the minimal swelling, which does not exceed 0.1% after the irradiation with fluence $5.5 \cdot 10^{21}$ cm$^{-2}$ (E >0.1 MeV) at 650-700°C.

After the irradiation at 130-180°C with fluence $5.5 \cdot 10^{21}$ cm$^{-2}$ (E >0.1 MeV) swelling in beryllium was not found.

Irradiation results in a significant change of mechanical properties of beryllium and its embrittlement. The lower the temperature of irradiation the higher an extent of embrittlement. Beryllium grades having the higher level of strength properties in initial condition and the less susceptibility to swelling (group I), demonstrate the less degradation of strength properties under irradiation. The advantage in initial elongation for the materials of the second group (S-65) disappeared under irradiation.

REFERENCES


8.7 Specific Features of Reactor or Cyclotron α-particles Irradiated Beryllium Microstructure


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249020, Obninsk, Russia

Studies were carried out into microstructure changes accompanying helium swelling of Be reactor neutron irradiated at 450°C or α-particles implanted in cyclotron to reach the same volume accumulation of He (6-8 ncm³He/cm³Be). The microstructures of reactor irradiated and implanted samples were compared after vacuum anneal at 600-800°C up to 50 h.

The irradiated samples revealed the etchability along the grain boundaries in zones formed by adequately large equilibrium helium pores. The width of the zones increased with the annealing time and after 50 h reached 30μ. Depleted areas 2-3μ dia were observed in some regions of near grain boundary zones.

The roles of grain boundaries and manufacturing pores as vacancies' sources and helium sinks are considered.

1. INTRODUCTION

Very often the existing classic understandings [1] of elementary phenomena determined swelling do not allowed to forecast the swelling value as were developed respectively to some ideal structures. Despondently to their technological prehistory, real metals have the wide spectrum of different structure particularities that may influence determinatively on the material's behavior under irradiation and high-temperature annealing.

In order to make sensible operations on the manufacturing technology' optimization of beryllium respectively to helium swelling, one needs some additional data about the mechanism of helium pores' forming around structure defects. The following materials consist of data for the grain boundaries and manufacturing pores received from the microstructure investigation of extruded beryllium after swelling under out-of-pile annealing.

It's shown that the above defects are the sources of vacancies and helium sinks and several proposals are suggested on beryllium structure's optimization in respect of helium swelling. The structure researches done using the method of the optical microscopy that stipulated by the necessity of studying of a process going on in rather large material volumes.

2. EXPERIMENTAL PART

2.1. Samples' material

The samples for irradiation have a cylinder shape (diameter - 7.5 mm, length - 15 mm) and were made from the bars of extruded distilled beryllium of the 400 μ powder. Beryllium content in the powder was 98%. Main impurities: oxygen (1.78%), carbon (0.1%), nitrogen (0.12%), iron (0.06%). Samples density - 1,845-1,848 g/cm³, porosity - 1.5-2%. Samples' structures formed by equiaxes grains with 3-40 μ diameter (fig. 1, 2). High heterogeneity of grains distribution by the size and porosity by the volume are the peculiarities of the structure. The low-porosity areas, consisted of the grains with 15-40 μ in diameter and have the lens-shape (diameter - 100-300 μ, length - 800-2000 μ), are “sunk” into material formed by the grains with the size less than 15 μ with the considerable amount of pores and oxide inclusions. Most of manufacturing pores presented in small grains’ accumulations, either continuous or stitched channel-typed formations with
1-3 μ thickness and several hundreds of microns length, that stretched along the extrusion axis. Such a material is very suitable for metallographic studying because the sections with different types of structure are existed within the polished field.

Figure 1. Areas with fine and coarse grains.

Figure 2. Unirradiated microstructure of beryllium after annealing 550°C, 1000 h.

2.2. Irradiation and annealing.

Reactor irradiation held in the periphery of a BR-5 reactor’s active zone at 450-475°C temperature that close to sodium temperature in the entrance of the reactor. For irradiation the samples placed instead of fuel along the fuel tubes made from stainless steal with 8,55 mm diameter. The internal diameter of the shell was 7,75 mm. Hermetization of the tubes with beryllium made in the open air. The samples irradiated within 12000 hours under the maximum neutron flux of $4.5 \times 10^{14}$ n/cm²*s to reach the maximum fluence $1.36 \times 10^{22}$ n/cm² and maximum helium accumulation 7-8 ncm³He/cm³Be. For the samples located near the fuel tubes ends during irradiation the helium content was 1-1.5 ncm³He/cm³Be. Helium content was determined by the mass-spectrometric method.

Out-of-pile annealing of the irradiated samples was held in the vacuum within 2-300 hours at the temperatures 600-800°C. The present research is mainly shows the results for the samples annealed within 2 hours at 600-750°C. Such a temperature-time range forms the optimal conditions for the usage of metallographic methodology of the vacancies’ sources determination that initiate helium swelling because the length of the diffusion source is comparable by the value with a view-field size of an optical microscope at the maximum increasing level.

2.3. Methodology of metallographic study of helium porosity in beryllium.

For finishing of the metallogphical methodology of helium pores determination the initial samples were irradiated by $\alpha$-particles in a cyclotron. Irradiation was made on a internal bunch at 20°C and on a deviated bunch at 500°C. The energy value for $\alpha$-particles was 40 Mev. The exposition was selected so to receive the average helium concentration in an braking area on the level of 6-8 ncm³He/cm³Be.

For helium pores determination a changed variant of the methodology [4] was used where helium pores presence is determinated by a specific etchability appearance in spirituous solution of hydrofluoric acid. The methodology was tested on the $\alpha$-particles irradiated and annealed samples (fig. 3). The selected etching regime allows to identify reliably the helium pores population in the manufacturing pores background. The methodology of metallographic data processing was also used that allowed to receive a size value of a separate isolated and big enough helium pore and helium pores population characteristics [6].

Vacancies’ sources determination under helium swelling [1] is stipulated by the fact that most of vacancies needed for a transportation of helium, located either in a solid solution or in initial supersaturated helium pores [7], to equilibrium pores population is generated on the structural...
defects but not homogeneously distributed along the whole volume. Because of that reason helium porosity formation during annealing first of all starts around the structural defects, that are the main vacancies' sources and that are distinguished by etchability' appearance around them. Helium pores' filling of the whole material volume during swelling goes up by increasing of the influence areas of the separate vacancies' sources.

Local helium sinks determination. Should the material has structural defects on which the sink of helium from solid solution is possible during irradiation or annealing, the areas appeared around them where the helium concentration drops down to a level not enough for helium pores' forming. Such an areas determinated under etching as a non-etching areas free from the pores.

2.4. Experimental results.

During irradiation at 450-475°C the helium sink into structural defects and primary supersaturated pores takes place. The main helium structural sinks are the manufacturing pores, that may decrease the effectiveness of them as the vacancies' sources.

The essential concentration of the small closed manufacturing pores in the researched metal was found, the pores that lost under irradiation the ability of vacancies' generation because the helium presence under the pressure that essentially bigger than equilibrium one. In the arias of the small grains the concentration of these pores is so high that the depleted zones located around crosses the whole material volume so all appeared under irradiation helium goes into these pores and no new pores appeared after the next annealing procedure. In the big grains accumulations where the concentration is rather small, helium is still located into the initial pores after irradiation.

Figure 4 shows the beryllium structure after irradiation up to helium accumulation 7-8 ncm$^3$He/cm$^3$Be. In comparison with the initial structure (fig.2) the strong etchability along grain boundaries and moderate on the grain's field appeared. The large grains' boundaries reveal like the complete knotty strips the thickness of that increases with etching time increasing. The small grains' boundaries reveal like double- and triple-out stripped lines not-made the complete contour along the grain perimeter. Within the fields of the small and the large grains the separate chaotic etching figures reveal having the size up to 1 μ with the surface density of $4 \times 10^{11}$ cm$^{-2}$. The observed changes corresponded to the appearance in the
structure of essential amount of the pores on the grain boundaries and not-essential pores - in the grain field with the diameter up to 0,1 μ. They do not lead to visible volume changes.

The main change in the structure after out-of-pile annealing at 600-800°C is the appearance of the areas of strong etching around grain boundaries and also manufacturing pores in big grains' accumulations, increasing in size with time and temperature increasing. First this effect was registered in helium implanted [8]. The main peculiarity of these areas in the studied samples is the essential variety of the thickness and the continuity along the sample volume.

Figure 5. Near boundary etching areas. Helium accumulation 7-8 ncm³He/cm³Be. Annealing 750°C, 2 h.

Fig. 5 shows the microphotoes of those structure fragments where the near boundary etching area has the approximately constant width 2-5 μ along the contour of all the boundaries. In the grains with the size of less than 15 μ met into the big grains' accumulations the width of the strip changes within the same limits. The field of grains with diameter less than 8-10 μ is etched fully in the most cases. The strip adjoins close or almost close to the grain boundary and the sharp boarder from the side turned to the grain center. The strip's boundary on the front smoothly repeats the grain's boundary shape.

Etching figures sizes and their surface density in the near boundary strip is approximately constant and have the values of 0,2-0,4 μ and (2-4)×10⁸ cm⁻² correspondingly. The corresponding values of the average pores size in the near boundary strip and the pores concentration are: 0,1 μ and (2-3)×10¹¹ cm⁻³. The porosity (swelling) estimated according to these values in the near boundary strip is 1-2 %. This value is essentially bigger than the swelling value outside the near-boundary fields that very close to the swelling value after irradiation and is 0,2-0,3 %.

Figure 6. Near grain boundary strip of variable width. 7-8 ncm³He/cm³Be. Annealing 750°C, 2 h.

From fig. 6 one can conclude the presence of a definite tendency in the strip's width changing within the volume of the big grains' accumulation that can not be explained by the difference in a inclination angle of the boundary's surface to the polished surface. The grains with the maximum width value of the near boundary strip located on the periphery of the big grains' accumulation. By moving away from the accumulation's boundary the width of the near-boundary strip is gradually decreasing with the each next grain.

Since the specific swelling in the near boundary strip is constant, its width is proportional to the average (during the annealing time) float of the vacancies from the grain's surface unit towards inside the grain. Change of the near boundary strip width along the grain boundary means that the effective capacity of the boundary vacancies' sources is not the attribute of the boundary itself and should stipulated by the other parameters not connected with the grain boundary. In [8] where this effect was found for the first time the grain boundary stress
jump and the external vacancies' sources influence were pointed as such a stipulating parameters. In the last case the grain boundary is considered as a vacancies' conductor capable to give a part of the circulated along it vacancies into the grain volume.

In the considered case the estimated stresses should be compensated not inside the very grain but inside the large grains' accumulations in general, and should change along the accumulation's radius. The appearance of such a stresses into the researched materials is possible, as it consists in essence of two phases - the large grains' accumulations and the small ones, that have the different swelling behavior under irradiation and annealing. In this case the appearance of the compressing or tensile stresses and the attendant effects, constant by the accumulation volume is inevitable. As the observed changes of the near boundary strip width do not correspond to this forecast, it can not be caused by the stresses. So summing up, the most possible is considered the conclusion that the grain boundaries are the conductors but not the vacancies' sources. In the large grains' accumulations the boundaries generate the vacancies at the account of the sources located outside of these accumulations.

This conclusion evidences the specific spreading of the vacancies' sources along the material volume. As the grain boundaries should be excluded from the vacancies' sources list, the other possible vacancies' sources are the manufacturing pores and the second-phases inclusions, almost fully concentrated into the small grains' accumulation areas. The half-width of the diffusion influence these sources in the aria of the large grains' accumulations is app. 25 μ (along the boundary, at T=750°C). The corresponding effective grain boundaries diffusion coefficient is 4,5×10⁻¹⁰ cm²/s. This value is lesser than the coefficient of the volume self-diffusion in Be 4,9×10⁻⁹ cm²/s [9] and the coefficient of vacancies' diffusion in Be 6,5×10⁻⁶ cm²/s at 750°C [10].

Figure 7 shows the fragments of a structure with helium content 1-1,5 ncm³He/cm³Be after annealing at 700°C, 2 h. It's obvious that in the large grains' accumulations the near boundary width is 3-4 times bigger than in the sample with helium concentration of 6-8 ncm³He/cm³Be and is

![Figure 7. He accumulation 1-1.5 ncm³He/cm³Be. Annealing 700°C, 2 h.](image)

5-6 μ. Besides, the pores size in the near boundary strip of this sample changes along its width from 0,04 μ -just near the boundary, up to 0,1 μ. With increasing of the annealing time - the strip width increases at the account of increasing the width of the area with the most big pores. Such a structure and evolution of the strip evidence about the helium concentration decrease near the boundary and also about the presence of the helium sink at a grain's boundary.

![Figure 8. Channel-type manufacturing pores with etchability strips (arrows). He accumulation 6-8 ncm³He/cm³Be. Annealing 700°C, 2 h.](image)

Figure 8 shows a structure' fragments consist of the manufacturing pores look like the prolonged channels paralleled to the extrusion axis. One can see a comparatively thick channel pore with a thickness of 1 μ that is the vacancies' source and the helium sink. Its quality as a vacancies' source is proved by the presence in its surroundings of an area of the increasing etchability, looks like a strip with the constant width, similar to the near boundary
strip, meanwhile in contradiction to the last one it situated on the distance of 1-2 µ from the pore's edge. As the material was not etched in the space between pore's back front and pore's edge and, therefore, is free of helium, the pore - is the helium sink. This impoverished area formed under irradiation, as its width does not depend either on time or annealing temperature.

In fig. 8 is seen that the helium outflow from the grain volume to the boundary is rather less than to the manufacturing pore, bit does not equal zero. The helium float from the grain's boundary to a crossing technological pore does not take place.

In [11] in order to describe a near boundary strip in helium implanted nickel the model is offered that developed in [12] to describe diffusion into an environment filled with the traps with the constant concentration along the volume. Vacancies are considered as a diffusing substance and the traps equalized with the not mobile initial helium pores. In such a case the estimation formula for a strip's width in the surrounding of the grain boundaries and a free surface has the view of:

\[ L = \sqrt{2DtC_0} / S \]

where:
- \( L \) - strip's width,
- \( D \) - self-diffusion coefficient,
- \( t \) - annealing time
- \( S \) - swelling in the strip,
- \( C_0 \) - vacancies concentration in the strip.

The formula is written respecting suggestion that \( S_0 \approx 0 \) and \( C_0 \gg C_0^* \) where \( S_0 \) and \( C_0^* \) - swelling and vacancies' concentration value outside the strip.

The comparison of the results under the formula (1) with received data can be not always correct because the initial conditions of a model used in gaining of this formula do not fulfilled notoriously for the separate parts of a sample. One can refer to them the central parts of the large grains' accumulations remote from the vacancies' sources and the manufacturing pores' boundaries, where the irregular spreading of helium takes place near the manufacturing pores' edge. Evidently, the most suitable in comparison with the estimates are the results received for the peripheral of the large grains' accumulations and near the channel-typed manufacturing pores(fig. 5,8). In such a case there is no absolute insurance in fulfillment of only one of the initial condition - the helium presence in the initial supersaturated pores after irradiation.

The results of the strip's width estimations are shown for two temperatures - 700 and 750°C for annealing time 2 hours, estimated value of the strip's width are 4,4 µ and 38 µ but the experimental - 1-2 µ and 30 µ correspondingly. At 750°C for 3 hour of annealing the estimated value of the strip' width is 9,3 µ, and experimental - 3-4 µ. During the estimation self-diffusion coefficients in Be were used, determinated in [9] and the meaning of the vacancies' concentration in Be, estimated in [10]. The swelling value was also used that equals 0,04. This value is not experimental. It corresponds to the full pump over of the manufacturing pore (0,02 of the sample volume) to the helium pores of the large grains' accumulations (0,5 of the sample volume). The estimated values are bigger than experimental but coincide with them by the order. As in the diffusion experiments such a difference is considered as a coincidence, the formula (1) describes rather good the strip's width changes depending to temperature and time.

In the formula (1) the dependence of the strip's width from helium concentration is stipulated by a \( S \). As know that swelling decreases with helium concentration' decrease, the formula (1) gives that strip's width dependence from helium concentration that observed experimentally.

Local depleted zones (LDZ). Fig. 3 and 9 shows the microstructure fragments where the local degradation of the near boundary strip is observed, with forming of the rounded areas with a diameter of 2-4 µ having one circle-like etching figure with 0,3-1 µ size in the center. These areas' material around the central figure does not etched under very strong etching. The research of the sample with the low helium content evidences that the helium content into observed areas of the near boundary strip (outside the central etching figure) is essentially lesser than 1 ncm⁻³He/cm³Be. Therefore, in these areas the impoverishing of helium no less than ten times took place.
The impoverishing took place, obviously, by the helium outflow to the one and only object located into the depleted area, that very object that is a pore by the etching character. 

The size of the LDZ does not depend on temperature and time of annealing. It is very close to the length of helium diffusion in Be, 0,7 μ, estimated for irradiated conditions with usage of the diffusion coefficient value of $3 \times 10^{17}$ cm$^2$/s, stipulated in [13]. From the following one may conclude that helium spreading in the LDZ was formed under irradiation, and outside the depleted area helium presents after irradiation in the low-moving form excluded the possibility of its migration to the distance of $\sim 1$ μ. This shows that during irradiation at $T=450-475°C$ whole helium comes out from the solid solution to the low-moving initial supersaturated helium pores. 

Sometimes one can see the regulate location of the LDZ that consists on that fact that the several LDZ situated like a stitch parallel to extrusion axis. If this stitch comes out to the grain's field outside the near boundary strip, the same etching figures are seen usually on it. Perhaps, the nature of this etching figures is the same and they present themselves the pores with the gas under pressure, that not-allowed the vacancies' generation. Only helium can be this very gas because before irradiation the similar pores were not seen in the Be structure. It's possible, that before irradiation their size was lesser than resolution limit of 0,04 μ. During irradiation their size increases up to 0,2 μ and they became seen as the etching figures in the fields of the large grains appeared in the structure after irradiation. During annealing their size decreased up to 0,4-0,5 μ. 

The estimation of the gas' pressure in the pore with the radius of 0,25 μ located in the center of LDZ with the radius 1,5 μ, was made respectively the suggestion, that all gas appeared in this very area has come to the central pore, and gives the value of 1,2-1,6 MPa. This value is essentially lesser than the value of equilibrium helium pressure in the pore (10$^3$ MPa). The difference can be explained by the mistakes in measurements of pore and LDZ radiuses. 

The LDZ' concentration into the separate grains can change within wide limits. Sometimes the concentration and spreading of the LDZ by the sample volume is so that the partial or full overlapping may take place. In such a cases the new filed with helium pores under annealing are not formed and the polished field is not etched in such a structure's fragments. 

The pores located in the center of the LDZ most likely have the technological origin. Perhaps, they appeared as a result of vacancies' condensation happened during annealing of the deformation' structure on the definite point of the technological cycle of the material manufacturing. 

In the samples with helium concentration of 1-1,5 ncm$^3$/He/cm$^3$Be the LDZ' radius is app. two time bigger than in the samples with helium concentration of 6-8 ncm$^3$/He/cm$^3$Be. It's possible that in the general case the LDZ' forming can not be described by a simple diffusion equation. From all above said it's followed that under irradiation conditions parallel to the helium outflow to the structural defects, the manufacturing pores & the grain boundaries, its outflow to the initial helium pores happens. Because of that the parameters of the LDZ around the structural defects depend on ratio of the structural & volume sinks capacity and the speed of helium accumulation. The LDZ boundary fixes the minimum level of helium concentration needed for the initial helium pores' forming. 

In fig 10 the typical fragment is shown, consisted of the small grains' accumulation. The small grains' boundaries are etched in the same manner as the boundaries of the large grains. They have the dark contrast and moderately increases its own thickness with the etching time increasing.
Perhaps, the size and concentration of the helium pores on the boundaries of both large & small grains is not differ essentially. The particularity of the small grains' accumulations is their low etchability: so the field of the small grains kept light even after long-time annealing accompanied by the essential volume changes. The reason of such a behavior is the presence in the even each small-sized grain of the etching figures with circle-like shape 0,5-0,8 μ in size. These etching figures are the imprint of the pores that, perhaps, identical with the central pores of the LDZ. Just to these pores almost all formed helium comes out. This conclusion is confirmed by the fact, that in several small grains of the accumulations where the pointed types of etching figures absent, the normal helium porosity is formed with no presence of the essential outflow to the grains' boundaries.

3. CONCLUSION

Offered results show the complex and not simple character of the influence of the porosity and the grain boundaries on helium swelling. Researched extruded material can be considered as if consist of two phases: a) small grained, contains the main part of the manufacturing pores and oxide inclusions and b) large grained, with the grain boundaries almost free from oxide inclusions; regarding the boundary of such a type one can at least state that they are the vacancies' conductors first of all but not the sources of them. Just because of such a structure, when the main vacancies, sources located mainly in the small grained areas, one can make the conclusion about the real role of the boundaries as the vacancies sources. So that means, the grain size plays the secondary role in helium swelling of beryllium.

Undoubtedly, the small pores located in the local depleted zones and in some small grains play an important role. It is necessary to clear the nature and conditions of appearance of them, for what the usage of more delicate methods of a research should be applied.

The questions connected with the role of dislocations as the possible vacancies' sources and helium condensation centers are left outside of the subject of this discussion as the applied methodology unfortunately does not allow to reveal a such a structure details.

The detected double influence of the manufacturing pores needs the quantitative estimation to define the actual role of them more precisely. Anyway, rather many ways of the influence on the porosity parameters exist within the powder technology of nowadays.

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8.8 The Effect of Helium, Radiation Damage and Irradiation Temperature on the Mechanical Properties of Beryllium

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In this work different RF beryllium grades were irradiated in the BOR-60 reactor to a dose of \~5-10 dpa at irradiation temperatures 350-420, 500, 800°C. Irradiation at temperatures of 350-400°C is shown to result in Be hardening due to the accumulation of radiation defect complexes. Hardening is accompanied with a sharp drop in plasticity at $T^\text{irrad} < 300°C$. A strong anisotropy in plasticity has been found at a mechanical testing temperature of 400°C and this parameter may be preferable when the samples cut are crosswise to the pressing direction.

