

EM 2002

**Transactions** Oral Presentations and Posters



6th International Topical Meeting on Research Reactor Fuel Management March 17 to 20, 2002, Ghent, Belgium Organised by the European Nuclear Society

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# **RRFM 2002**

6<sup>th</sup> International Topical Meeting on

# **Research Reactor Fuel Management**

Cultural Center of Ghent University 'Het Pand', Ghent, Belgium

March 17-20, 2002

Organised by the European Nuclear Society

in cooperation with the International Atomic Energy Agency

Invited and Contributed Papers Oral and Poster Presentations Chairman: Pol Gubel, SCK/CEN Mol, Belgium

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## Session 1: Fissile materials supply

# Past and present Supply of enriched Uranium for Research Reactors in the European1UnionH. Müller, RWE NUKEM GmbH, GermanyA new Standard Specification for Uranium Metal intended for Research Reactor Fuel4FabricationJ. Laucht, RWE NUKEM GmbH, GermanyThe revised IAEA Regulations and their Impact on the Transport of Research8

Page

**Reactor Fuel** R. Christ, F. Hilbert and M. Kübel, NCS GmbH, Germany

# Session 2: Fuel development, qualification, fabrication and licensing

<b>RERTR Progress in MO-99 Production from LEU (Invited Paper)</b> G.F. Vandegrift, C. Conner, S. Aase, A. Bakel, D. Bowers, E. Freiberg, A. Gelis, K.J. Quigley, and J.L. Snelgrove, Argonne National Laboratory, USA	11
An Update on the LEU Target Development and Conversion Program for the Maple Reactors and new Processing Facility (Invited Paper) G.R. Malkoske, MDS Nordion, Canada	18
Experimental Irradiation of UMo Fuel: Pie Results and Modeling of Fuel Behaviour A. Languille, D. Plancq, F. Huet, B. Guigon, CEA Cadarache, France; P. Lemoine, P. Sacristan, CEA Saclay, France; G. Hofman, J. Snelgrove, J. Rest, S. Hayes, M. Meyer, Argonne National Laboratory, USA; H. Vacelet, CERCA Framatome, France; E. Leborgne, Technicatome, France; G. Dassel, NRG Petten, The Netherlands	26
Status as of March 2002 of the UMo Development Program JM. Hamy, Framatome-ANP; A. Languille, B. Guigon, CEA, Cadarache; P. Lemoine, CEA Saclay; C. Jarousse, CERCA, Framatome-ANP; M. Boyard, Technicatome; JL. Emin, COGEMA; France	33
Advances and Highlights of the CNEA Qualification Program as High Density Fuel Manufacturer for Research Reactors P. Adelfang, <u>L. Alvarez,</u> N. Boero, R. Calabrese, P. Echenique, M. Markiewicz, E. Pasqualini, G. Ruggirello and H. Taboada, Comisión Nacional de Energía Atómica (CNEA), Argentina	40
Status of the Qualification Program for atomized U-Mo Dispersion Rod Type Fuel in Korea C.K. Kim, K.H. Kim, J.M. Park, D.S. Sohn, Fuel Design Technology Development Dept., K.H. Lee, H.T. Chae, HANARO Center, Korea Atomic Energy Research Institute, Korea	45
Progress in Development of Low-Enriched U-Mo Dispersion Fuels G.L. Hofman and J.L. Snelgrove, Argonne National Laboratory, Argonne, USA S.L. Hayes and M.K. Meyer, Argonne National Laboratory, Idaho Falls, USA	50
MTR Fuel Plate Qualification Capabilities at SCK-CEN E. Koonen, P. Jacquet, SCK-CEN, Belgium	59

Session 3: Reactor operation, fuel safety and core conversion	
Progress of the RERTR Program in 2001 (Invited Paper) A. Travelli, Argonne National Laboratory, USA	63
Progress of the Russian RERTR Program: Development of new-type Fuel Elements for Russian-built Research Reactors (Invited Paper) A.V. Vatulin, Y.A. Stetskiy, V.A. Mishunin, V.B. Suprun, I.V. Dobrikova, VNIINM, Russia	69
Use of Highly Enriched Uranium at the FRM-II (Invited Paper) K. Böning, Technische Universität München, Germany	73
Characteristic Differences of LEU and HEU Cores at the German FRJ-2 Research Reactor R. Nabbi, J. Wolters and G. Damm, Research Center Jülich, Germany	78
A preliminary Study on the Suitability of High Density LEU Fuel Elements to maintain BR2 Reactor Operational Characteristics A. Beeckmans and E. Koonen, SCK-CEN, Belgium	83
<b>Behaviour of UMo with Cladding Failure UMUS test in the HFR</b> J. Guidez, S. Casalta, JRC Petten, The Netherlands; F.J. Wijtsma, G. Dassel, P.M.J. Thijssen, NRG Petten, The Netherlands; C. Jarousse, CERCA Framatome, France; A. Languille, CEA Cadarache, France	88
Best Safety Practices for the Operation of Research Reactors H. Böck, M. Villa, Atominstitute of the Austrian Universities, Austria	93
Session 4: Spent fuel management, corrosion and degradation	
Influence of the Fuel operational Parameters on the Aluminium Cladding Quality of Discharged Fuel S. Chwaszczewski, W. Czajkowski, E. Borek-Kruszewska, Institute of Atomic Energy, Poland	99
MIR Reactor Fuel Assemblies Operating Experience A.F. Grachev, A.L. Izhutov, A.E. Novosyolov, M.N. Svyatkin, Z.I. Chechetkina, V.V. Aleksandrov, State Scientific Centre of Russia Research Institute of Atomic Reactors, Russia	104
New Storage Mode for spent Fuel at the Budapest Research Reactor T. Hargitai, I. Vidovszky, KFKI Atomic Energy Research Institute, Hungary	109
Storage, Inspection and SIP Testing of spent nuclear fuel from the HIFAR Materials Test Reactor	114
H. Selwyn, R. Finlay, P. Bull and A. Irwin, ANSTO, Australia	
Session 5: Back-end options and transportation	
Progress of the United States Foreign Research Reactor spent Nuclear Fuel Acceptance Program (Invited Paper) D.G. Huizenga, M. Clapper, U.S. Department of Energy and A.W. Thrower, Science Applications International Corporation, USA	119
Management of the Acceptance Process of RTR Aluminide Type Spent Fuel P. Auzière, J. Thomasson, COGEMA, France	123
Development of Melt Dilute Technology for Disposition of Aluminium based spent Nuclear Fuel W.F. Swift, Westinghouse Savannah River Company, USA	132
Spent Fuel Management Plans for the FiR 1 Reactor S.E.J. Salmenhaara, Technical Research Centre of Finland, Finland	136
The Russian Federation Legislation. The new Laws. Prospects for International Cooperatio	<b>n.</b> 139

.

The Russian Federation Legis A.Ye. Lebedev, Tenex, Russia

L

# Posters

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1	In-Reactor Behaviour of centrifugally atomized U <sub>3</sub> Si Dispersion Fuel irradiated up to high burn-up for HANARO K.H. Kim, D.G. Park, C.S. Lee, S.J. Oh, C.K. Kim, Korea Atomic Energy Research Institute, Korea	145
2	Boro Silicate Glass: The proven Conditioning of RTR Ultimate Waste O. Bartagnon and V. Petitjean, COGEMA, France	150
3	Consortium NCS/GNS: Disposal of spent Nuclear Fuel from the DKFZ (Heidelberg, Germany) R. Vallentin, GNS; B. Jünger, DKFZ; T. Schmidt, NCS, Germany	155
4	U.S. Spent Nuclear Fuel Acceptance Policy 2006 and beyond J. Edlow and K.R. Brown, Edlow International Company, USA	160
5	MR-6 Type Fuel Elements cooling in natural Convection Conditions after the Reactor shut down K. Pytel, W. Bykowski, A. Moldysz, Institute of Atomic Energy, Poland	165
6	Encapsulation Technology of MR6 spent Fuel and Quality Analysis of the EK-10 and WWR-SM spent Fuel stored more than 30 years in wet Conditions E. Borek-Kruszewska, W. Bykowski, S. Chwaszczewski, W. Czajkowski, M. Madry, Institute of Atomic Energy, Poland	170
7	Study on HANARO Core Conversion using U-Mo Fuel K.H. Lee, C.S. Lee, C.G. Seo, S.J. Park and H. Kim, Research Reactor Analysis Dept., and C.K. Kim, Fuel Design Techn. Devel. Dept., Korea Atomic Energy Research Institute, Korea	175
8	Study on possibly Usage of 36 % or 20 % enriched UO <sub>2</sub> TWR-S Fuel Type at RB Heavy Water Critical Assembly M. Pesic, N. Dasic and V. Ljubenov, The VINCA Institute of Nuclear Sciences, Yugoslavia	179
9	Powder Production of U-Mo Alloy, HMD Process (Hydriding-Milling-Dehydriding) E.E. Pasqualini, J. Helzel García, M. López, E. Cabanillas and P. Adelfang, Comisión Nacional de Energía Atómica, Argentina	183
10	Improvements in irradiated Fuel Handling at Dhruva for enhanced Safety H. Gujar and S. Agarwal, Dhruva, Bhabha Atomic Research Centre, India	188
11	Examination of U-9 % Mo Alloy Powder Microstructure in its initial Condition and after Fuel Pin Fabrication R.Kh. Gibadullin, A.D. Karpin, Yu.M. Pevchikh, V.V. Popov, V.N. Sugonyaev, V.M. Troyanov, Institute for Physics and Power Engineering, Russia	193
12	Study of in-Reactor Creep in the Alloys employed as Structural Materials for Research Reactor Core Components and Fuel Pins M.G. Bulkanov, A.S. Kruglov, Yu.M. Pevchikh, V.V. Popov, V.M. Troyanov, Institute for Physics and Power Engineering, Russia	197
13	Management and Inspection of Integrity of spent Fuel from IRT Mephi Research Reactor V.G. Aden, S.Yu. Bulkin, A.V. Sokolov, Research and Development Institute of Power Engineering, and A.V. Bushuev, A.F. Redkin, A.A. Portnov, Moscow State Engineering Physics Institute - Technical University, Russia	or 201
14	Monte Carlo Simulation of Irradiation of MTR Fuel Plates in the BR2 Reactor using a full-scale 3-D Model with inclined Channels V.V. Kuzminov, E. Koonen and B. Ponsard, SCK-CEN, Belgium	206
15	Neutronic Design of the RSG-Gas' Silicide Core T.M. Sembiring, I. Kuntoro and H. Hastowo, BATAN, Indonesia	211
16	Spent Fuel Management and Site exploitation at IFIN-HH, Bucharest-Magurele C.A. Dragolici, A. Zorliu, C. Petran and I. Mincu, National Institute of Research and Development for Physics and Nuclear Engineering 'Horia Hulubei' IFIN-HH, Romania	216
17	Fuel Management Optimization for the WWR-M Research Reactor in Kiev Y.P. Mahlers, Institute for Nuclear Research, Ukraine	221

# **Session 1**

**Fissile materials supply** 

# PAST AND PRESENT SUPPLY OF ENRICHED URANIUM FOR RESEARCH REACTORS IN THE EUROPEAN UNION

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# ABSTRACT

In the last decade research reactor operators have focused mainly on the issues of disposal of spent research reactor fuel and the development of high density fuels. The safe supply of fresh uranium did not receive as much attention. This is surprising since the United States - who was the main supplier for LEU and HEU since the late 1950's - stopped supplying non-US research reactors with enriched uranium a decade ago. The reason for this stop of supply is described in this paper.

This paper explains how research reactors in the EU continued to operate during the last decade , in spite of the fact that their primary supply source had not provided LEU and HEU over the same period.

#### 1. Historical background of supply of enriched uranium for research reactors

The timely availability of sufficient enriched uranium with the suitable U-235 assay is mandatory for the individual research reactor to guarantee continuous operation of its research reactor. In the former Eastern and Western world mainly the weapon states USA and Russia are supplying enriched uranium with higher U-235 assays as a spin-off of their military programs.

In the Western world the standard U-235 assays are either 19.75 % (LEU) or 93 % (HEU); in the former Soviet Union and its satellites the common U-235 assays were 36 and between 80 and 90 %. From the sixties of the 20th century until the mid eighties the USA supplied mainly HEU to research reactors outside the USA under the relevant bilateral agreements for cooperation, until 1974 even under lease conditions. This means, that until 1974 HEU remained the property of the USA while it was outside the USA for use in research reactors and later return as spent fuel to the USA.

In 1977, however, US President Carter became concerned about the "wide spread of weapons usable uranium" in research reactors worldwide. He initiated the International Fuel Cycle Evaluation (INFCE) and especially its working group 8 to minimize the traffic and use of HEU.

As a result of the working group 8 it was decided that the ideal U-235 assay in the sense of the nonproliferation should be less than 20 %, namely 19.75 %. The development of such high density fuel with the target of maintaining the same geometry of the fuel elements was initiated by the Reducing Enrichment in Research and Test Reactors (RERTR) Programme of the US-Department of Energy (DOE) with international cooperation. Declared preliminary target of the USA was at that time to reduce an export of 450 kg of HEU per year to 150 kg HEU outside the USA.

#### 2. Stop of supply of LEU and HEU by the USA in the nineties

In 1992 supplies of LEU and HEU by the USA came almost to a complete stop. For supplies of HEU the U. S. Energy Act of 1992 limited exports according to the following criteria:

1.) There is no alternative nuclear fuel or target enriched in the isotope 235 to a lesser percent than the proposed export, that can be used in that reactor.

2.) The proposed recipient of that uranium has provided assurances that, whenever an alternative reactor fuel or target can be used in that reactor, it will use that alternative in lieu of highly enriched uranium; and

3.) The United States Government is actively developing an alternative reactor fuel or target that can be used in that reactors.

This new legislation stopped de facto US supplies of HEU to Europe which were so far the main customers with about 150 kg HEU per year. Only limited quantities of HEU have thereafter been supplied to the Canadian AECL which apparently met at this time the new US criteria for HEU supplies outside the USA.

In 1994 the USA had to stop as well completely the supply of LEU for several years due to a safeguards problem at the Y-12 plant in Oak Ridge.

The research reactor community having a demand of enriched uranium of US origin in order to return spent fuel to the USA faced more or less a disaster by the lack of US supplied LEU and HEU.

#### 3. NUKEM's role in bridging the gap of US supplies

Since 1973 NUKEM, nowadays RWE NUKEM GmbH, plays a major role in the trading to nuclear fuels both for power and research reactors worldwide. The Fuel Cycle Department of RWE NUKEM GmbH is today the largest private trader of nuclear fuels which is its core business.

Although RWE NUKEM gave up manufacturing of fuel elements for research reactors in 1998 and handed over this business to the French company CERCA, NUKEM continued to be active in the field of LEU and HEU supplies after this date due to its experience in the external fuel cycle for research reactors. RWE NUKEM bridged the supply-gap in the nineties and supplied LEU to research reactors in the Western hemisphere and HEU to high flux reactors in the EU.

