LA-UR- 99-58
CONF-990109--

The Development of Beryllium Plasma Spray Technology for the International Thermonuclear Experimental Reactor (ITER)

R.G. Castro[a], K.E. Elliott[a], A.H. Bartlett[c], K.J. Hollis[a] and R.D. Watson[b]

[a]Los Alamos National Laboratory, Material Science and Technology Division
Los Alamos, NM 87545 USA
[b]Sandia National Laboratory, Fusion Technology Department,
Albuquerque, NM 87185, USA
[c]Norsam Technologies Inc., Los Alamos, NM 87544 USA

Abstract

Over the past five years, four international parties, which include the European Communities, Japan, the Russian Federation and the United States, have been collaborating on the design and development of the International Thermonuclear Experimental Reactor (ITER), the next generation magnetic fusion energy device. During the ITER Engineering Design Activity (EDA), beryllium plasma spray technology was investigated by Los Alamos National Laboratory as a method for fabricating and repairing and the beryllium first wall surface of the ITER tokamak. Significant progress has been made in developing beryllium plasma spraying technology for this application. Information will be presented on the research performed to improve the thermal properties of plasma sprayed beryllium coatings and a method that was developed for cleaning and preparing the surface of beryllium prior to depositing plasma sprayed beryllium coatings. Results of high heat flux testing of the beryllium coatings using electron beam simulated ITER conditions will also be presented.

1.0 Introduction

The International Thermonuclear Experimental Reactor (ITER) represents a major step toward realizing the benefits of magnetic confinement fusion energy. Four International Parties; the European Communities, Japan, the Russian Federation (assuming the role of the former Soviet Union), and the United States are collaborating on the design of ITER and the development of relevant technology. As a potential energy resource for the twenty-first century, magnetic confinement fusion could be the source of large amounts of electricity for world use without contributing to global warming or acid rain. Fusion is the process by which the sun and other stars produce energy. The fusion of two light atoms, such as hydrogen, into a heavier atom, such as helium, results in the release of excess energy. On earth, however, fusion reactions only occur under carefully created conditions. Fusion research focuses on using strong magnetic fields to contain an extremely hot plasma of light atoms. The challenge is to hold the fast-moving particles together long enough for fusion reactions to occur and in sufficient numbers to produce useful energy. Major fusion experiments confine the plasma within a doughnut-shaped device called a tokamak. The high-power electromagnets used in a tokamak are arranged so that their magnetic fields

contain and control the plasma. ITER is being designed as an engineering test reactor to demonstrate the scientific and technical feasibility of magnetic fusion energy. A major objective is to demonstrate controlled ignition and extended plasma burn with steady-state operation as an ultimate goal. Another objective is to demonstrate technologies needed for fusion energy and to serve as an integrated test bed for high heat-flux components.

Over the past six years, the United States has participated in the Engineering Design Activities (EDA) for ITER. The design of the ITER device and its auxiliary systems and facilities have been investigated during this time. Developmental testing of key components and research and development of critical technologies has also been accomplished during the EDA. Los Alamos National Laboratory (LANL) has been involved in the development of plasma spray coating technology for initial fabrication and repair and of the beryllium first wall surfaces inside of the ITER reactor. Research investigations at LANL focused on a number of critical areas including parametric studies of the plasma spray process to maximize the through thickness thermal conductivity of plasma sprayed beryllium coatings, developing a method for preparing the surface of beryllium prior to depositing beryllium by plasma spraying and high heat flux performance testing of the plasma sprayed beryllium coatings. This paper will summarize key results that were achieved during the EDA.