High-temperature irradiation ($T^\text{irrad} = 780°C$) gives rise to large helium pores over the grain boundaries and smaller pores in the grain body. Fracture is brittle and intercrystallite at $T^\text{test} \geq 600°C$. Helium embrittlement is accompanied as well with a drop in the Be strength properties.

1. INTRODUCTION

Beryllium is presently considered as the most likely candidate material for the plasma facing components of the ITER first wall and divertor [1]. The main advantage of beryllium is its low $Z$ and a weak effect on plasma pollution. When substantiating the workability of beryllium for its use as a plasma facing heat flux material for the ITER divertor and first wall, a number of problems arises. The first is the lack of data on Be radiation resistance in the operational temperature range of 300-800°C. The matter is that Be was used previously in nuclear engineering solely at low temperatures (50-100°C), hence, the bulk of available data refers to these temperatures. But there is one more, no less important problem associated with applicability of available data obtained in fission reactors to substantiate the use of Be in a fusion reactor, i.e. fusion-fission correlation. This problem is particularly critical for Be, as its embrittlement and swelling depend heavily on helium accumulation [1,2]. At the same time, since helium in Be is accumulated due to the threshold reactions (\textbf{E}_{\text{threshold}} \sim 1.5 \text{ MeV}) , a considerable rise in the helium accumulation rate should be expected in Be under ITER irradiation, when a fraction of hard neutrons is considerably higher than in fission reactors.

The results of investigations into the effect of neutron irradiation and the related radiation damage and the effect of helium accumulation on the mechanical properties and structure of various RF Be grades are presented in this paper.

The experimental data on neutron irradiation effect on the Be properties in this temperature range is scanty. In the latest reviews [4, 5] some data can be found on Be radiation effects. But still the available data do not cover the entire dose-temperature range proposed for Be to be used in the ITER. It is worth noting that to date there is no data base sufficient to understand the main radiation effects limiting the Be life time as an ITER material. To answer these questions this experiment was set up in RF on irradiation of several commercial Be grades with the aim of providing the information on Be behavior throughout the temperature range corresponding to the Be operation temperatures in ITER. The DShG-200 and TShG-56 types of beryllium considered in RF as alternatives to S-65 B(C) were chosen as materials for investigation. In this paper we deliberately exclude the swelling problem, as the papers to be followed will be devoted to this subject.
2. EXPERIMENT

Various modifications of RF beryllium, i.e. DShG-56, DShG-200, TR-30, TShG-56, and TShG-200, were chosen for investigation. Blanks for samples were produced by the hot stamping method from distilled or technical powder, the maximum size of particles being less than 56 μm or 200 μm. Small tensile cylindrical samples, when being stamped, were cut transverse (T) and lengthwise (L) in the direction of blank deformation.

The samples were irradiated in the BOR-60 reactor in stainless ampoules filled with helium. One batch of Be samples was irradiated in the BOR-60 reactor to a dose of $0.7 \times 10^{22}$ n/cm$^2$ ($E > 0.1$ MeV) at $T_{irr} = 350^\circ C$. Three other batches of Be samples were irradiated in one capsule of BOR-60 to doses of 0.8 - 1.1 $\times 10^{22}$ n/cm$^2$ ($E > 0.1$ MeV) in three subcapsules at three temperatures - 400, 500, 780°C, respectively. The average damage dose in Be samples was ~5 dpa and the average helium concentration was calculated as ~500 appm.

After irradiation the samples were tested for tension in vacuum ($10^{-2}$ Pa) at $T_{test} \sim 20-800^\circ C$ at a deformation rate of $1.3 \times 10^{-3}$ s$^{-1}$. The irradiation and testing technique was described in detail in [6,7]. The TEM and SEM investigations were carried out on the JEOL 2000 FX microscope.

3. RESULTS OF EXPERIMENT

3.1. Mechanical properties

Figs.1 a), b) present typical dependencies of mechanical properties on $T_{test}$ for two Be grades DShG200 and TR-30. As follows from Fig. 1, Be DShG200, which is not so strong and more ductile in the initial state, demonstrates, when irradiated in the temperature range of 350-400°C, a satisfactory resistance to radiation embrittlement, when tested in the temperature range of 200-700°C. But, under irradiation at higher temperatures (550-800°C) high-temperature ductility of DShG200 at $T_{test} = 600-800^\circ C$ drops to zero.

Stronger Be modification, TR-30, has nearly twice as high level of strength properties as DShG200, but this alloy is susceptible to radiation embrittlement to a greater extent than DShG200. At $T_{test} \sim T_{irr} \sim 550-800^\circ C$ the TR-30 alloy has a low ductility, when irradiated (but still different from zero). Note, that the temperature dependencies of uniform elongation of irradiated Be samples have the distinct maximum $T=400^\circ C$. For DShG200 the level at $T=400^\circ C$ for all $T_{irr}(350-800^\circ C)$ is ~7.10%.

Effect of anysotropy

Figs. 2 a), b) presents the dependencies of ultimate strength and total elongation of Be DShG200, TShG200 and TShG56 on testing temperature in the initial and irradiated states. As follows from Fig.2, samples cut crosswise and lengthwise the blank compaction direction have, when unirradiated, a rather slight difference in ductility. In this case samples cut along the compaction direction have lower ductile properties. The strength properties of samples cut in different directions are practically the same.

After irradiation to 5 dpa at 350°C a difference is observed in Be resistance to radiation embrittlement depending on a direction of sample cutting. Irradiated samples cut in the transverse direction have a satisfactory level of total elongation at $T_{test} = 400-600^\circ C$ ($\delta_{tot} > 20\%$). At the same time, samples of the same materials cut in longitudinal direction have $\delta_{tot} = 0\%$ throughout the range of $T_{test}$.

Consequently, a relatively moderate anisotropy in properties of Hot pressed Be in the initial state is drastically increased after irradiation.

The effect of irradiation temperatures on the Be mechanical properties.

Low-temperature irradiation ($T_{irr} < 0.45T_{mp}$) -

Figs. 3 a), b) show the yield strength and uniform elongation of the Be alloys DShG-200T and TShG-56T as a function of testing temperature before and after irradiation to a dose of ~5 dpa at 350°C and 400°C. It is evident that a relatively low-temperature irradiation $T_{irr} = 350^\circ C$ results in Be hardening in the testing temperature range of 400 - 600°C. Note, that higher $T_{irr} = 400^\circ C$ results in a lesser level of hardening.

In line with the $\epsilon_t$ behavior the uniform elongation of samples irradiated at 400°C is somewhat higher at $T_{test} = 400 - 600^\circ C$ (i.e. in the hardening temperature range) than in samples irradiated at 350°C. At $T_{test} \geq 700^\circ C$ the yield strength of irradiated and unirradiated samples is equalized (Figs. 3 a, b). In
Fig. 1 The effect of neutron irradiation (5 dpa) on total elongation and ultimate strength of DShG200T and TR-30 Beryllium.
Fig. 2 The effect of relatively low temperature irradiation ($T_{ir}<0.45 T_{mol}$) on ultimate strength and total elongation of DShG200 and TShG200 Beryllium.
In this case the samples fracture at extremely low levels of plasticity-1%.

**High-temperature irradiation** \( (T_{irr} > 0.5T_m) \) – Irradiation at 800°C does not practically result in beryllium hardening. Moreover, as follows from Figs. 4 a), b) an increase in the irradiation temperature causes a drop in the strength characteristics to ~10 MPa \( (T_m \sim T_{test} \sim 800°C) \) (less than in reference samples) and a decrease in plasticity to the zero level at \( T_{test} \geq 700°C \). An increase in \( T_m \) intensifies helium embrittlement. Thus, at \( T_{test} = 600°C \) the samples irradiated at 800°C have both strength and plasticity lower than those irradiated at 520°C. Irradiation at 520°C hardens beryllium at \( T_{test} \sim 400-700°C \) and causes a decrease in the plasticity at all testing temperatures. At \( T_{test} \sim 800°C \) both strength and ductility characteristics of Be irradiated at 520°C are lower than those of unirradiated Be. A simultaneous drop of the strength and plastic characteristics in irradiated metals is typical of helium embrittlement [7–10]. The calculations performed demonstrate that irradiation in BOR-60 to a dose of \( 1 \times 10^{22} \text{ n/cm}^2 (E > 0.1 \text{ MeV}) \) should result in an accumulation of \( \sim 500 \text{ appm He} \) [9]. Needless to say, this amount of helium is more than adequate for helium embrittlement in Be at \( T_{test} > 0.5 \ T_m \).

### 3.2. Irradiation effect on the Be structure

**Low-temperature irradiation.** – The TEM investigations of the TShG-56T Be samples irradiated at 400°C to a dose of \( 1 \times 10^{22} \text{ n/cm}^2 (E > 0.1 \text{ MeV}) \) demonstrated that the TShG-56T structure contains abundant, relatively small precipitation of 100-150 nm in size (Fig. 5a). These precipitates are mainly plates, but still rather large global precipitation up to 1000 nm in size are encountered. The density of the latter is considerably lower. The comparison of the material structure and precipitation morphology in irradiated and unirradiated samples allow the conclusion to be made that irradiation at 400°C does not practically affect the type and density of the secondary phases in TShG-56T.

The TEM investigations did not detect the radiation defect complexes in the irradiated sample. We can to state only that these complexes are less than 10 nm at \( T_m = 400°C \).

The Be alloy DShG-200T had the similar structure after the low-temperature irradiation with the only difference that the density of the secondary precipitation was very smaller and the grain size was considerably larger, for this material, consistent with its manufacturing technology. DShG-200T is produced from a distilled beryllium powder with less content of impurities, which is why the density of the secondary phases therein is sure to be considerably lower. The larger grain size in DShG-200T is also attributed to the manufacturing technology, as DShG-200T is made from a powder larger in size than TShG-56T.

**High-temperature irradiation** – As follows from Figs. 5 b, c, d, where the TEM structure of a DShG-200 specimen irradiated to a dose of \( 1.15 \times 10^{22} \text{ n/cm}^2 \) at \( T_m = 790°C \) is presented, helium porosity develops in the Be structure. The density of pores over the boundaries is great. As seen from Fig. 5 b, the pores cover \( 1/2 - 1/3 \) of the grain boundary area. The average size of grain-boundary pores is \( <d> \approx 120 \text{ nm} \). In this case the average distance between pores \( <\lambda> \approx 200 \text{ nm} \), i.e. close to the pore diameter. The prevailing form of grain-boundary pores is an irregular octahedron.

In the matrix the form of pores is quite different. Observed mainly are parallelepipeds with the axis ratio \( a/b \sim 0.3 \) where \( a \sim 20 \text{ nm} \) and \( b \sim 60 \text{ nm} \). Large pores are closer to a cubic form, i.e. \( a/b \sim 0.8 \). Pores in the matrix are practically always observed on dislocations or subgrains boundaries (Figs. 5 b, c). The pore density is great and the distance between pores is commensurable to their sizes. As seen in Fig. 5c, the dislocations are pushed through helium pores. In doing so the pushing direction coincides with the large axis of helium pores.

The TEM investigations are underway. The final information on the type of phases in alloys and on the defect orientation will be presented in the later papers.

### 3.3. The character of material fracture.

**Low-temperature irradiation.** – The study of fracture surface of the TShG-56T sample irradiated at 400°C and tested at 200°C demonstrates (Fig. 6a) that the material is fractured in a brittle way by a combined mode, i.e. partially by a transgranular cleavage mode, partially over the intergranular facets. Dimple rupture typical of an unirradiated material is not seen. The brittle cleavage fracture and practically unaffected facets of grain are easily observable in Fig. 6a. The TShG-56T grain size is
Fig. 3 The effect of relatively low temperature irradiation ($T_{irr} < 0.45 T_{mel}$) on yield strength and uniform elongation of DShG200T and TShG56T Beryllium.
Fig. 4 The effect of relatively high temperature irradiation ($T_{irr} > 0.5 T_{mat}$) on yield strength and uniform elongation of DShG200T and TShG56T Beryllium.
Fig. 5  Microstructure of TShG56T Be alloy irradiated up to 5 dpa at 400°C a), and DShG200T Be alloy irradiated up to 5 dpa at \( T_r = 790°C \) b), c), d).
Fig 6 SEM fracture surface photographs after tensile tests for (a) TShG56T Be alloy irradiated up to 5 dpa, at 400°C ($T_{\text{test}}=200°C$) and (b, c, d) DShG200T Be alloy irradiated up to 5 dpa, at 790°C ($T_{\text{test}}=600°C$).
small, i.e. 10 μm. The density of precipitation on the grain boundary surface is rather high.

High-temperature irradiation. - After irradiation at 790°C and testing at 600°C the fracture surface of the DSgG-200T sample is a faceted intergranular fracture (Figs. 6 b, c, d). The density of grain-boundary precipitation in DSgG-200 is essentially lower than in TShG-56T, the grain size being much larger, i.e. ~60 μm. The study of intergranular facets surface with an ~20000-fold magnification reveals that all facets are covered with pores. The average size of a pore on the fracture surface is ~250 nm, i.e. it correlates well with the distance between pores observed in TEM (κ200 nm).

On the whole it can be noted that the results of TEM investigations of the irradiated sample structure and the SEM investigations of the irradiated sample fracture surface correlate well with each other.

3.4. Swelling

Density of technical beryllium TShG-56 and TShG-200 was increased slightly under irradiation, whereas the volume of distilled DSgG-56 and DSgG-200 was not changed. At an annealing temperature of 600°C all beryllium samples swelled, the maximum value, equal to 0.8%, being observed in beryllium DSgG-200. With the annealing temperature increased to 700°C, a slight increase in beryllium density is observed.

4. DISCUSSION

The results presented demonstrate that two major radiation effects control the Be life time. These are LTE in the temperature range T < 0.4T_m and helium embrittlement in the high temperature range (T > 0.5T_m).

The irradiation temperature effect on the basic properties of beryllium. - Of interest is to compare to what extent our results correlate with the data obtained in other experiments. When constructing the dependence of the yield strength and total elongation on the irradiation temperature (at T_{net}=T_{ir}) using the data of this work and those from [2,10], it can be seen that an appropriate qualitative correlation is observed in the behavior of different Be grades (Figs. 7 a,b). Beryllium produced by the hot pressing and HIP methods, when irradiated to doses of 0.5–5 dpa, demonstrate the similar dependence of the strength and plasticity properties on the irradiation temperature. In the low temperature range, LTE causes Be hardening and embrittlement. In the high temperature range the helium embrittlement is responsible for a decrease in both the strength and plasticity properties of beryllium.

It is immediately apparent that the irradiation temperature range, where irradiated beryllium has a satisfactory level of properties, is rather narrow: 300°C ≤ T ≤ 500°C. This is sure to pose certain problems for designers of the ITER divertor and first wall. Open remains the question as to the effect the helium concentration (irradiation dose) has on helium embrittlement. Typical of austenitic steels is the shift of the helium embrittlement to the range of lower testing temperature [11] with a growth in the helium concentration. The data presented in Fig. 7 are indirect evidence in favor of this assumption being also true for beryllium. Thus, the beryllium samples TIP-30 [10] after ~1 dpa (C_{He}~150 appm) have at T_{net}=T_{ir}=750°C the plasticity different from zero (1.7%), whereas the TShG-56T and DSgG-200T samples irradiated to 5 dpa (C_{He}~500 appm) have the zero plasticity at T_{test}=T_{ir}=700°C.

5. CONCLUSION

The investigations on the beryllium radiation resistance allow some conclusions to be made on the major factors controlling its workability as applied to the ITER operation conditions. Beryllium hardens and embritles in the range of relatively low temperatures (T<300°C). The degree of embrittlement depends on the direction of sample cutting. The investigations of beryllium performed after irradiation in the BOR-60 reactor have not revealed an appreciable advantage to any beryllium modification depending on the grain size and the technology of the initial powder production. A strong anisotropy in plasticity has been found at a mechanical testing temperature of 400°C and this parameter may be preferable when the samples cut are crosswise to the pressing direction. In this case the fracture character, the dependence of hardening and embrittlement on the irradiation temperature are typical of the LTE manifestation in metals with a HCP lattice.

In the high temperature range (T>500°C) the helium embrittlement is the major factor determining the level of plasticity, strength properties and the Be fracture character. Prior to irradiation the grain size and purity of different Be grades can profoundly
Fig. 7 The irradiation temperature effect on the yield strength and total elongation of different Beryllium grades: TShG56T, DShG200T [this work], Be HP [2], Be unknown grade [2], TIP and TIP30 [10]; open symbols - unirradiated, filled symbols - irradiated.
affect the plasticity of alloys. But after irradiation these distinctions are equalized. All Be grades have practically the zero plasticity at $T_{irr} - T^* < 300^\circ C$ and $T_{irr} - T^* > 600^\circ C$ after irradiation to 0.5..5 dpa.

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8.9 Behavior of Beryllium Pebbles under Irradiation

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Beryllium pebbles are being considered in fusion reactor blanket designs as neutron multiplier. An example is the European "Helium Cooled Pebble Bed Blanket." Several forms of beryllium pebbles are commercially available but little is known about these forms in response to fast neutron irradiation. Commercially available beryllium pebbles have been irradiated to approximately $1.3 \times 10^{22} \text{ n/cm}^2 (E > 1 \text{ MeV})$ at 390°C. Pebbles 1-mm in diameter manufactured by Brush Wellman, USA and by Nippon Gaishi Company, Japan, and 3-mm pebbles manufactured by Brush Wellman were included. All were irradiated in the below-core area of the Experimental Breeder Reactor-II in Idaho Falls, USA, in molybdenum alloy capsules containing helium. Post-irradiation results are presented on density change measurements, tritium release by assay, stepped-temperature anneal, and thermal ramp desorption tests, and helium release by assay and stepped-temperature anneal measurements, for Be pebbles from two manufacturing methods, and with two specimen diameters. The experimental results on density change and tritium and helium release are compared with the predictions of the code ANFIBE.

1. Introduction

A large number of beryllium specimens have been irradiated in US DOE Office of Fusion Energy Sciences experiments in the Experimental Breeder Reactor II (EBR-II) in Idaho Falls, ID and the Fast Flux Test Facility (FFTF) in Richland, WA. With the increased difficulty for performing irradiation experiments, due in part to the shut down of FFTF and EBR-II and other reactors worldwide, these specimens have become even more valuable. The European fusion materials development program centered at Forschungszentrum Karlsruhe has been concentrating efforts on design of a fusion blanket using beryllium pebbles as neutron multiplier [1]. The EBR-II test, COBRA-1A, included four beryllium pebble product forms, provided by JAERI, and thereby provides fast neutron irradiated materials for postirradiation testing. Therefore, the Forschungszentrum Karlsruhe has collaborated with the Pacific Northwest National Laboratory of Richland, Washington, USA, to conduct postirradiation testing of some of these specimens. Post-irradiation test results are presented on density change measurements, tritium release by assay, stepped-temperature anneal, and thermal ramp desorption tests, and helium release by assay and stepped-temperature anneal measurements, for Be pebbles from two manufacturing methods, and with two specimen diameters. The experimental results on density change and tritium and helium release are compared with the predictions of the code ANFIBE[2]. This paper provides selected results from this testing program [3].

2. Description of the Materials, Irradiation and Fluence

The COBRA-1A vehicle, irradiated in EBR-II starting on November 26, 1992, contained a number of beryllium specimens in canisters C03 and D03 located in below-core positions in capsules B-390 and B-391, respectively. Both capsules were of sweeper design, so that reactor coolant was in direct contact with each canister, and canisters were helium filled to provide a heat transfer medium. Irradiation temperatures were 388°C for the C03 and 395°C for the D03 capsule. Experimental details of the capsules were described previously [4]. The capsules were removed from the reactor upon removal of the COBRA-1A2 vehicle on September 26, 1994. At that time, EBR-II ceased to operate. Neutron dosimetry and damage calculations for these irradiations were described previously [5]. Canisters contained beryllium in several product forms including 1-mm pebbles in D03 and 3-mm pebbles in C03 from Brush Wellman, and 1-mm pebbles in C03 from Nippon Gaishi Co. (NGK). 5-mm pebbles from
Brush Wellman were also irradiated but will not be discussed here. Pebble details, as supplied by the manufacturer are provided in Table 1. The production of \(^4\)He and tritium has been calculated for beryllium irradiated in the COBRA-1A2 experiment in the Experimental Breeder Reactor II and details are summarized in Table 2. Reaction rates were based on adjusted neutron spectra determined from reactor dosimetry measurements at the specific elevations in the region of the beryllium capsules. The table corrects the tritium and \(^3\)He values for decay to May 1997.

3. Experimental Details

3.1 Optical Microscopy (OM) and Scanning Electron Microscopy (SEM)

To gain an understanding of pebble to pebble conformity and determine surface degradation due to irradiation, several specimens were selected for optical examination and one 3-mm pebble of each of the irradiated and unirradiated conditions was then examined by SEM on a JSM 840.

3.2 Swelling Measurements

Four irradiated pebbles from the C03 capsule were selected for density measurement and compared with four unirradiated specimens. Specimens were weighed in air and in water at least three times, until reasonable consistency was obtained because imperfect wetting and air bubble adherence were found to be problems.

3.3 Transmission Electron Microscopy (TEM)

Several 3-mm pebbles were ground to disks approximately 0.12-mm thick and thinned by electropolishing in a solution of 75 parts nitric acid, 15 parts sulfuric acid, 15 parts hydrochloric acid in 750 parts ethylene glycol at 9-13 V with the solution cooled to -20°C. Pitting presumed to be due to internal flaws and impurities was found to be a problem, but suitably thin areas were obtained in several specimens prepared for examination following brief repolishing of pitted specimens. Two unirradiated specimens and two irradiated specimens were thinned and polished providing one good irradiated specimen and one poor unirradiated specimen. Examinations were on a JEM 1200EX.

3.4 Tritium Assay and Residuals

The tritium assay and residuals results were determined by the method of high-temperature sweep gas extraction. The basic method has been described previously [6], but has been modified using sweep gas instead of vacuum [7]. The sweep gas method is used instead of the vacuum method due to 1000x lower method detection limits of the existing system. The method has an estimated precision and accuracy of ±5%. Briefly, the weighed sample is melted, at 1700°C, in a sweep gas of He+0.1% H\(_2\) at 100 SCCM, with the released hydrogen isotopes oxidized on copper oxide, collected in a pair of water bubblers, and measured by liquid scintillation counting, traceable to NIST.