RWE NUKEM's ability to supply LEU and HEU was due to the availability of large stocks of no longer needed research reactor fuels from O-experiments which de facto had no or only slight burn-up after termination or shut down of those research or demonstration reactors in Europe, mainly in Germany. The no longer used fuels were treated in various uranium chemistry plants in the EU according to the requirements of the research reactor operators; i.e. mainly to produce LEU or HEU in the form of metal.

While the Western research reactors community in the last decade of the 20th century concentrated on issues of the back-end (open again the blocked path of returning of spent fuel elements to the USA) and the further development of high density fuels under the RERTR program, RWE NUKEM provided a reliable supply of LEU and HEU to research reactors in Europe, Canada, Japan and South-East Asia. All of the supply contracts were concluded with the concurrence of the Euratom Supply Agency. Otherwise many of the research reactors would have been forced to shut down due to lack of LEU and HEU of US origin.

#### 4. Some details on RWE NUKEM's supply of HEU and LEU

In order to maintain its expert knowledge and to conserve its know-how, RWE NUKEM played an active role in the development of common and unified specification for enriched uranium for research reactors. The company is a member of the relevant ASTM-committee.

RWE NUKEM also contributed considerably to the solution of the so-called "U-232 problem" which was of relevance during the handling and processing of no longer needed uranium fuels in the EU.

The interruption of HEU supplies by the USA after 1992 caused serious difficulties to the high-flux reactors in the European Union because the USA had supplied HEU only as annual quantities for the individual reactors.

The inventory of HEU of the shut-down German demonstration reactor THTR received more than 1 tonne of HEU from the USA secured operation of all high-flux reactors in the EU. RWE NUKEM organized the recovery of the non-irradiated graphite spheres containing the HEU. The separation of the contained Thorium from the HEU was a complicated process however was completed successfully.

It is an open secret that Russia also contributed to the operation of high-flux reactors in the European Union. RWE NUKEM also played an important role on the securing of the HEU for the operation of the new Munich research reactor FRM-2.

With regard to bridging the gap of US supplies of LEU, RWE NUKEM organized in the EU the processing of several tonnes of no longer used LEU of US origin. The available material, stemming mainly from a O-experiment, and available as 20 and 35 % enriched uranium in the form of nickel plated metal platelets was denickelized and remelted. The 35 % enriched uranium was then blended down in induction furnaces to an enrichment of 19.75 % U-235.

The motive and motto under which RWE NUKEM acted in the past and continues in the field of research reactors is to contribute to the undisturbed operation of research reactors by reliable fuel supplies and related services.



## A NEW STANDARD SPECIFICATION FOR URANIUM METAL INTENDED FOR RESEARCH REACTOR FUEL FABRICATION

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#### ABSTRACT

The American Society of Standards and Testing Materials (ASTM) Committee C-26 on Nuclear Fuel Cycle released the final standard specification for low enriched uranium metal for the use in Research Reactor Fuel Fabrication in mid 2001. This specification is the result of discussions between members of different companies and organizations dealing with research reactor uranium or fuel production for research reactors. The specification is a consensus, typical for all ASTM specifications, reflecting the material and interests of all participants of the fuel cycle of research reactor uranium. It has been achieved that all uranium, independent from where it was obtained is defined as commercial grade enriched uranium metal and shall be treated under the same specification C 1462. Both, "buyer" and "seller" could now refer to this specification with a simple reference number in order to define their requirements more easily.

#### 1. Introduction

For over 10 years RWE NUKEM has recommended to the research reactor community to agree upon a standard specification for low enriched uranium metal for the use in research reactor fuel fabrication. Finally, the ASTM Committee C-26 on Nuclear Fuel Cycle released a standard specification for uranium metal of the enrichment between 15 - 20 % U-235 which is used for Research Reactor Fuel for most of the Research Reactors. We already discussed draft #5 of this specification available at the RERTR Meeting in Sao Paulo 1998. Changes have been made during the following committee meetings (held every six months) to get a consensus between all parties involved in uranium production and fuel fabrication. This paper shall report about the changes made and shall discuss the final specification to inform the research reactor community about the now available specification. The approved ASTM specification could be referred under the designation C 1462-00.

### 2. Terminology

The ASTM standard C 1462 applies to all uranium metal derived from commercial natural uranium, recovered uranium, or uranium obtained from the blending of highly enriched uranium with commercial natural uranium, recovered uranium, or depleted uranium.

Commercial natural uranium is defined as any form of natural unirradiated uranium (containing 0.711 + 0.004 % U-235).

Depleted uranium is any form of unirradiated uranium with an assay less than commercial natural uranium.

Highly enriched uranium is any form of uranium having a U-235 content equal or in excess of 20 %. Recovered uranium is any form of uranium having been exposed in a neutron facility and either later chemically separated from fission products or may be used as is, due to low irradiation levels.

#### 3. Health physics requirements

It is recognized that different limits would be necessary to accommodate different fuel histories when recovered uranium is intended to be used for utilizing fuel element production for research reactor fuel. This may depend on the burn up of the utilizing spent fuel elements used in reprocessing. The specification defines the burn up with up to 50% although – according to our experience – this value is in most of all cases in the range of 20%.

For the defined commercial grade enriched uranium metal, the gamma activity of all fission products shall not exceed 600 Bq/gU (Measurements as per ASTM C 1295 or equivalent). During the late discussions of that standard one manufacturer intended to weaken this value up to a level of 6,000 Bq/gU. But all other members of the committee refused to accept such high value in the standard due to problems caused to other manufacturers. A compromise was found with a side note that in case the fabricator could accept gamma activities as high as 6,000 Bq/gU, the material may be used for fabrication.

For commercial grade enriched uranium metal the total alpha activity from transuranic elements shall be less than 250 Bq/gU. This value was set as high as 500 Bq/gU in the previous draft and is with the 250 Bq/gU limit now a suitable compromise, reflecting the real material situation as well as health physics requirements in our fuel cycle.

Emitter / irradiation	Activity
Fission products /gamma	600 Bq/gU
Transuranic Elements / alpha	250 Bq/gU

Fig 1. Limits for fission products and transuranic elements

#### 4. Isotopic requirements

For the isotopic requirements no changes have been made since discussion of the draft in 1998. Following limits have been set in C 1462:

U-isotope	Value
U-232	2 ppb
U-234	1%
U-236	4 %

Fig 2. Uranium isotope limits

U-236 is still on the high side taking into account that most recovered uranium after reprocessing is being downblended to less than 20% and subsequently having typically U-236 contents of less than 1%. The future will show whether such relaxed value is necessary.

#### 5. Physical requirements

The uranium content of commercial grade enriched uranium metal shall be greater or equal to 99.85 weight percent.

The product form to be delivered caused long discussions during the Committee meetings. It was proposed that the product shall come in regular pieces with a mass of 150 to 300 grams. However one American supplier wanted to have its broken pieces as well in line with the specification. Subsequently, a consensus was reached and the product is now defined as follows: The product shall be supplied as spherical, cubical, cylindrical or broken pieces with dimensions

between 10 and 40 mm. The mass of each individual piece shall be between 130 and 300 gram.

#### 6. Sampling

One representative sample of sufficient size to perform tests prescribed shall be taken from each individual lot. The lot size shall not be greater than 10 kg.

#### 7. Impurities

After much consideration the impurities of the uranium are now limited according to following table:

Element	Value (µg/gU)	Element	Value (µg/gU)
Al	150	Mg	50
В	1*	Mn	50**
Be	10	Mo	100
С	800**	Ni	100
Ca	100	Р	100
Cd	1*	Si	250**
Cr	50	Na	25
Со	10**	Sn	100
Cu	50	Va	30**
Fe	250	W	100
Pb	10	Zr	250
Li	10		
Rare Earths	3.0*	Total	1500
N(DyEu,Gd,Sm)			

Fig 3. Chemical impurity limits

\* Those elements have been added to the elements contained in the previous draft specification \*\* Those elements have exceeding limits in comparison with the previous draft specification The levels of five individual impurities have been elevated after a serious of discussions between the committee members and fuel fabricators that were consulted:

- The carbon content has been raised from 550 µg/gU to 800 µg/gU
- Cobalt is limited with 10 instead of 5 µg/gU
- The Manganese limit was raised from 25 to 50 µg/gU
- A higher silicon content of  $250 \,\mu g/gU$  instead of  $100 \,\mu g/gU$  is allowed
- The permitted vanadium content has been elevated by the factor of 3 from 10 to  $30 \mu g/g$

As commented by RWE NUKEM during the RERTR meeting in Sao Paulo, the Committee members accepted to add a limitation for the total impurities of  $1500 \,\mu g/gU$  which meets the requirements of the most known customer specifications.

Following RWE NUKEM's proposal, another step forward was made to add a boron equivalent (EBC) to the specification. For research reactor use, the total boron equivalent shall not exceed 4.0  $\mu$ g/gU. The list of elements to be considered for the EBC shall be agreed upon the buyer and the seller. The limit of 4.0  $\mu$ g/gU for the EBC seems to be a little bit on the high side, but the parties in the fuel cycle may consider that as a reasonable value.

#### 8. Summary

After many years of discussions a standard specification with the designation C 1462 could be concluded by the members of ASTM Committee C-26. As a unified specification it covers all uranium products used for research reactor fuel element production including uranium derived from reprocessing and downblending of spent HEU. This "one for all" specification provides the advantage to all of us to use one specification for our research reactor uranium only. The limits set in this specification are agreed upon between all major manufacturers, reflecting the current needs for such specification. However, if the manufacturers and fuel customers find out that some values needs to be corrected, everyone of us is invited to give their comments to the members or directly to the ASTM Committee C-26. In case of no urgent need for revision, revisions are normally made every five years in order to keep standards updated.

#### 9. References

[1] ASTM, Uranium Specification for Uranium Metal Enriched to More than 15 % and Less Than 20 %  $^{235}\mathrm{U}$ 

[2] Juergen Laucht, A New Standard for Uranium Metal Intended for Research Reactor Fuel Fabrication, RERTR Conference, Sao Paulo, 1998



# THE REVISED IAEA REGULATIONS AND THEIR IMPACT ON THE TRANSPORT OF RESEARCH REACTOR FUEL

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# ABSTRACT

The IAEA Transport Regulations are the basis for most shipments of radioactive material. During the year 2001 the requirements of the 1996 revised edition have been implemented into international and national regulations. The revised version contain new and more stringent requirements concerning radiation protection, package performance and criticality safety in case of air transport. Whereas the transport of spent MTR fuel remains practically unaffected, there might be restrictions of package capacity in case of air shipments of fresh MTR fuel. In future, the IAEA transport regulations will be reviewed at 2 years intervals. Attempts are made to avoid a negative impact on the validity of package approvals.

#### 1. Introduction

The IAEA "Regulations for the Safe Transport of Radioactive Materials" are recommendations which have been developed by experts from IAEA and its member states since the early sixties. They are reviewed and revised from time to time in order to reflect the evolution of safety philosophy and technology.

The latest revision, published in 1996 (initially called ST-1, then TS-R-1) replaces the 1985 version (called Safety Series No. 6). During the year 2001 the requirements of this edition have been introduced and put into force by national authorities and international organisations in the framework of their regulations for the transport of dangerous goods. Since the beginning of 2002 the revised version is mandatory for international shipments by air and sea as well as for transport by road and rail in Europe.

There are some countries where the implementation of the last revision is delayed. Among them are the USA and Russia. Nevertheless these countries accept the application of the new regulations in case of import/export shipments.

#### 2. Features of the new regulations

Numerous modifications and new features are built into the new version. The most important ones are listed and commented below.

There is a requirement for all organisations involved in the transport of radioactive material to establish a "Radiation Protection Program". At the same time the annual dose limit for transport workers without individual dose monitoring has been drastically reduced. Whereas this is not a problem for companies specialised in the transport of radioactive cargo, there is concern among carriers who only occasionally ship such materials. They need and they will receive assistance from their contractors in implementing such programs. For the first time special packaging requirements have been introduced for uranium hexafluoride to assure that the chemical hazard of this material is properly taken into account. Most requirements are already met by the existing packagings but a new fire test for packages containing natural or depleted UF6 will make additional fire protection means necessary in the future.

In view of the consequences of air transport accidents with high toxicity radioactive materials a new package type has been introduced, the so-called type C package. It has to not only to survive the well-known type B tests, but in addition a high speed impact test and an extended fire test. This package type will for example be necessary for air transport of plutonium products in industrial quantities.

The review of air transport conditions also lead to new requirements concerning the criticality safety analysis of packages carrying any fissile material by air. It has to be assumed that the package is exposed to the additional tests mentioned above to assure criticality safety also in cases of severe and sudden geometrical changes. Water in-leakage has not be considered.

As concerns so-called low weight/ low density packages, the crush test requirement is now applicable to all packages containing fissile material, not only to packages for the transport of plutonium products.

#### 3. Impact on the transport of fresh MTR fuel

There exist packagings for the transport of fresh fuel which are in the affected low weight/low density range. During a transition period of 3 years such packages can continue to be used for surface transport (road, rail, sea). In the meantime it has to be investigated by tests and/or calculations if the existing packages are able to meet the crush test requirements or if they have to be modified.

There is no transition period for the requirement of assuming the new air transport accident scenario concerning the criticality safety. This requirement is in force since 1 July 2001. For existing package designs new criticality safety calculations have to be performed taking into account conservative assumptions concerning the expected damage from the additional test criteria ( It is probably too expensive to perform the tests themselves ). Depending on the package design features the outcome could be a reduction of capacity compared to the previous package approval.

#### 4. Impact on the transport of spent MTR fuel

Packages and the transportation of spent fuel including MTR spent fuel are not affected, as the relevant regulatory requirements are not modified in a negative sense.

#### 5. Future revisions of the IAEA regulations.

In 1998 the IAEA decided to introduce a 2-years review/revision cycle compared to the previous revision intervals of about 10 years. Background of the new procedure is to bring the revision process in line with the revision intervals of the UN requirements for the transport of dangerous goods.

At the moment the review/revision of the IAEA 1996 edition is in an advanced stage. The next edition will be published in 2003. As far as can be judged today this edition will not present substantial changes. The next review cycle will start soon with the aim of publishing a further edition in 2005.

In the opinion of the affected industry the new revision cycle of the transport regulations will have a negative impact on package licensing, package approval continuity and on the validation of foreign package approvals. But even more important, it will bring an element of uncertainty concerning the investment into the design and manufacturing of new packages. Fortunately the problems are now understood and efforts are on the way to find reasonable solutions.

# **Session 2**

Fuel development, qualification, fabrication and licensing

#### **RERTR PROGRESS IN MO-99 PRODUCTION FROM LEU**

by

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#### RERTR PROGRESS IN MO-99 PRODUCTION FROM LEU

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#### ABSTRACT

The ANL RERTR program is performing R&D supporting conversion of <sup>99</sup>Mo production from HEU to LEU targets. Irradiation and processing of LEU targets were demonstrated at the Argentine Ezeiza Atomic Center. Target irradiation and disassembly were flawless, but the processing is not fully developed. In addition to preparing for, assisting in, and analyzing results of the demonstration, we performed other R&D related to LEU conversion: (1) designing a prototype production dissolver for digesting irradiated LEU foils in alkaline solutions and developing means to simplify digestion, (2) modifying ionexchange columns used in the CNEA recovery and purification of <sup>99</sup>Mo to deal with the lower volumes generated from LEU-foil digestion, (3) measuring the performance of new inorganic sorbents that outperform alumina for recovering Mo(VI) from nitric acid solutions containing high concentrations of uranium nitrate, and (4) developing means to facilitate the concentration and calcination of waste nitric-acid/LEU-nitrate solutions from <sup>99</sup>Mo production.