2.0 The Effect of Processing Parameters on the Thermal Conductivity of Plasma Sprayed Beryllium Coatings

2.1 Processing Gases

Addition of hydrogen gas into a conventional DC argon plasma torch is commonly used throughout the thermal spray industry to improve the as-deposited density and overall quality of plasma sprayed coatings. The selection of a plasma generating gas for spraying a given material depends primarily on the gas energy, gas reactivity and cost. The energy content of nitrogen and hydrogen plasmas is considerably higher then for argon or helium due to the dissociation reactions in the nitrogen and hydrogen prior to ionization. Generally about 1 to 10% hydrogen is mixed with argon to increase the heat content and improve the heat transfer characteristics of the primary arc gas. Hydrogen also acts as a reducing agent in the plasma. The gas selection should provide a plasma source capable of heating the injected particles to a molten or near molten state so that particle flow will occur upon impact. Investigations were performed to observe the effect of secondary processing gas additions on the through thickness thermal conductivity of plasma sprayed beryllium coatings [1]. The micrographs in Figure 1 illustrate the effect of helium and hydrogen gas additions on the as-deposited microstructures of plasma sprayed beryllium. Large columnar grains were observed through the thickness of the beryllium coatings when using hydrogen gas additions. Increasing the amount of hydrogen gas from 1 standard liter per minute (slm) to 4 slm showed a substantial increase in the columnar grain size from approximately 50 to 250 μm. Coatings produced with helium gas additions showed the presence of unmelted beryllium particles embedded in a fine-grained beryllium matrix. The corresponding thermal conductivity of the beryllium coatings produced with helium and hydrogen secondary gas additions of 1 and 4 slm were approximately 115, 160 and 190 W/mK. Higher thermal conductivity's were associated with beryllium coatings which had larger elongated columnar grains through the thickness of the coating.

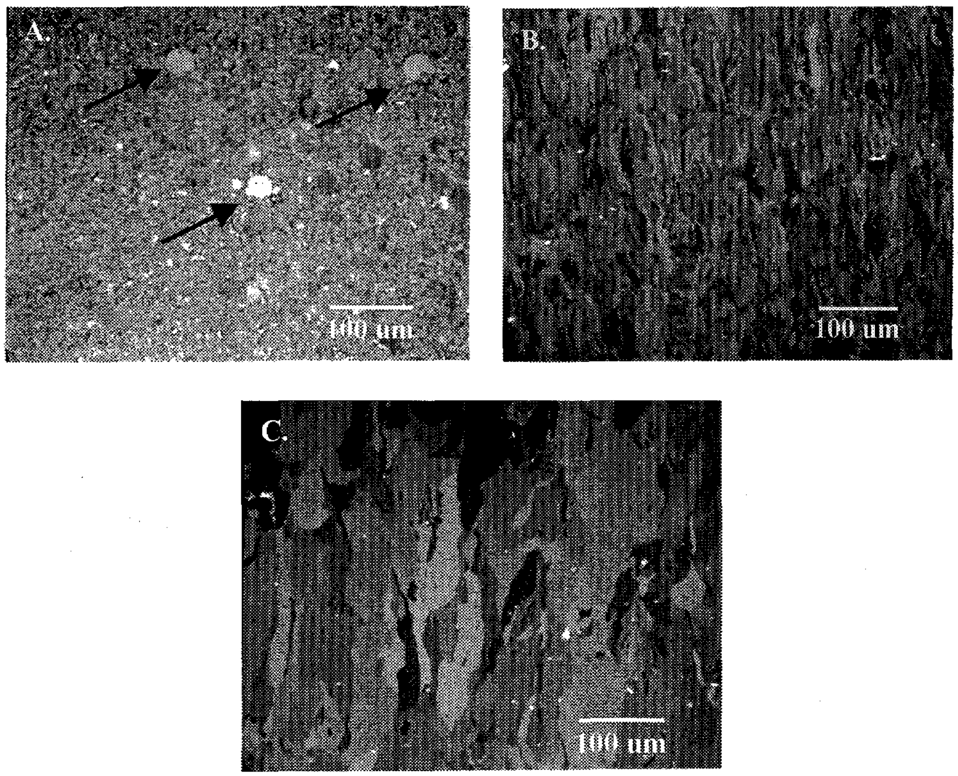


Figure 1. Through thickness microstructures of plasma-sprayed beryllium using a) 10 slm of helium b) 1 slm of H_2 and c) 4 slm of H_2 as a secondary processing gas.

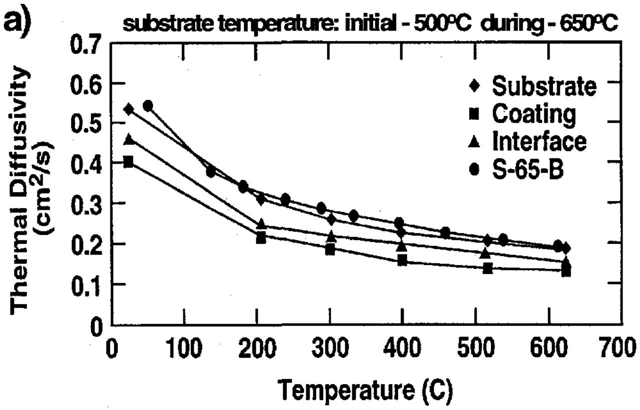
2.2 Substrate Temperature

The influence of substrate temperature on the thermal diffusivity and bond strength of plasma sprayed beryllium coatings deposited on beryllium substrates was investigated. Beryllium substrate temperatures were varied from 500 to 800°C in steps of 100°C [2]. During the plasma spray process the substrate temperatures reached a steady-state temperature of 650, 700, 900 and 1000°C, respectively. Plots of the thermal diffusivity as function of temperature (from room temperature to 600 °C) of the beryllium plasma sprayed coatings, the beryllium substrate, and the interface between the beryllium coating and the substrate are given in Figure 2.