3.5 Tritium Release by Stepped-Temperature Anneal

The stepped-temperature anneal tritium release method has been described previously [8,9]. The test temperature range is 350-850°C. Briefly, the sample is heated in sweep gas of He+0.1%H\(_2\) at 100 SCCM, the released tritium measured in real-time by ion chamber, oxidized on copper oxide and collected in a pair of water bubblers, and measured by liquid scintillation counting. Therefore two independent tritium measurements are obtained. The C03 1-mm pebble test used 5 pebbles with total sample weight of 0.0066g, while the D03 1-mm test used 10 pebbles with total sample weight of 0.0098g.

3.6 Tritium Release by Thermal Linear Ramp Desorption

The linear thermal ramp heating method and apparatus have been described previously [10]. This release kinetics test method was performed on C03 1-mm pebbles using 15 pebbles for each of three tests. The three tests were conducted over a decade range of ramp rates at 0.5, 2.0, and 5.0°C/min. The temperature range for all tests was 300-900°C. A ramp maximum temperature of 850°C was initially planned for the tests. However, a scoping test was performed, and the results indicated a \(T_{\text{max}}\) for this Be of >850°C, therefore, the ramp maximum was increased to 900°C. The sample was heated in a sweep gas of He+0.1%H\(_2\) at 100 SCCM, the released tritium measured in real-time by an ion chamber, then oxidized on copper oxide and collected in a pair of water bubblers, and finally measured by liquid
scintillation counting. The ramp rates were preselected as specified. After loading the weighed sample pebbles, the system was purged before heating was begun. After the temperature ramp was completed, the sample temperature was held at the maximum of 900°C for a period of time ranging from 4 to 18 hours, as seen in the plots.

3.7 Helium Assay and Stepped-Temperature Anneal Release Methods

Helium released from a single or several pebbles was determined by isotope-dilution gas mass spectrometry during stepped-temperature anneal heating or high-temperature vaporization in a resistance-heated graphite crucible in the mass spectrometer system's high-temperature vacuum furnaces [11]. Assay for total helium was measured on single pebbles, while stepped-temperature anneal tests for release rate measurements were conducted on batch sizes of 15 pebbles. The absolute amount of \(^4\)He released was measured relative to a known quantity of added \(^3\)He spike. The \(^3\)He spikes were obtained by expanding and partitioning a known quantity of gas through a succession of calibrated volumes [12]. The mass spectrometer was calibrated for mass sensitivity during each series of runs by analyzing known mixtures of \(^3\)He and \(^4\)He.

4. Experimental Results

4.1 OM and SEM

Figure 1 provides examples of 1 mm and 3 mm pebbles using optical microscopy (OM). It demonstrates that beryllium pebbles vary significantly in size and many contain surface irregularities, including depressions and seams. Also, stereo pairs were produced using scanning electron microscopy (SEM), and quantitative depth measurements can be obtained for surface features of interest. Examples at higher magnification are shown for both an unirradiated specimen and an irradiated specimen in Figure 2. It was found that both specimens contain two types of shallow surface depressions, on the order of 100 \(\mu\)m in diameter and on the order of 5 \(\mu\)m in diameter. Evidence for fine surface porosity due to irradiation was not found.

4.2 Swelling Measurements

Density measurements following irradiation (V) gave 1.795 ± 0.011 g/cm\(^3\) and prior to irradiation (V\(_o\)) 1.810 ± 0.033 g/cm\(^3\). The resultant swelling (V-V\(_o\))/V\(_o\) was 0.83%, smaller in magnitude than the sample-to-sample variability for the unirradiated density (1.86%) but similar to the variability in the irradiated samples (0.6%).

4.3 Tritium Assay

The measured tritium assay results are shown in Table 3, and provide interesting comparisons to produced or predicted values. These assay results, all measured in duplicate, are decay-corrected to May 1997. For each material type, the ratio of measured-to-produced tritium was calculated and is shown, with the produced value being as calculated by Greenwood. Note the 1-mm pebble measured/produced ratios, for both C03 and D03 materials, are low, at a consistent 0.71-0.73, with the 3-mm measured/produced ratio much higher at 0.93. The less-than-1 ratios indicate an in-situ diffusive loss of tritium during irradiation at temperature in reactor. A comparison of the ratios provides evidence that in-reactor loss of tritium is dependent upon pebble diameter size, with greater loss in the 1-mm pebbles than for the 3-mm pebbles.

4.4 Tritium Stepped-Temperature Anneal

The C03 stepped anneal tests results, using a sample size of 5 pebbles, are shown in Table 4. The
The total cumulative released at 350-850°C was 75% of the total inventory. The anneal times ranged from 6 hrs to 25 hrs at each temperature step. An additional anneal step was added at 850°C, in order to increase the cumulative fraction released at that temperature. The residual tritium remaining after all the anneal steps, measured by melting the sample at 1700°C, was about 25% of total inventory. Therefore, the cumulative plus residual total adds to 10.38 appm, slightly higher than the measured assay results of 9.03-9.96 appm.

For the D03 stepped anneal test, a larger sample of 10 pebbles, and longer-time anneal steps was used, with the final 850°C step held for 72 hours, as shown in Table 5. This D03 test resulted in a much higher cumulative total released than the C03 test. At 350-750°C, 20% was released, with 94% released by the end of the 850°C step. The residual tritium, at 6% of total inventory, brought the cumulative plus residual total of 9.38 appm into excellent agreement with the assay results of 9.05-9.29 appm.

Note for both stepped anneal tests, less than 2% of inventory was released at 350-550°C. The D03 material showed an order of magnitude still lower release rates at these lower temperatures, a fact which may be a great importance for in-reactor operation. A very consistent cumulative total of about 8.7% was released after 650°C for both materials. After 750°C, the cumulative released was 20-28% for both tests. The major difference between the two tests is seen at the 850°C step, though time duration at temperature is definitely a factor.

The plot of the stepped-anneal C03 test is shown in Figure 3. The IC cumulative release curve in MBq/g, calculated from the IC release rate curve in Bq/s, was normalized to the collection bubbler results, by adjusting the IC baseline and/or IC calibration factor, as has been done in previous work[8]. This method assumes the collection bubbler liquid scintillation results are always reliably accurate, therefore, the IC curve must be adjusted or normalized to obtain agreement.

The plot of the stepped anneal D03 test is shown in Figure 4. The total cumulative released tritium at 350-850°C was 1050 MBq/g or 94% of the total inventory of 1120 MBq/g.

4.5 Tritium Release by Thermal Ramp Desorption

Tritium release kinetics used the method of linear thermal ramp heating at three linear ramp rates. The plot of the thermal ramp test data for the C03 Be material is shown in Figure 5. The figure shows all three test curves of release rate versus time, showing release rate on linear axis, and with the corresponding temperature profiles, for comparison purposes. The difference in the release curve shapes and peak widths can be seen clearly in this plot.
Table 6 shows release kinetics parameters as determined from this data. The parameter \( P \), in Bq/g, is the "production" rate as measured by the assay test reported earlier, not as calculated from the fluence prediction; \( R \), in Bq/g, is the measured total release from the sample as determined from the bubbler liquid scintillation results; \( R/P \), in percent, is the fractional release at end of test; the temperature, \( T_{\text{max}} \), corresponds to the temperature of peak maximum release rate; and \( DF \) is the factor by which the release rate decreases, at 900°C in 3 hours. As seen in the table, the fractional release, \( R/P \), is high in these 1-mm pebbles, >92% in all cases. The \( T_{\text{max}} \) for the three cases shows a slight increase with increasing ramp rate, from 882°C to 900°C, as expected. The \( DF \) is consistently low for this material, ranging from 3.1 to 11.4, even at the high hold temperature of 900°C, indicating only gradual continuing release at the hold temperature. These \( DF \) numbers are low compared with other studied beryllium specimens [13].

### 4.6 Helium Release

Helium assay for total helium was performed on single pebbles each of both 1-mm C03 and D03 material. Stepped-temperature anneal tests were performed on 15-pebble batch sizes for both the C03 and D03 pebbles. For the stepped-temperature anneal tests, multiple sequential helium measurements were taken at each temperature, with increment steps in sample temperature of approximately 100°C, from either 500°C-1100°C for scoping tests, or about 800°C-1100°C, for the final tests. Each temperature was held until some leveling off of \(^4\text{He}\) was observed, at which point the temperature was raised to the next level. After the anneal tests, residual helium in the samples was determined by vaporization runs.

Helium release as a fraction of the total helium in the sample was markedly different for the C03 and D03 material, shown in Figures 6 and 7. The C03 sample released virtually all of its helium after approximately 30 minutes at 1000°C. Sample D03, on the other hand, released only 62% of its helium even after about 1 hour at 1100°C. Absolute helium release was correspondingly lower for D03 at each temperature level.

![Figure 5: Tritium Release by Thermal Ramp Desorption for C03](image)

![Figure 6: Helium Release by Stepped-Temperature Anneal for C03](image)

![Figure 7: Helium Release by Stepped-Temperature Anneal for D03](image)

Helium assay results are given in Table 7. In terms of helium concentration, the \(^4\text{He}\) totals for C03 are 2941 and 2724 appm. The corresponding totals for D03 are 2574 and 2459 appm. The averages agree with predicted concentrations within ±5%.
4.7 Microstructure

TEM examinations revealed several noteworthy features. In the irradiated specimens, grain size was found to be large, with a low dislocation density but containing many small features tentatively identified as bubbles lying as plate-like features on the basal plane. Several examples were also found of large regions as big as 5 μm in diameter with amorphous diffraction patterns. Such regions were found to be rich in Si, Al, Mg and Mn, indicating that impurities were non-uniformly distributed and these impure regions became amorphous due to irradiation. In comparison, the dislocation density was greater in the unirradiated condition, with evidence of subgrain development. Therefore, irradiation is found to reduce the dislocation density significantly, to form a fine distribution of unusually shaped bubbles, and to amorphize large regions of impurity.

Examples of the bubble structure found following irradiation are shown in Figure 8. The sequence uses void contrast showing the features edge-on in 8(a), tilted off 7° in 8(b), and tilted off 12° in 8(c). Figure 8(d) provides the unusual situation where the features are shown in strong contrast with the features tilted edge-on in a manner similar to that used to measure strain around precipitate particles and can be used to demonstrate that the features are probably internally stressed. It can therefore be concluded that the weaker features are probably unusually shaped helium bubbles.

The effects of irradiation on microstructure can be understood as follows. It was previously assumed that irradiation would cause microporosity, and c-type loop formation [14]. The added complexity of neutron damage near pebble surfaces was expected to enhance transmutation, possibly leading to enhanced surface microporosity. The present SEM examination appears to indicate that surface roughening due to irradiation is negligible to resolutions on the order of 100 nm. TEM did not provide microstructural information regarding cavitation at grain boundaries, but it now appears that swelling in the form of helium bubbles takes on an unusual highly non-equiaxed geometry of thin platelets on the basal plane. Careful review of earlier micrographs [14] shows examples of contrast similar to that found in the present experiment. The differences in interpretation may be compatible if it is possible that c-type loops accumulate gases such as helium and tritium, eventually becoming highly anisotropic bubbles.

In the current work, the bubbles were measured to provide diameter, number density and an estimate of swelling. It was found that the bubbles ranged in diameter from 5 to 25 nm, with a mean diameter of 11.9 nm at a number density of 2.0x10^16 cm^-3. If one were to assume spherical voids with these diameters, this corresponded to 2.5% swelling; but as the bubble shape was flattened, with an aspect ratio for thickness to diameter of about 4/25 or 0.16, the actual swelling can be estimated at about 0.4 of the spherical value or about 1%. Therefore, the measured density change of 0.83% appears to be due to the development of internal bubbles estimated at 1% swelling.

Swelling may also originate from the transformation of impurity regions from a crystalline to an amorphous phase. It is difficult to estimate the volume fraction of impurity regions or to estimate the resultant swelling due to phase transformation, but based on the agreement between measured density change and swelling demonstrated above, the magnitude of this swelling change is probably small.

If much of the helium and tritium is bound in bubbles, then release of these gases from heated specimens can be expected to occur only after bubble coarsening, and therefore will be observed mainly at
higher temperatures. Examination of heated specimens may allow verification of such prediction.

5. Comparison with ANFIBE

In order to better understand the physical mechanisms governing swelling and tritium and helium release behavior in beryllium, as well as to generate confidence in the results provided by the code ANFIBE [2] and to assess its prediction capabilities, experimental results on density change and tritium and helium release obtained in the present work have been compared with the code predictions.

Swelling calculations were performed for both 1-mm and 5-mm C03 and D03 material as well as for 3-mm C03 material. As shown in Table 8, the calculated volumetric swelling for C03 material ranges from 0.61% to 0.91% depending on the respective helium content. On the other hand, the resultant calculated swelling in case of D03 material is up to about 25% higher than in case of C03 material and ranges from 0.83% to 0.98%. The agreement between the calculated swelling data and the experimental estimated value of 0.83% is satisfactory. The calculated diameter of the homogeneously distributed intragranular helium bubbles is 16.8 nm and the corresponding bubble density is $2.4 \times 10^{15}$ cm$^{-3}$. These values are in reasonable agreement with those estimated by means of TEM examinations (mean bubble diameter 11.9 nm, mean bubble density $2.0 \times 10^{16}$ cm$^{-3}$). This fact confirms the assumption that beryllium swelling under neutron irradiation is predominantly due to the development of internal helium bubbles, so that void formation from radiation damage can be neglected.

Helium release calculations were also performed on pebbles of both 1-mm C03 and D03 material in the same way as described in the Sect. 4.6. The calculated helium inventory after annealing as a percentage of total helium in the sample was 7% and 29% for C03 and D03 material respectively. The compares with the measured values of 2% and 38%, respectively, for C03 and D03 material.

A comparison in terms of cumulative tritium release as a function of the annealing time between experimental data and predictions of ANFIBE for the C03 stepped-annal test described in Sect. 4.4 is shown in Fig. 9. In agreement with previous studies [15,16] it has been found that the release starts at about 500-550 °C and becomes significant for temperatures higher than 700-750 °C. The total cumulative released tritium at 350-850 °C has been calculated as 883 MBq/g, which represents 71% of the 1240 MBq/g total inventory. This compares with the measured cumulative released tritium, shown in Table 4, of 931 MBq/g, or 75% of the total inventory. The ANFIBE predicted value, therefore, was 95% of the measured result.
6. Conclusions

Post irradiation examinations of COBRA-I A beryllium pebbles irradiated in the EBR II fast reactor at neutron fluences which generated 2700-3700 appm helium at 390°C, have been performed. PIE results have been presented on tritium and helium assay and release measurements, and on density change for beryllium pebbles from two manufacturing methods and with two specimen diameters. These results are all in reasonable agreement with the predictions of the code ANFIBE.

References

### Table 1
#### Pebble details

<table>
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<th>Type</th>
<th>Supplier &amp; Batch #</th>
<th>Size</th>
<th>Major Impurities</th>
<th>Minor impurities (in ppm)</th>
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<td>Brush Wellman 826-64-5</td>
<td>1 ± 0.2 mm -16+20 mesh</td>
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<td>750 Mg, 565 Fe, 380 Si, 365 Al, 110 Cr, 105 Zr, 95 Mn, 90 Ti, 80 Ni, 70 Zn, etc.</td>
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<tr>
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<td>1.51 wt% BeO</td>
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<td>0.023 % C</td>
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### Table 2
#### Fluence Calculations for Be Pebbles in COBRA-1A2 at 1 May 1997

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<tr>
<th>Be Pebbles</th>
<th>Ht,in.</th>
<th>²⁴He, appm</th>
<th>³⁵H, appm</th>
<th>Fast Fluence &gt; 1 MeV E+22 n/cm²</th>
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### Table 3
#### Tritium Assay Results

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<th>sample</th>
<th># of Pebbles</th>
<th>sample weight (g)</th>
<th>Measured (Bq/g)</th>
<th>Measured (appm)</th>
<th>Produced (appm)</th>
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Table 4
1-mm C03 Stepped-Anneal Tritium Release Results

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<th>Sample</th>
<th>Temp, C</th>
<th>Time (hr)</th>
<th>Bubbler Result (Bq/g)</th>
<th>Cumulative (Bq/g)</th>
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Table 5
1-mm D03 Stepped-Anneal Tritium Release Results

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<tr>
<th>Sample</th>
<th>Temp, C</th>
<th>Time (hr)</th>
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### Table 6
Trinitium Ramp Test Release Parameters

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<tr>
<th>Material</th>
<th>Ramp rate</th>
<th>Sample wt [g]</th>
<th># of Pebbles</th>
<th>P [Bq/g]</th>
<th>R [Bq/g]</th>
<th>R/P [%]</th>
<th>T_{max} [°C]</th>
<th>DF</th>
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<tr>
<td>1-mm C03</td>
<td>0.5°C/min</td>
<td>0.0188</td>
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<td></td>
<td>2.0°C/min</td>
<td>0.0178</td>
<td>15</td>
<td>1.13 E+09</td>
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<td>887</td>
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<td></td>
<td>5.0°C/min</td>
<td>0.0179</td>
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<td>1.07 E+09</td>
<td>95%</td>
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### Table 7
Helium Assay Results

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<th>Sample</th>
<th>Sample wt (mg)</th>
<th>^3He (appm)</th>
<th>^4He (atoms)</th>
<th>Measured ^4He (appm)</th>
<th>Predicted ^4He (appm)</th>
<th>Avg ^4He Meas/Prod</th>
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<tr>
<td>1-mm C03</td>
<td>1.633</td>
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<td>2.59E17</td>
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<td>1-mm D03</td>
<td>1.206</td>
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<td>1.472</td>
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### Table 8
Calculated Volumetric Swelling for Be Balls in Position C03 and D03 of COBRA-1A2

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<tr>
<th>Position</th>
<th>Be Ball Diameter</th>
<th>^4He,appm</th>
<th>^3H,appm</th>
<th>Fast Fluence &gt; 1 MeV xE+22 cm^2</th>
<th>Fast Fluence &gt;0.1 MeV xE+22 cm^2</th>
<th>Be dpa</th>
<th>Calculated Volumetric Swelling %</th>
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<td>C03</td>
<td>1 mm</td>
<td>2723</td>
<td>13.1</td>
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<td>3595</td>
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<td>5 mm</td>
<td>3595</td>
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<td>1.60</td>
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<tr>
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<td>1 mm</td>
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ABSTRACT

This paper reports the results of an experimental effort to anneal irradiated beryllium specimens and characterize them for steam-chemical reactivity experiments. Fully-dense, consolidated powder metallurgy Be cylinders, irradiated in the EBR-II to a fast neutron (>0.1MeV) fluence of ~6x10^{22} n/cm^2, were annealed at temperatures from 450°C to 1200°C. The releases of tritium and helium were measured during the heat-up phase and during the high-temperature anneals. These experiments revealed that, at 600°C and below, there was insignificant gas release. Tritium release at 700°C exhibited a delayed increase in the release rate, while the specimen was at 700°C. For anneal temperatures of 800°C and higher, tritium and helium release was concurrent and the release behavior was characterized by gas-burst peaks. Essentially all of the tritium and helium was released at temperatures of 1000°C and higher, whereas about 1/10 of the tritium was released during the anneals at 700°C and 800°C. Measurements were made to determine the bulk density, porosity and specific surface area for each specimen before and after annealing. These measurements indicated that annealing caused the irradiated Be to swell, by as much as 14% at 700°C and 56% at 1200°C. Kr gas adsorption measurements for samples annealed at 1000°C and 1200°C determined specific surface areas between 0.04 m^2/g and 0.1 m^2/g for these annealed specimens. The tritium and helium gas release measurements and the specific surface area measurements indicated that annealing of irradiated Be caused a porosity network to evolve and become surface-connected to relieve internal gas pressure.

1.0 INTRODUCTION

Evaluation of the safety hazards for the International Thermonuclear Experimental Reactor (ITER) includes an assessment of the failures of plasma-facing-component (PFC) materials due to various accident scenarios. One scenario is a loss-of-coolant accident (LOCA) in which a waterline break injects steam into the torus vacuum vessel. This type of accident is of particular concern for the current ITER design because steam interactions with hot beryllium, the proposed first-wall PFC material, can produce hydrogen via the following reaction.

\[ \text{Be} + \text{H}_2\text{O} \rightarrow \text{BeO} + \text{H}_2 \]  

Assessment of the consequences of such LOCAs is typically done by means of accident model-simulations that use experimentally-derived, chemical-reactivity data for the material. Such data are sparse for irradiated Be.

This paper is part of an effort to provide steam-chemical reactivity data for irradiated Be. Its primary purpose is to present the results of annealing experiments and material characterization measurements for irradiated Be specimens. A companion paper at this workshop presents the results of steam-chemical reactivity experiments for these specimens. Both papers relate to an effort to evaluate the influence of neutron irradiation effects and swelling on steam-chemical reactivity behavior for Be. The results presented in this paper include measurements of tritium and helium release...
behavior during annealing of irradiated specimens and measurements of the bulk densities, swelling, porosity, and specific surface areas for the annealed material. Although many studies have investigated swelling and mechanical properties (5-6), swelling evolution (7-10) and gas-release behavior (11-14), all phenomena that are related to the presence of entrapped tritium and $^4$He produced in neutron-irradiated Be via nuclear transmutation reactions, few have studied these phenomena with respect to steam-chemical reactivity behavior.

2.0 EXPERIMENTAL DETAILS

2.1 Specimen Characterization
All of the Be specimens used in this work were fabricated using consolidated powder-metallurgy (CPM) techniques and Brush-Wellman SP-200-F powder [ < 44 micron size particles with the following chemical composition in weight percent: Be(99.1%), BeO(0.9%), Fe(0.09%), C(0.08%), Al(0.04%), Si(0.03%), Mg(0.02%), and other metals(<0.04%) ]. To achieve fully-dense material, the powder was cold isostatic pressed, sintered in a vacuum furnace at 1250°C for 4 hours and hot isostatic pressed at 1000°C for 3 hours with a pressure of 103 MPa. Cylindrical test specimens were made by dry-machining, yielding specimens with a 0.76-cm diameter and three different lengths, 0.635 cm, 2.032 cm, and 3.051 cm. No chemical etching was used to remove surface deformation layers from machining.