#### 1. Introduction

To reduce nuclear-proliferation concerns, the U.S. Reduced Enrichment for Research and Test Reactors (RERTR) Program is working to limit the use of high-enriched uranium (HEU) by substituting low-enriched uranium (LEU) fuel and targets. Low-enriched uranium contains  $<20\%^{235}$ U. Technetium-99m, the daughter of <sup>99</sup>Mo, is the most commonly used medical radioisotope in the world. Currently, most of the world's supply of <sup>99</sup>Mo is produced by fissioning the <sup>235</sup>U in HEU targets, generally 93% <sup>235</sup>U. Targets for the production of <sup>99</sup>Mo are generally either (1) miniature Al-clad fuel plates or pins containing U-Al alloy or UAl<sub>x</sub> dispersion fuel or (2) a thin film of UO<sub>2</sub> cn the inside of a stainless steel tube. After irradiation, the <sup>99</sup>Mo is recovered from the irradiated uranium and purified.

To yield equivalent amounts of <sup>99</sup>Mo, an LEU target must contain approximately five times the uranium as an HEU target. Consequently, substituting LEU for HEU requires changes in both target design and chemical processing. Three major challenges have been identified when substituting LEU for HEU: (1) modifying the targets and purification processes as little as possible, (2) assuring continued high yield and purity of the <sup>99</sup>Mo product, and (3) limiting economic disadvantages.

#### 2. Progress

Since reporting R&D results at the 2000 International RERTR Meeting [1,2], we have made progress in four R&D areas aimed at the conversion of <sup>99</sup>Mo production to LEU targets. Those areas are (1) developing and demonstrating the recovery and purification of <sup>99</sup>Mo from an LEU target in Argentina, (2) developing a production dissolver for digesting irradiated LEU foils by alkaline solution, (3) testing new inorganic sorbents for their ability to recover molybdenum from acidic uranium nitrate solutions, and (4) developing means to facilitate calcination of acidic uranium nitrate waste solutions.

<u>Processing of Irradiated LEU Foils by CNEA</u> A number of current producers dissolve/digest uraniumaluminide/aluminum-dispersion plates in alkaline solution as an initial step to recovering fissionproduced <sup>99</sup>Mo from irradiated HEU. These producers include Argentine Comisión Nacional de Energía Atómica (CNEA), Institut National des Radioéléments (IRE), Mallinckrodt, and the South African Nuclear Energy Corporation Limited (NECSA). Argonne National Laboratory (ANL) is actively cooperating with one of these producers, CNEA, to convert its process to low-enriched uranium (LEU). The CNEA process has been described in the literature [3] and has much in common with the Mallinckrodt process; both processes are based on that developed by A. Sameh [4]. In this process, the irradiated targets are heated in sodium hydroxide solution. The aluminum cladding and meat in the targets are dissolved to form soluble sodium aluminate, and the uranium is digested, forming a mixed precipitate of  $UO_2$  and  $Na_2U_2O_7$ . Molybdenum is soluble in alkaline solutions as the molybdate ion, but the actinides and many of the metallic fission products precipitate as hydroxide salts. Following filtration, the filtrate is fed to an anion-exchange column, which retains molybdenum and some other anionic species. After it is eluted from this column, the molybdenum is purified to meet pharmaceutical standards by a series of separation processes.

The LEU-modified process begins with a two-step digestion of irradiated LEU foils. Because the digestion of LEU foils generates less than 10% of the solution volume of that from dissolving HEU targets, the size of the primary Bio-Rad AG MP-1 (hydroxide form) and Bio-Rad Chelex 100 anion-exchange columns are significantly smaller than for the current HEU process. This advantage creates far less liquid waste and cuts processing time considerably; however, development is required to downsize equipment and specify process conditions. Early in 1999, ANL and CNEA began active cooperation with the goal of enabling CNEA to convert to LEU at the end of three years. It is a multifaceted program, and many steps are required to modify targets and the current process to allow the use of LEU targets. Progress during 2000 was described earlier [1]. Irradiations and process demonstrations were performed in December 2000 and May 2001.

Two targets containing four foils were irradiated May 3-8, 2001, for ~120 hours at a flux of 4-6x10<sup>13</sup> n/cm<sup>2</sup>/s. Following irradiation, the targets were allowed to cool in the reactor pool for 10 hours and then transported to a processing hot cell in the <sup>99</sup>Mo Production Facility. After arrival at the <sup>99</sup>Mo production facility, the targets were disassembled and inspected. An irradiated 9-g LEU foil with a 40-µm aluminum fission recoil barrier was processed to recover <sup>99</sup>Mo using the two-step alkaline digestion and a slightly modified anion exchange process. The foil was loaded into the dissolver. The atmosphere inside the dissolver was evacuated, and 50 mL of sodium hydroxide solution was injected. The dissolver was then heated to dissolve the aluminum barrier and to convert the uranium foil into a solid uranium oxide product. During this initial digestion step, the temperature of the dissolver was controlled to limit the pressure in the dissolver to 700 psig (4.8 MPa). After 45 minutes, the dissolver was cooled, and hydrogen gas and the released fission gasses were vented to a vacuum tank, where they were stored until the fission gases decayed. To ensure that all of the <sup>99</sup>Mo was released to the sodium hydroxide solution, a second digestion step was performed by pressurizing the dissolver with 100-psig (0.7-MPa) oxygen and heating the dissolver again. This step converts the solid uranium dioxide into a solid sodium diuranate product; it also converts iodide to iodate, which sorbs far less strongly on the AG MP-1 column. Again, the temperature was controlled to limit the pressure in the dissolver to 700 psig (4.8 MPa). Following digestion, the suspension in the dissolver was filtered to separate the solid sodium diuranate product and alkaline-insoluble activation and fission products from the solution containing <sup>99</sup>Mo and other alkaline-soluble fission products.

The initial digestion of the foil and the oxygen conversion of the oxide proceeded smoothly. We used a sintered metal filter that CNEA provided to filter the suspension from the digester. The solution passed easily through the filter; however, some solids appeared in the filtrate. We are not sure what the composition of the solids was (i.e., precipitated uranium that passed through the filter or corrosion products from the sintered metal filter) or what impact they may have had on the subsequent ion exchange process. However, the solids appeared to collect in the glass-wool packing at the top of the anion exchange column. Following filtration, the <sup>99</sup>Mo (as  $MOQ_4^{2-}$ ) in the 175 mL of solution (dissolving solution plus dissolver rinses) was recovered on the AG MP-1 anion exchange column, which was considerably smaller than that required for an equal amount of <sup>99</sup>Mo from dissolving the current HEU targets. Any I<sup>-</sup> in the feed solution would also be sorbed. Wash solution was then passed through the column to remove impurities. Less than 0.03% of the <sup>99</sup>Mo escaped the column during loading and rinsing. The <sup>99</sup>Mo was eluted from the column in 72 mL of solution, which was then prepared for the second <sup>99</sup>Mo-purification step using Chelex 100. Because of the considerably smaller than that used in HEU-target processing. The strip-solution volume for removing molybdenum from the Chelex 100 column was 50 mL. The yield and purity of the molybdenum effluents in both columns were measured by gamma spectroscopy. Results were qualitatively as expected, but problems associated with sample dilution preclude a quantitative description of results. The next CNEA demonstration is planned for Spring 2002. For this demonstration, a new prototype production dissolver has been designed and fabricated at ANL and is being tested. Studies are underway to size both the AG MP-1 and Chelex 100 columns and set maximum flow rates through the columns. We are also developing a one-step digestion process using potassium permanganate to oxidize uranium to U(VI) and iodine to iodate.

<u>Prototype Dissolver for CNEA Production</u>: To allow for early testing of the alkaline digestion of irradiated LEU foil at CNEA, we used the ANL dissolver designed for nitric-acid dissolution of foil in the modified Cintichem process demonstrations performed in Indonesia [5]. Although the design was awkward to use in the CNEA hot cells, it did allow us to test the digestion and processing of LEU foils. Early work showed that the material of construction, 304 stainless steel, undergoes minimal corrosion when contacted with sodium-hydroxide solution alone, however, when the reaction was run with 100-psig (0.7-MPa) oxygen overpressure, corrosion was severe. A series of corrosion tests was conducted to compare the corrosion rates of 304 stainless steel, Hastelloy C-276, and Inconel 600 during the second digestion step of the LEU target dissolution (i.e., in the presence of oxygen). These tests were conducted in a vessel, containing one of the coupons, sodium hydroxide solution, and 100-psig (0.7 MPa)  $O_2$ , that was heated to ~250°C (470 psig total pressure) for about 100 hours. After the test, the coupon was removed, rinsed, and dried.

Visual examination of the corroded coupons showed that the 304 stainless steel coupon was much more affected by these conditions than either the Hastelloy C-276 or the Inconel 600 coupon. The corroded 304 stainless steel coupon had developed a thin red/brown layer in the liquid phase, a somewhat thicker layer in the vapor phase, and a much thicker layer at the vapor-liquid interface. In addition, significant pitting was observable in the interface region. The corrosion of the welded metal was similar to that of the rest of the coupon. Examination of the corroded 304 stainless steel coupon with scanning electron microscopy (SEM) showed that the maximum thickness of the oxidation layer (at the vapor-liquid interface) varied from 50 to 100  $\mu$ m. Oxidation-layer thickness developed in the liquid phase varied from 10 to 20  $\mu$ m, and that in the vapor phase varied from 20 to 50  $\mu$ m.

The corroded Hastelloy coupon had developed a very thin green-brown layer in the vapor phase, a thin gray layer at the vapor-liquid interface, and no observable layer in the liquid phase. The corroded Inconel 600 had developed a very thin yellow/brown layer in the vapor phase, a thin gray layer at the vapor-liquid interface, and no observable layer on the section exposed to the liquid phase. The oxidation layers on the corroded Hastelloy C-276 and Inconel 600 coupons were not observable with SEM and, therefore, were  $<1\mu$ m. Welded metal showed the same corrosion as the rest of the coupon.

Based on our observations, the corrosion rate of the 304 stainless steel would be as much as 1  $\mu$ m/hr during the second step of foil digestion. On the other hand, the corrosion rates for the Hastelloy C-276 and Inconel 600 under the same conditions would be <0.01  $\mu$ m/hr. Therefore, 304 stainless steel is not an acceptable material of construction for the two-step digestion; however, both Hastelloy C-276 and Inconel 600 are acceptable.

The design of the production dissolver is being developed. To allow testing of various operations in a hot-cell environment, a prototype was fabricated from 304 stainless steel. Heating will be accomplished using heat-tape tracing; cooling will be performed by blowing air. Figure 1 is a photograph of the prototype body and a schematic of the entire unit. This unit will be able to accommodate  $\sim$ 70 g of LEU foil ( $\sim$ 14 g of <sup>235</sup>U) and has five lateral connections for accommodating pressure gauges, a pressure relief valve, a vacuum port and solution inlets. The bottom drain will be connected to a ball valve and filter. The top has a bayonet fitting for ease of closure. The unit is designed and has been tested to hold up to 1500 psig (10.3 MPa) pressure at 300°C.

The two-step process is effective but (1) having two warm-up, reaction, and cool-down periods doubles digestion time and (2) using pressurized oxygen leads to safety concerns. Therefore, we are

testing the addition of KMnO<sub>4</sub> to the digestion solution to perform the oxidation to U(VI) and I(V). Thus far, results have been very promising. Adding potassium permanganate dissolved in sodium or potassium hydroxide solution converts all the uranium to an alkali diuranate, iodide is oxidized, and molybdate is not sorbed on the MnO<sub>2</sub> precipitate that is generated. Further tests will be performed with low-burnup uranium foils and then with fully irradiated targets in Argentina.



Fig. 1. Prototype and Conceptual Design cf ANL-Designed LEU-Foil Digester

<u>CNEA-Process Column Sizing</u>: R&D is being undertaken to size the AG MP-1 and Chelex 100 columns and to set optimum conditions for their use in the LEU process. The molybdate-sorption kinetics of AG MP-1 are extremely fast. Equilibrium in static tests is reached in 15-30 minutes. The partitioning coefficient for Mo(VI) is inversely dependent on the hydroxide concentration and will be in the 100-200 mL/g range for hydroxide concentrations contemplated for the process; AG MP-1 has a 2.3 meq/g loading capacity for molybdate. Distribution ratios for MoO(SCN)<sub>5</sub><sup>2-</sup> on Chelex 100 under process conditions are extremely high ( $\geq 10^7$  mL/g) once the Mo(VI) has been reduced and complexed. The conversion is relatively slow at 22°C, taking 80 minutes to reach completion. Future work will measure the effects of temperature on this reaction. All of these activities will be completed for implementation during the CNEA demonstration in spring 2002.

<u>Mo-Specific Sorbents for Solutions with High Corcentrations of HNO<sub>3</sub> and UO<sub>2</sub>(NO<sub>3</sub>)<sub>2</sub></u>: Two inorganic sorbents were tested for their ability to recover molybdenum from nitric acid solutions with significant concentrations of uranyl nitrate. These patented sorbents were produced in Russia for Technology Commercialization International Inc. (TCInternational). They are 0.4-1.0 mm spherical particles with high mechanical strength. Reported below are the results of our initial screening of these two materials. In the first series of tests, the effects of contact time, nitric acid concentration, and uranyl nitrate on the partitioning coefficient of <sup>99</sup>Mo (K<sub>d</sub>, mL/g) were studied for these two sorbents (designated as R-1 and R-2) and compared with alumina (Aldrich aluminum oxide, activated, acidic, Brockmann 1, ~150 mesh). From data presented in Table 1, several observations can be made:

- Both R-1 and R-2 have higher affinity for Mo(VI) than does alumina at all solution compositions and contact times.
- Their affinity is especially higher from solutions of concentrated uranium.
- When the concentration of uranium is high, the R-2 sorbent has ~100 times higher affinity for Mo(VI) than does alumina.
- Sorbent R-1 appears to have better kinetic properties than do R-2 and alumina, and thus will allow faster feed flow rates through the column.

In separate column tests, we found that molybdenum is easily stripped from these sorbents in the same manner used for alumina. The loading capacity for both sorbents is  $\sim 3 \text{ meq/g}$ . Based on these preliminary results, we feel that use of either of these materials should (1) allow a more efficient

recovery of molybdenum than the current alumina column and (2) generate a waste-solution volume for LEU processing equal to, or perhaps less than, that for current HEU target processing.

Table 1. Partitioning coefficient ( $K_d$ ) for sorption of tracer <sup>99</sup>Mo for three sorbents vs. contact time and concentrations of nitric acid and uranyl nitrate. All contacts were performed at 22-25°C while test tubes containing 2 mL of solution and 0.1 g of sorbent were slowly rotated to provide mixing.