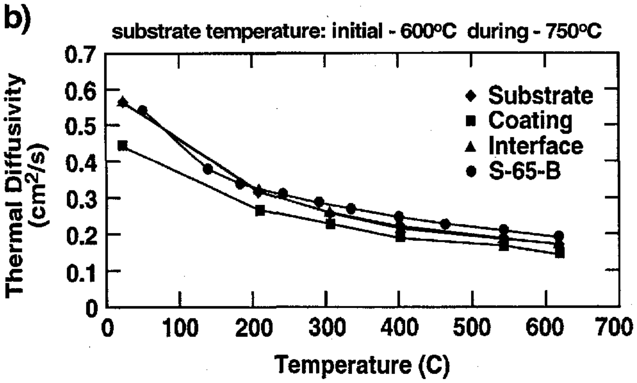
The effect of substrate temperature on the thermal properties of the plasma sprayed beryllium coatings can be summarized as follows:

- The thermal resistance at the interface between the beryllium coating and the beryllium substrate was minimal and showed little dependence on substrate temperature.
- The through thickness thermal diffusivity of the plasma sprayed beryllium coatings increased with increased substrate temperature. Thermal diffusivity measurements of beryllium coatings deposited on substrates with temperatures of 500°C and 800°C were approximately 67% and 94% of the reported values for commercial grade S-65B beryllium material measured from room temperature to 600°C.
- The coating/substrate interface samples that were produced on substrates with temperatures of 500°C and 800°C had thermal diffusivity values approximately 79% and 93% of commercial S-65B beryllium measured from room temperature to 600°C.
- The substrate material showed a thermal diffusivity of approximately 92% of the commercial grade S-65B. Although, the history and composition of the beryllium substrates used in this investigation were not known, it has been reported that the room temperature thermal conductivity can vary depending on the composition and processing history [3].

a)



b)



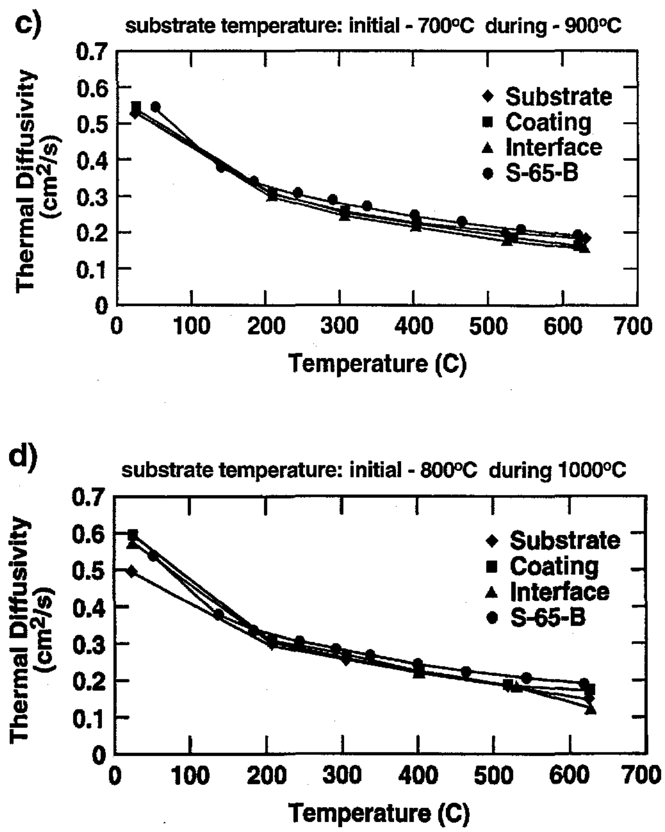
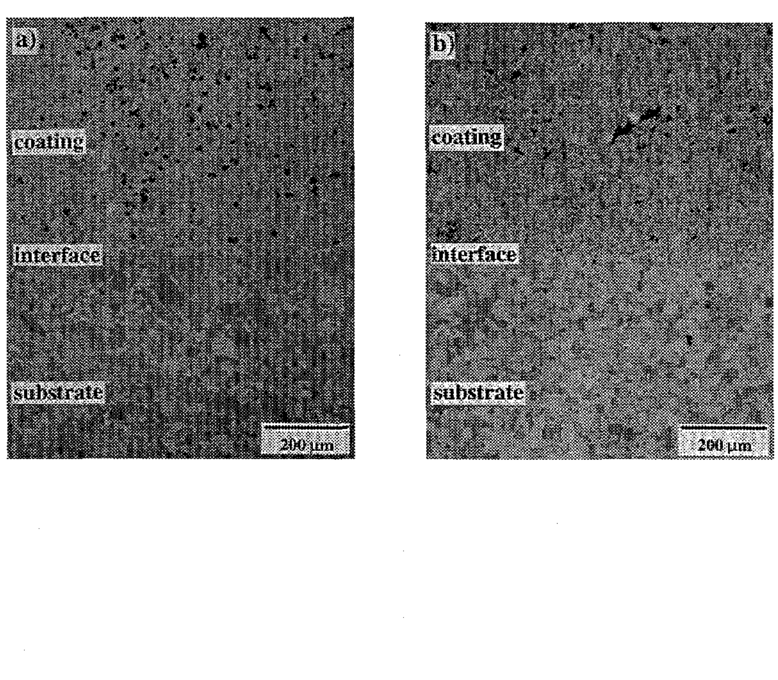