Irradiation of 0.635-cm and 2.032-cm long Be specimens took place in position 2B1 (near the core center) of the Experimental Breeder Reactor, EBR-II, a fission reactor with a fast neutron environment.[15] Nominal fast neutron fluences (>0.11 MeV neutrons) were estimated to range from 5.2x10$^{22}$ n/cm$^2$ to 6.7x10$^{22}$ n/cm$^2$, values that are approximately a factor of ten higher than those expected for Be components in ITER. The nominal irradiation temperature was 400°C.

2.2 System for Annealing Experiments
Annealing and gas-release measurements for the irradiated Be specimens were conducted with a flow-through, integrated assembly set up in a laboratory hood. This system, as illustrated in Figure 1, consisted of an inlet gas manifold, a molybdenum-lined tube furnace, in-line instruments downstream from the furnace for analysis of the gas-stream composition and an in-line oxidizer and ethylene-glycol traps at the end of the process line. An Ar carrier gas with approximately 1% H$_2$ was used in the annealing tests. Measurements of the gas composition in the process line were made as a function of time with an on-line mass spectrometer. Tritium measurements were made with an in-line ion chamber and by post-test analyses (liquid scintillation counting) of the tritium content in ethylene glycol traps at the end of the process line.

2.3 Material Property Characterization
Measurements were made to characterize the density, porosity and specific surface area for the test specimens, both before and after annealing tests. Densities were obtained by two techniques; (1) measurements of the masses and of the diameters and lengths of the specimens and (2) water immersion density measurements. Porosity was determined from the water immersion measurements. Specific surface areas for the test specimens were measured using a gas adsorption technique, with Kr as the adsorptive gas. The gas adsorption measurements were made with an automated Micromeritics ASAP 2010 system and the data were analyzed by the method of Brunauer, Emmett and Teller [16], commonly referred to as the BET method.

3.0 MEASUREMENTS AND RESULTS

3.1 Gas-Release Measurements
Two sets of annealing experiments were done for the irradiated Be, one for the 0.635-long specimens and another for the 2.032-cm long specimens. The small cylinders were annealed at temperatures of 450°C, 600°C, 700°C, 800°C, 1000°C and 1200°C, with the thermal history patterned after that used in chemical reactivity tests for the irradiated Be [4, 17]. For these annealing experiments, the thermal ramp from ambient temperatures to the anneal temperature consisted of up to seven ramp segments, with the ramp rates for each segment selected to simulate the temperature rise in the chemical-reactivity system. Typical temperature ramp rates ranged from 33°C/min at the start to 2°C/min in the final segment. Approximate annealing times were
as follows: 450°C (300 minutes), 600°C (1440 minutes), 700°C (260 minutes), 800°C (185 minutes), 1000°C (100 minutes), and 1200°C (80 minutes). These times include ramp-up times of ~60 minutes for the two high temperature anneals and ~30 to 40 minutes for the other anneals. In addition, both long and short cylinders were annealed at a temperature of 1100°C for about 35 minutes, following a linear thermal ramp of 20°C/minute. In all cases presented here, the temperatures given correspond to the control temperatures of the annealing furnace. A 100 cm³/min argon carrier gas with approximately 1% H₂ was used for the anneal experiments to facilitate tritium transport.

Tritium release rates were measured with an in-line ion chamber and ⁴He release rates were measured with a quadrupole mass spectrometer (QMS). QMS signatures were mass-2 for H₂, mass-4 for ⁴He and mass-40 for ⁴₀Ar. Based on earlier studies of gas release from irradiated Be [14], the contribution of HT to the QMS mass-4 peak was negligible. The QMS was calibrated with standard gas mixtures flowing in the process line, and the in-line ion chamber was calibrated with a tritiated gas mixture that was standardized using ethylene glycol traps and liquid scintillation counting.

Our gas analysis measurements indicated that tritium and helium gas-release behavior was a complex function of both temperature and time at temperature. Some results of these measurements
were reported earlier for the small Be specimens [4] and we include them here for completeness.

We first focus on the gas release behavior for low temperature anneal experiments. No measurable tritium and helium were observed from the specimens annealed to temperatures of 600°C and below. Although tritium was released from the specimen at 700°C, no helium release was observed, probably because of insufficient detection sensitivity in the QMS analysis setup. As shown in Figure 2, tritium release from the small specimen annealed at 700°C exhibited a small steady release from the specimen, once it reached 700°C, followed by a rise to a substantially higher level, after an incubation time of about 60 minutes at 700°C.

In contrast, the tritium release behavior for specimens annealed at temperatures of 800°C and above was characterized by relatively sharp release peaks (gas burst release). Measurements of the tritium and helium released from a small irradiated specimen annealed at 800°C are shown in Figure 3. The largest peak corresponded to the time that the specimen first reached 800°C, and smaller release peaks occurred about 20 minutes and 40 minutes after the initial release transient, again indicating incubation times associated with the gas-burst release behavior. Although the observation of helium is weak, Figure 3 clearly shows that the release of tritium and helium is concurrent in the gas-burst peaks.

Figure 2. Tritium release from specimen D03T4 annealed at 700°C.

For the specimen annealed at 1000°C, a prominent gas-burst release peak was observed. Figure 4 shows the tritium and 4He gas release data that were measured for D03T6, a 0.64-cm long specimen, heated on a segmented ramp and annealed at 1000°C. The gas release behavior is characterized by small gas-burst release peaks on the heat-up and by a large gas-burst release peak after reaching 1000°C. Concurrent release of tritium and helium is shown by the measurements in Figure 4.

Figure 3. Tritium and helium release from specimen D03T6 annealed at 800°C.

Figure 4. Tritium and helium release from specimen D03T6 heated on a segmented ramp to an anneal temperature of 1000°C.
Similarly, sharp gas-burst release peaks were observed for D03M4, a 0.64-cm long sample, heated on a segmented ramp to an anneal temperature of 1200°C, as shown in Figure 5. Tritium and helium were observed to be released concurrently, primarily in a sharp release peak at about 1000°C during the heat-up phase.

![Figure 5. Tritium and helium release from specimen D03M4 heated on a segmented ramp to an anneal temperature of 1200°C.](image)

In Figure 6, we show the tritium and helium release data for specimen D03B3, a 2.03-cm long cylinder, that was heated on a linear ramp of 20°C/min to an anneal temperature of 1100°C. Similar to the data for the 1200°C anneal shown in Figure 5, tritium and helium were released concurrently in a peak at about 1000°C on the heat-up phase. In spite of the differences in specimen size and in the heat-up rates, gas-release behavior was very similar for the 1100°C and 1200°C tests.

Comparable quantities of tritium were released from specimens annealed at 700°C and 800°C, and these quantities were about a factor of ten below that released from the specimens annealed at 1000°C and 1200°C. The high temperature anneals released the entire inventory of tritium and helium.

We have derived total quantities of tritium and helium for the small samples annealed at 1000°C and 1200°C. The results are shown in Figure 7, in which they are compared to predictions of the gas production rates that are referenced to neutron dosimetry for the COBRA-1A2 irradiation experiment in EBR-II [18]. The helium results from the present work are in reasonable agreement with the predicted values but the derived tritium values are somewhat higher than the predicted ones.

![Figure 6. Tritium and helium release from specimen D03B3 heated on a linear ramp to an anneal temperature of 1100°C.](image)

![Figure 7. Comparison of tritium and helium quantities derived from our experiments with values predicted based on neutron dosimetry.](image)

### 3.2 Material Characterization Measurements

Measurements of the mass, density, and physical dimensions (length and diameter) were made both before and after the anneal tests. Based on these measurements, the density of the unirradiated Be was 1.854 g/cm³ and the density of the irradiated Be was 1.825 g/cm³, prior to anneal tests, indicating a density loss of 1.6% during the EBR-II irradiation. For the irradiated specimens that were annealed,
densities were derived both from immersion-density measurements and from dimensional measurements.

The results of density measurements, presented in Figure 8, show that the irradiated Be experienced a significant density loss with annealing because of swelling. As shown in the figure, both immersion-density and dimensional measurements were made for the short (-IS) specimens annealed on segmented thermal ramps and only dimensional measurements were made for the short (-2S) and long (-2L) specimens annealed using a linear thermal ramp to achieve the anneal temperature. The results obtained from the two anneal campaigns are in reasonable agreement. Differences in the values from the two measurement approaches are most likely due to inaccuracies in the dimensional measurements for the highly-swelled specimens with non-uniform dimensional changes.

![Figure 8. Bulk densities for irradiated Be specimens that have been annealed at temperatures from 450°C to 1200°C.](image)

Volumetric swelling for the irradiated-annealed specimens is presented as a function of anneal temperature in Figure 9. With $V_o$ representing the volume of unirradiated material and $V$ representing the volume of irradiated-annealed material, volumetric swelling is defined here as $(V-V_o)/V_o$. It was computed as $[(\rho_0-\rho)/\rho] \times 100\%$, where $\rho_0$ is the density of unirradiated Be and $\rho$ is the density of Be after irradiation and heat treatment. Annealing at temperatures from 450°C to 600°C resulted in little change in the swelling, relative to that observed for the as-received irradiated material. However, based on the swelling results derived from immersion-density measurements, annealing to temperatures above 600°C resulted in swelling, relative to unirradiated material, that ranged from 14% at 700°C to 56% at 1200°C.

![Figure 9. Volumetric swelling of irradiated Be specimens as a function of anneal temperature.](image)

Measurements of surface-connected (open) porosity were attempted for the annealed-irradiated Be by the liquid immersion approach. However the results indicated no appreciable open porosity, even though the density changes were large.

Specific surface area measurements were made for the annealed-irradiated specimens and for unirradiated CPM-Be specimens of varying density. The results are presented in Figures 10 and 11. Figure 10 presents both bulk density and specific surface area as a function of open porosity for unirradiated CPM-Be. In this case immersion density measurements were successfully used to characterize the porosity of the unirradiated Be. The figure shows that for bulk densities of about 92% and higher, the porosity is comprised of both open and closed fractions. Furthermore, for bulk densities less than 92%, the porosity is open, with a very small if any closed component. The figure also shows that, for the unirradiated CPM-Be, the specific surface area is a linear function of the open porosity, and it reaches values that are comparable to those for the powders that comprise the consolidated product.
Measurements for long and short specimens annealed at 1100°C gave a specific surface area of about 0.005 m²/g for this material. This is a very surprising result, considering that the swelling is comparable to that of the specimens annealed at 1000°C to 1200°C. This difference suggests that the difference in the thermal ramp rates for the two anneal types, segmented versus linear, could have influenced the character of the surface-connected porosity network that evolved during the heat treatment. Although both thermal ramps reached the target anneal temperatures in about the same time, the segmented ramp had a faster ramp rate initially and a much slower one toward the end of the ramp.

4.0 DISCUSSION AND CONCLUSIONS

In this paper we have presented the results of annealing experiments for CPM-Be specimens that were irradiated to a high fluence in the EBR-II. The results include both gas-release measurements during annealing and material characterization measurements for irradiated specimens that have been annealed, as compared to unannealed material.

We found that tritium and helium release from irradiated Be during annealing was a complex function of the temperature ramp rate, the anneal temperature and the time at the anneal temperature. No significant tritium release was observed for material annealed at 600°C and below. For irradiated Be annealed at 700°C, we observed a delayed tritium release after an incubation time of about 60 minutes. For anneal temperatures of 800°C and higher, tritium and helium release was concurrent and the release behavior was characterized by gas-burst peaks. Essentially all of the tritium and helium were released in prominent gas-burst peaks at about 1000°C for irradiated samples annealed at 1000°C and higher. In contrast, the quantities of tritium released during the anneals to 700°C and 800°C were about 1/10 of the expected inventory in the material.

Material characterization measurements for irradiated-annealed Be demonstrated that annealing caused the material to swell and develop a porosity network that became surface-connected to relieve the internal gas pressure. In effect, the gas-burst release of tritium and helium are a signature for this process. Swelling, relative to unirradiated Be,
increased by 14% at 700°C and by 56% at 1200°C for the irradiated test specimens.

Measurements of the specific surface areas for the irradiated-annealed material provided some important information concerning the porosity network that evolves during annealing. For 0.64-cm long irradiated specimens that were annealed at 1000°C and 1200°C, the measured specific surface areas were orders of magnitude greater than the geometric surface areas for irradiated material that was not annealed. However, the specific surface areas were much less than those measured for CPM-Be with a bulk density of 88% theoretical. The specific surface area for the low density CPM-Be product approaches that of the unconsolidated powder equivalent in which the surfaces of each powder particle are exposed to the adsorption gas. We infer from this that the porosity network that evolves during annealing of irradiated Be does not expose internal surfaces on a scale commensurate with individual powder particles.

Kr gas-adsorption measurements of specific surface areas for irradiated Be subjected to different thermal ramp rates revealed large differences in the specific surface areas for the treated specimens. The results suggest that irradiated Be that undergoes a fast thermal ramp could develop a surface-connected porosity network with a higher specific surface area than that for irradiated Be heated on a slower ramp.

As discussed in the companion paper to this workshop, the results of this work have provided information important to understanding the steam-chemical reactivity for irradiated Be that has been annealed [3]. We have shown that the surface area associated with the open porosity network that evolves during annealing of irradiated Be can account for increased reactivity of the irradiated-annealed material. However, we have discovered that specific surface area characteristics of the evolving network may be significantly influenced by the thermal ramp rate that the irradiated material is subjected to. This issue warrants additional research.

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REFERENCES


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9. Health and Safety Issues
9.1 Protection of Air in Premises and Environment Against Beryllium Aerosols

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First and foremost, the danger of beryllium aerosols concerns a possibility of their inhalation. The situation is aggravated with high biological activity of the beryllium in a human lung. The small allowable beryllium aerosols' concentration in air poses a rather complex and expensive problem of the pollution prevention and clearing up of air. The delivery and transportation of beryllium aerosols from sites of their formation are defined by the circuit of ventilation, that forms aerodynamics of air flows in premises, and aerodynamic links between premises. The causes of aerosols release in air of premises from hoods, isolated and hermetically sealed vessels can be vibrations, as well as pulses of temperature and pressure. Furthermore, it is possible the redispersion of aerosols from dirty surfaces. The effective protection of air against beryllium aerosols at industrial plants is provided by a complex of hygienic measures: from individual means of breath protection up to collective means of the prevention of air pollution. As a whole, for good safety of the person in air environment, and inside premises, depending on function of latter, it is expedient the following:

- Applying of individual means of breath protection (expendable or reusable with forced delivery of air or without it, filtering or isolating, hose arranged or self contained).
- Sheltering, isolation or hermetic sealing of sources of probable allocation of impurity, use of stationary and independent means of local ventilation, ensuring effective air extraction and filtering of released beryllium aerosols.
- Zoning of premises, application of the effective circuits of general ventilation (lamination and generation of directed air flow), repeated use of air.

The prevention of air environment pollution provides the following:

- Avoidance of an unorganized exhaust of air from considerably polluted premises.
- The greatest possible reduction of volume of air, exhausted in an environment, due to repeated use of air and creation of closed systems of ventilation in premises of a certain type.
- Clearing up of industrial and ventilation discharges.
- Creation of functionally various independent circuits of general and local ventilation.

1. INTRODUCTION

In ITER to protect the air environment and the site personnel we should take into account beryllium (Be) and tritium (T) release potential. Measures to avoid air contamination depend on Be aerosols production and spread mechanisms and may include:
- sheltering, isolation or hermetic sealing of Be sources; compact location and isolatoin of working areas with harmful release risks;
- local ventilation;
- zoning of working premises and general ventilation;
- special zones tailored for repair operations, waste handling, etc.

2. BERYLLIUM TOXICITY AND MAXIMUM PERMISSIBLE CONCENTRATION

The toxic effect of Be aerosols on humans may be general and local. The general one can lead to allergic reactions and accompanying changes in parenchymatous organs. The local one manifests itself in respiratory system affection. The response, general or local, depends on the form of a Be compound and dispersion of its aerosols. The form of a Be compound governs its chemical properties. Its dispersion governs the dimensions of aerosols particles. In ITER only insoluble Be compounds are expected. They pose hazards first of all due to their dispersion parameters.

The general toxic effect occurs due to aerosols accumulated in the alveoli, with basically submicron-sized particles. Particles greater than 1 micrometer in
size do not reach the alveoli and deposit in convolutions of the airway. It is no mere chance that at melting and welding operations causing aerosols generation with mainly submicron-sized particles there was a time span of less than 3 years between the exposure and allergic reactions. At mechanical treatment of Be when disintegration-produced aerosols with larger particles are prevailing, allergic reactions were not observed in 5 years.

Available information on aerosols dispersion at conditions which can be extrapolated to their production processes in ITER shows strong evidence for the possibility of generation of considerable amount of submicron fractions. It would be reasonable to solve the problem of Be aerosol submicron particles toxicity experimentally, wouldn't it?

In ITER one expects to see tritiated Be. Health consequences of its toxic effects are unknown. There may be one of three types of effect: independent, magnification or weakening of the simultaneous impact of Be and T.

Tritium is an inert gas for respiratory tract. Thus the independent effect might be expected. At the same time it is reasonable to evaluate the health risks of exposure to Be-T combination in terms of its toxicity. Taking into account of Be and T hazards air protection should be provided first of all against Be aerosols. At the present stage the maximum permissible Be concentration in the air of working premises is established to be 1 mkg/m³.

3. BERYLLIUM AEROSOLS PRODUCTION AND TRANSPORT

Air protection in working premises against Be aerosols depends on mechanisms of their generation and transport by air flows. Be aerosols are generated first of all during technological process in the vacuum vessel. They might arise during repair work and other similar operations. Aerosols generated in the reactor may penetrate to adjacent premises during opening the reactor after filling it by inert gas or air. Fine-dispersed aerosols enter the air from blanket and diverter components surfaces as a result of turbulent diffusion and mechanical effects (inert gas or air flows, local air flows provoked by different components replacement and vibrations).

Equipment vibration may cause aerosols in the air of premises and their releases out of different sealed spaces even at rarefaction conditions. The release of a medium out of sealed spaces within equipment may take place only if there is excess pressure in it. A sharp increase in pressure and releases out of equipment working at vibration have been observed. Probably this is the result of resonance occurred in sealed spaces or equipment.

3.1 Model, conditions and research technique

As a model there has been used a thin-walled cylinder made of organic glass with the following dimensions: 224 mm - inner diameter, 466 mm - length, 8 mm - wall thickness. The cylinder has been fastened by one end to a chuck plate of a vibrating table. VED-200 vibrating electrodynamical stand has been used together with its standard control systems (see Fig. 1).
handling zones can also contribute to the production of Be aerosols and their spread in the air of working premises.

Fig. 2 Effect of the vibration frequency on the static pressure and its pulsation in confined space at \( P_n = 0 \).

Fig. 3 The same mentioned Fig. 2 for velocities: \( U_1' \)- longitudinal, \( U_2' \)- transverse.

**4. AIR PROTECTION AGAINST CONTAMINATION**

To prevent air contamination in premises adjacent to the reactor vessel after its filling with air or inert gas it would be reasonable:

1. to fill the vacuum vessel with air or inert gas with its simultaneous partial pumping into purification system followed by return of cleaned gas into the vacuum vessel, or venting. This will permit to reduce the total volume of the air used, releases into the air environment and contamination of the air in the vacuum vessel.

Fig. 4 Effect of the vibration frequency on the static pressure and its pulsation with the opened hole in the end-face wall.

Fig. 5 The same mentioned Fig. 4 for velocities: \( U_1' \)- across hole; \( U_2' \)- inside volume behind.

(2) filling the vessel with inert gas or air and opening is followed by air removal from the vacuum vessel into the atmosphere. This makes it possible to reduce aerosols releases from the vessel to adjacent
But some leakage of aerosols due to diffusion in the boundary layer and contaminated transportation facilities and components removed from the vacuum vessel for repair and replacement work is inevitable.

(3) to limit contaminated air transport and spread throughout the adjacent premises it is necessary to provide for an additional room between the vacuum vessel and transportation galleries. Such a buffer zone will permit to reduce aerosols movement to transportation area.

Vacuum vessel (VV)

Interacent room (IR)

Filter

To the atmosphere

— : Contaminated air
— : Decontaminated
Δ : Clean

Fig. 6 Ventilation scheme in the vacuum vessel.

Zone layout of workrooms should prevent contaminated air from leaving aerosols production areas. Zone layouts together with ventilation allow to come to the following air protection coefficients:

4.1 One-zone layout

Fig. 7 One-zone layout.

\[ K_p = \frac{L_u \cdot K_s}{V} \]  

(1)

where \( L_u \) - quantity of incoming air, \( m^3/s \); \( V \) - room volume, \( m^3 \); \( K_s \) - coefficient of effectiveness of ventilation scheme accepted for the room discussed.

\[ K_s = \frac{C_s}{C_u} \]  

(2)

where \( C_s \) - contaminant concentration in room air, \( mg/m^3 \); \( C_u \) - contaminant concentration in exhaust air, \( mg/m^3 \).

\[ C_u = \frac{G}{L_u \cdot C_u} \]  

(3)

where \( G \) - contaminant quantity entering the room, \( mg/s \).

4.2 Two-zone layout

The air protection coefficient for workroom with contamination source is defined by analogy with the above:

\[ \frac{g_1 \cdot V_1}{K_p^1} = \frac{L_u \cdot K_s \cdot V_1}{V_1} \]  

(4)

\[ K_p^2 = \frac{G \cdot L_u}{g_2 \cdot V_2} \cdot K_s^2 \]  

(5)

where \( V_1 \) and \( V_2 \) - room volumes, \( m^3 \); \( K_s^1 \) and \( K_s^2 \) - coefficients of effectiveness of ventilation schemes (see Eq. 2); \( g_2 \) - contaminant quantity entering the air of adjacent room, \( mg/s \).

\[ g_2 = L_u \cdot C_{s2} \]  

(6)

where \( C_{s2} \) - contaminant concentration in adjacent room.