		1	Sorbent R-2			Sorbent R-1			Al <sub>2</sub> O <sub>3</sub>	
Conditions			Kd(Mo) mL/g			Kd(Mo) mL/g			Kd(Mo) mL/g	
[HNO3], M	[U], g/L	4 hr	24 hr	48 hr	4 hr	24 hr	48 hr	4 hr	24 hr	48 hr
1	0	9.79E+03	6.33E+04	1.80E+05	1.04E+04	4.88E+04	4.38E+04	4.87E+02	3.52E+03	6.83E+03
2	0	4.97E+03	2.95E+04	9.31E+04	5.76E+03	2.77E+04	3.03E+04	9.63E+01	7.46E+02	1.41E+03
4	0	2.18E+03	1.79E+04	3.39E+04	2.47E+03	6.75E+03	1.07E+04	2.43E+01	3.00E+01	2.92E+02
1	80	2.76E+03	2.28E+04	5.06E+04	4.09E+03	2.04E+04	1.49E+04	4.19E+01	2.92E+02	6.11E+02
2	80	1.99E+03	1.54E+04	3.40E+04	2.19E+03	9.62E+03	1.15E+04	1.77E+01	1.11E+02	1.87E+02
4	80	1.11E+03	1.17E+04	2.18E+04	1.40E+03	1.03E+04	5.69E+03	9.32E+00	4.88E+01	8.91E+01
1	160	9.07E+02	7.10E+03	1.42E+04	5.28E+02	4.80E+03	4.25E+03	6.99E+00	5.55E+01	6.07E+01
2	160	5.88E+02	4.93E+03	1.09E+04	3.76E+02	2.39E+03	2.38E+03	4.85E+00	4.43E+01	4.32E+01
4	160	4.70E+02	3.43E+03	8.08E+03	2.48E+02	1.38E+03	1.29E+03	3.33E+00	5.16E+01	3.64E+01
1	320	1.98E+02	1.66E+03	2.93E+03	7.56E+01	2.21E+02	2.88E+02	2.20E+00	3.27E+01	3.27E+01
2	320	1.16E+02	1.03E+03	1.48E+03	3.59E+01	1.33E+02	9.49E+01	1.96E+00	6.38E+01	2.87E+01
4	320	1.23E+02	1.18E+03	1.68E+03	3.29E+01	1.76E+02	8.51E+01	1.73E+00	2.63E+01	2.83E+01

<u>Chemically Facilitated Calcination of Uranium-Nitrate/Nitric-Acid Waste Solutions</u>: Argonne is cooperating with MDS Nordion, Atomic Energy of Canada Ltd. (AECL), and SGN to develop technical means for converting <sup>99</sup>Mo production at Chalk River, Ontario, Canada to LEU [6]. When LEU targets are dissolved in nitric acid, the dissolver solution will contain approximately five times more uranium than HEU targets for the same <sup>99</sup>Mo yield. Consequently, the waste solution from initial molybdenum recovery will have far higher concentrations of uranium and perhaps a greater volume than that generated from the current process. For disposal, the waste solution must be stabilized as a solid. Direct calcination to a uranium/fission-product oxide product has been accepted as a suitable long-term storage form [6,7]. The technical challenge being addressed is developing means to use the same equipment and facilities to deal with LEU waste as were designed to treat HEU waste. The barrier(s) in concentrating and calcining these solutions are:

- Higher evaporation rates may be required (if LEU conversion increases waste-solution volume).
- Following evaporation of liquids, formation of approximately five times more UNH (uranyl nitrate hexahydrate) will test the capacity of the equipment and storage containers.
- Dehydration and denitration of molten UNH can be messy. Pairing this with its larger volume in the case of LEU targets makes contamination of equipment a great concern.

The path Argonne chose to address these potential problems is to (1) study the limits of direct evaporation/calcination of the nitric-acid/UNH solution and (2) use chemical means (precipitation of uranyl oxalate) to eliminate the formation of UNH. Assisted calcination with oxalic acid has been attempted by two means.

In the first method, the uranium-nitrate/nitric-acid solution and a saturated oxalic-acid solution (at a 1.1 oxalate/uranyl mole ratio) are simultaneously fed to the calciner at the same rate as liquid evaporates. Experiments begin with an initial charge of 100 mL of the uranium solution.

In the second method, the calciner cup is initially filled with solid oxalic acid and 100 mL of the nitricacid/UNH solution. The uranium solution is fed to the calciner at the same rate that condensate is collected until 1 mole of uranium is added per 1.1 mole of oxalate already in the cup. Both methods produce the same product, a mixture of  $UO_2$  and  $U_3O_7$ . The material is easily removed from the cup in the form of powder.

Addition of the aqueous 1 <u>M</u> oxalic acid solutions requires significantly more evaporation than adding solid oxalic acid; thus, adding solid  $H_2C_2O_4 \cdot 2H_2O$  to the cup before startup is the preferred option. Argonne is developing a chemical and engineering understanding of both direct and oxalate-assisted

calcination using a simplified calciner system, glassware studies, and solid, liquid, and off-gas analyses. This information will allow SGN to develop a testing program to pilot these processes in a duplicate of the calcining unit in place at the New Processing Facility at Chalk River.

#### 3. CONCLUSIONS AND FUTURE WORK

Argonne continues to partner with <sup>99</sup>Mo producers to convert targets from HEU to LEU. The cooperation with CNEA is on schedule, and we are planning additional demonstrations in spring 2002 for all aspects of production--from target fabrication to the final <sup>99</sup>Mo product. Our work on the Russian sorbents for molybdenum recovery will continue; we are looking for a partner to test them under production conditions. Our R&D on facilitated calcination has not yet demonstrated an assured winner or loser; pilot testing by SGN will be required to make that decision. The next phase of development work is being discussed with our MDS Nordion partners; Argonne research will support the pilot demonstrations by SGN. The extremely successful cooperation with Indonesia, which is nearing completion, was inactive in 2001. As soon as the world political situation improves, we will visit our colleagues in Indonesia to observe them (1) demonstrate that LEU-produced <sup>99</sup>Mo will achieve the same <sup>99m</sup>Tc yield and purity from a generator as HEU-produced <sup>99</sup>Mo and (2) fabricate LEU-foil targets for irradiation. We will continue to pursue the building of active programs with other <sup>99</sup>Mo producers.

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# AN UPDATE ON THE LEU TARGET DEVELOPMENT AND CONVERSION PROGRAM FOR THE MAPLE REACTORS AND NEW PROCESSING FACILITY

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## ABSTRACT

Historically, the production of molybdenum-99 in the NRU research reactors at Chalk River, Canada has been extracted from reactor targets employing highly enriched uranium (HEU). A reliable supply of HEU metal from the United States used in the manufacture of targets for the NRU research reactor has been a key factor to enable MDS Nordion to develop a secure supply of medical isotopes for the international nuclear medicine community. The molybdenum extraction process from HEU targets provides predictable, consistent yields for our high-volume molybdenum production process. Each link of the isotope supply chain, from isotope production to ultimate use by the physician, has been established using this proven and established method of HEU target irradiation and processing to extract molybdenum-99.

To ensure a continued reliable and timely supply of medical isotopes, MDS Nordion is completing the construction of two MAPLE reactors and a New Processing Facility. The design of the MAPLE facilities was based on an established process developed by Atomic Energy of Canada Ltd. (AECL) – extraction of isotopes from HEU target material. However, in concert with the global trend to utilize low enriched uranium (LEU) in research reactors, MDS Nordion has launched a three phase *LEU Target Development and Conversion Program* for the MAPLE facilities. Phase 1, the Initial Feasibility Study, which identified the technical issues to convert the MAPLE reactor targets from HEU to LEU for large scale commercial production was reported on at the RERTR- 2000 conference.

The second phase of the *LEU Target Development and Conversion Program* was developed with extensive consultation and involvement of experts knowledgeable in target development, process system design, enriched uranium conversion chemistry and commercial scale reactor operations and molybdenum production. This paper will provide an overview of the Phase 2 Conversion Development Program, report on progress to date, and further discuss the challenges in converting the MAPLE facilities to molybdenum production from LEU targets.

#### 1. Introduction

It is important to step back and consider the context in which MDS Nordion's MAPLE facility will operate. MDS Nordion is the world's leading supplier of medical isotopes. The company is in the business of supplying isotopes used to conduct some 34,000 nuclear medicine procedures every day around the world, such as determining the severity of heart disease, the spread of cancer and diagnosing brain disorders.

There are over 100 medical applications for radioisotopes and some 80% of nuclear medicine procedures rely on just one isotope, molybdenum-99. Moreover, some of these procedures are performed using medical isotopes as soon as 41 hours after leaving the reactor. This is a real just-in-time business. As the radioisotope decays, MDS Nordion must get the product to the customer as quickly as possible. This is also a global endeavor. For example, some 5000 hospitals in North America depend on this supply each week; in Germany, its 850 hospitals; in Japan, its over 1000 hospitals and in Argentina, over 250 hospitals rely on our supply.

MDS Nordion's medical isotope business is also providing an exciting new platform in radioimmunotherapy. For example, novel ways to use a radioisotope to treat disease, such as for non-Hodgkin's lymphoma, a blood-borne cancer, are being developed. This exciting platform will expand the horizon for applications of medical isotopes.

At the end of the day, the MAPLE story, its planning, construction and the development work relating to LEU conversion, is about securing the supply of medical isotopes required by the international nuclear medicine community, and, ultimately, the thousands of patients who expect to receive their daily medical procedures.

#### 2. The MAPLE Facilities – Securing the World Supply of Medical Isotopes

#### Why build the MAPLE facilities?

There are compelling reasons to build the MAPLE facilities. MDS Nordion supplies the majority of the world's isotopes. Notably today molybdenum-99 is the most extensively used isotope. However, new medical techniques are providing opportunities for iodine-131, and the utilization of iodine-125 and xenon-133 are growing. The NRU reactor owned by AECL at Chalk River has been operating since 1957 and producing molybdenum since the early 1970's. Today NRU also supplies other medical isotopes, including cobalt-60.

Molybdenum-99 supply in the pre-1980 era was supported by four capable suppliers: Cintichem and GE in the United States, IRE in Belgium, and MDS Nordion in Canada. MDS Nordion obtained our medical isotopes in the NRU and NRX reactors, owned by AECL. Some of the reactors used for isotope production have encountered insurmountable circumstances that resulted in their permanent shutdown. The GE reactor was found to be located on a seismic fault and it was shutdown in 1980; the Cintichem reactor experienced a contamination incident that resulted in its permanent shutdown in 1990; the NRX reactor in Canada was shut down in March 31, 1993 due to safety concerns. The global nuclear medicine community had serious concerns about supply reliability, which was created by the shutdown of these reactors, as there was no viable backup supply of medical isotopes.

A number of initiatives to broaden molybdenum-99 supply were taken. Mallinckrodt established a contract with HFR at Petten with MDS Nordion's help; IRE increased its production capacity to provide a larger volume back up; NECSA in South Africa began to produce molybdenum-99 in 1993. None-the-less, the concern of a supply shortage continued. If NRU were to shutdown, the nuclear medicine

community would not be able to obtain the volume of molybdenum-99 that it required from the aggregate of other producers.

In response to concerns the nuclear medicine community had about the long term, secure supply of molybdenum-99, in 1996 MDS Nordion and AECL announced an agreement that will ensure reliable and economic availability of radioisotopes to hospitals and clinics worldwide. The agreement provided for construction of two MAPLE reactors and a high volume, commercial, first stage processing facility at AECL's Chalk River Laboratories. MDS Nordion will own the reactors and processing facility and be responsible for managing the business and developing the isotope production planning activities. AECL have been contracted to design, build, and operate the facilities on behalf of MDS Nordion. The MAPLE reactors will be the only reactors in the world totally dedicated to the commercial production of medical radioisotopes. The significant investment being made by MDS Nordion at the AECL site at Chalk River, will capitalize on the extensive infrastructure, expertise and experience of AECL and MDS Nordion for the reliable, continuous supply of isotopes.

#### What are the MAPLE Facilities?

The MAPLE Facilities consist of two reactors and a processing facility to extract isotopes and manage the process waste. The MAPLE reactors are 10 Mw, open pool, light water reactors. The reactor has a compact core, about the size of a 90 litre (20 gallon) drum, and is surrounded by heavy water reflector tank. The reactor assembly consists of five major components: the inlet plenum; the grid plate; the core structure consisting of vertical flow tubes containing low enriched uranium (LEU) driver fuel bundles and high enriched uranium (HEU) target assemblies for medical isotope production; the heavy water reflector tank; and the chimney. The light water primary coolant enters the inlet plenum, flows upward through the grid plate, the flow tubes and fuel and target assemblies, and is directed back to the suction of the primary cooling pump via the outlet arms of the chimney.



Figure 1. Reactor Vessel

The MAPLE reactor is composed of 13 hexagonal and six circular flow tubes. Four of the 13 hexagonal flow tubes are used for irradiating HEU targets; the remaining nine contain 36 element LEU driver fuel assemblies. The six circular flow tubes contain 18 element driver fuel assemblies.

Two reactors were built to ensure a secure and continuous supply of medical isotopes. Isotopes will be produced in one reactor while the other is being maintained and on stand-by for its next production run.

The operating cycle of each reactor will have a minimum operating period of up to four weeks depending on isotope demand. HEU targets irradiated in the MAPLE reactors will be transferred in shielded containers to the processing facility for isotope extraction. The radioactive waste from the extraction process will be solidified within the processing facility and transferred to the waste management area on the Chalk River site for storage in concrete canisters.

The New Processing Facility (NPF) will extract the radioisotopes produced in the HEU feedstock target material, process the residuals, and transfer the product to containers for shipment to MDS Nordion's Ottawa facility. There, the isotopes are further processed, packaged, and distributed to MDS Nordion's nuclear medicine customers around the world. The timeline from start of target processing to product delivery to the hospital can be as little as 41 hours.

Overall, completion of the MAPLE project was planned to be about fifty months in duration. These new, one-of-a-kind facilities had several challenges to meet during the execution of the project. Advanced technology, a new licensing environment and a compressed schedule created challenges in licensing, design, construction and commissioning. Construction of the facilities had to take place on a crowded site. Existing buildings had to be removed and the ZEEP reactor had to be decommissioned and dismantled. So far several key milestones have been achieved.

•	Project Start	September	1996
•	Environmental Approval	April	1997
•	Construction Approval	December	1997
•	MAPLE 1 Reactor		
	- Operating License	August	1999
	- First Sustained Nuclear Reaction	February	2000
•	New Processing Facility		
	- Operating License	August	1999
•	MAPLE 2 Reactor		
	- Operating License	June	2000

Figure 2. MAPLE Project Key Milestones

An environmental assessment for the facilities was completed in April 1997 and construction approvals for these facilities were granted in December 1997. MAPLE 1 achieved its first sustained nuclear reaction on February 19, 2000. This was a significant milestone in the project. The MAPLE 1 reactor had achieved all of its performance objectives at the 2 kW level and power was increased to the 500 kW level. Also, the operating license for MAPLE 2 was received in June 2000. During the past eighteen months, there has been a pause in commissioning to address licensing issues related to the shut-off rod safety shutdown system. At a recent meeting of the Canadian Nuclear Safety Commission (CNSC), AECL received authorization to proceed with the project, subject to satisfactory completion of specific licensing pre-requisites. We are pleased that the project has resumed and commissioning is underway. Assuming satisfactory completion of the licensing issues and operating license hold points, nuclear commissioning and acceptance testing of the MAPLE 1 reactor and New Processing Facility are expected to be complete later this year.