Figure 2. Plots of the thermal diffusivity as a function of temperature (from room temperature to 600 °C) of the beryllium coating, the beryllium substrate and the interface between the beryllium coating and the substrate [2].

The thermal diffusivity values for the beryllium coatings showed very good correlation with both the beryllium substrate and commercial grade S-65B beryllium when measured from room temperature to 600°C. The observed differences are most likely due to differences in the residual porosity and grain size in the coatings.



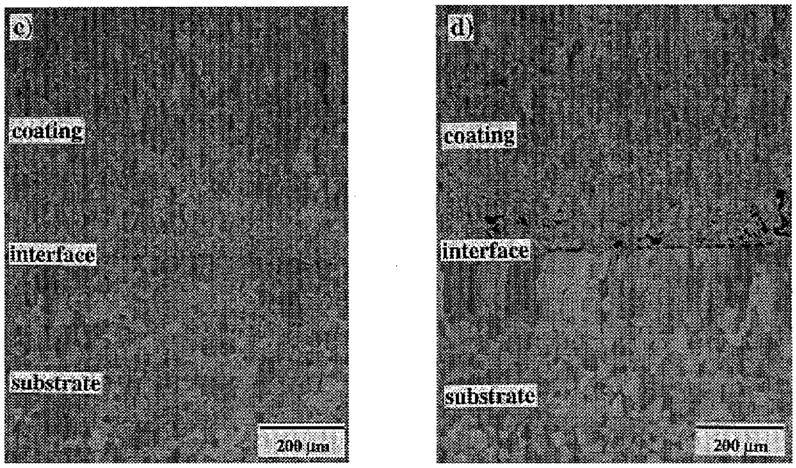


Figure 3. Microstructures of the plasma sprayed beryllium coatings, interface and beryllium substrates at initial substrate temperatures of a) 500 °C, b) 600 °C, c) 700 °C and d) 800 °C [2].

Water immersion density measurements and metallographic image analysis revealed that the density of the beryllium coatings, produced at an initial substrate temperature of 500°C, were approximately 90% (1.67×10^3 kg m^{-3}) of theoretical density (1.85×10^3 kg m^{-3}) with an open porosity of 7.7%. The beryllium coating which was produced at an initial substrate temperature of 800°C had a density of approximately 98% of theoretical (1.81×10^3 kg m^{-3}) with an open porosity of 0.21 %. The corresponding microstructures of the plasma sprayed beryllium coatings on the beryllium substrates at the different substrate temperatures are given in Figure 3. Melting of the surface of the beryllium substrate, during negative transferred-arc heating/cleaning, was observed in Figure 3d.