Thus the air protection coefficient for two-zone layout is as follows:

\[ K_p^* = K_p^1 \cdot K_p^2 \]  

(7)
4.3 Three-zone layout

The air protection coefficient for three-zone layout is calculated similarly, using \( V_3 \), \( g_3 \), \( C_{s3} \), and \( K_{s3} \) respectively. The air protection coefficient for three-zone layout is as follows:

\[
K_{p^3} = K_{p1} \cdot K_{p2}
\]  

(8)

Working premises arrangement should lead to controlled air flows. In its term ventilation scheme should provide for minimal contaminant concentrations in the air of work place and maximum ones - in the exhaust air. The distance between the source of emissions or contaminant generation and the exhaust outlet should be as minimal as possible.

Approximate values of air protection coefficients for different types of workroom layout are given in Table 1.

Table 1

<table>
<thead>
<tr>
<th>Type of layout</th>
<th>Air protection coefficient</th>
</tr>
</thead>
<tbody>
<tr>
<td>One - zone</td>
<td>( 10^3 )</td>
</tr>
<tr>
<td>Two - zone</td>
<td>( 10^2 \sim 10^3 )</td>
</tr>
<tr>
<td>Three - zone</td>
<td>( &gt; 10^4 )</td>
</tr>
</tbody>
</table>

The air protection coefficient is not constant and depends on the degree of air turbulence in workroom as well as on energy dissipated by air flows. Fig.9 shows the air protection coefficient at two-zone layout as a function of air velocity in the opening between two zones.

5. THE PROTECTION OF OUTDOOR AIR

Under normal conditions the problem of environmental safety of enterprises is solved by using a number of measures. They include:
- technology;
- environment;
- industrial premises and their planning;
- protection measures.

Protection measures to prevent environmental contamination are the following:
- ventilation;
- air purification;
- protection zone.

The amount of contaminants released into the atmosphere with ventilation air:

\[
G_a = Lu \cdot Cu \cdot (1 - K_o)
\]  

(9)

This value must be less than the maximum permissible one (\( G_{mp} \))

\[
G_{mp} = C_d \cdot Lu
\]  

(10)

where: \( K_o \) - air purification coefficient; \( C_d' \) - allowable air contaminant concentration in the environment under normal conditions.

If there is a protection zone

\[
G_a = C_d' \cdot X^2 \cdot (1 + U)
\]  

(11)

where: \( X \) - distance between an enterprise and a protection zone; \( U \) - velocity of the wind.

The accumulation of contaminants on filters

\[
G_n = Cu \cdot Lu \cdot K_o \cdot T
\]  

(12)

where \( T \) - duration of filters work.

Under accident conditions in ventilation and air purification systems \( G_n \) should not exceed \( G_{mp} \). If \( G_n < G_{mp} \),

\[
G_{mp} = C_{d'} \cdot Lu \cdot T
\]  

(13)

where \( C_{d'} \) - allowable air contaminant concentration in the environment under accidents.

If \( G_n > G_{mp} \), then the necessary number of independent filters

\[
N = \frac{Cu \cdot K_o}{C_d}
\]  

(14)
6. CONCLUSION

The problem of air protection in premises adjacent to the reactor vessel may be solved by using the following measures:

1. Ventilation of the vessel during the process of its filling with inert gas and after finishing it up to its sealing.
2. The use of local ventilation and interjacent rooms.
3. The air protection coefficient should exceed the level of possible contamination.
4. In order to reduce the unfavourable phenomena risks bound with vibration discussed above it is necessary to:
   - divide large internal spaces into a number of smaller ones;
   - increase the rigidity of vibration walls;
   - provide absorption means for vibrating elements of constructions.
5. To achieve required protection of outdoor air we need to:
   - reduce amount of contaminants released to the air of working premises;
   - reduce amount of contaminants accumulated on filters used for air purification;
   - several independent filtration systems must be used.

Fig. 10 Dependency of air protection coefficient in two-zone layout on the dissipated energy rate.
1 - velocity in the channel connecting two zones is equal to 0.1 m/s.
2 - velocity in the channel connecting two zones is equal to 0.2 m/s.
9.2 Study of the Chemical Interaction between the Beryllium Powders of Different Particles Size and the Air in the Temperature Range 500-1000°C form the Viewpoint of ITER Safety

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ABSTRACT

Under an effect of some factors characteristic for the ITER- operating condition a dense beryllium facing plasma can transit into various forms, changing its structural states. As a result of the bombardment of beryllium plasma facing components by ion fluxes, the production of a dust including the particles from a few micrometers to a few millimeters in size is possible.

The specific features in the behaviour of various beryllium forms under emergency conditions are of an essential interest from the viewpoint of ITER safety.

Some grades of powders of different average particles size (14-31 micron) have been produced in a given study, and their chemical interaction at high temperatures with air (500-1100°C), test duration effects simulating the emergency situation at ITER in the first approximation have been studied.

The temperature dependence of beryllium powders (different particles size after disc abraded) interaction with air in the temperature range 500-1000°C at the exposure of 5 hours long for each temperature and kinetic dependence of interaction of these powders with air at 800°C for the exposure from half an hour to 7 hours long were studied.

An analysis of granulometric weight fraction in the metallic and oxidized beryllium powders with different particles size has been done by the photodimensional technique with the instrument "Analysette 20".

Construction of a mathematical model for the chemical interaction of beryllium powders with air at high temperatures have been carried out.

INTRODUCTION

Under an effect of some factors typical for the ITER- operation condition, a dense beryllium facing plasma can transit into various forms, changing its structural states. As a result of the bombardment of beryllium plasma facing components by ion fluxes, the production of a dust including the particles from a few micrometers to a few millimeters in size is possible [1-4].

The specific features in the behaviour of various beryllium kinds under emergency conditions, such as LOFA, LOVA, LOCA are of an essential interest from the viewpoint of ITER safety.

Some grades of beryllium powder of different grain size have been produced in a given study, and their chemical interaction with air at high temperatures, test duration effects simulating the emergency situation at ITER in the first approximation have been studied.

1. STUDY OF CHEMICAL INTERACTION BETWEEN THE BERYLLIUM POWDERS OF DIFFERENT PARTICLE SIZE AND THE AIR IN THE TEMPERATURE RANGE 500-1000°C.

1.1. Characteristics of initial beryllium powders.

The powders of metallic beryllium of various particle size, produced by the technique of disk abraded were used in the studies.

The chemical composition, average size of the particles, specific surface and the bulk density of the beryllium powders used in the studies are represented in Table 1.
The studies of powder-air interaction processes at high temperatures were performed in the special crucibles made of beryllium oxide with the dimensions: 25 mm in diameter, 43 mm high, with the walls, 2.5 mm thick, and 13 cm³ in volume. The crucible density is 2.90 g/cm³ at practically-zero open porosity.

The powder-air interaction surface was equal about 3 cm², at the inner crucible diameter of about 20 mm.

The reacted beryllium fraction was found from a weight gain of the samples. The calculation of the reacted beryllium percent was done, using the chemical reaction 2Be + O² → 2BeO.

The reacted beryllium powder fraction was found by dividing the weight gain by the coefficient 1.775. An average value of 3-5 experiments (done at the same conditions) was used for calculations. The mean square deviation (MSD) of the experimental results was also calculated.

In the report [5] the technique and instrumentation for the studies have been discussed in more detail.

1.2. EXPERIMENTAL RESULTS.

1.2.1. Temperature dependence of the beryllium powder interaction with air.

The temperature dependence of beryllium powder (of different particle size after disc abrasion) interaction with air was studied in the temperature range 500-1000°C, at the exposure of 5 hours long for each temperature. The filled powder layer height was varied in the range of 2-8 mm. As a result of studying the temperature dependence, it was found out that:

- start-up of beryllium powder interaction was registered at 500°C, the fraction of the reacted beryllium was 1.5-1.8% for the powders, 20µm in the particle size and 1.2-1.4% for the powders, 30µm in the particle size;
- the tendency to a reduction in the powder interaction at an increase in the filled layer thickness was detected moreover, at 700°C the percent of the reacted powder is 18-25% for smaller particle size powders and 12-15% for larger particle size powders;
- at 800°C, due to an increase in the produced oxide film thickness and, as a result of this,
1.2.2. Kinetic dependence of beryllium powder interaction with air.

The kinetic dependence of beryllium powder (of various particle size) interaction with air was studied at 800°C in 4 mm thick powder layer, at exposure from a half an hour to 7 hours long. The temperature 800°C was chosen for the experiments as a maximum-permissible one for beryllium components facing plasma in ITER under operating conditions.

As a result of studying the kinetic dependence of beryllium powder interaction with air it has been found out the following:

Fig. 1. Temperature dependence of interaction of the various particles size beryllium powders with air during 5 hours (powders layer thickness 4-4.5 mm).

<table>
<thead>
<tr>
<th>Test temperature, °C</th>
<th>500</th>
<th>600</th>
<th>700</th>
<th>800</th>
<th>900</th>
<th>1000</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1600</td>
<td>1500</td>
<td>1400</td>
<td>1300</td>
<td>1200</td>
<td>1100</td>
</tr>
</tbody>
</table>

Table 1
Characteristics of initial beryllium powders

<table>
<thead>
<tr>
<th>Element</th>
<th>Be</th>
<th>Si</th>
<th>Fe</th>
<th>Mn</th>
<th>Mg</th>
<th>Cr</th>
<th>Ni</th>
<th>Al</th>
<th>Cu</th>
<th>C</th>
<th>O</th>
</tr>
</thead>
<tbody>
<tr>
<td>Grades of beryllium powders</td>
<td>CBP-56-1</td>
<td>CBP-56-2</td>
<td>CBP-56-3</td>
<td>CBP-30-1</td>
<td>CBP-30-2</td>
<td>CBP-30</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Chemical composition, % wt.</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Be</td>
<td>99.13</td>
<td>99.00</td>
<td>98.81</td>
<td>98.61</td>
<td>97.73</td>
<td>98.9</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Si</td>
<td>0.013</td>
<td>0.014</td>
<td>0.013</td>
<td>0.014</td>
<td>0.013</td>
<td>0.013</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fe</td>
<td>0.11</td>
<td>0.15</td>
<td>0.14</td>
<td>0.18</td>
<td>0.10</td>
<td>0.026</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mn</td>
<td>0.0038</td>
<td>0.012</td>
<td>0.0043</td>
<td>0.0084</td>
<td>0.004</td>
<td>0.0026</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mg</td>
<td>0.008</td>
<td>0.011</td>
<td>0.008</td>
<td>0.0093</td>
<td>0.0004</td>
<td>0.002</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cr</td>
<td>0.026</td>
<td>0.036</td>
<td>0.028</td>
<td>0.064</td>
<td>0.028</td>
<td>0.0054</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ni</td>
<td>0.009</td>
<td>0.014</td>
<td>0.011</td>
<td>0.017</td>
<td>0.011</td>
<td>0.004</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Al</td>
<td>0.014</td>
<td>0.017</td>
<td>0.015</td>
<td>0.024</td>
<td>0.013</td>
<td>0.004</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cu</td>
<td>0.004</td>
<td>0.004</td>
<td>0.005</td>
<td>0.0071</td>
<td>0.0043</td>
<td>0.0026</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>0.086</td>
<td>0.077</td>
<td>0.088</td>
<td>0.07</td>
<td>0.079</td>
<td>0.077</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>O</td>
<td>0.6</td>
<td>0.66</td>
<td>0.88</td>
<td>1.0</td>
<td>1.01</td>
<td>0.96</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Average particle size, μm

| Average particle size, μm | 31.3 | 30.1 | 29.1 | 14.0 | 19.1 | 20.5 |

Specific surface, m²/g

| Specific surface, m²/g | 0.43 | 0.65 | 0.46 | 0.90 | 1.00 | 1.03 |

Bulk density, g/cm³

| Bulk density, g/cm³ | 0.68 | 0.62 | 0.68 | 0.61 | 0.57 | 0.57 |
- a considerable beryllium powder oxidation is observed during the first half an hour, the fraction of the reacted beryllium is equal to 16-21% for the 20μm powder particles and it is equal to 9-16% for the 30μm particles. The enhanced interaction rate for the powder of DBP-30-Type (up to 30%) is explained by the technology of its production and by the chemical composition of the material.

The interaction during the first three hours occurs at the reduced rate, as a result of the dense protective surface layer formation with continuous growth of the layer thickness. During the subsequent two hours the rate of interaction rises and attains a maximum after the exposure time of 5h that can be explained by a rupture in the thickened oxide layer and by the emergence of new surfaces of non-oxidized beryllium. The further increase in the time of oxidation up to seven hours insignificantly affects the interaction rate as a result of a reduction in the non-oxidized metal surface.

For a finer beryllium powders the percent of the oxidized material, after 5h of exposure, is about 83-86%; for a larger one-about 76-80%.

These results show that finer beryllium powders having a great specific surface are more active in the interaction with air, producing a finer-disperse oxide. It is confirmed by the studies of granulometric oxide structure, as well as by enhanced oxide activity under subsequent high temperature sintering manifested in the formation of denser structures.

Thus, the oxide produced under interaction of an initial 14μm beryllium powder with air, after sintering in vacuo at 1940°C, has the density 2.78 g/cm³, meanwhile the initial beryllium powder with the 31μm particles, has the density 2.63 g/cm³ only.

The results of studying the kinetic dependencies are represented in Fig.2.

Fig. 2. Kinetic dependence of interaction of the various particles size beryllium powders with air at temperature 800°C (powders layer thickness 4-4.5 mm)

2. Conclusion:

1. Start-up of the powder interaction with air in freely-filled layer begins at the temperature below 500°C, and a fraction of the reacted material is in the range from 1.2 to 1.8%. The rise is related with a reduction in the initial particle size of powders;

2. Interaction of the powders with air up to 94-98% is attained at 1000°C. The oxidation less 100% is probably related with the presence of some amount of oxygen in the initial beryllium powders and with accuracy of the technique;

3. At 800°C, the interacting fraction up to 9-13% is attained during the first half an hour of the contact between the powders and the air, it rises with reduction in the particle size of the initial beryllium powder. During the next 3-4 hours the rate of interaction is moderated, and the oxidized material fraction is equal to about 23-30%. After 5h of contact with air, a steep rise in the interaction rate is observed and the reacted powder fraction attains 75-
86%. In this case, the distilled powders, DBP-30-Type, interact with air more intensely;

4. An experimental technique for beryllium powder oxidation - partly-modelling the emergency situation - has been developed. It has been shown that the optimal layer thickness for production of a reliable information on the powder oxidation in air should not exceed 4 mm. The mean square deviation of the experimental results expressed in the percent of the oxidized powder fraction is 0.3-4.5%, dependent on the temperature of the tests;

5. A partial packing of the interaction products (conglomeration of the oxide powder at 900-1000°C, more pronounced at the initial beryllium powder layer thickness of 8 mm) has been registered [5].

3. Construction of a mathematical model for chemical interaction of beryllium powders with air in the temperature range from 500 to 1000°C

A statistical analysis of an experimental data file including kinetic and temperature dependences of a weight gain, as a result of beryllium interaction with air, has been done. Along with the test temperature and the exposure time, the apparent (loosened) density, specific surface and an average particles size were varied. The ranges of changes in these factors are represented in Table 2.

<table>
<thead>
<tr>
<th>Factor</th>
<th>Range of changes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Apparent density, g/cm³</td>
<td>0.57-0.68</td>
</tr>
<tr>
<td>Specific surface, m²/g</td>
<td>0.43-1.00</td>
</tr>
<tr>
<td>Average particles size, µm</td>
<td>14.0-31.3</td>
</tr>
<tr>
<td>Test temperature, °C</td>
<td>500-1000</td>
</tr>
<tr>
<td>Exposure time, h</td>
<td>0.5-7</td>
</tr>
</tbody>
</table>

In the construction of a model for the heterogeneous processes rate of which is controlled by diffusion to long distances, the semiempirical kinetic equations [8]:

\[
\frac{dW}{dt} = K \cdot W^a \cdot (W_N - W)^b
\]

where \( W \) is the weight gain [mg/g]; \( W_N \) is the maximum-possible weight gain [mg/g];

\( t \) is the exposure time [h];

\( K \) is the process rate constant;

\( a, b \) are the constants.

If one assumes that \( b = 1 \), \( a = \frac{n}{n-1} \) and takes account of the temperature dependence of \( K \) in the Arrhenius representation:

\[
K = \exp\left(-\frac{Q}{R \cdot T}\right) \cdot t^n,
\]

the relationship for kinetic and temperature dependences of an weight gain has the form:

\[
W = W_N \cdot \left(1 - \exp\left(-\exp\left(-\frac{Q}{R \cdot T}\right) \cdot t^n\right)\right)
\]

where \( T \) is the test temperature, K;

\( R \) is the universal gas constant (\( R = 8.3143 \) J/mole×K);

\( Q \) is the apparent activation energy of the beryllium oxidation process, J/mole;

\( n \) is the index of a power (weight gain kinetics rate);
C - is the index of the weight gain change rate.

In the construction of the regressive model, the dependences of C-parameter of the equation (1) on some factors: apparent density, specific surface, average particles size - were checked. All these factors are closely related with each other, and, although the factor "specific surface" only was included into the final model, involvement of the factors "apparent density" or "average particle size" into the model, instead of that factor, gives close results, but somewhat worse from the viewpoint of statistic characteristics.

As a result, a regressive model [9] of the weight gain dependence on the test temperature, exposure time, as well as on the specific surface \( Y \) \( [m^2/g] \), has been produced:

\[
W = 1774.85 - \exp\left(\frac{14.864 + 0.6867 \cdot Y}{R \cdot T}\right) \cdot \exp\left(-\frac{152234}{J \cdot t}\right) + 15.
\]

A theoretical value of \( W_n = 1774.85 \) [mg/g] is used in this relationship.

3.1. Statistical characteristics of the model

The model parameters in the relationship are given together with the corresponding 95%-reliability ranges. One can see that the model is statistically useable with the reliability level higher than 95%.

An average deviation of the model-calculated weight gain from the experimental data is equal to 150 mg/g.

3.2. Graphical model representation

The comparison between the calculated and experimental kinetic dependences for temperature of 800°C is given in Fig.3.

Fig. 3. Comparison of the kinetic of calculated data with experimental ones for \( T=800 \) °C

The comparison between the calculated and experimental temperature dependences for \( t = 5h \) within the coordinates \( 1/T - \ln (W) \) is given in Fig.4.
Fig. 4. Comparison of temperature calculated dependencies with experimental ones for τ=5 hours in the coordinates 1/T - ln(W)

One can see a satisfactory agreement between the experimental and calculated data.

References


9.3 Some Results of Medical Researches at Ulba Metallurgical Plant

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The results of 45-years medical researches at beryllium production of Ulba Metallurgical Plant are summarized in this report. Statistic data on different kinds of occupational diseases, related to beryllium production and the dynamics of changing occupational diseases with the development of beryllium production, are given there. Data on average duration of life of occupational disease patients are presented in the report. It includes the description of problems, related to berylliosis diagnosis. Issues, connected to beryllium production effect on health of man, located nearby beryllium production are also discussed there as well.

One of the reasons, which prevents a wide application of beryllium, is its high toxicity, which causes an occupational diseases of lungs, skin, mucous membranes of human body.

Numerous investigations on this problem were carried out in the USSR. Considerable study is being given to berylliosis problem in the USSR since the early 70-s. The main researches were executed on the base of Ulba Metallurgical Plant under the supervision of Leningrad R&D Institute of Sea Transport Hygiene (nowadays it is R&D of Industrial and Sea Medicine, Sankt-Petersburg) and Institute of Biophysics (Moscow). Investigations involved the range of issues, such as epidemiology, diagnostics, treatment and prophylactics of Be disease as well as the health problem of personnel and people, live near Be processing plant. Recommendations were made for all above described directions, as the result of accomplished work.

Epidemiology of beryllium injury at the facility. There are 3 stages in evolution of Be manufacturing and processing production:

- 1951-59 - putting into production;
- 1960-69- reconstruction;
- 1970 up to the present day post-reconstruction period.

During the 1-st stage the technology of Be production with sulfate-ammonium and chloride methods were developed. Average concentrations of soluble and insoluble Be compounds exceeded the top permissible concentration (TPC) for standard work shift in 10...100 times; maximum values came up to 400 TPC. From 1960 through 1964 shops for manufacturing of metal, BeO and their products were put into operation sequentially. Reconstruction of the operating shops was completed before 1969. This period is characterized by deterioration of a sanitary and hygienic situation. The average concentrations of Be compounds in air of production rooms were 2...600 mg/m$^3$ and peak values exceeded TPC in 5000 times. The 3-d stage of production development involves the time since 1970 up to the present day. During this period a significant improvement in labor conditions was made due to a betterment of technology, individual protection means and because the sanitary requirements were adhered to. But yet some of areas indicated a high level of Be compounds concentration. The number of such areas successfully goes down as a
consequence of executed work. At present the average concentrations in the various shops are 1...3 TPC.

Within 1985-1990 98...150 cases of exceeding TPC were registered at the plant.

In compliance with the above mentioned, the nature of occupational sickness varied. Within the 1-st stage a general toxic effect of Be compounds occupied the 1-st place. It led to a development of numerous injuries in the form of acute toxic bronchitis, broncho-bronchiolitis, pneumonitis, as well as conjunctivitis and dermatitis. Disease incidence increase correlates to the worsen of sanitary situation, owing to either putting into operation new shops, or reconstruction of an old ones.

Over a period of putting the production into operation, the labor conditions were very disadvantageous and the lack of individual protection means along with Be high toxicity caused a mass disease incidence of staff (acute bronchitis, pneumonitis, conjunctivitis, dermatitis).

After 1965 a scheme of occupational disease incidence changed entirely. The first case of granulomatosis was registered in 1967, and since 70-s chronic forms of lungs pathology have occupied the first place. Acute conjunctivitis and dermatitis have not been registered since 1988. An acute lung injuries were liquidated by 1970. Five cases of such injury took place, but it was correlated with emergencies.

Within 1951-1995 1810 cases of disease incidence were identified at the plant, which were connected with Be effect. An acute Be injuries have comprised 71 %, chronic- 29% (tabl.1).