#### 3. Converting the MAPLE Facilities to LEU Targets

Tremendous progress has already been made by AECL and MDS Nordion to convert medical isotope production to LEU based material. The MAPLE reactors were designed to operate with LEU fuel, thus achieving a significant technological advancement in conversion from HEU to LEU. This in itself has

reduced the reliance on HEU and thus makes the MAPLE technology unique at this point in time. The leadership taken by AECL and MDS Nordion to use LEU fuel in the MAPLE reactors for commercial production of medical isotopes is a substantial accomplishment in the Reduced Enrichment for Research and Test Reactors (RERTR) program.

HEU target technology is an integral part of the reactor operating system. It is a proven and reliable process for molybdenum production in the reactors operated by all commercial isotope producers. Predictable, consistent yields of molybdenum from HEU targets are the foundation for a reliable supply. Furthermore, all of the requisite licensing has been approved by national nuclear regulators and by health care regulators such as the U.S. Food and Drug Administration (FDA), and European national authorities. These links provide a secure chain of medical isotopes for the international nuclear medicine community. For this reason, the isotope production process has been designed using HEU targets clad in zirconium alloy.

Although HEU target technology is well established and consumes a relatively small quantity of material, proliferation concerns have caused significant international interest in reducing reactor reliance on HEU feedstock material. Today, the only known commercial sources of HEU are the United States and Russia. South Africa has their own supply of HEU used for molybdenum-99 production. Concerns with international safeguards and non-proliferation of HEU material have caused the U.S. to enact legislation in that will encourage reactor operators to convert to LEU and create increased difficulty for non-U.S. organizations to access HEU from the United States. Known as the Schumer Amendment, changes were made to the Atomic Energy Act in 1992 that imposed restrictions on exports of HEU fuel and targets from the United States, with certain qualifying statements related to the technical and economic viability of converting to LEU.

Furthermore, there is today increasing difficulty in the logistics of sourcing HEU material worldwide. Generally, HEU material is government controlled, lead-time to access material is long, and the reliability of supply is uncertain. The process of obtaining an export license is complicated and protracted. All of the above has created a public perception that favours the use of LEU material. This has caused MDS Nordion to initiate a program to examine the feasibility of, and address the issues to converting those reactors and their associated processing facilities to operate and process LEU targets. A significant challenge will be to convert the MAPLE facilities from HEU to LEU targets while they are operating; to ensure there is no disruption in the supply of molybdenum-99 and other radioisotopes used annually in millions of essential medical procedures.

To convert the MAPLE facilities to LEU targets in a logical manner, MDS Nordion established a threephase *LEU Target Development and Conversion Program*. They are:

- Initial Feasibility Study
- Conversion Development Program
- Conversion Implementation Program

At the completion of each of the first two stages, an assessment will be done of the technical and economic issues that will be involved in the implementation phase.

It is important to understand that the *LEU Target Development and Conversion Program* must be accomplished within certain conditions. The first condition is that there must be minimum change to the MAPLE reactor design and operation, as well as the downstream processing system, all of which have been designed and built based on HEU target technology. A second condition is that isotope production capacity must be maintained to ensure the production capability exists to meet market demand. As a consequence of these two conditions, the same number of targets will be used. As a

result of this, the mass of uranium in the LEU targets will be 4.7 times greater than in an HEU target. The processing facility must be able to handle the increased uranium mass from the LEU targets and achieve acceptable performance characteristics in the areas of uranium dissolution, molybdenum-99 recovery yields, waste solidification and waste storage. Any incremental operational burden placed on the isotope production and processing system must ensure that the rigorous equipment preventative maintenance program for these facilities is not compromised.

The Initial Feasibility Study has been completed. A design for a LEU target for the MAPLE reactors has been produced and it has been determined that operation of the MAPLE reactors with LEU targets is technically feasible. The Initial Feasibility Study also identified the key Canadian regulatory conditions that must be met to use LEU targets in those reactors. Before LEU targets may be used in the MAPLE reactors, the Canadian Nuclear Safety Commission (CNSC) must review and approve environmental assessments and safety analyses performed by AECL, including critical heat flux tests and irradiation tests. It is anticipated that a public licensing process could be carried out by the CNSC in connection with its consideration of whether the MAPLE reactors will be authorized to use LEU targets. Based on consultation with the CNSC, it is expected that completion of their licensing process could require a minimum of three years. In addition, the drug certification requirements of the FDA and the European national authorities must be satisfied for production of molybdenum-99 from a new target source material comprised of LEU.

The situation with the New Processing Facility is more challenging and complex. Assuming that LEU targets could be irradiated in the MAPLE reactors, 4.7 times more uranium than in HEU targets will be chemically processed in the NPF to extract a similar quantity of medical isotopes. This additional mass must also be calcined to solidify the waste in stable form for long-term storage. The isotope processing hot-cell system and equipment, as noted earlier, is custom designed and solely dedicated to the processing and extraction of molybdenum-99 from HEU targets. Therefore a thorough assessment of the processing facility was required to explore the technical viability of converting to LEU.

MDS Nordion commissioned an assessment of the New Processing Facility with the following three key objectives:

- determine whether the equipment designed for the NPF can process LEU targets;
- determine the production capacity with LEU targets; and
- determine changes that should be implemented prior to the introduction of radioactivity into the NPF.

The Initial Feasibility Study indicated the following key areas of concern in the New Processing Facility:

#### Liquid Waste Storage Capacity

The NPF has been built with adequate tanks to store the waste to reduce decay heat prior to the start of calcination. To improve production capacity, two areas have been identified: reduction of decay time and improvement of the calcination process.

#### Waste Calcination

The process used is a continuous batch process with the calcination taking place in a storage can that will be sealed and disposed of as solid waste. The calcination process requires the liquid from the target dissolution and alumina column washes to be metered into a can so free water can be evaporated on-line in a controlled manner. The system must operate under stable conditions to maintain process control; this will retain the residual material in the storage can, minimize sputtering which could cause fouling of the calcining equipment and minimize entrainment of the off-gas. Once the free water has been evaporated, thermal energy is applied to the residual material and a solid, cinder-like mass is left in the

storage can. The storage can is then seal-welded, loaded in a basket, and transported in a shielded container to a storage silo in the waste management area. Because of the sputtering observed after the removal of free water, the total amount of uranium in the can is the limiting factor. With greater amounts of uranium arising from processing LEU targets, there was danger of sputtering not being contained in the can and causing fouling of the calcining system. The Initial Feasibility Study confirmed that a significant number of additional waste cans would be required if the change to LEU was made and the calcination process performed in a controlled manner.

A key step in the conversion feasibility evaluation is for MDS Nordion to gain operational experience processing HEU through the calcination and waste handling system to determine any changes that could be made to manage the increased mass of uranium that will be in the LEU targets. In addition, an evaluation will be made to determine if the process could be carried out more quickly. Furthermore this facility was built with an expected useful life of 40 years. In the natural course of events it is expected that MDS Nordion and AECL will find ways to improve the process capacity of the facility to accommodate any additional process demands to meet market growth projections.

#### Solid Waste Storage

Because of the processing considerations mentioned earlier, the number of waste containers will increase. As there will be less uranium per container, the arrangement of these containers in the concrete waste storage silos may be different and it is expected that the number of silos required will be greater. This, in turn, could have an impact upon the storage capacity of the existing waste storage site. The licensing and environmental requirements to address this issue will need to be defined.

Overall, the Initial Feasibility Study demonstrated that a LEU target could be processed and a similar molybdenum-99 extraction efficiency could be obtained as when compared to an HEU target. However, although there would be a greater volume of liquid waste required to obtain a suitable extraction efficiency, the limiting factor in system capability was identified to be the speed at which waste from the LEU target could be calcined. It was determined that this could best be addressed by establishing a process development program aimed at improving the cycle time to process the LEU waste stream. Essentially the key issue is the capacity and capability of the calcination system. This assessment was discussed with and reviewed by Argonne National Laboratory (ANL) who are co-operating with MDS Nordion and AECL on the LEU conversion program.

#### 4. Status of the Phase 2 Conversion Development Program

With completion of the Initial Feasibility Study, MDS Nordion has proceeded with Phase 2, the Conversion Development Program. The Conversion Development Program is examining ways to address the two main obstacles to LEU conversion. These are:

- 1. high volumes of solution to separate the molybdenum-99 from the LEU targets because of the greater amount of uranium in solution; and
- 2. approximately five times more uranium will be present in the wastes to be calcined.

The Phase 2 program essentially consists of a waste process development program which is examining the technical, regulatory and economic implications for managing the increased volume of waste arising from processing LEU targets in the NPF. The objective of the Conversion Development Program Phase is to identify and evaluate improvements to the calcining system capacity and capability to process the LEU targets. The waste processing development program will:

- 1. identify throughput and cycle time improvements to the calcining system and equipment;
- 2. identify possible process improvements in the NPF to reduce the waste arising from processing LEU targets; and
- 3. ensure adequate equipment operation and maintenance cycles for commercial production.
The Phase 2 Program will consist of technical evaluation, bench chemistry precipitation studies, and operational experience to identify process improvements. An important condition is that the throughput capability of the converted NPF should be able to process the contracted design basis for maximum production quantities per week of bulk molybdenum-99. The Phase 2 Program will also establish the regulatory milestones for implementing a conversion program. The primary requirements to be determined are the Canadian nuclear regulations as defined in the *Nuclear Safety and Control Act* and the FDA regulations related to use of drug products.

## 5. Progressing Forward – Next Steps

Extensive progress has been made in Phase 2 on experimental and development work, examining ways to improve throughput and capacity of the calciner and waste management systems. Argonne National Laboratory (ANL) have played a key role in examining various precipitates as a means to improve throughput. This work has been substantially supported by other partners in Phase 2 Conversion Development Program. The designers of the waste management system, SGN, are examining technologies to improve calciner throughput. The process system integrators, AECL, are providing their overall expertise to ensure the entire process can continue viably as a full scale, continuous commercial operation.

The precipitation options have been narrowed down to one that is potentially viable – oxalic acid. More experimental work is underway at ANL to examine some of the technical problems that have so far occurred.

Although significant progress has been made by SGN and AECL, in the calcining and waste management systems, it is too early to report on the outcomes on the feasibility of their implementation.

## 6. Conclusion

MDS Nordion will continue to provide a secure, reliable source of isotopes to the nuclear medicine community. To comply with the spirit and intent of policy and legislation intended to reduce reliance on HEU material, we have already made a significant contribution to the RERTR Program by having the MAPLE reactors designed to operate using LEU fuel. Furthermore, we are proceeding diligently with our *LEU Target Development and Conversion Program*. A key outcome will be the evaluation of the technical and economic feasibility of conversion program options. The option chosen to convert to LEU targets must be both technically and economically feasible, and must ensure the reliable supply of medical isotopes, particularly molybdenum-99.

In the end, wherever our LEU conversion process leads, MDS Nordion will not lose sight of the fact that there are millions of individual patients that depend on our supply of medical isotopes every year.



# EXPERIMENTAL IRRADIATION OF UMo FUEL: PIE RESULTS AND MODELING OF FUEL BEHAVIOUR

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# ABSTRACT

Seven full -sized UMo plates containing ~8 g.cm<sup>-3</sup> of uranium in the fuel meat have been irradiated since the beginning of the French UMo development program. The first three of them with 20% <sup>235</sup>U enrichment were irradiated at maximum surfacic power under 150 W/cm2 in the OSIRIS reactor up to 50% burn-up and are under examination. Their global behaviour is satisfactory : no failure and a low swelling.

The other four plates were irradiated in the HFR Petten at maximum surfacic power between 150 and 250 W/cm2 with two enrichments 20 & 35%. The experiment was stopped after two cycles due a fuel failure. The post-irradiation examinations were completed in 2001 in Petten. Examinations showed a correct behaviour of 20% enriched plates and an abnormal behaviour of the two other plates (35%-enriched) with a clad failure on the plate 4.

The fuel failure appears to result from a combination of factors that led to high corrosion cladding and high fuel meat temperatures.

## 1. Introduction

The objective of the French UMo Fuel Development program is to qualify high density UMo fuel at 8 gU.cm<sup>-3</sup>. A series of two experiments have been performed in two reactors: the IRIS experiment in the OSIRIS reactor and the UMUS1 experiment in the High Flux Reactor (HFR) under higher irradiation conditions. The target was to irradiate these plates to an average burn-up up to 50% [1]. But UMUS1 was stopped after two cycles due to a fuel failure.

Two years ago, an agreement of collaboration on UMo fuel qualification based on an complete exchange of experimental results was signed between the French UMo Group and ANL in order to put in common the R&D effort on UMo fuel modeling [2]. On this basis, a close collaboration between CEA and ANL fuel experts has developed, starting first by a detailed exchange of PIE observations performed on RERTR experiments and French experiments and second by exchange of different fuel behaviour models and sharing the work of developing a common approach. The final objective is to have a common understanding of the in-reactor fuel behaviour. This collaboration has been instrumental in assessing the cause of the failure of the 35%-enriched UMo plates in the HFR.

The purpose of this paper is to give the major results obtained from the two French irradiation experiments performed on full-sized plates and to explain the failure that occurred in HFR.

## 2. Plate Manufacturing

The manufacturing of the UMo plates at CERCA was described two years ago [3]. CERCA used its proprietary advanced process, developed for the manufacture of highly loaded  $U_3Si_2$  plates with densities up to 6 gU.cm<sup>-3</sup>. So the UMo powders were produced by grinding. Thus the powders so produced have irregular shapes and contained two separated classes of particles with a large surface-to-volume ratio. The volume fraction of fuel particles in both plates was 50%, so that the uranium densities in the fuel meats were greater than 8.0 g cm<sup>-3</sup> for the two types of UMo alloys used (7 and 9% Mo mass content). The results of fuel-plate inspection (based on micrographs, X-ray, blister test,...) indicated that the plates conformed to the specifications. However, even though, the high volume loadings of these ground powders resulted in substantial fabrication voids. Consequently, the fuel meat contained as little as 35% by volume of matrix aluminum, and it is likely that there were some undetected local areas with thick fuel meat (and thin cladding). Figure 1 shows a section of fuel meat in an un-irradiated plate.



Fig. 1. Micrograph of asfabricated UMo fuel

The main characteristics of the seven plates fabricated by CERCA with ground powder and tested in reactor are given in Table 1.

Reactor	OSIRIS			HFR			
Plate number	1	2	3	1	2	3	4
%Mo in alloy	7.6	8.7	8.7	7.6	6.6	8.7	9.6
Enrichment (%)	19.65	19.66	19.66	19.65	34.54	19.66	34.86
U (g)	150.38	145.47	145.59	149.99	150.35	145.34	144.43
<sup>235</sup> U (g)	29.55	28.60	28.62	29.47	51.93	28.57	50.35
Meat density (gU.cm <sup>-3</sup> )	8.3	8.1	7.9	8.2	8.1	8.0	8.2
Porosity (%)	12.4	10.9	13.0	13.5	14.7	11.5	10.8

Table 1 : Characteristics of UMo plates

## 3. Overall Presentation of the Experiments

## IRIS experiment

In this first experiment of the program, three plates with 20% <sup>235</sup>U enrichment have been irradiated to different burn-up levels in the IRIS device in a peripheral location of the OSIRIS reactor core. Table 2 gives some irradiation conditions of these plates at three times during their irradiation. At the maximum flux mid-plane, the local highest burn-up is 65% in the U7Mo plate.