3.0 Negative Transferred-Arc Cleaning as a Surface Preparation Method

To coat a beryllium surface by plasma spraying, some type of surface preparation is required to clean and roughen the surface in order to maximize the coating adhesion. Grit blasting is the conventional method used to prepare most surfaces prior to plasma spraying a coating. Application of this surface preparation technique is limited inside of the ITER tokamak. Negative transferred-arc cleaning (TA), which has been used extensively in the aerospace community for preparing surfaces prior to applying thermal barrier coatings, can clean damaged and contaminated surfaces as a result of the cathodic cleaning that occurs on the surface of the material to be coated. The use of this technique can result in a factor of two increase in the bond strength between plasma sprayed coatings and the underlying material [4]. The basic operation of negative transferred-arc cleaning results from applying a voltage between the plasma spray torch nozzle and the surface to be coated, Figure 4.

a)

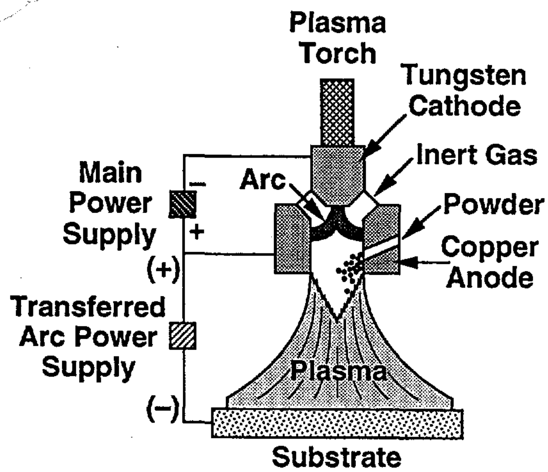


Figure 4. Schematic of the negative transferred-arc cleaning process. The front of the plasma spray torch is (+) and the substrate is (-). Electron flow is from the substrate surface to the face of the plasma torch.

The mechanism of cleaning results from the formation of microscopic cathode arc spots on the thin dielectric oxide layer present on the metal surface [5,6]. Processing conditions which can effect the performance of the negative transferred-arc process are the composition of the plasma gas, the torch to substrate distance, power levels associated with the plasma torch and the transferred-arc power supply, and the operating pressure in the plasma-spraying chamber. The negative transferred-arc process is described in more detail in reference [7]. The effect of negative transferred-arc cleaning of beryllium surfaces prior to plasma spraying was investigated by cleaning eight beryllium samples which had been sputter coated with 10 to 100 monolayers of tungsten and carbon [8]. This combination of materials was chosen to simulate a contaminated surface similar to what might be expected in ITER where carbon, tungsten and beryllium are used as plasma-facing materials.

Rutherford Backscattering Spectrometry (RBS) using 1.6 MeV He ions was used to characterize the surface of the eight beryllium samples before and after negative transferred-arc cleaning. The surface analysis included detection of carbon, tungsten, oxygen, argon and copper. All measurements were done at the centerpoint of each beryllium sample. Analysis of the carbon and tungsten levels before and after the TA cleaning process for the eight-beryllium samples is given in Table 1. The carbon present on the surface of beryllium was reduced below the detection limit ($< 10^{13}$ atoms/cm^2) of the RBS analysis technique and the sputter coated tungsten was reduced by an order of magnitude below the initial starting levels.

Table 1. 1.6 MeV He RBS at centerpoint of Be tile samples prior to negative transferred-arc cleaning (10^{15} atoms/cm^2).

Sample	C-before	C-after	W-before	W-after
1	43.49	n.d	13.45	1.10
2	114.10	n.d	15.97	0.68
3	50.41	n.d	19.12	3.00
4	152.10	n.d	21.59	0.41
5	37.58	n.d	17.78	7.00
6	96.16	n.d	12.73	9.60
7	68.55	n.d	10.54	0.71
8	116.80	n.d	6.73	1.30

n.d.-below the minimum detection limit of RBS analysis ($< 10^{13}$ atoms/cm^2)

The chamber pressure can significantly effect the degree of surface roughening and contamination removal during the TA cleaning of beryllium. The microstructures in Figure 5 illustrate the different degrees of surface roughening which can be achieved when TA cleaning beryllium at chamber pressures of .004, .012 and .033 MPa using a standard operating condition. The average surface roughness measured by surface profilometry was 5.5 μm at .004 MPa, 4.42 μm at .012 MPa and 1.75 μm at .033 MPa. Increasing surface roughness will improve the mechanical interlocking/bonding of the coating to the substrate.