The analysis of lungs pathology was based on the statement that all its forms were interrelated and indicated one disease-berylliosis. It was confirmed by the fact, that the first diagnosis of chronic occupational bronchitis in a patient was followed by identification of lungs granulomatosis at which clinico-roentgenologic indications of bronchi injury took place in a greater or lesser extend [1...3].

Chronic lungs diseases, caused by Be effect, are represented at the facility in 3 major forms:
- chronic toxic-chemical bronchitis (CTCB) as the result of soluble Be components effect;
- primary chronic toxic-chemical bronchitis;
- lungs granulomatosis.

About 34,6 % of patients, exposed to a strong effect of Be compounds, got a chronic toxic-chemical bronchitis. 6,3 % of acute intoxication cases ended in lungs granulomatosis. But the analysis of these workers activity within the followed years gives no confirmation that an acute injury directly transforms to granulomatosis. The matter is that the half of the workers continued to contact with Be within some time after getting intoxication; another half of them worked in a supplementary shops at a production site.

Table 1. Occupational disease incidence at the facility (1951...1995 years).

<table>
<thead>
<tr>
<th>Name of a disease</th>
<th>Acute</th>
<th>Chronic</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>Occupational lungs disease</td>
<td>252</td>
<td>459</td>
<td>711</td>
</tr>
<tr>
<td>Occupational dermatitis</td>
<td>602</td>
<td>42</td>
<td>644</td>
</tr>
<tr>
<td>Occupational conjunctivitis</td>
<td>432</td>
<td>23</td>
<td>455</td>
</tr>
<tr>
<td>TOTAL:</td>
<td>1286</td>
<td>524</td>
<td>1810</td>
</tr>
</tbody>
</table>

The possibility of acute Be injury in the future can not be excluded. Emergency situation, or breaking of industrial sanitary requirements and safety rules can cause such injuries.

Primary-chronic toxic-chemical bronchitis developed among people, who started their work during putting-into-production and reconstruction stage. They were exposed to a strong and continuous effect of soluble Be compounds and to a range
of irritate agents (ammonia, hydrofluoric acid, nitrogen oxide) mainly. The complex examination of Be production workers, which was being carried out together with NII GMT specialists up to 1992, allowed to identify practically all cases of chronic bronchitis. It is unlikely, that this pathology will develop in the future [4...7].

Examinations which were accomplished on the base of the Register of occupational disease patients and Be production workers, have showed that lungs granulomatosis is a leading pathology at present and it will be leading in the future.

It is determined mainly among the experienced workers, who was exposed to Be before 1970, but it is also possible to identify berylliosis among those, who started their work after 1970. Cohort research method have showed that 3.2 % of people, who started to work at the plant in 1970-1974, had a lungs granulomatosis. 1986 year prognosis about the growth of disease in 2...4 times was not confirmed by the additional research of 1992 [5...7, 8]. According to different authors evidence, it is possible, that 1...10 % of employers, been exposed to the high Be concentrations could have berylliosis [9].

Analysis of Be disease incidence have presented, that 95.5% of patients were exposed to insoluble Be compounds (in Be, BeO and its products manufacturing shops).76.4 % of main workers (instrument control man, operatives, mechanics) and 23.6 % of the others (electricians, engineers, inspectors) were among patients. By the moment of making a diagnosis, an experience of working under the exposure to Be was the following:

- 5 years- 15.3 %;
- 6-10 years- 19 %;
- 11-15 years -24.4 %;
- 16 years and more -41.3 %.

Consequently, with the increase of exposure time the risk of granulomatosis development also grows [4...7].

An occupational lungs pathology is often accompanied by serious attendant diseases and complications. Lungs granulomatosis grows progressively with a restrictive infringement of pulmonary ventilation, which could ended in a pulmonary-heart insufficiency. Patients with a chronic occupational bronchitis have a predominance of the obstructive infringement of pulmonary ventilation; a progress of a diseases, which could cause pulmonary- heart is rare. Such patients more oftenly get heart ischemia disease and hypertension.

Liver, biliary tract and stomach diseases, are berylliosis accompanying pathologies. Joints disease of polyarthritis type, which occurs frequently among patients with any kind of Be injury of lungs, engages one's attention. Sometimes the prognosis for man life and work is significantly deteriorated by addition of diabetes mellitus.

Analysis of occupational disease patients mortality. Throughout the whole lifetime of the facility 156 patient have died. The analysis was made in lungs pathology, as far as a skin and mucosal pathology does not change a duration of patients life and their capacity for work. However, the exception is made by skin Be granulomas, which indicate a general berylliosis and a prognosis for them is made on a base of lungs granulomatosis growth.

Table 2. The mortality of occupational disease patients

<table>
<thead>
<tr>
<th>Death reasons</th>
<th>Be-s</th>
<th>%</th>
<th>CTC</th>
<th>%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diseases</td>
<td>63</td>
<td>95.5</td>
<td>77</td>
<td>85.6</td>
</tr>
<tr>
<td>including:</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Malignant tumor</td>
<td>5</td>
<td>7.6</td>
<td>17</td>
<td>18.9</td>
</tr>
<tr>
<td>Blood circulation</td>
<td>5</td>
<td>7.6</td>
<td>21</td>
<td>23.3</td>
</tr>
<tr>
<td>Lungs dysfunction</td>
<td>46</td>
<td>69.7</td>
<td>20</td>
<td>22.2</td>
</tr>
<tr>
<td>Other diseases</td>
<td>7</td>
<td>10.6</td>
<td>19</td>
<td>21.1</td>
</tr>
<tr>
<td>Injuries, intoxication</td>
<td>3</td>
<td>4.5</td>
<td>13</td>
<td>14.4</td>
</tr>
<tr>
<td><strong>TOTAL</strong></td>
<td>66</td>
<td>100</td>
<td>90</td>
<td>100</td>
</tr>
</tbody>
</table>
Reasons of death, an average age, a time interval from the moment of making a diagnosis depend on a form of occupational lungs pathology.

The average life period with lungs granulomatosis is 9.2 years after diagnosis is made and with chronic toxic-chemical bronchitis (CTCB) - 13.5 years. The average age of patients, died from berylliosis is 54.7 year; from CTCB - 56.6 years. This data testify that the growth of a disease is very hard and the prognosis for berylliosis is unfavorable. Life period of diseased woman, is 3 years less than of man. This comes in a contradiction with the common tendency of mortality depending on sex and indicates the harder growing of pathology among woman [10].

The main reason of death among granulomatosis patients is pulmonary-heart insufficiency. CTCB patients die to the same extend from cardiovascular, respiratory systems diseases, oncology disease and etc. The percentage of those who died from injuries and intoxication are very high. Oncology pathology turns to be a reason for CTCB patients death more than twice as often as that for lungs granulomatosis patients (18.9% and 7.6 % accordingly). Lungs cancer forms more than half of all cases (granulomatosis -60%; CTCB -58.8%). Be is recognized as a suspected carcinogen for man, so with Be injuries no question comes up about the linkage between a profession and oncological pathology. The other localization of oncological process occur in a single case.

Berylliosis diagnosis presents some difficulties because clinical, roentgenological and some other changes caused by berylliosis are compatible to these caused by sarcoidosis and other granulomatosis diseases of occupational and non-occupational etiology [11, 12]. It is necessary to make a correct diagnosis for the following reasons:

• it is necessary to provide a patient with the other work, which is not connected with Be.

Generally, the diagnosis of Be granulomatosis is based on the following criteria:

• the effect of insoluble Be compounds with a high concentration within a lasting period of time;
• clinical indications of lungs injury (cough, dyspnea, etc.);
• specific changes in lungs, determined by roentgenological examination of initial stage of disease (X-ray photograph of enlarged lungs);
• external respiration dysfunction of a restrictive type;
• loss of lungs diffusion capacity;
• positive skin tests with soluble Be compounds;

The following immune-allergic tests for identifying a sensibility to Be, are the additional methods of diagnosis:

• determination of Be antibodies in compliment binding reaction;
• determination of lymphocytes with ceptors to Be;
• specific agglomeration of leukocytes under Be effect (RSAL);

The other researches (protein exchange, infringement, cytochemical changes, etc.) are of a lesser diagnosis importance. Among immunology confirmations of Be effect there are no researches of liquid, obtained from bronchial lavage. The major immune examinations are being carried out with the peripheral blood cells.

The diagnosis of a chronic occupational bronchitis is based on the continuance of a contact with toxic compounds and irritants and on the immune researches, which confirm a sensibility to Be. It is very difficult to make a diagnosis of a chronic bronchitis because there are no specific changes and a lot of nonspecific factors which could also cause a chronically bronchitis (smoking, infections, etc.) are available.
Health state of people located nearby beryllium facility. The researches of people state of health were being carried out with 10 years repetition rate. The objective of such researches was to find out the changes in state of health, which could be caused by Be effect. Recommendations on environment improvement, people making healthier, primary and secondary prophylactics were the result of that work. Researches included disease incidence analysis, examination of the hematological and biochemical indexes, tests for determination of specific Be effect (immunological researches, skin allergic tests with beryllium, cobalt, nickel, chromium) [8, 10, 13, 14].

Workers of different plants, pupils, pensioners were under examination. Separate study was accomplished on the evaluation of children health. It was noticed in all researches that townspeople are exposed to wastes of 176 enterprises. More than 60 chemical agents and compounds are ejected to environment, including high-toxic and carcinogenic stuff.

It is very complicated to separate the effect of some single agent in such a situation. So, Be forms $1.0 \times 10^{-3}$ share of UMZ wastes and $1.0 \times 10^{-6}$ of the whole town wastes. It is necessary to point out that within the past 30 years Be ejection into the open environment have been decreased approximately in 50 times.

As the result of a complex research the high level of disease incidence of the population was found generally at the expense of internal organs and nervous system pathology. A level of upper respiratory tract pathology is also very high. It should be mentioned that a personnel, which works with Be and is exposed to higher concentrations, has a lower percent of disease incidence if to compare with the whole town and other plants levels. Hence, there is a reason to think that a high disease incidence in the city does not interrelate with Be impact on people.

Results of monitoring researches allow to trace the changes in specific characteristics. So, a comparison of Be content in environment and in a human body within past years shows its reduction towards the natural content. It should be particularly noticed that in 90...91 Be content was near normal and rarely exceeded 1 mg/l in urine. It indicates the reduction of Be effect and decrease of its content in a body. It is also confirmed by the examination: in 1974 - 4% of inspected people had a positive skin test on Be; in 1982 and 1991- no positive tests were found even among Be plant workers.

Immunological researches allowed to determine that RSAL frequency and intensity have reduced from 48% to 28%. No biochemical changes existed earlier, were found. For example, the reduction of alkaline phosphates in periphery blood wasn't determined.

It could be concluded Based on this data, that Be wastes and its emission into environment during the last years had a minimal effect on people health and did not cause any noticeable changes. Researches having been carried out confirmed again the absence of Be carcinogenic effect.

SUMMARY

The results of researches have allowed to study the development of principal forms of lungs pathology. After 1965 acute lungs injuries were practically liquidated. Now, acute toxic injuries can take place either in emergency situations, or when sanitary and safety requirements are broken.

During the plant workers examination it was found out, that the primary chronic toxic-chemical bronchitis had people, started their work before 1970. It is possible that these diseases will grow in the future in patients, having been exposed for a long time to toxic and irritate stuff in concentrations that exceeded TPC tenfold.

The main occupational lungs pathology in the future 20 years will be a lungs granulomatosis (LG). It will be found in workers, started to work in a contact with Be before 1970. Among individuals, started to work with insoluble compounds after 1970.
the growth of granulomatosis is possible if their work length of service is more than 10 years. Long-standing observations over Be inflow and accumulation in a body indicate a significant increase of its content in lung tissue and bronchi-pulmonary lymphonodus.

There are no direct dependence in granulomatosis between a quantity of Be in a body and disease growth, but an excessive accumulation of Be could cause a growth of sensitization, and exhaustion of body compensatory possibilities. With the fail of body protection specific pathology has a chance to grow.

Be effect on townspeople goes down. This could be confirmed by the examination of a specific immune reaction. There were no berylliosis cases among townspeople registered within the whole period of Be production operation.

People health examinations are to be continued, with the special concern to children. Health of Be workers children is noteworthy. Be, found in children's organism in quantities, equal to what workers got during the work with the material, points to the fact that significant portions are being carried out with clothes, footwear, on skin, what is intolerable.

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10. Hot News

10.1 Development of Be/Glidcop Joint Obtained by Hot Isostatic Pressing Diffusion Bonding for High In-service Temperature

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This paper addresses some aspects of the beryllium-Glidcop\textsuperscript{®} joining by Hot Isostatic Pressing diffusion Bonding. The quality of a joint is mainly dependent on the interface microstructure. Thus, as Be/copper direct bonding is not recommended, the choice of interlayers is a critical point. The joining process parameters, i.e. temperature, pressure and time, must take into account the in-service requirements, the mechanical and metallurgical properties of each material. The Be/Glidcop\textsuperscript{®} joining process developed at CEA/Grenoble is presented here.

1. INTRODUCTION

For several years, research on reliable joining techniques of beryllium onto copper alloys has been carried out in the frame of the ITER fusion reactor project. Various joining alternatives have been studied: Be coating technologies [1], brazing [2-4] and diffusion bonding [4-7]. Even though mechanical attachment is simple [8], it does not seem to be considered any more because of the poor thermal transfer through the joint. Depending on the location of beryllium used as plasma facing material, the maximum Be surface heat flux considered for the design of components is very different: 0.5 MW/m\textsuperscript{2} for primary wall modules, 8 MW/m\textsuperscript{2} for limiters and 3 MW/m\textsuperscript{2} for baffle modules. Thus, joining solutions may be different from a component to another.

For instance, as the heat flux is low for primary wall modules, the use of Thermal Bond Layer (TBL) is under consideration [1]. Thixotropic brazing Al-Ge alloys are potential candidates as TBL [1,9,10]: these alloys allow the in-situ replacement of damaged component because of their medium melting point. Moreover, these alloys are designed to be compliant for reducing the interface thermal stresses.

On the contrary, the beryllium-copper alloy junctions must withstand high in-service temperature for limiters and baffle modules.

Diffusion bonding is a process that produces solid-state joining by diffusion under pressure and temperature. Hot Isostatic Pressing diffusion Bonding (HIPB) is particularly suitable for large surface to be joined and complex or intricate geometry. Moreover, HIPB allows high quality junction that is necessary for a good heat transfer through the interface. Direct diffusion bonding between dissimilar materials may be sometimes difficult to achieve. Thus, interlayers are used to facilitate bonding or to prevent the formation of brittle phases at the interface [11].

HIPB of Be to Glidcop\textsuperscript{®} was firstly developed at CEA/Grenoble for low in-service temperature: interlayers were used to avoid interdiffusion between Cu and Be [5]. As pure Al associated with diffusion barriers was used for the bonding, the in-service temperature was restricted to about 200-300°C.

This paper reports the development of beryllium to Glidcop\textsuperscript{®} joining technique using HIPB for high in-service temperature.

2. JUNCTION DESIGN

The thermal and mechanical properties of a joint are mainly dependent on the interface microstructure and its stability under service conditions. Thus, the process, mechanical and metallurgical aspects have to be considered for the design of high resistance Be/Glidcop\textsuperscript{®} joints.

The reactivity between Be and Cu is very high: it has been shown with diffusion couples that reactive layer begins to appear at the interface at 400°C and becomes very large at temperature above 700°C [12]. It is recommended that temperature should be lower than 620°C for direct Be/Cu diffusion bonding. It should be noted that the joint temperature may be higher than 400°C during transient events. Thus, it
seems that interlayers have to be used to avoid the excessive growth of brittle Cu-Be intermetallics. The use of silver as interlayer is out of consideration because of its transmutation to cadmium under neutron flux [2,4].

2.1. Process point of view

The ITER baffles or primary wall modules are made of succession of different joined materials: Be as plasma facing material, copper alloy as heat sink material and 316LN stainless steel as supporting structure.

The Be to copper alloy joining operation will be performed after the copper alloy to 316LN stainless steel joining. As 316LN stainless steel is sensitive to stress corrosion cracking when heat-treated between, roughly, 650°C and 720°C [13], this region of temperature should be avoided for Be/Glidcop® HIPB. For a low carbon content (about 0.03 wt%), the sensitisation region is reached in 100 hours but may appear sooner for higher carbon content.

The maximum HIPB temperature is limited to 860°C: the Be-Cu phase diagram shows a minimum liquidus temperature at 860°C [15]. The temperature range for the Be/Glidcop® HIPB process is then: below 650°C or, between 720°C and 860°C.

For temperature higher than 860°C, the joining process and interlayers used have to guarantee that no Be will be in contact with Cu anytime at temperature above 860°C. Otherwise, Be-Cu liquid will appear and the joining may be difficult to control.

As HIPB temperature precision is about 10°C, it seems reasonable to consider the following HIPB temperature range: below 640°C, between 730°C and 850°C as extremes. Below 640°C solution based on aluminum can be used [see for instance 5].

In the following, we will only consider the 730-850°C temperature range for HIPB.

2.2. Mechanical point of view

One of the main problems in dissimilar joining is the occurrence of residual stresses during the cooling down due to the difference of the Coefficient of Thermal Expansion (CTE). The residual stresses were calculated on Be/Glidcop® junction using an elastic-plastic model [16] with the following dimensions for the Be and Glidcop® plate:

\[
\sigma_i = \Delta T (\alpha_2 - \alpha_1) \frac{E_1 E_2}{E_1 + E_2} 
\]

where \( \sigma_i \) is the residual stress in material \( i \), \( \Delta T \) is the difference between the joining temperature and the room temperature, \( E_1 \) and \( E_2 \), \( \alpha_1 \) and \( \alpha_2 \) are respectively the Young modulus and the CTE for materials 1 and 2.

In (1), the two joined materials have the same dimensions and geometry. The residual stress is then only a function of the CTE and the Young modulus. The CTE difference between Be and Glidcop® is not very large above 200°C (see fig. 1). The residual stress difference \( \sigma_2 - \sigma_1 \) is minimized for \( E_1 = E_2 \) and will increase with the \( E_1 - E_2 \) difference. The Young modulus difference between Be and Glidcop® is

Figure 1. Be and Glidcop® CTE as function of temperature.

Figure 2. Be and Glidcop® Young modulus (GPa) as function of temperature.
very large at low temperature (see fig.2) [18,19]. Thus, we think that interlayers may have intermediate Young modulus in order to smooth the very large Be/Glidcop® Young modulus difference.

The linear elongation or contraction of a material with temperature is given by:

\[ \frac{\Delta L}{L} = \alpha \Delta T \]  

(2)

where \( \Delta L \) is the length variation and \( L \) is the reference length.

Figure 3 which gives the Be and Glidcop® contraction as function of temperature is deduced from figure 1. The reference is taken at 850°C: the joint is supposed to be achieved at this temperature. In figure 3, the largest contraction difference between the two contraction curves is within the temperature range 200-350°C. This can be seen in figure 4 where the contraction difference between Be and Glidcop® during the cooling down is given by \( \Delta T(\alpha_2 - \alpha_1) \). Thus, a residual stress relaxation cooling cycle should be defined to take into account this behaviour in that temperature range.

2.3. Metallurgical point of view

In order to have one or several layers with lower Young modulus than the Be one, the interlayer placed between Be and Glidcop® should promote the formation of intermetallics with Be and with Cu. Beryllide intermetallics occur with most of the transition elements. Thus, the choice is mainly dependent on the formation of Cu intermetallics. Most of the transition elements have no solubility in Cu, or are immiscible with Cu, or does not form any intermetallics with Cu [14], or are not acceptable for fusion applications. A certain amount of solubility between the interlayer and, Be and Cu is necessary for the diffusion process. Titanium has an intersolubility of few percent with copper at temperature above 800°C [14]. The metallurgical aspect of Be/Ti/Glidcop® joint is developed in [20].

3. EXPERIMENTAL PROCEDURE

The experimental procedure, sample and container preparation is described elsewhere [20]. The HIPB cycle is: heating up to 850°C and 120 MPa, plateau with these parameters during 3 hours, cooling down to 400°C and 3 MPa, plateau with these parameters during 2 hours followed by the final cooling to room temperature.

4. RESULTS

Interdiffusion at the Be/Ti/Glidcop® joint has led to the formation of six different layers and a Cu-Ti solid-solution with precipitates in the Glidcop®. The joint microstructure is given in figure 5. The interpretation of the different phases was obtained from X-microprobe profile analysis [20] and phase

![Figure 3. Be and Glidcop® contraction as function of temperature with reference point at 850°C.](image3)

![Figure 4. Contraction difference (%) between Be and Glidcop® as function of temperature with reference point at 850°C.](image4)

![Figure 5. Microstructure of the Be/Ti/Glidcop joint.](image5)
diagrams. As the Be-Ti-Cu ternary phase diagram does not exist, Cu-Ti and Ti-Be binary phase diagrams were only considered [14,15].

5. CONCLUSION

Titanium interlayer may be used as interlayer for the fabrication of HIPB Be/Glidcop® joint. The HIPB parameters have been chosen regarding the process, mechanical and metallurgical aspects for the design of high resistance Be/Glidcop® joints. This joining technique was successfully used for the fabrication of two mock-ups: one with a large Be plate and the other one with Be tiles [21,22].

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1. Introduction

In the fusion blanket, beryllium pebble will be used as neutron multiplier. From points of temperature control of tritium breeder region, etc., it is important to estimate the thermal irradiation behavior in beryllium pebble bed [1-8]. Therefore, preliminary irradiation test on thermal irradiation behavior in beryllium pebble bed was started with Japan Materials Testing Reactor (JMTR).

Fig. 1 Technical flow of investigation of radial temperature distribution in Be pebble bed

2. Experiment

The technical flow of investigation of radial temperature distribution in Be pebble bed is shown in Fig.1. As first step, Be pebbles and irradiation capsule were fabricated, and neutron irradiation test was started. Finally, thermal irradiation behavior in Be pebble bed will be investigated, and thermal analysis method in-pile functional test of fusion blanket will be establishment.