Irradiation time (days)			Maxim N	um bur MFP (%	n-up at )	Maximum surfacic power (W cm <sup>-2</sup> ) at BOL		
U9Mo	U9Mo	U7Mo	U9Mo	U9Mo	U7Mo	U9Mo	U9Mo	U7Mo
1	2	3	1 2 3			1	2	3
46	155	241	16.1	41.6	54.0	132	138	121

#### Table 2: Plate power of OSIRIS plates

## UMUS experiment

In this second experiment, four experimental UMo plates with 7 and 9% Mo by mass and with 19.75 and 35% enrichment in  $^{235}$ U have been irradiated in a special device in the HFR, in the in-core position D2. A fission-product release was detected near the end of the second cycle (48.4 <u>Full Power Days</u>) leading to an early unloading of the device. The irradiation conditions are given in Table 3.

Irradiation time	Maxi	mum bu (9	irn-up ai %)	t MFP	Maximum surfacic power (W.cm <sup>-2</sup> ) at BOL			
Plates	U7Mo 20%	U7Mo 35%	U9Mo 20%	U9Mo 35%	U7Mo 20%	U7Mo 35%	U9Mo 20%	U9Mo 35%
Number	1	2	3	4	1	2	3	4
54	15.6	13.6	14.5	14.7	170.0	241.6	136.6	236.4

#### Table 3. Plate power of UMUS plates

## 4. Post-irradiation Examinations

#### IRIS experiment

Non destructive examinations were performed last trimester and destructive examinations are in progress. The major results obtained thus far are the following:

- Nothing abnormal on visual observation,
- A quite-normal increase of the plate thickness with the irradiation time (70 µm increase)
- Confirmation by quantitative  $\gamma$  spectrometry analysis of the average burn-up given in table 1.
- <u>UMUS experiment</u>

A comprehensive post-irradiation examination program, performed at NRG/Petten, was completed in September 2001. The most-significant observations are:

- The plate surfaces show different degrees of damage (large spots with colored zones especially on plates 2 and 4 at high <sup>235</sup>U enrichment).

- Plate thickness measurements show a very slight increase in thickness of plate 1 (less than  $20 \,\mu\text{m}$ ) and a more significant increase in the thickness of the plate 2 (more than  $80 \,\mu\text{m}$  at the maximum flux plane)—Fig. 2.
- Another important fact is the presence of an oxide layer (boehmite) on the cladding surfaces of the two plates examined (less than 25 µm for plate 1 and about 60 up to 80 µm for plate 2 (at the maximum flux plane location) —Fig. 3.
- A transverse cladding failure (17 mm length) on plate 4 (highly enriched U9Mo) is observed at the maximum flux plane—Fig. 4.
- Optical micrographs of a section at the maximum flux plane location on the U7Mo plate 1 show a fuel with an interaction layer of 4 to 5  $\mu$ m and with a few percent of aluminum remaining in the meat—Fig. 5.
- However, Fig. 6 shows an abnormal behaviour of plate 2, which was irradiated in very similar conditions to plate 4, which failed. First, there is a clear open gap between the cladding and the meat. Second, the interaction layer on the fuel is significantly thicker, and practically all aluminum has reacted with UMo particles.



Fig. 2. Thickness measurements of plate 2





Fig. 4. Clad failure on plate 4



Fig. 5. Micrograph of UMo plate 1

Fig. 6. Micrograph of UMo plate 2

# 5. Thermo-mechanical Calculations

During the past two years, ANL has put considerable effort into modeling the evolution of the temperature of dispersion fuel plates during irradiation [4]. CEA started the development of a thermal-mechanical code early in 2001. The major models for fuel-matrix interaction and fuel-meat thermal-conductivity change developed at ANL have been implemented in the French code.

These codes were used to perform preliminary calculations for the plates of the French experiments. The comparison between ANL and CEA calculations shows very similar results. The two codes calculate early depletion of matrix aluminum. Taking into account the measured boehmite layer, the cladding temperature at the end of irradiation is close to 185°C, and the central fuel temperature is around 220°C in plate 2. First preliminary calculations of stresses in the cladding have been performed with the CEA code using a cladding creep law; however, this work must be validated in the future.

# 6. Explanation of the UMUS Failure

At a meeting at Cadarache in late January, CEA and ANL fuel experts carefully considered the results of the post-irradiation examinations of the UMUS plates, the results of the initial modeling calculations, and the results from irradiations of mini-plates performed by the RERTR program at ANL. A number of potential failure scenarios were critiqued and eliminated from further consideration. It was concluded that the 35%-enriched UMUS plates 2 and 4 failed owing to the cumulative effects of several factors that led to high fuel and cladding temperatures and to weakened cladding:

Excessive boehmite formation, up to 80  $\mu$ m at the peak power location, where the failures occurred, resulted in an additional ~80°C increase in fuel and cladding temperatures.

Local thickening of the fuel meat owing to the high fuel volume loading and high specific surface area of the ground fuel powders resulted in additional local fuel and cladding temperature increases and temperature-gradient-induced stresses.

Locally thin areas of cladding resulted from the local thickening of the fuel meat and the consumption of cladding by the fuel/matrix interaction and the boehmite formation. Local thermal stresses were apparently great enough to cause the debonding of the cladding and the reacted meat at the cladding meat interface. This was the primary failure of UMUS plates 2 and 4. In plate 4, the cladding was thin enough in one area for the thermal stresses to cause the cladding to fracture.

The fact that several 20%-enriched plates operated successfully at similar power densities in the RERTR-5 experiment appears to rule out the overall power of UMUS as the primary cause of the failure.

A similar failure occurred in mini-plate Q8003I, which was irradiated in the RERTR-5 experiment in the ATR. This 20%-enriched mini-plate, as well as several other 8-gU.cm<sup>-3</sup> mini-plates in the

experiment, operated at power densities comparable to those of UMUS plates 2 and 4. Mini-plate Q8003I was similar to UMUS plates 2 and 4 in several respects:

- It was one of two low-enriched U-7Mo, 8-gU cm<sup>-3</sup> plates in the RERTR-5 test made with ground powder. The sibling plate, which operated at higher power, did not fail.
- It had severe meat thickening of ~60% at the failure location (in the dogbone region). Calculations show that early in life, when Q8003I appears to have failed, the local fuel and cladding temperatures were, respectively, 70 and 50°C higher than at adjacent nominal regions.
- Post-irradiation examination revealed extensive cladding/fuel meat debonding and showed that the cladding cracked in an area of severe local cladding thinning---Fig. 7.

There were also some dissimilarities:

- The ground U-Mo powder in Q8003I was produced by a more-sophisticated process and the powder shape resulted in a smaller specific surface area.
  - The boehmite layer on the plate surfaces was only a few micrometres thick, owing to the water chemistry of the ATR and to plate surface treatment before irradiation.



Fig. 7. Micrographs of failed plate Q8003I **RERTR-5** experiment: in cladding fracture, (a) through showing very thin cladding: (b) laterally away from cladding fracture. showing cladding/fuel meat debonding

#### 7. Conclusions

Two principal conclusions have been drawn from the results of the French (and ANL) high-density UMo fuel irradiation experiments:

- Most importantly, the behaviour of the 20%-enriched plates irradiated in the IRIS experiment in OSIRIS to high burn-up under rather low irradiation conditions is rather satisfactory. Similarly, the two 20%-enriched plates of UMUS exhibit a correct behaviour even if the boehmite layer is quite high, taking into account the short irradiation time.
- The 35%-enriched UMUS plates appear to have failed owing to the <u>cumulative effect</u> of thick boehmite and local factors (such as local higher power and local thick fuel meat) leading to local cladding debonding and subsequent cladding rupture.

It appears that the shape and specific surface area of the fuel particles is an important factor in the behaviour of UMo fuel plates. Since one of the goals of the French UMo Fuel Development program is to provide a high-density fuel that can operate at even higher surface powers than those of UMUS plates 2 and 4, one must limit the amount of boehmite formation during irradiation.

The French MTR Group has decided to fabricate new experimental plates for the second series of experiments of its future experimental program using only atomised powder.

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## STATUS AS OF MARCH 2002 OF THE UMo DEVELOPMENT PROGRAM

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#### ABSTRACT

The French program for the development of UMo fuel has been launched in 1999 in close collaboration with five partners [5][6][9]. The aim of this program is to develop a high performance and reprocessable UMo fuel and to obtained a world wide qualified fuel before the end of the present US return policy.

The very first step of this program is the experimental irradiation of fuel plates. Three full size plates (20% enrichment, 8gU/cm<sup>3</sup> density) have been irradiated in OSIRIS reactor between September 1999 and January 2001. This paper gives the results already obtained.

Four full sized plates (20% & 35% enrichment, 8 gU/cm<sup>3</sup> density) have been irradiated in HFR reactor during two cycles ; the irradiation was interrupted due to a plate failure. All PIE, non destructive and destructive, were completed in 2001. This paper gives some comments about the results of theses examinations.

The French development program is covering complementary full-sized plates irradiation tests and experimental irradiation of fuel size U-7%Mo elements will be started on the basis of the results obtained with plates. This paper presents the next steps of the UMo development program, and the time schedule focused on the milestone of 2006.

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#### 1. Introduction

CEA, CERCA, TECHNICATOME, Framatome-ANP, COGEMA make up the French working group that supports the development program of a high performance LEU UMo fuel. The team is acting intensively to meet the milestone of May 2006 induces by the deadline of the US return policy of spent fuel.

The UMo alloys offer the opportunity to reach density higher than  $6gU/cm^3$  [3] with better reprocessing capacities than the existing LEU fuel [3].

The objective of the French development program is focused on the qualification of LEU U-7%Mo with a high density of 8gU/cm<sup>3</sup> to cover the needs of a wide range of reactors and anticipate the high performance expected for the MTR next generation like the RJH project in France.

The first step concerning the manufacturing of UMo fuel plates has been issued by CERCA since 1997 [1], and 7 full-sized plate have been irradiated since fall of 1999 [9][10] : three in the OSIRIS reactor and 4 plates in HFR, all with a density of 8gU/cm<sup>3</sup> (the feasibility of the manufacturing of UMo plates up to 9gU/cm<sup>3</sup> has already been obtained).

The second step of the program based on plate irradiation experiments in OSIRIS and HFR is rather completed (PIE are still in progress for the OSIRIS plates) and the further irradiation tests have been defined. They include both UMo plates and fuel elements irradiation.

This program, of first importance for the MTR community also intends to re-enforce the international collaboration as part of its success.

#### 2. Irradiation in the OSIRIS reactor

The special irradiation device IRIS has been used in OSIRIS for the irradiation of 3 UMo plates (one in U-7%Mo and two in U-9%Mo). This device has the same external geometry of a  $U_3Si_2$  standard fuel element and can be loaded with 4 plates. It allows the removal of the plates during the inter-cycle phases for swelling measurements and examination of the plates, on a facility designed for the control of irradiated plates implemented in OSIRIS.

The swelling measurements are made using automatically motorised probes that produces horizontally and vertically thickness profiles of the examined plates that are immerged under 3 meters of water due to their activity.

This measurement facility provides a fine evaluation of the swelling versus the burn-up of the irradiated plates.

The irradiation conditions on the plates have been confirmed through quantitative  $\gamma$  spectrometry analyses and post-evaluations based on neutronic computations taken into account the operating conditions of the reactor.

The maximum surface heat flux value was about 130 W/cm<sup>2</sup> with a maximum cladding temperature of 75°C.

The mean burn-up value obtained was about 48%.

Under these irradiation conditions, a good behaviour of the two irradiated plates was obtained, with no significant difference between UMo 7% and UMo 9% alloys.

A low swelling has been measured with an average value of  $37\mu m$  for the U-7%Mo plate and a maximum value of 57  $\mu m$ , that is fully satisfactory as regard to the burn-up level reached.

These results confirmed the choice of the U-7%Mo alloy to conduct the further steps of qualification of the UMo fuel.

#### 3. Irradiation in the HFR reactor

The special irradiation device UMUS, developed for HFR, has been used for the irradiation of four plates. The plates are loaded in a cassette along the diagonal of the device. The removal of the plate is not possible during the inter-cycle phase. The control of the water channels width can be made only using reference gauges.

The objective of this irradiation is to test UMo fuel under more severe conditions using both two types of Mo concentration and two enrichment levels.

The table below recalls the main characteristics of the four plates loaded in the UMUS device :

% Mo in alloy	7.6	6.6	8.7	9.6
Enrichment (%)	19.65	34.54	19.66	34.86
Meat density (gU/cm <sup>3</sup> )	8.2	8.1	8.0	8.2

The UMUS device was inserted in the D2 peripheral position in the core. The irradiation experiment has to be unload from the core at the end of the second cycle corresponding to a burn-up level of about 10%, due to the failure of the high enriched U-9%Mo plate [10].

The main conclusions of non destructive examinations are the following :

- only the two 35% enriched plates were damaged,
- the increase of the thickness reached 100 mm on the U-7%Mo 35% enriched plate,
- the behaviour of the two 20% enriched plate was conformed with a low thickness increase (lower than 20 μm in accordance with the flux distribution).

The destructive examinations have induced a lot of complementary actions. In order to get the best possible interpretation of the fuel behaviour, a close collaboration between fuel experts has been organised. Different models for fuel behaviour prediction have been upgraded to take into account the irradiation conditions of the UMo plates in HFR.

The comparison between the numerical predictions and the PIE issued from RERTR experiments or irradiation of CERCA's plates, and the interpretation of the HFR UMUS test are reported in [11]. This work is based on the collaboration between CEA and ANL.

In addition, different models have been used to re-evaluate the irradiation conditions of the HFR experiment of the core and to get a better understanding of the results.

The synthesis of the irradiation conditions are the following :

- heat flux density : 170 W/cm2
- maximum cladding temperature : > 90 °C
- burn-up : 13% at the end of the second cycle for the 20% enrichment plates.

This irradiation test in HFR with the failure of one of the high enrichment plates induces work extension for the examinations (non destructive and destructive) and for the interpretations through the use of fuel behaviour models. This experiment will increase the knowledge of the UMo fuel behaviour under high irradiation conditions for high enriched plates.

#### 4. Further steps of the French UMo Development Program

On the basis on the different results obtained with the irradiation experiments in OSIRIS and HFR, only the U-7%Mo is retained for the further irradiation test, with a maximum enrichment value of 20%. The density will be kept to 8 gU/cm<sup>3</sup>.

At least two irradiation experiments on full-sized UMo plates are under preparation at present. The first one has the objective to confirm the behaviour of the UMo fuel under a maximum surface heat flux of about 250 W/cm<sup>2</sup> with a maximum cladding temperature of 130°C. The second experiment will cover higher irradiation conditions with a maximum surface heat flux value that overcomes 300W/cm<sup>2</sup>.