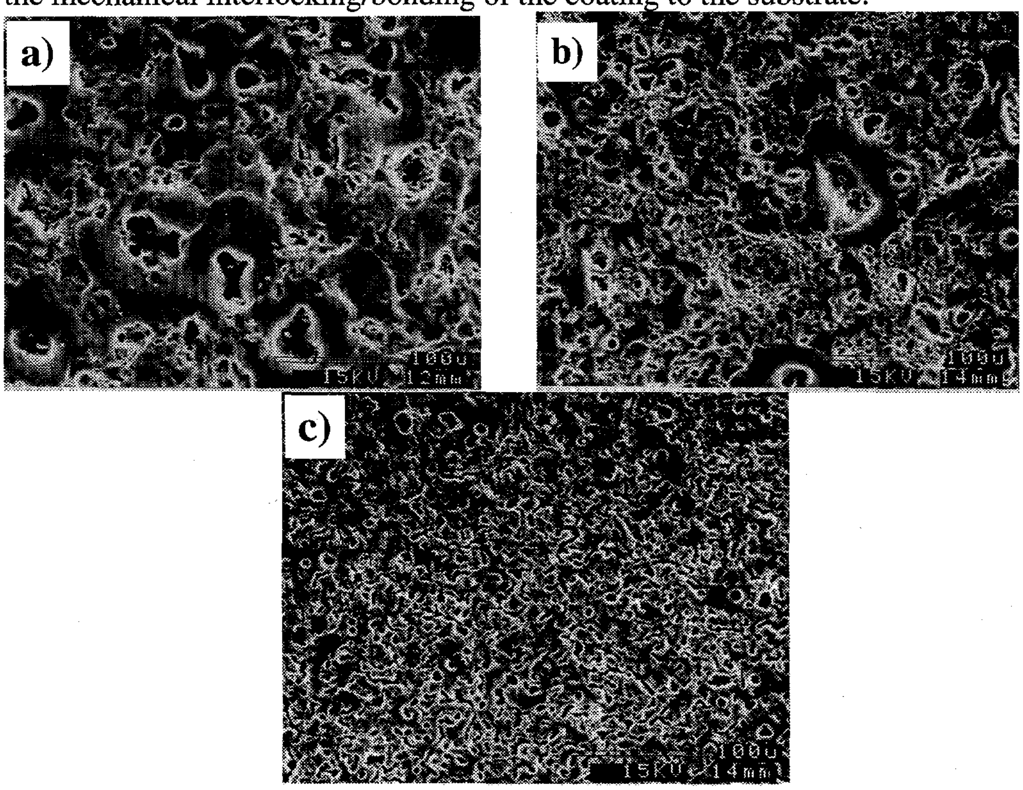


Figure 5. Negative transferred-arc cleaned surfaces of beryllium at various chamber pressures a) .004 MPa, b) .012 MPa and c) .033 MPa [8].

3.1 Interfacial bond strength of plasma sprayed beryllium on beryllium

The interfacial bond strength between the plasma sprayed beryllium coatings and the beryllium substrates, at the different substrate temperatures, was measured using four-point bend testing, Figure 6. Details associated with the testing procedure can be found in reference [9]. The surfaces of the beryllium substrates were TA cleaned prior to depositing the beryllium.

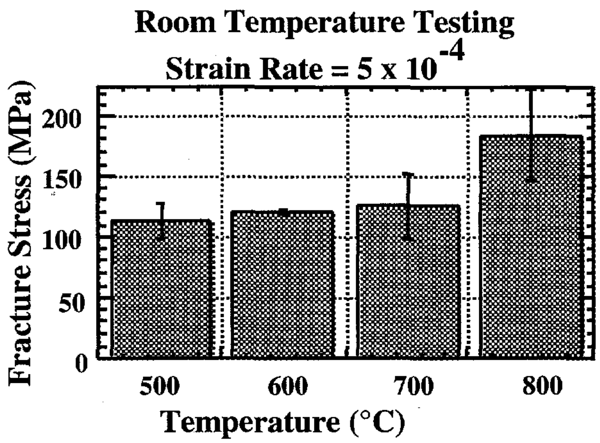


Figure 6. Four-point bend bond strength for beryllium coatings on beryllium substrates at initial substrate temperatures of 500°C, 600°C, 700°C and 800°C.

The tests were performed at room temperature at a strain rate of 10^{-4} /sec. Loading of the test samples was applied parallel to the direction of the beryllium coating/substrate interface. Failure occurred at the substrate/coating interface. The bond strength of the beryllium coating was typically greater than 100 MPa. Mechanical locking between the coating and the transferred-arc cleaned beryllium substrate was the main contributor to the observed bond strength, Figure 7. Beryllium coatings which were deposited at an initial substrate temperature of 800°C show a much higher interfacial strength suggesting the possibility of metallurgical bonding across the interface.