The diameter and density of beryllium pebble are 1mm and about 100%T.D., respectively. This pebble was fabricated by Rotating Electrode Method [9]. The irradiation capsule with Be pebble is shown in Fig.2. The packing density of beryllium pebble is about 63% and beryllium pebble bed is packed only by 1mm pebbles [10]. The diameter and height of beryllium pebble bed were 30mm and 80mm, respectively. The irradiation hole in JMTR for irradiation capsule is shown in Fig.3.
3. Results

The measurement results of temperature distribution in Be pebble packing region is shown in Fig.4. The neutron irradiation data of radial temperature distribution in Be pebble bed was obtained under neutron irradiation.

\[ \Delta t = -32^\circ C \text{ at } 0.3 \text{ W/g (heating rate) in } \phi 30\text{mm pebble bed} \]

The 3-dimensional nuclear and thermal calculation will be carried out for comparison between measured value and calculated value. And, finally, thermal analysis method for in-pile functional test of fusion blanket will be established.

Table 1  The neutron irradiation conditions

<table>
<thead>
<tr>
<th>Reactor</th>
<th>JMTR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Irradiation hole</td>
<td>1-3</td>
</tr>
<tr>
<td>Neutron flux</td>
<td></td>
</tr>
<tr>
<td>Thermal, ( \phi_t (&lt;0.683\text{eV}) )</td>
<td>( 3 \times 10^{13} \text{n/cm}^2/\text{s} )</td>
</tr>
<tr>
<td>Fast, ( \phi_f (&gt;1\text{MeV}) )</td>
<td>( 4 \times 10^{12} \text{n/cm}^2/\text{s} )</td>
</tr>
<tr>
<td>Neutron fluence</td>
<td></td>
</tr>
<tr>
<td>Thermal, ( \phi_t (&lt;0.683\text{eV}) )</td>
<td>( 1.3 \times 10^{20} \text{n/cm}^2 )</td>
</tr>
<tr>
<td>Fast, ( \phi_f (&gt;1\text{MeV}) )</td>
<td>( 1.5 \times 10^{19} \text{n/cm}^2 )</td>
</tr>
<tr>
<td>Irradiation temperature</td>
<td>200-300°C</td>
</tr>
</tbody>
</table>

Fig.3 The irradiation hole in JMTR for irradiation capsule

Fig.4 The temperature distribution in Be pebble packing region

References
10.3 Beryllium Coating Produced by Evaporation-Condensation Method and Some Their Properties

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Abstract

The method of vacuum evaporation-condensation for deposition of beryllium coatings on metal substrates, considered in the paper, side by side with a plasma-spray method is attractive for ITER application. In particular this technique may be useful for repair the surface of eroded tiles which operate in a strong magnetic field. The possibility of deposition of beryllium coatings with the rate of layer growth 0.1-0.2 mm/h is shown. The compatibility of beryllium coating with copper or stainless steel substrate is provided due to intermediate barrier.

The results of examination of microstructure, microhardness, porosity, thermal and physical properties and stability under thermal cycling of beryllium materials are presented. The value of thermal expansion coefficient and thermal conductivity of condensed beryllium are approximately the same as for industrial grade material produced by powder metallurgy technique. However, the condensed beryllium has higher purity (up to 99.9-99.99 % wt.).

1. INTRODUCTION

Beryllium due to its physical, chemical, thermal and mechanical properties is considered as lining material with good prospects for lining of working surfaces of components, including first wall of thermonuclear fusion reactor.

Deposition of beryllium coating presents doubtless interest among various possible approaches to the solution of lining problem. That is why plasma spray method for beryllium deposition intensively investigated during the last years, e.g. [1,2].

Taking into account, on the one hand, the fact that lining of ITER components requires rather thick coating (several mm), and on the other hand - that deposition process for healing of defects, appearing during reactor service, should be conducted in a strong magnetic field, in the present paper the results of investigation of deposition of beryllium coating with use of the evaporation-condensation method in vacuum are considered.

2. EXPERIMENTAL TECHNIQUE

Investigation of process of beryllium evaporation and its further condensation on a metal substrate was conducted with use of experimental facility (Fig. 1). Scrap and waste products of TSP-grade beryllium were used as an initial material.

Process of evaporation was conducted with use of beryllium oxide crucible loaded by 500-600 g of beryllium. Before the beginning of evaporation the facility was evacuated up to residual pressure not greater than 1 \times 10^{-4} \text{torr}, and during evaporation it was 5 \times 10^{-4} - 5 \times 10^{-5} due to high gettering properties of beryllium.

Various metals (beryllium, copper, stainless steel, vanadium) were used as a substrate material in dependence of the objectives of experiments. As a rule, for deposition of beryllium coatings on metal substrates the latter were made of stainless steel, beryllium, and copper alloys. The substrate prepared from powder beryllium was used for preparation of condensed beryllium plates.
3. RESULTS AND DISCUSSION

Choice of Material of Intermediate Layer Between Coating and Substrate

Preliminary investigations have shown that the quality of beryllium coating prepared by evaporation-condensation method depends strongly on the substrate temperature. In our experiments, temperature of a substrate was chosen 550-650 °C. At lower temperatures, the coating was friable, and at higher - it had coarse-grained structure, passing into separately growing dendrites. In addition, in a temperature range of 550-650 °C interaction of beryllium with a substrate material was observed with formation of intermetallic phases due to high beryllium chemical activity. Investigations were conducted to choose a material of an intermediate layer to eliminate or to decrease to a minimum this chemical process. Some results of estimation of beryllium phases thickness formed at 500-900 °C during 10,000 hours are presented in Table 1.

Presented results show that in the temperature range of 500-600 °C beryllium has a better compatibility with yttrium, titanium, and tantalum, and in the temperature range of 800-900 °C - with vanadium and tantalum. Among substrate materials which can be used in ITER (stainless steel, vanadium, copper) only vanadium and stainless steel are comparatively compatible with beryllium in the temperature range of 550-650 °C. Copper actively reacts with beryllium with formation of three beryllide phases. Therefore, an intermediate layer is necessary to prevent interaction between copper substrate and beryllium coating.

Beryllium coatings 1-2 mm in thickness were deposited on substrates made of stainless steel, vanadium, and chromium (better to say chromium layer 70 μm in thickness deposited on copper and stainless steel) to check experimentally. Duration of deposition process was 8-10 h at the substrate temperature 600 °C. Microstructure of specimens cross section is presented in Fig. 2.
Table 1. Beryllide phases thickness in Be-Me systems formed at 500-900 °C after 10 000 hours

<table>
<thead>
<tr>
<th>Be-Me Systems</th>
<th>Temperature, °C</th>
<th>500</th>
<th>600</th>
<th>700</th>
<th>800</th>
<th>900</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Be-Y</td>
<td>3</td>
<td>29</td>
<td>169</td>
<td>706</td>
<td>2259</td>
<td></td>
</tr>
<tr>
<td>2. Be-Ti</td>
<td>4</td>
<td>34</td>
<td>200</td>
<td>837</td>
<td>2732</td>
<td></td>
</tr>
<tr>
<td>3. Be-Zr</td>
<td>380</td>
<td>1400</td>
<td>4100</td>
<td>9400</td>
<td>20000</td>
<td></td>
</tr>
<tr>
<td>4. Be-V</td>
<td>59</td>
<td>100</td>
<td>163</td>
<td>270</td>
<td>454</td>
<td></td>
</tr>
<tr>
<td>5. Be-Nb</td>
<td>52</td>
<td>137</td>
<td>323</td>
<td>678</td>
<td>1308</td>
<td></td>
</tr>
<tr>
<td>6. Be-Ta</td>
<td>7</td>
<td>27</td>
<td>92</td>
<td>274</td>
<td>726</td>
<td></td>
</tr>
<tr>
<td>7. Be-Mo</td>
<td>31</td>
<td>100</td>
<td>266</td>
<td>625</td>
<td>1420</td>
<td></td>
</tr>
<tr>
<td>8. Be-Fe</td>
<td>41</td>
<td>142</td>
<td>412</td>
<td>1082</td>
<td>2371</td>
<td></td>
</tr>
<tr>
<td>9. Be-Ni</td>
<td>50</td>
<td>217</td>
<td>700</td>
<td>1835</td>
<td>4056</td>
<td></td>
</tr>
<tr>
<td>10. Be-Cr</td>
<td>94</td>
<td>170</td>
<td>296</td>
<td>491</td>
<td>779</td>
<td></td>
</tr>
</tbody>
</table>

Fig. 2. Microstructure of boundary between beryllium coating and substrate of various materials
a) - stainless steel; b) - vanadium; c) - chromium (on copper and stainless steel); d) - copper with chromium coating (x100).
Comparison of microstructure at the boundary beryllium coating-substrate showed that interaction layer in realized conditions of deposition was not found only in a case of beryllium-chromium couple. Therefore chromium layer with thickness up to 70 μm was used as an intermediate layer during manufacturing of specimens with beryllium coating ensuring compatibility of beryllium coating with copper and stainless steel substrates. During deposition of beryllium coating on a beryllium substrate intermediate chromium layer was not used.

To improve coupling of beryllium coating with a substrate in a number of cases an artificial relief was formed by milling of a substrate surface or by welding of copper or steel grid.

Some thermophysical properties of beryllium coating, its purity, microstructure and stability during thermal cycles were studied in the present work. The total content of 15 typical impurities in a beryllium layer, including oxygen and carbon, did not exceed 0.1 % wt.

Deposited beryllium is characterized by coarse-grained (50-100 μm) columnar structure. Microhardness of condensed beryllium at room temperature is in the range of 220-330 kg/mm². Microporosity of a beryllium layer (with pore sizes not greater than 30 μm) were determined by a Hg-porometry method and was not greater than 0.9-1.2 % rel.

Coefficient of thermal expansion (CTE) of beryllium was measured with use of quartz dilatometer EAD equipped by the inductive sensor of movement of model 214, Kalibr. Measurement error of CTE value is 10 % at temperatures <300 °C; in the temperature range of 300-800 °C it does not exceed 5 %.

The results of measurements of CTE of specimens of condensed and powder TSP-grade beryllium are presented in table 2. The level of anisotropy of condensed and industrial (powder) beryllium was estimated comparing CTE measurements in two directions in relation to a surface of condensation for the first material and in the direction of powder compaction for the second one.

The condensed metal has higher CTE values and is a material with a more expressed anisotropy of thermal expansion in comparison with industrial TSP-grade beryllium. Minimum CTE values of condensed beryllium are closest to the data [3] of cast and extruded metal. CTE of condensed beryllium is close to CTE of stainless steel and copper, that facilitates production of beryllium coatings with a necessary level of resistance to thermal cycles.

**Thermal conductivity of beryllium coating**. It is reasonable to introduce such definitions as thermal conductivity and thermal resistance of coating, determined by the following relations:

\[ \lambda_{coat} = \frac{h_{Be}}{\xi + \frac{h_{Be}}{\lambda_{Be}}} \]  

(1)

\[ R_{coat} = \xi + \frac{h_{Be}}{\lambda_{Be}} \]  

(2)

Thus the coating is considered as unified heat object consisting of beryllium layer itself and interface at the boundary of coating-substrate (contact layer). Thermal conductivity and thermal resistance of coating were calculated using experimental values of thermal conductivity of specimens and substrate material:

\[ \lambda_{coat} = \frac{h_{Be}}{h_{spc} \frac{h_{sub}}{\lambda_{spc} \frac{h_{sub}}{\lambda_{sub}}}} \]  

(3)

\[ R_{coat} = \frac{h_{spc}}{\lambda_{spc} \lambda_{sub}} \]  

(4)

Where \( h_{spc}, h_{sub}, \lambda_{spc}, \lambda_{sub} \), \( \lambda_{Be}, \lambda_{coat} \) are thickness and thermal conductivity of a sample (coating+substrate), substrate, beryllium layer and coating (beryllium layer+contact layer), respectively.

\( \xi, R_{coat}, R_{spc} \) are thermal resistance of contact between beryllium and substrate, thermal resistance of coating and complete thermal resistance of a sample (substrate+coating) respectively.
Samples for measurements of thermal conductivity differentiated one from another by their sizes, material compositions and technology of preparation of a substrate surface (Table 3). Thermal conductivity of samples was measured on installation IT-200 by a relative stationary axial heat flow method in the temperature range of 50-400 °C in vacuum. Measurement error of thermal conductivity decreases with temperature increase and is ±30 % and ±15 % at 50 °C and 460 °C respectively.

The data on thermal conductivity and thermal resistance of coatings are presented in Table 4. To estimate thermal conductivity of condensed beryllium in coating and relative contribution of an intermediate contact layer to thermal resistance of a sample, thermal conductivity of "substrate-coating" system in a sample No.5 was measured in addition on a site 4.1 mm in thickness, including part of a substrate and coating thicknesses, as well as boundary between them.

Thermal conductivity of beryllium in coating (without contact layer) calculated on the basis of obtained experimental data was 120 W/m.K. This value is not less than 70 % of thermal conductivity of hot pressed powder-grade beryllium at 350 °C [4].

Table 2. CTE of condensed and TSP-grade industrial beryllium

<table>
<thead>
<tr>
<th>Interval of temperatures, °C</th>
<th>Average CTE. 10⁻⁶ K⁻¹ (parallel/perpendicular)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Condensed beryllium</td>
</tr>
<tr>
<td></td>
<td>TSP-grade industrial Be</td>
</tr>
<tr>
<td>20 - 100</td>
<td>10.2/13.0</td>
</tr>
<tr>
<td>20 - 200</td>
<td>12.2/14.0</td>
</tr>
<tr>
<td>20 - 300</td>
<td>13.6/16.0</td>
</tr>
<tr>
<td>20 - 400</td>
<td>14.6/16.8</td>
</tr>
<tr>
<td>20 - 500</td>
<td>15.5/17.6</td>
</tr>
<tr>
<td>20 - 600</td>
<td>16.2/18.2</td>
</tr>
<tr>
<td>20 - 700</td>
<td>16.8/18.6</td>
</tr>
<tr>
<td>20 - 800</td>
<td>17.4/19.2</td>
</tr>
</tbody>
</table>

Table 3. Characteristic of samples for thermal conductivity measurements.

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Substrate characteristics</th>
<th>Preparation of substrate surface</th>
<th>Thickness, mm</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Sample</td>
<td>Coating</td>
</tr>
<tr>
<td>1.</td>
<td>Steel 12X18N10T</td>
<td>Milling of grooves for relief</td>
<td>7.2</td>
<td>2.7</td>
</tr>
<tr>
<td>2.</td>
<td>Substrate thickness 4.5 mm</td>
<td></td>
<td>10.3</td>
<td>5.8</td>
</tr>
<tr>
<td>3.</td>
<td>Welding of a steel grid to a substrate</td>
<td></td>
<td>12.7</td>
<td>8.2</td>
</tr>
<tr>
<td>4.</td>
<td>Copper M1</td>
<td>Welding of a steel grid to a substrate</td>
<td>10.2</td>
<td>5.7</td>
</tr>
<tr>
<td>5.</td>
<td>Substrate thickness 4.0 mm</td>
<td></td>
<td>10.5</td>
<td>6.5</td>
</tr>
<tr>
<td>6.</td>
<td>Welding of a copper grid to a substrate</td>
<td></td>
<td>9.0</td>
<td>5.0</td>
</tr>
<tr>
<td>7.</td>
<td></td>
<td></td>
<td>12.8</td>
<td>8.8</td>
</tr>
<tr>
<td>8.</td>
<td></td>
<td></td>
<td>10.8</td>
<td>6.8</td>
</tr>
</tbody>
</table>

* Note. TMP means that after coating deposition thermomechanical treatment of a sample was carried out. The used measurement technique has not allowed to reveal dependence of thermal conductivity of condensed beryllium on temperature in the temperature range of 50-400 °C.

Analysis of data presented in Table 4 on the basis of measured level of thermal conductivity of condensed beryllium leads to conclusion that value of thermal resistance of an intermediate contact layer is comparable with those of the beryllium layer on a copper substrate and can essentially exceed it, if substrate is steel. Thermomechanical processing (TMP) of specimens with coating essentially (up to 1.5 time) decreases contact thermal resistance.

Stability of beryllium coating during thermal cycling was investigated in vacuum in the temperature range of 25-650 °C. Heating (cooling) rate of specimens was 25-30 °C/min. Samples with porous coating, as well as with coating deposited on substrates of copper and stainless steel with thickness 0.05-8 mm with artificial relief were tested during thermal cycling. After 108 cycles in the range from room temperature to 650 °C delaminations and any destruction of coatings was not found out.

As the conclusion we should like to note that condensed beryllium does not essentially differ from industrial-grade beryllium from the point of view of investigated properties, however, it is characterized by
higher purity (up to 99.9% wt. Be). Deposition of beryllium coatings by evaporation-condensation method in vacuum is effective especially with reference to application of details of complicated shape, as well as alternative of repair technology for redressing of sites of coating degraded during operation.

Table 4. Thermal conductivity and thermal resistance of coatings (with a joined contact layer).

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Thermal conductivity; W/m. K</th>
<th>Thermal resistance, cm² K/W</th>
<th>Temperature, °C</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>50</td>
<td>100</td>
<td>150</td>
</tr>
<tr>
<td>1.</td>
<td>1.05</td>
<td>2.06</td>
<td>3.44</td>
</tr>
<tr>
<td>2.</td>
<td>5.9</td>
<td>7.8</td>
<td>9.9</td>
</tr>
<tr>
<td>3.</td>
<td>22.6</td>
<td>28.6</td>
<td>32.0</td>
</tr>
<tr>
<td>4.</td>
<td>3.62</td>
<td>2.87</td>
<td>2.56</td>
</tr>
<tr>
<td>5.</td>
<td>78.5</td>
<td>79.8</td>
<td>82.0</td>
</tr>
<tr>
<td>6.</td>
<td>0.828</td>
<td>0.814</td>
<td>0.792</td>
</tr>
<tr>
<td>7.</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>8.</td>
<td>112</td>
<td>117</td>
<td>120</td>
</tr>
<tr>
<td></td>
<td>0.605</td>
<td>0.583</td>
<td>0.566</td>
</tr>
</tbody>
</table>

Notes: Coating of sample No 4 has high thermal conductivity (>80 W/m K) and thermal resistance which is negligible in comparison with thermal resistance of substrate. Exact evaluation of these characteristics was impossible because of insufficient accuracy of experimental data.

REFERENCES

11. Summary

The Third International Energy Agency International Workshop on Beryllium Technology for Fusion was held on October 22-24, 1997, at Sangyou Kaikan in Mito City. 68 participants, who attended from the Europe (18), the Russian Federation (9), the Kazakstan (4), the United States (7) and Japan (30) took part in the workshop. Cochair of the workshop were H.Kawamura [Japan Atomic Energy Research Institute (JAERI)], G.R.Longhurst [Idaho National Engineering and Environmental Laboratory (INEEL)], M.Dalle Donne [Forschungzentrum Karlsruhe (FZK)] and V. Shestakov [Science Technical Center of Controlled Thermonuclear Fusion "CTF"]. This is the third workshop in this area conducted under the auspices of the International Energy Agency (IEA). The first was held at Karlsruhe, Germany, in October of 1993. The second was held at Jackson Lake Lodge, the United States, in September of 1995. Prior to IEA sponsorship, similar workshops were held at the INEEL in 1988 and at Clearwater Beach, Florida, in 1991.

The workshop was divided into ten technical sessions. Technical sessions addressed the general topics of (a) Beryllium applications for ITER; (b) Production and characterization; (c) Chemical compatibility and corrosion; (d) Forming and joining; (e) Plasma/tritium interactions; (f) Beryllium coating; (g) First wall applications; (h) Neutron irradiation effects; (i) Health and safety issues; and (j) Hot news. Highlights of the various sessions follow.

In the Beryllium applications for ITER session, V. Barabash presented the status of the design of the ITER Plasma Facing Components. The general understanding of the Be behavior as plasma facing material has very much improved. Issues still to be investigated at a later stage, and additional R&D needs were discussed. S.Tanaka presented the "ITER Materials R&D Data Bank". As for thermo-mechanical properties of Be, tensile properties of unirradiated and neutron irradiated S65C, S-200 and so on are available in the data bank. Tensile properties, shear strength and bending strength of various kinds of Be/Cu joints are available in the data bank.

In the Production and characterization session, M.Dalle Donne presented experiments to determine the thermal conductivity and the heat transfer coefficient between the pebble bed and the containing walls. The experiments allowed researchers to determine the effects of the bed temperature, of the interference between bed and containing walls and of the pressure of the helium flow. A.M. Khomutov reported that there is a simple linear dependence between the magnitude of residual thermal microstresses and the ultimate tensile strength. In the framework of the concept under development one could explain formerly not fully understandable effects, which were characteristic of polycrystalline beryllium such as the unexpected course of the Petch-Stro curve, the role of twinning etc.. T. Iwadachi presented some production methods of beryllium pebbles proposed as the neutron multiplier for the pebble-bed-type tritium breeding blanket. Production of each size of beryllium pebbles by the rotating electrode method and some properties were discussed in detail. D.E.Dombrowski presented mechanical properties of S65-C grade beryllium at elevated temperature. The effect of strain rate was examined. E.M.Dvinskykh reported data on beryllium production at the Ulba metallurgical plant. Beryllium production was showed to have extended technological opportunities in manufacturing semi-products and in production of structural beryllium and parts. The plant had a satisfactory experience in solving ecological problems. Dvinskykh also presented processes used for sanitary-hygienic and ecological control of beryllium production at the...
Ulba metallurgical plant. Problems on designing devices for a rapid analysis of beryllium in air were briefly discussed. Effects of beryllium production on the ecological situation in the town were analyzed.

In the Chemical compatibility and corrosion session, A.M.Chekhonadskich reported chemical beryllium-steam interaction studies to validate computer codes for fusion applications. Currently, experiments simulating LOCA-type accidents in minor scale enable the ITER Joint Central Team to forecast accident consequences based on experimental data and dependencies of temperature variation for chemical beryllium-water steam interaction. A.M.Zimin reported erosion resistance of beryllium for armour. Erosion characteristics, the dispersive composition of the eroded particles, some structural changes and redeposition of the sputtered atoms upon different elements of the chamber were determined. R.A.Anderl presented the influence of neutron irradiation-induced swelling and porosity on the chemical reactivity of Be specimens irradiated in the EBR-II to fast neutron fluence of $5 \times 10^{27}$ n/cm$^2$ at 400°C. Initial experiments for unirradiated Be specimens and annealed, irradiated Be specimens indicated that for specimens annealed above 600°C, steam/Be chemical reactivity is significantly enhanced because of the development of a surface-connected porosity in the annealed, irradiated specimens.