Provisions have been made as regard the plate manufacturing aspects to cover, if necessary, complementary irradiation tests representative of high performance expected for future MTR projects like the RJH.

In parallel, the non destructive and destructive examination of the OSIRIS plates will continue at CEA Grenoble and at the end at CEA Cadarache. The technical synthesis of these results will be issued during in the middle of this year.

The definition of the full-sized test elements is in progress. The density will be  $8gU/cm^3$  and the enrichment 20%. The irradiation experiment is still scheduled for 2003, in order to get the main PIE results during the second semester of 2004.

Irradiated material is also available to start the validation of reprocessing treatment and get the final results with respect of the global time schedule of the project.

On the basis on the full-sized plate irradiation experiments already done, the French development program schedule of work in well defined at present. It takes the benefit of all the physical examinations and complementary models focused on irradiated UMo fuel behaviour and the developing support of international fuel experts.

#### 5. UMo Development Time Schedule

The time schedule of the UMo fuel development program is presented below. This program is in accordance with the initial objective to qualify the 8gU/cm<sup>3</sup> UMo fuel in 2005.

									-
MTR high density fuel program	1997	1998	1999	2000	2001	2002	2003	2004	2005
Fabrication									
Preliminary test		i							
Fabrication optimisation	1								
Feed-back ANL PIE results			_				-		
Full-sized plate program									
Irradiation in OSIRIS reactor									
preparation									
experimental irradiation up to 50% burr	n-up				_				
cooling time & transport									
PIE					_				
Irradiation in HFR									
preparation									
experimental irradiation up to 13% burr	n-up			-					
PIE						-			
Irradiation with heat flux of 250 W/cm <sup>2</sup>				-					
preparation									
irradiation									
PIE									
Irradiation with heat flux >300 W/cm <sup>2</sup>									
preparation									
irradiation									
PIE									
Lead test elements program	1								
fabrication									
preparation									
irradiation									
transport									
PIE									
Reporcessing									
preliminary test									
irrdiated									
Evalution report & qualification			_						
qualification									

#### 6. Conclusions

The French UMo development program is dedicated to the qualification of LEU U-7%Mo fuel, with a density up to  $8gU/cm^3$  in 2005, with respect to the important milestone linked with the US return policy of spent fuel.

This program takes benefit of the results already obtained through the two irradiation experiments handled at OSIRIS and HFR concerning full-sized plates. A lot work has been already done, based on the PIE and the correlation with fuel behaviour models and core thermal hydraulic or neutronic computations.

The failure of one high enriched plate of the UMUS experiment leads to a close collaboration of fuel experts of CEA and ANL for the interpretation of UMo behaviour under severe thermal conditions for the fuel meat.

The UMo development program is ready to launch the further full-sized plates irradiation experiments and has already defined the full-sized elements irradiation conditions with the support of reactor utilities.

This program has the ability to enlarge an international collaboration and the support of the MTR community to better anticipate the effort needed to convert the reactors to UMo before the deadline of May 2006.

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# ADVANCES AND HIGHLIGHTS OF THE CNEA QUALIFICATION PROGRAM AS HIGH DENSITY FUEL MANUFACTURER FOR RESEARCH REACTORS

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# ABSTRACT

One of the main objectives of CNEA regarding the fuel for research reactors is the development and qualification of the manufacturing of LEU high-density fuels.

The qualification programs for both types of fuels, Silicide fuel and U-xMo fuel, are similar. They include the following activities: development and set up of the fissile compound manufacturing technology, set up of fuel plate manufacturing, fabrication and irradiation of miniplates and plates, design and fabrication of fuel assembly prototypes for irradiation, post-irradiation examination and feedback for manufacturing improvements.

This paper describes the different activities performed within each program during the last year and the main advances and achievements of the programs within this period.

The main achievements may be summarized in the following activities:

- Continuation of the irradiation of the first silicide fuel element in the RA3
- Completion of the manufacturing of the second silicide fuel element, licensing and beginning of its irradiation in the RA3
- Development of the HMD Process to manufacture U-Mo powder (PUMA project)
- Set up of fuel plates manufacturing at industrial level using U-Mo powder
- Preliminary studies and the design for the irradiation of miniplates, plates and full scale fuel elements with U-Mo and 7 gU/cm3
- PIE destructive studies for the P-04 silicide fuel prototype (accurate burnup determination through chemical analysis, metallography and SEM of samples from the irradiated fuel plates)
- Improvement and development of new characterization techniques for high density fuel plates quality control including US testing and densitometric analysis of X-ray examinations.

The results obtained in this period are encouraging and also allow to foresee a wider participation of CNEA in the international effort to qualify U-Mo as a new material for the manufacturing of research reactor fuels.

#### 1 Introduction

CNEA, the National Atomic Energy Commission of Argentina is focusing its activities regarding fuel for research reactors on the development and qualification of LEU high-density fuel manufacturing, mainly based on uranium silicide and U-xMo alloy.

As described in previous presentations [1-4], the qualifications programs for both types of fuels,

Silicide fuel and U-xMo fuel, include similar activities. These activities are: development and set up of the fissile compound manufacturing technology, set up of fuel plate manufacturing, fabrication and irradiation of miniplates and plates, fabrication of fuel assembly prototypes for irradiation, post-irradiation examination and feedback for manufacturing improvements.

The purpose of the program is to demonstrate that CNEA's technology to manufacture high density fuels (namely U3Si2 and UMo) for research reactors is sufficiently proved and stabilized and, thus, that CNEA stands as a qualified supplier of this type of fuel elements

This paper describes the different activities performed within each program during the last year and outlines the main advances and achievements within this period.

#### 2.1 Silicide fuel activities

The main activities regarding silicide fuels were the manufacturing of the second fuel element of the qualification program, its licensing and the irradiation of both fuel elements in the RA3 reactor.

#### 2.2 Second silicide fuel element (P-07)

During this year the design and fabrication of the second fuel element belonging to the U3Si2 fuels qualification program were completed. This fuel element includes thinner fuel plates, with thinner cladding and thicker meat and also a larger number of fuel plates. As reported in [1], before the fabrication it was necessary to set up and fine-tune the manufacturing of the thinner fuel plates.

Contents of U and Si in the U3Si2 powder range within 91,55-91,82 % and 7,47-7,61 % respectively. The next table shows the typical content of relevant impurities for the different batches of U3Si2 used for P-07 fabrication. These batches were obtained in quasi-industrial stable conditions [5].

Impurity	В	Cd	Co	Cr	Hf
Average Content [ppm]	0,3	1	1,8	8	14

The density of the resulting powder ranges from 12,02 to 12,08 g/cm3. The average content of particles under 50  $\mu$ m is 38 %(w).

The following Table shows typical numbers for plate thickness distribution

		Specification (Nominal)	Average	Мах	Min
Fuel plate	External	1,475	1,47	1,49	1,44
ruei piate	Internal	1,325	1,32	1,35	1,3

The picture shows the result of the metallographic examination of the meat and a typical distribution of silicide particles after rolling.



The next two pictures show the final shape of the fuel element P-07.



## 2.3 Irradiation of the silicide fuel elements

The irradiation of the fuel elements P-06 and P-07 is carried out in the RA3 reactor. The irradiation of P-06 started in a peripheral position because of a requirement of our licensing authority, but now both fuel elements are in highly rated positions and the average burnup at the beginning of 2002 is 19 % for P-06 and 6 % for P-07. The irradiation of these fuel elements will be completed by the end of 2002 for P-06 and in March 2003 for P-07. The target burnup in both cases is 55 %.

#### 3 U-Mo Fuel Activities

The main activities that constitute CNEA's program to develop and qualify the technology for the production of high-density LEU fuel elements using U-xMo alloy are the following:

- **Procurement of the UMo alloy**
- Hydriding, Milling and Dehydriding
- UMo Powder characterization
- Set up of miniplates and fuel plates manufacturing
- Fuel plates manufacturing
- Design, fabrication and licensing of one UMo fuel element
- Irradiation in the RA3 reactor

#### 3.1 Development of the HMD Process to manufacture U-Mo powder

A massive hydriding of U-7%(w)Mo in  $\gamma$  (gamma) phase has been achieved [6]. The fragile compound allows the grinding of the hydride to the desired granulometry with less than 50% fines. After removal of the hydrogen, molybdenum homogenization is enhanced and no traces of the as cast structure remains. The development of this manufacturing process called HMD is being completed with the final tuning of the particles internal morphology and the particles size distribution. Process automation is being performed before going to an industrial full-scale production.

The following pictures show from left to right the shapes of both the hydride powder and the U-Mo powder



The next picture provides a typical X-ray diffraction diagram of the U-Mo particles obtained with the HMD process.



#### 3.2 Set up of mini-plates and full scale fuel plates manufacturing using U-Mo powder

To continue with the set up of the fuel plates manufacturing process eight miniplates were fabricated recently. Four with U-7Mo powder supplied by KAERI and prepared by centrifugal atomization. The others with HMD powder. In both cases the loading was 7 gU/cm<sup>3</sup>. The volume fraction of U-Mo in the meat is 44 %.

The result of the comparison between both materials is slightly favorable to the HMD powder, mainly because the shape of the particles is comparable with the silicide particles shape. However additional work will be necessary to improve the homogeneity of the U distribution and the amount of stray fuel particles outside the meat zone.

Two full-scale fuel plates with powder supplied by KAERI and a loading of 7 gU/cm<sup>3</sup> were also manufactured. In this case the result of the U distribution shows that this property is clearly dependent on the procedure to fill the die and on the expertise of the operator. This work was performed under a cooperation agreement with ANL.

## 4 PIE destructive studies for the P-04 silicide fuel prototype

The works in the hot cells continued during this period with the development and procurement of samples from irradiated fuel plates of the P-04 silicide fuel element for the accurate burnup determination through chemical analysis and for the metallographic and SEM examinations. The picture shows the punching machine inside the hot cells to obtain samples for absolute burnup determination.



# 5 Improvement and development of new characterization techniques for high density fuel plates quality control

To improve the quality control procedures of fabricated fuel plates, the densitometric analysis of X-ray examinations was set up and implemented during the last year. The calibration of the method was performed using samples where the U content was checked through chemical determination. P-07 fuel plates were tested using this technique. The following chart shows as an example the distribution of U density in the central zone of the fuel plates manufactured for P-07.



Works for the development of ultrasonic testing of fuel plates are also in progress.

## 6 Conclusions

CNEA has continued in the high-density fuels area with the qualification as  $U_3Si_2$  fuel manufacturer, which is now in the irradiation phase. Significant advances were obtained in the development of the HMD process to fabricate U-Mo powder and in the set up of fuel plates manufacturing with this kind of fissile material. The results obtained in this period are encouraging and also allows to foresee a wider participation of CNEA in the international effort to qualify U-Mo as a new material for the manufacturing of research reactor fuels.

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#### STATUS OF THE QUALIFICATION PROGRAM FOR ATOMIZED U-Mo DISPERSION ROD TYPE FUEL IN KOREA

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#### Abstract

Atomized U-Mo powder produced by KAERI has been applied to the development of very high uranium density fuel. The reprocess availability and the reactor performance upgrade possibility by higher uranium density induced the qualification of atomized U-Mo dispersion rod type fuel, which is available for HANARO. Since 1999 the characterization of atomized U-Mo dispersion fuel has been performed. Based on the obtained out-pile results and the RERTR test results, an irradiation test had been performed in the OR hole of HANARO from June 2001. The irradiation test fuel was discharged due to the occurrence of failures in the U-7wt.%Mo dispersion fuel of 6 g-U/cc in August 2001. A preliminary investigation represented that the reasons could be attributed to the corrosion attack of cooling water on the interface of the deforming aluminum flows in the cladding extrusion under the tensile stress influence from the interaction expansion of fuel meat and the acceleration effect of the high cladding temperature.

#### 1. Introduction

The new research reactor fuel having more than 8 g-U/cc had been requested to replace highly enriched uranium fuel of high performance research and test reactors with low enrich uranium fuel[1]. U-Mo alloy with about 16.8 g-U/cc has been assessed to be acceptable for the above fuel from the stable in-reactor behavior as shown in the ATR irradiation tests[2]. In connection with the end of the FRRSNF Acceptance Program of spent research reactor fuel in May 2006, an alternative available disposal was requested[3]. Uranium silicide dispersion fuel was investigated to be difficult for the reprocessing process, while U-Mo fuel is reprocessable[4]. In order to replace the current fuel with the reprocessable fuel, an accelerated program to qualify a U-Mo fuel was initiated by the RERTR program in 1999.

In case of using high uranium density fuel of U-Mo dispersion in HANARO in Korea, it is assumed that some beneficial effects could be obtained such as versatility in spent fuel management, fuel life extension, higher neutron flux, and availability to use a few of the driving fuel sites for irradiation holes[5]. A qualification program of U-Mo dispersion rod type fuel applicable to HANARO was launched in 1999. An investigation on the thermal properties of fuel material such as compatibility, conductivity, and reaction heat and kinetics had been done for various Mo contents and U loading densities. Also tensile tests for various fuel meats had been done to compare mechanical property with the uranium silicide fuel being used in HANARO at present. In general, U-Mo dispersion fuel had been evaluated to be better than  $U_3$ Si dispersion fuel. Based on these results irradiation tests on U-Mo dispersion fuel in HANARO were planned. The irradiation tests for the qualification of U-Mo fuel consist of three phases, which are a preliminary test for various U-Mo content and U-density fuel meats, a qualitative test for the candidate fuel meat in the previous test, and a demonstration test for the qualifying fuel. A fuel assembly containing 10 fuel rods had been fabricated for irradiation and had been irradiated in HANARO from June 26, 2001. In the middle of August, an abnormal phenomenon of increasing radioactivity in cooling water was observed. The irradiation fuel assembly was discharged from reactor to cooling pool. A sipping test for the irradiation fuel assembly indicated that a failure happened. In order to find out the failure cause an examination on the irradiated fuel rods was initiated from the beginning of January 2002. In this paper, the status of qualification program for U-Mo rod type fuel in KAERI is reported.

#### 2. U-Mo Dispersion Rod type Fuel Fabrication for Irradiation Test

4 different types of fuel rods were selected for the irradiation test as shown in Table 1. Two different Mo contents of 7 wt.% and 9 wt.% and two different uranium loadings of 3.4 and 6 g-U/cc were applied. 3.4 g-U/cc of U-Mo dispersion fuel is equivalent to the uranium density of current HANARO fuel of  $U_3Si$  dispersion, the density of which is 3.15 g-U/cc. Since fuel meat diameter of standard HANARO fuel is 6.35 mm, the transfer distance of fission heat in fuel meat is quite long comparing with plate type fuel. In order to reduce the center temperature of fuel meat in the safety point of view the smaller diameter fuel of 5.49 mm was designed. For each fuel type it was planned that three fuel rods were fabricated for the irradiation test. However, quality inspection activities allowed 10 fuel rods for the irradiation test. The porosities of fuel meat rods with low uranium density were measured to be very small as shown in Table 1.

In the first irradiation test it was purposed to burnup the candidate fuels in the moderate power condition and the burnup of 40 at.%. So it was decided to use an OR hole in HANARO for the irradiation test. In OR hole, normally circular type fuel assembly consisting of 18 fuel rods is used for irradiation test. An irradiation fuel assembly of cylindrical type was fabricated in arranging 10 fuel rods at the outer ring sites.