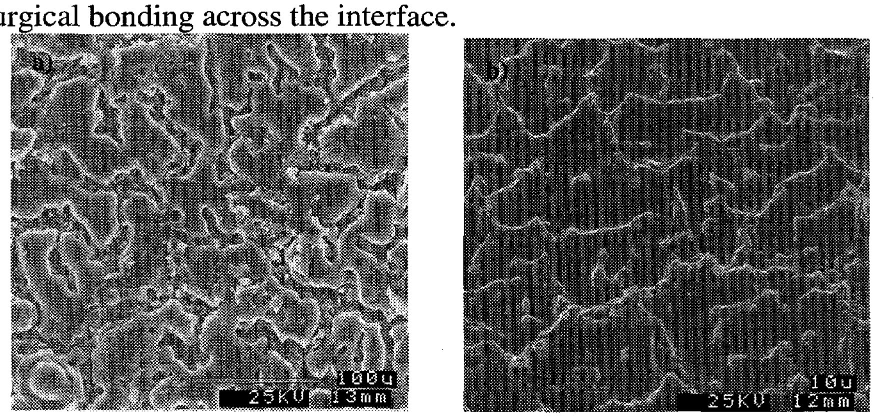


Figure 7. Fracture surfaces of the beryllium coating/substrate interface a) substrate and b) coating [8].

4.0 Electron Beam High Heat Flux Testing

Plasma sprayed beryllium on copper heat sink materials was evaluated for initial fabrication of the beryllium first wall armor for ITER [10]. Electron beam high heat flux testing, using the 30 kW Electron Beam Test System (EBTS) at Sandia National Laboratory in Albuquerque, New Mexico was done to determine the performance of the plasma sprayed beryllium coatings at ITER first wall conditions. Plasma sprayed beryllium/copper mockups were fabricated and subsequently thermal cycled in the EBTS facility. Thick beryllium coatings (> 15mm) were deposited on the surface of copper heat sink materials. The beryllium coating was subsequently machined to 5 and 10mm thick individual beryllium tiles for high heat flux testing. Plasma sprayed beryllium/copper mockups for electron beam high heat flux testing are shown in Figure 8.

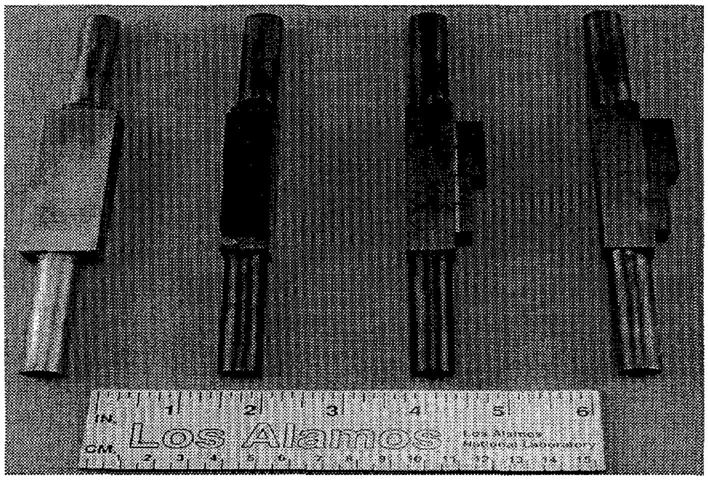


Figure 8. Plasma sprayed beryllium/copper mockups with 5 and 10mm thick individual beryllium tiles used for electron beam high heat flux testing.

Thermal fatigue testing of the beryllium/copper mockups was performed initially at 1 MW/m^2 which is twice the expected heat flux for the ITER primary first wall modules (.5 MW/m^2). Subsequent thermal cycling was performed at 3 and 5 MW/m^2 heat fluxes until the beryllium/copper mockups failed. Failure was indicated by a large increase in the surface temperature of the beryllium plasma sprayed coating. Testing at 1 MW/m^2 was done with a water inlet temperature of 160 ^{o}C, a pressure of 4 MPa and a velocity of 1 m/s. Pulse lengths of 20 seconds on and 20 seconds off were sufficient to reach 90% of steady-state temperature profiles within the mock-up. The fabrication and testing of the beryllium/copper mockups is described in more detail in reference [10]. Efforts were made to maximize the surface contact between the underlying heat sink material and the plasma sprayed beryllium coating and to prevent the formation of brittle beryllium/copper intermetallics at the coating/heat sink interface. A knurled surface produced by Electro Discharge Machining (EDM) was investigated as an effective method for increasing the surface areas of the copper heat sink/beryllium coating interface. The knurl was .076 mm deep with a radius of 0.125 mm.