In the Forming and joining session, E.Abramov presented the characterization of welded beryllium specimens. The Fused Zone (FZ) has the typical microstructure of a pure component eutectic, i.e., very fine dispersion of primary beryllium and continuos aluminum phases. Aluminum from the FZ extends along grain boundaries into the base metal Heat Affected Zone (HAZ) to form "aluminum stringers". The FZ and HAZ were free from porosity and inclusions. F.Muecklica presented first principle investigations for formation of Be-Cu gradients in the millimeter scale. For technology of production, segregation mechanisms were examined by stimulated powder rearrangement in the gravitational field. It is conceivable to determine the later chemical gradient completely via the characteristics of this precursor powder. The entire microstructure formation of course was driven by the sintering process. During sintering, phase formations in the Be-Cu system played an essential role. T.Makino presented diffusion bonding tests of beryllium with dispersion strengthened copper (Be/DSCu) with various interlayers for the selection of the suitable interlayer materials. The bonding of Be/DSCu was achieved by hot pressing with an interlayer of Al, Ni, Nb, Ti, Zr and Be-Cu alloy. As a result of the shear strength tests, Be/DSCu joints showed strength of about 200 MPa. By using Be-Cu alloy interlayer, Be/DSCu diffusion bonding tests at temperatures from 600°C to 850°C have been conducted to identify the effect of bonding temperature on interface formation and strength. The shear strength at room temperature does not depend on the thickness of the BeCu layer but on the formation of Be$_2$Cu layer. R.Giniatulin presented the results of metallographic analysis of Be/Cu joints, i.e. divertor mockups that have beryllium tiles with different dimensions of castellation and non-flat brazing profiles with copper alloy heat sink. The description of mockups design, the special geometry of armour tiles that increases joint strength, and temperature and stress calculations were presented. The results of thermocyclic tests screening of these mockups were presented. B.C. Odegard presented a bonding technique for joining beryllium armor tile to copper base heat sink for fusion reactor applications. Several different processing schedules were evaluated during the course of this study. Bonding assemblies were produced with 100% joint efficiency. D.E.Dombrowski presented bonding Be to Cu alloys using a proprietary Brush Wellman process. The process is compatible with current manufacturing capabilities and can be readily scaled up from lab scale to production scale. Previously published data indicated bond
strengths useful for ITER applications. Present results show the effect of process variations, and the effect of bond cycle on copper substrate strength.

In the Plasma/tritium interactions session, W.Eckstein presented erosion of Be and deposition of C and O due to bombardment with C* and CO*. Bombardment of Be with CO* and C* results in a complicated behavior of composition profiles, sputtering, and reflection with the bombardment fluence. Experimentally, the monoenergetic bombardment was performed with C* and CO* ions at normal incidence to study the deposition with different incident C* and CO* fluences and energies. A.V.Fedorov presented thermal desorption of deuterium from Be and Be with helium bubbles. Deuterium desorption measurements carried out on a single-crystalline beryllium sample were presented. Results of deuterium interaction with helium bubbles at various stages of evolution during thermal annealing of 30-keV helium implanted beryllium were presented. Y.V.Chikhray presented experimental studies and modeling of processes of hydrogen isotopes interactions with beryllium. Analysis of literature data showed hydrogen interaction parameters' dependence on beryllium surface condition including oxide film. The goal of this work was to investigate the influence of in-pile irradiation and plasma glow discharge on surface beryllium oxide and on hydrogen-beryllium interaction characteristics. A.V.Markin presented the degassing of beryllium loaded with deuterium by heating in D$_2$ atmosphere. It has been found that thin oxide layers grown on the surface of beryllium samples under heating in D$_2$ retain a great amount of deuterium. Therefore, in all experiments, degassing rate did not depend on the diffusion of deuterium in beryllium. Samples of larger sizes and ion sputtering of the surface oxide are currently used with the aim of obtaining the diffusion-limited regime of degassing. V.M.Sharapov presented the deuterium accumulation in TIP-30 beryllium under atomic deuterium exposure at 740K. It was observed that deposited tungsten influences deuterium accumulation. When the tungsten amount does not exceed (3-4)x10$^{15}$ at/cm$^2$, the deuterium content increases depending on the exposure time, and the amount of tungsten does not influence accumulation. A tungsten layer decreases the rate of beryllium oxide film growth. G.R.Longhurst discussed an improved mathematical model for interactions of hydrogen plasmas with beryllium surfaces that accounts for the saturation behavior observed in experiments. Reviews of various results and modeling of recent TPE experiments confirmed that surface layers develop high porosity with the result that implanting ions and atoms are readily returned to the plasma once saturation has been achieved.

In the Beryllium coating session, A.M.Zimin presented the technological aspects of the Be-coating on various materials investigated with the experimental facility "MAGRAS". This facility allows studying Be-sputtering under intense bombardment of its surface by hydrogen ions. Be-films possessing good adhesion to backings of beryllium, copper, steel, glass, paper, lavsan, were produced. In a number of cases, the films produced in such a way possess unique properties. Zimin also described sputtered Be-layers produced by bombardment with low energy hydrogen-ions. As a result of the beryllium cathode etching by ions, the beryllium grains were manifested differently. In some of them the bubbles within the collapsed shells were seen. At the grain boundaries the production of pores and cracks was observed. R.G.Castro presented characterization of microstructure, thermal and mechanical properties of the beryllium plasma sprayed mockups. The results of high heat flux (HHF) testing of the beryllium plasma sprayed mockups at 5 MW/m$^2$, which resulted in cracking within the beryllium plasma sprayed armor, were reported. The crack initiation sites could be a result of unmelted beryllium particles within the armor.
In the First wall applications session, M.Rödig described thermal shock tests on small scale test coupons and on actively cooled divertor modules. Single and multiple shot experiments have been performed on different bulk beryllium grades and on plasma sprayed coatings. Detailed in-situ diagnostics and post mortem analyses have been applied to quantify the resulting material damage. Rödig also presented thermal cycling tests of actively cooled beryllium copper joints. Mock-ups produced by brazing with silver and silver-free braze materials, hot isostatic pressing, diffusion bonding at low temperatures were tested. Brazing with and without silver seems to be the best solution. Hot isostatically pressed (HIPed) and diffusion bonded Be/Cu modules have not yet reached the standards for application in divertor components.

K.Ashida presented changes in the co-existing system of Be and C with vacuum heating with XPS, SIMS, XRD and TDS. Be,C was formed by vacuum heating above 600°C. A large amount of molecule hydrogen was desorbed from the carbon film. Be-carbide has little ability to capture/trap hydrogen isotope atoms in it. S.N.Korshunov presented energy dependencies of sputtering yields of Be, Be-C and Be-W films by Be+-ions. The experimental values of the sputtering yield by Be+-ions on Be, Be-C, Be-W redeposited films were near by those for TShP-type Be and near calculated data. I.Mazul presented in-pile thermocycling testing and post-test analysis of beryllium divertor mockups. Two mockups with different beryllium grades were mounted face to face, and a tantalum heater was placed between them. Metallographic analyses were performed to investigate the beryllium and beryllium/copper joint structures.

In the Neutron irradiation effects session, C.H. Wu presented the influence of neutron irradiation on tritium retention in beryllium. For carbon-based materials tritium retention increased with neutron damage in the range ≤0.1 dpa by one to three orders of magnitude. Tritium traps were generated by fast neutrons, and retained tritium in irradiated and unirradiated samples was higher than in carbon-based materials.

F.Scaffidi-Argentina presented mechanical performance of irradiated beryllium pebbles. The microstructure, the porosity distribution, the impurity content, the behavior under compression loads and the compatibility with lithium orthosilicate during the in-pile irradiation were presented and critically discussed. F.Moons presented mechanical data on four different neutron irradiated beryllium species. Tensile tests results and fracture toughness measurements were discussed. F.Scaffidi-Argentina presented tritium release from neutron irradiated beryllium pebbles and the present status of beryllium post-irradiation examinations. Tritium release by chemical traps and/or physical traps was discussed. The consequences of direct contact between beryllium and ceramics during irradiation, causing tritium implanting in a surface layer of beryllium and leading to an additional inventory were presented. E. Ishitsuka presented microstructure and mechanical properties of beryllium irradiated in JMTR. Obvious decrease of the fracture stress was observed for high-temperature irradiated and small grain-size specimens, which was caused by helium bubbles on the grain boundaries. I.B.Kupriyanov presented mechanical properties, swelling and microstructure of six neutron irradiated beryllium grades. All the samples were irradiated in the active zone of the SM-3 reactor. Beryllium grades having the smallest grain size and highest beryllium oxide content demonstrated minimal swelling, which was no more than 0.2 % at 700°C and a fluence of 5.5×10¹¹ n/cm². A.M.Khomutov presented microstructure changes accompanying helium swelling of Be irradiated at 450°C or α-particle implanted in a cyclotron at 100°C to reach the same volume accumulation of He. It seems feasible to produce Be with the initial structure promoting the complete absorption of helium into manufacturing pores under in-pile conditions. S.A.Fabritsiev presented neutron
irradiation and helium effect on structure and mechanical properties of various types of beryllium. Neutron irradiation was in the BOR-60 reactor at 350, 500 and 750°C up to a fluence of $7 \times 10^{17} \text{n/cm}^2$. An increase in the irradiation and testing temperature to 700°C resulted in an abrupt embrittlement of all irradiated samples. D.S. Gelles presented the behavior of beryllium pebbles irradiated to approximately $4 \times 10^{12} \text{n/cm}^2$ ($E > 0.1 \text{ MeV}$) at 380°C. Post-irradiation results were presented on density change measurements, tritium and helium assay, stepped-anneal, and thermal-ramp measurements. R.A. Anderl presented the results of experiments to measure (1) tritium and helium retention and release behavior and (2) volumetric swelling for fully-dense Be specimens irradiated in the EBR-II. Concurrent release of tritium and helium was observed for temperatures of 800°C and higher. Volumetric swelling results were derived from liquid-immersion density measurements for unirradiated, irradiated, and annealed/irradiated specimens.

In the Health and safety issues session, N.Z. Bitkolov presented protection of air in working premises and in the environment against beryllium aerosols. Techniques for effective protection at industrial plants and the prevention of air environment pollution were discussed. A.M. Khomutov (for D.A. Davydov) presented study of the chemical interaction between the beryllium powders of different particle sizes and the air in the temperature range 500-1000°C from the viewpoint of ITER safety. The temperature dependence of beryllium powders interaction with air were studied. V.V. Savchuk presented results of 45-years of medical research in beryllium production at the Ulba metallurgical plant. Issues connected with beryllium production effects on health of people, located near beryllium production facilities were discussed.

In the Hot news session, F. Saint-Antonin presented joining of beryllium onto copper alloy by hot isostatic press bonding (HIPB) and the structure of joints performed with the help of HIPB at temperature higher than 800°C. The influence of the HIP cycle pressure and temperature on the properties of the different joints have been systematically studied. H. Kawamura presented the preliminary irradiation test on thermal irradiation behavior in a beryllium pebble bed tested in JMTR. A part of the analysis results for thermal irradiation behavior of the beryllium pebble bed were presented. A. Chernikov presented the method of vacuum evaporation-condensation for deposition of beryllium coating on metal substrates. The compatibility of beryllium coating with copper or stainless steel substrate, microstructure, microhardness, porosity, thermal and physical properties and stability under thermal cycling of beryllium materials were presented.

In summary, it was recognized that swelling, He/T release of beryllium pebble irradiated to high He content, effective thermal conductivity of pebble beds, tritium permeation, and coating and fabrication costs of pebble were important for the pebble beds type blankets. For plasma facing components, life time estimation of beryllium and beryllium coating due to erosion, dust and particle production, joining, waste, mechanical properties and deformation by swelling are recognized as significant issues by those in attendance. Especially, it was recognized that comparative evaluation by another researchers was important because the reliability of data is increased. Therefore, as a first step of collaboration, comparative evaluation of beryllium pebbles was proposed as a world wide collaboration by the workshop International Organizing Committee, and accepted by all participants. Methods of collaboration, etc. will be presented by Organizing Committee in near future. It was also decided that the next workshop on beryllium technology will be held in Europe in 1999.
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T. Terai
Appendix I  Program of the Third IEA International Workshop on Beryllium Technology for Fusion

WEDNESDAY, OCTOBER 22

<table>
<thead>
<tr>
<th>Time</th>
<th>Session</th>
<th>Speaker(s)</th>
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<tbody>
<tr>
<td>8:30</td>
<td>WELCOME</td>
<td>M. Okamoto, H. Kawamura</td>
</tr>
<tr>
<td>8:35</td>
<td>OPENING</td>
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<tr>
<td>8:45</td>
<td>[A] BERYLLIUM APPLICATIONS FOR ITER</td>
<td></td>
</tr>
<tr>
<td>9:55</td>
<td>(BREAK)</td>
<td></td>
</tr>
<tr>
<td>9:25</td>
<td>[B] PRODUCTION AND CHARACTERIZATION (1)</td>
<td></td>
</tr>
<tr>
<td>11:10</td>
<td>COMMEMORATIVE PHOTOGRAPH</td>
<td></td>
</tr>
<tr>
<td>11:30</td>
<td>LUNCH</td>
<td></td>
</tr>
<tr>
<td>13:00</td>
<td>[C] PRODUCTION AND CHARACTERIZATION (2)</td>
<td></td>
</tr>
<tr>
<td>13:25</td>
<td>[C2] Beryllium Production at Ulba Metallurgical Plant</td>
<td>E. M. Dvinskykh, V. V. Savoluk, Y. V. Tuzov</td>
</tr>
<tr>
<td>14:15</td>
<td>[D] CHEMICAL COMPATIBILITY AND CORROSION</td>
<td></td>
</tr>
<tr>
<td>15:05</td>
<td>[D2] On Erosion Resistance Test Technology of Be-Armor for ITER</td>
<td>T. Makino, T. Iwadachi</td>
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<tr>
<td>15:05</td>
<td>(BREAK)</td>
<td></td>
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<tr>
<td>16:05</td>
<td>[E] FORMING AND JOINING (1)</td>
<td></td>
</tr>
<tr>
<td>17:20</td>
<td>[E3] Interface Structure of Be/DSCu Diffusion Bonding</td>
<td>T. Makino, T. Iwadachi</td>
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<tr>
<td>16:55</td>
<td>[D] CHEMICAL COMPATIBILITY AND CORROSION</td>
<td></td>
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<tr>
<td>16:55</td>
<td>[D3] Chemical Reactivity for Irradiated beryllium</td>
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**THURSDAY, OCTOBER 23**

<table>
<thead>
<tr>
<th>Time</th>
<th>Event</th>
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<tbody>
<tr>
<td>8:30</td>
<td>[G1] Beryllium-Copper Diffusion Bonding for an ITER First Wall Application</td>
</tr>
<tr>
<td>8:55</td>
<td>[G2] Bonding Be to Cu Alloys Using a Proprietary Brush Wellman Process</td>
</tr>
<tr>
<td>9:00</td>
<td>(BREAK)</td>
</tr>
<tr>
<td>9:20</td>
<td>[H1] Beryllium Coating Application by Sputtering in a Magnetron</td>
</tr>
<tr>
<td>9:35</td>
<td>[H2] Study of the Sputtered Be-layers Produced by Bombardment with Hydrogen Low-energy Ions</td>
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<tr>
<td>10:00</td>
<td>[H3] Characterization of Plasma Sprayed Beryllium ITER First Wall Mockups</td>
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**HOT NEWS**

<table>
<thead>
<tr>
<th>Time</th>
<th>Event</th>
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<tr>
<td>10:50</td>
<td>[HN1] Development and characterization of Be/Glidcop joint obtained by Hot Isostatic Pressing for high temperature working conditions</td>
</tr>
<tr>
<td>11:00</td>
<td>[HN2] Radial Temperature Distribution in Beryllium Pebble Bed under Neutron Irradiation</td>
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<tr>
<td>11:10</td>
<td>[HN3] Beryllium Coating Produced by Evaporation Condensation Method and Some Their Properties</td>
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<tr>
<td>11:20</td>
<td>(LUNCH)</td>
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</table>

**FIRST WALL APPLICATIONS**

<table>
<thead>
<tr>
<th>Time</th>
<th>Event</th>
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<tbody>
<tr>
<td>12:50</td>
<td>[I1] Response of Beryllium to Severe Thermal Shocks-Simulation of Disruption and Vertical Displacement Events in Future Thermonuclear Devices</td>
</tr>
<tr>
<td>13:15</td>
<td>[I2] Thermal Cycling Tests of Actively Cooled Beryllium Copper Joints</td>
</tr>
<tr>
<td>13:40</td>
<td>[I3] Be,C Formation in Beryllium-Carbon Binary System by Vacuum Heating</td>
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<tr>
<td>14:05</td>
<td>[I4] Energy Dependence of Sputtering Yields of Be, Be-C and Be-W Films by Be- ions</td>
</tr>
<tr>
<td>14:30</td>
<td>[I5] In-pile Thermocycling Testing and Post-test Analysis of Beryllium Divertor Mockups</td>
</tr>
<tr>
<td>15:00</td>
<td>KASAMA TOUR</td>
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<tr>
<td>18:30</td>
<td>RECEPTION</td>
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<tr>
<td>19:00</td>
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</table>
FRIDAY, OCTOBER 24

(J) NEUTRON IRRADIATION EFFECTS (1)

[J1] Influence of Neutron Irradiation on the Tritium Retention in Beryllium
Chairman: G. Longhurst
R. Roll, S. Rubel, H. Werle, C.H. Wu
8:30 - 8:55

[J2] Mechanical Performance of Irradiated Beryllium Pebbles
Chairman: G. Longhurst
F. Scalfiddi, Argentina, M. Dalle Donne, H. Werle
8:55 - 9:20

[J3] Neutron Irradiated Beryllium: Mechanical Strength, Swelling and Microstructure
Chairman: G. Longhurst
F. Moones, R. Chaboudi, J.L. Puzzolante, L. Sannen, A. Rahn, M. Verwerft
9:20 - 9:45

[J4] Tritium Release from Neutron Irradiated Beryllium Pebbles
Chairman: G. Longhurst
F. Scalfiddi, Argentina, H. Werle
9:45 - 10:10

[J5] Microstructure and Mechanical Properties of Neutron Irradiated Beryllium
Chairman: G. Longhurst
E. Ishitsuka, H. Kawamura, T. Terai, S. Tanaka
10:10 - 10:35

(BREAK)

(K) HEALTH AND SAFETY ISSUES

Chairman: V. Barabash

[K1] Protection of Air in Premises and Environment Against Beryllium Aerosols
Chairman: V. Barabash
N. Z. Birkulov, E. P. Vishnevsiki, A. V. Kupchik
10:45 - 11:10

Chairman: V. Barabash
D. A. Davydov, V. A. Gorokhov, Yu. V. Konovalov, V. B. Levin, G. M. Chekhlatov
11:10 - 11:35

[K3] Some Results of Medical Researches at Ulba Metallurgical Plant
Chairman: V. Barabash
G. I. Artemieva, V. G. Novikov, V. V. Savchuk
11:35 - 12:00

(LUNCH)

(L) NEUTRON IRRADIATION EFFECTS (2)

Chairman: T. Terai

[L1] Neutron Irradiated Behavior of ITER Candidate Beryllium Grades
Chairman: T. Terai
I. B. Kuplyansky, V. A. Gorokhov, R. R. Melder, V. P. Chakun, V. S. Mikhailov, G. N. Nikolaev
13:30 - 13:55

[L2] Specific Features of Reactor or Cyclotron α-particle Irradiated Beryllium Microstructure
Chairman: T. Terai
13:55 - 14:20

[L3] The Helium Embrittlement of Beryllium After Neutron Irradiation
Chairman: T. Terai
M. Dalle Donne, D. L. Baldwin, D. S. Geteps, L. R. Greenwood, H. Kawamura, B. M. Oliver, F. Scalfiddi, Argentina
14:20 - 14:45

[L4] Behavior of Beryllium Pebbles under Irradiation
Chairman: T. Terai
M. Dalle Donne, D. L. Baldwin, D. S. Geteps, L. R. Greenwood, H. Kawamura, B. M. Oliver, F. Scalfiddi, Argentina
14:45 - 15:10

[L5] Tritium and Helium Retention and Release from Irradiated Beryllium
Chairman: T. Terai
B. A. Andert, G. R. Longhurst, M. A. Dales, R. J. Pawelko
15:10 - 15:35

(BREAK)

[M] PLASMA / TRITIUM INTERACTIONS (2)

Chairman: K. Ashida

[M1] Degassing Measurements for Beryllium Exposed to Deuterium Atmosphere
Chairman: K. Ashida
A. V. Markin, R. Kh. Zalavudinov, A. E. Gorodetsky, A. P. Zakharov (V. M. Sharapov)
15:45 - 16:10

[M2] Effect of Deposited Tungsten on Deuterium Accumulation in Beryllium in Contact with Atomic Deuterium
Chairman: K. Ashida
V. M. Sharapov, L. E. Gavrilov, V. S. Kuiekushas
16:10 - 16:35

Chairman: K. Ashida
G. R. Longhurst, R. A. Anderl
16:35 - 17:00

SUMMARY

H. Kawamura, M. Dalle Donne
G. Longhurst, V. Sheshtakov
T. Terai
17:00 - 18:00
Appendix II  List of Participants for the Third IEA International Workshop on Beryllium Technology for Fusion

<table>
<thead>
<tr>
<th>NAME</th>
<th>AFFILIATION / ADDRESS</th>
<th>TEL</th>
<th>FAX</th>
<th>e-mail</th>
</tr>
</thead>
<tbody>
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Appendix III  Picture of Participants for the Third IEA International Workshop on Beryllium Technology for Fusion
国際単位系 (SI) と換算表

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