Plasma sprayed beryllium/copper mockups successfully survived 3000 thermal fatigue cycles at 1 MW/m^2 on all four of the individual beryllium 5 and 10mm tiles with no damage except for a discoloration of the surface and a slight increase in the surface temperature of one of the sub-castellated section. The temperature of the interface between the beryllium armor and the copper heat sink was typically 240 °C. The peak surface temperatures of the beryllium tiles remained below 550 °C during the 3000 thermal fatigue cycles. After completing the thermal cycles at 1 MW/m^2, the tested area was reduced to two 5 mm thick tiles and tested at a heat flux of 3 MW/m^2. After 10 cycles at this heat flux, both tiles showed a substantial increase in surface temperature indicating that some type of tile delamination had occurred. Post-mortem analysis of the beryllium/copper mockup using scanning electron microscopy indicated that cracking had occurred in the beryllium tile with no evidence of cracking at the beryllium/copper interface.

4.0 Conclusion

The research on beryllium plasma spray technology that was performed during the ITER-EDA resulted in a number of advances and recommendations:

- Beryllium plasma sprayed coatings have been produced with thermal conductivity properties equivalent to bulk beryllium (~ 190 W/m^2).
- Negative transferred-arc cleaning (TA) has been shown to be an effective method for preparing the surface of beryllium and copper prior to depositing beryllium by plasma spraying.
- Interfacial bond strengths between plasma sprayed beryllium coatings and beryllium surfaces were greater than 100 MPa when tested in four-point bending.
- Electron beam testing of plasma sprayed beryllium survived 3000 thermal fatigue cycles at a heat flux of 1 MW/m^2.

Beryllium plasma spray technology has been recommended by the ITER Joint Central Team for *in-situ* repair and initial fabrication of the beryllium plasma-facing armor.

Acknowledgments

The author would like to acknowledge the following people for their contributions to the research that was performed during the ITER-EDA: R.D. Fields, R.U. Vaidya, P.W. Stanek, L.A. Jacobsen, H. Kung, J. Montoya, P. Papin, and R. Brunner from Los Alamos National Laboratory, M. Ulrickson, D.L. Youchison and D.S. Walsh from Sandia National Laboratory, K. Slattery from the Boeing Company and Brush Wellman Inc., for machining operations. The author would also like to acknowledge the DOE Office of Fusion Energy for their support of this research.

References

1. R.G. Castro, P.W. Stanek, K.E. Elliot, D.L. Youchison, R.D. Watson, and D.S.Walsh, "The structure, properties and performances of plasma sprayed beryllium for fusion applications" Phys. Scripta T64 (1996)77-83.
2. R.G. Castro, A.H. Bartlett, K.J. Hollis and R.D. Fields, "The Effect of Substrate Temperature on the Thermal Diffusivity and Bonding Characteristics of Plasma Sprayed Beryllium," Fusion Engineering and Design, 37, (1997) 243-252.
3. I.W. Dunmur, Plasma-Sprayed Beryllium, in Beryllium Science and Technology Vol. 2, D.R. Floyd and J.N. Lowe Eds. Plenum Press New York, (1979) 135-175.
4. A. Itoh, K. Takeda, M. Itoh and M. Koga, "Pretreatment of substrates by using reversered transferred-arc in low pressure plasma spray," Proc. of the 3rd Natl. Thermal Spray Conf., Long Beach, CA (1990) 245-251.
5. A.E. Guile, and A.E. Juttner, IEEE Trans. on Plasma Science, PS-8, (3) (1980) 259-269.
6. A.E. Guile, "Electric Arc-Phenomena", Proc. IEE, IEE Reviews, 118 (9R)(1971) 1131-1154.
7. Muehlberger, E., U.S. Patent No. 4,328,257 (1982).
8. R.G. Castro, K.J. Hollis and K.E. Elliott, "Roughening and removal of surface contamination from beryllium using negative transferred-arc cleaning," Proc. of ISAPS'97, Advances in Applied Science, September 22-26, (1997), Los Angeles, CA, 99-106.
9. R.G. Castro, A.H. Bartlett, K.J. Hollis and R.D. Fields, "The Effect of Substrate Temperature on the Thermal Diffusivity and Bonding Characteristics of Plasma Sprayed Beryllium," Fusion Engineering and Design, 37, (1997) 243-252.
10. R.G. Castro, K.E. Elliott, R.D. Watson, D.L. Youchison and K.T. Slattery, "Fabrication and high heat flux testing of plasma sprayed beryllium ITER first wall mock-ups" J. of Nuc. Mat. 258-263 (1998) 252-257.