Туре	Mo contents (wt.%)	Mo contents (wt.%) Targeted uranium density (g-U/cc)		% to theoretical density	Designation of fuel rod
				99.4	428A-L1
1	7.0	3.4	6.35	99.1	428A-L4
				99.8	439M-L7
		6.0		98.3	428A-H1
2	7.0		5.49	97.6	428A-H2
			]	96.9	428A-H5
3	9.0	3.4	6.35	99.9	429A-L2
				97.7	429A-H1
4	9.0	6.0	5.49	95.9	429A-H3
			[	95.5	429A-H4

Table 1. Fuel rods loaded in OR hole for irradiation

#### 3. Irradiation

The fuel assembly containing 10 U-Mo dispersion rods had been loaded in OR-5 hole in HANARO on June 26, 2001 and irradiated under the generating power of 20 MW. It was planned for the irradiation test to be performed up to 40 at.% burnup. The maximum linear power was estimated to be about 80 kW/m. The delayed neutron flux of HANARO core was monitored to increase little by little from the end of July. Around the middle of August, rapid increase of delayed neutron flux as well as radioactivity of cooling water was measured and a gamma spectrum analysis of cooling water radioactivity represented a leakage of Tc-99m. After

the shutdown of reactor, sipping tests showed that a failure occurred on U-Mo dispersion fuel. Immediately the U-Mo fuel was discharged from HANARO core to the cooling pool on August 27, 2001. It was calculated that the U-Mo fuel had been irradiated for 26.6 full power days.

In the cooling pool, observations were made on the failed fuel rods using water-immersion camera as shown in Figure 1. On the cladding of the highest power generating fuel rod a longish defect like an axial cleavage was found at 30% to 60 % positions in the vertical direction from the bottom in spite of thicker cladding of 1.19 mm than standard thickness of 0.76 mm. Black spots were observed on the cladding surfaces of uranium high-density fuel rods irradiated at high linear power. It is assumed that the spots were formed from pitting Al cladding by cooling water. As the linear power of fuel rod increases, the spots were found more densely. The swelling of fuel rod could not be observed optically on an examination using the water-immersion camera.



Fig. 1. Photographs on the damaged parts of irradiated U-Mo dispersion fuel rods; (a) spots on the cladding surface, (b) cleavage-like defect on cladding, (c) More severely damaged defect.

The linear power calculations were conducted for the vertically divided positions of all irradiation fuel rods. The maximum powers of the failed fuel rod before and after the failure occurrence were obtained to be 105.9 kW/m and 107.1 kW/m, which are much higher than the expected value of 80 kW/m. It was investigated that this phenomenon happened from the influence of the newly loaded fuels in the core fuel sites situated near the irradiating fuel. The more closely situated fuel rods toward the reactor core were shown having higher linear power. Additionally the burnups for the positions were calculated. As the linear power is higher among the same loading density fuels, the burnup increases. The lower loading density fuels appeared to have higher of burnup. The peak burnup of the failed fuel rod was 12.9 at.%. In the fuel rod the highest linear power was shown at a little lower position than the middle of fuel rod because control rods were inserted from the top. That is a reason why the most severely damaged part on the failed fuel rod was observed at a little lower position than the middle of the fuel rod on the camera observation.

#### 4. Discussion on Failure Occurrence

It could be considered that failure would be induced from the reaction of Al cladding with fuel powder dispersed in fuel meat or cooling water. Supposing all fuel powder of U-Mo reacts with aluminum in fuel meat firstly and then reacts with aluminum in cladding additionally, the volume fraction of the remaining aluminum in fuel meat and the reacted cladding thickness were calculated for the failed fuel rod of U-9wt.%Mo with 6 g-U/cc. The results were obtained as shown in Table 2. Generally as the temperature in the fuel goes up, the interaction rate increases. If the temperature in the fuel meat went up enough highly in the irradiation test, all aluminum in the fuel meat would be consumed completely and then additionally U-Mo would

interact with aluminum in cladding. Therefore the temperatures of fuel meat as well as cladding were calculated using thermal conductivity of the partially interacted fuel meat with 10 vol. % of product and supposing the formation of aluminum hydroxide on the cladding surface. And then the temperature profile from the surface to the center of fuel rod was depicted as Fig. 2.

Fuel	g-U/cc	Vo in fue	l.% l meat	Supposing the reacted	Vol.% of the remaining Al	Thickness of the reacted
		U-Mo	Al	product	in fuel meat	cladding
			61.6	UAl <sub>2</sub>	0.96	
LI Ort 9/Ma	60	20.26		UAl <sub>3</sub>	None	0.35 mm
U-9wt.%Mo	0.0	38.30		UAL <sub>4</sub>	None	0.54 mm
				UAl <sub>5</sub>	None	0.65 mm

Table 2. Calculations for the remaining aluminum in fuel meat and the reacted cladding thickness supposing all fuel powder of U-Mo reacts with aluminum in fuel meat firstly and then reacts with aluminum in cladding additionally



Fig. 2. Temperature profiles on the cross sections of U-Mo fuels under irradiation in HANARO

It is considered that the fuel meat would be interacted with aluminum in fuel meat almost because the fuel meat temperature rose enough highly up to about 270 °C. However, the cladding temperature was shown to be between 120 °C and 147 °C. Presumably such temperature would not be enough high to interact fuel powder and cladding but could induce the Interaction of cladding with cooling water. The cladding of rod type fuel is generally performed by extrusion-forming aluminum ingot at 500 °C. When aluminum metal flows with covering from one side surface of fuel meat to other side surface, two metal flows contacts each other at

the other side and should be bonded each other. The contacted interface would be mechanically bonded with remaining a defect as misfits of atoms because the temperature of 500 °C is not enough high. Presumably the interface would be more easily attacked by cooling water than other sound surface of cladding. The corrosion reaction rate is accelerated exponentially by the increasing temperature of fuel meat. In addition, when volume expansion of fuel from the interaction between fuel powder and aluminum matrix acts a tensile stress at the interface, the corrosion rate of the interface will be more speeded up. Accordingly it is assumed that this failure of U-Mo dispersion fuel rod could be attributed to the corrosion attack of cooling water on the interface of aluminum flowing contact in the cladding extrusion under the tensile stress influence from the expansion of fuel meat and the acceleration effect of the high cladding temperature.

#### 5. Future Plan

The post-irradiation examination on the failed fuel rod was initiated around the end of January 2002. It is expect 3 months to take for carrying out the PIE. First of all, all efforts will be made on finding the reasons for the failure occurrence. Then the next irradiation test will be planned with considering the PIE results. The maximum uranium density will be selected in the view points of converting some driving fuel sites to irradiation test holes using high uranium-density. The preliminary study revealed 5 g-U/cc applicable for the above purpose. Fuel rods will be inserted for investigating the effect of particle size on the fuel performance. It is expected the next irradiation fuel assembly to be loaded in HANARO around summer season of this year.

#### 6. Conclusions

KAERI has carried out an irradiation test for U-Mo dispersion rod type fuel in HANARO. A failure happened on the high-density fuel rod with the highest linear power. A preliminary investigation represented that the reasons could be attributed to the corrosion attack of cooling water on the interface of the deforming aluminum flows in the cladding extrusion under the tensile stress influence from the interaction expansion of fuel meat and the acceleration effect of the high cladding temperature. The next irradiation test will be planned considering the PIE results. It is expected the next irradiation fuel assembly to be loaded in HANARO around summer season of this year.

#### Acknowledgement

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# PROGRESS IN DEVELOPMENT OF LOW-ENRICHED U-MO DISPERSION FUELS

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# ABSTRACT

Results from postirradiation examinations and analyses of U-Mo/Al dispersion miniplates are presented. Irradiation test RERTR-5 contained mini-fuel plates with fuel loadings of 6 and 8 gU cm<sup>-3</sup>. The fuel material consisted of 6, 7 and 10 wt.% Mo-uranium-alloy powders in atomized and machined form. The swelling behavior of the various fuel types is analyzed, indicating athermal swelling of the U-Mo alloy and temperature-dependent swelling owing to U-Mo/Al interdiffusion.

#### 1. Introduction

For the past several years the focus of the fuels area of the U.S. Reduced Enrichment for Research and Test Reactors (RERTR) program has been the development of aluminum-based dispersion fuels that will accommodate uranium densities in the fuel meat of 8 to 9 gU cm<sup>-3</sup> [1]. Our primary focus has been on determining the irradiation behavior of candidate fuels. Thus far, data are available from irradiation tests of very small fuel plates—RERTR-1, -2, and -3—and most recently for larger "miniplates"—RERTR-5. The first two tests resulted in the identification of U-Mo alloys with Mo contents of at least 6 wt.% as very promising candidates [2]. The third test was focused on the behavior of the U-Mo fuels under high-temperature (up to ~250°C) irradiation conditions [3].

The miniplates irradiated in RERTR-5 contained either atomized or machined fuel particles ranging in composition from, nominally, 6 wt.% Mo to 10 wt.% Mo. The fuel plates in the RERTR-5 test measured 100 mm x 25 mm x 1.40 mm; the meat was in a rectangular zone nominally 0.64 mm thick and contained, nominally, 6 and 8 gU cm<sup>-3</sup> of fuel meat. The experiment was irradiated in the Advanced Test Reactor (ATR) for 116 EFPD (effective full power days).

#### 2. Postirradiation data

The volume of the miniplates was measured with the customary immersion method after removal of the boehmite surface layers. The meat swelling was calculated by subtracting the known cladding volumes. The resulting data plotted against beginning-of-life (BOL) peak meat temperatures are shown in Fig. 1. A linear normalization to 50% burnup (Bu) was applied to the swelling values to aid comparison. The BOL temperatures plotted in Fig. 1 only serve to illustrate the trend in swelling with increasing temperature. The actual fuel temperature is a complex function of irradiation time and position in the fuel meat. Not only are there temperature gradients in the fuel meat, but the temperature changes during the irradiation as a result of the competing effects of the decreasing thermal conductivity and U-235 burnup. This issue was treated in detail previously [4] and remains the subject of continuing study. Apart from the obvious temperature dependence of the meat swelling, there is a clear difference in swelling magnitude between atomized and machined fuel. The lower swelling of the plates containing machined fuel powder can be attributed to the higher as-fabricated

porosity, 4 to 8%, in the meat of the plates, compared to 0 to 2% in plates containing atomized fuel powder. This effect of fuel swelling accommodation by as-fabricated porosity is also clearly evident for the three  $U_3Si_2$  miniplates which measured ~0% meat swelling and contained ~7% as-fabricated porosity. The apparent differences in swelling in plates with 6 g cm<sup>-3</sup> and 8 g cm<sup>-3</sup> uranium loading, or plates containing either 6 or 10 wt.% Mo fuel primarily results from differences in the amount of fuel-aluminum interaction phase formed during irradiation. The amount of interaction phase formed during irradiation, and the temperature history of each particular plate. Detailed characterization of the dependencies requires extensive metallographic examinations and modeling, which is an ongoing effort. However, to date we have completed the examination of a set of U-10Mo plates covering both machined and atomized fuel, 6 and 8 g cm<sup>-3</sup> uranium loading, and high and low irradiation temperatures. The results of this examination are discussed below.



Fig. 1. Meat swelling of miniplates from experiment RERTR-5 as a function of <u>Beginning-Of-Life</u> peak meat temperature.

#### 3. Metallographic Analysis

Metallographic samples taken from the center of the six U-10Mo miniplates listed in Table I were analyzed in order to characterize their swelling behavior. Examples of the meat microstructure of high- and low-temperature machined and atomized fuel are shown in Fig. 2. The effect of irradiation temperature on the extent of fuel-aluminum interdiffusion (represented by the light gray phase) and the diminution of aluminum matrix are the most prominent features.



600AH

600V





600D

600C



Low Temperature, 6 g cm<sup>-3</sup>

Fig. 2. Optical metallographic examples of machined and atomized U-10Mo fuel irradiated at ~100 to 200°C BOL temperature.

Quantitative image analysis was performed, yielding postirradiation volume fractions of the reaction product, unreacted U-Mo fuel, residual matrix aluminum, and porosity. These data were compared to preirradiation data from sibling miniplates to obtain the irradiation-induced changes in the meat.

# 3.1 Fuel – Aluminum Interactions

Characterization of the magnitude and kinetics of the interaction phase formation is an important item in the analysis of the irradiation behavior. Fuel-aluminum interaction is a major contributor to meat swelling and, as will be discussed in the next section, is the source of the temperature dependence of the meat swelling illustrated in Fig. 1. Moreover, the reaction product, which, based on available phase diagrams and out-of-reactor diffusion studies, appears to be a uranium aluminide compound (UAl<sub>x</sub>), which has a very low thermal conductivity of ~6 W cm<sup>-1</sup> K<sup>-1</sup>. As it replaces matrix aluminum, with a conductivity of ~200 W cm<sup>-1</sup> K<sup>-1</sup>, it significantly increases the meat temperature during the course of the irradiation [4].



Fig. 3. Volume U-10 Mo/Al interaction phase versus volume of aluminum consumed in the formation of the interaction phase.

The measured quantities of interaction phase formed versus matrix aluminum consumed are plotted in Fig. 3, along with the calculated value for the highest aluminide compound known to exist in the U-Al system. The plot indicates that there is substantially more aluminum consumed than needed for the formation of (U-10Mo)Al<sub>4</sub>. This observation was made previously during the analysis of RERTR-3 samples. The "missing" aluminum was accounted for by assuming that the aluminide compound was not stoichiometric, i.e., x = 4.4, and that a large amount of aluminum had diffused into the unreacted U-Mo fuel, residing there as solid solution [3]. The first assumption remains a possibility because UAl<sub>4</sub> is known to have a solubility range; it can, however, account for only a minor fraction of the missing aluminum. The second assumption requires postulation of a fission-induced solubility limit of aluminum in U-Mo that is several times higher than the maximum equilibrium value of 0.5 wt% for  $\gamma$ -U. Recent microprobe measurements indicate that the maximum aluminum content in the unreacted fuel is indeed only approximately 0.5 wt.%. This leaves the major fraction of the missing aluminum unaccounted for.

The explanation offered here is that, because of fuel and interaction product swelling, matrix aluminum is "squeezed" out to the periphery of the meat. This effect was indeed included in the DART code [5] to account for the observed irradiation behavior of  $U_3Si_2$  dispersion fuel. The question can only be definitively settled when complete microprobe and planned neutron diffraction data become available.

#### 4. Swelling Behavior

The swelling behavior was calculated in two ways: 1) using quantitative metallography data only and 2) using a combination of these data and the measured meat swelling data.

#### 4.1 Reaction Product Swelling (U-10Mo)Al<sub>4</sub>

The reaction 0.78U+0.22Mo+4Al results in a net volume increase of  $\frac{V^{I}}{V^{F} + V^{Al}} = 1.16$  or 16%. Fission-induced swelling of (U-10Mo)Al<sub>4</sub> is assumed to be equal to that of UAl<sub>x</sub>, and amounts to 0.05%  $\Delta V$  /% U-235 Bu [6].

# **4.2** Fuel Swelling $\Delta V^F$ (U-10Mo)

1) 
$$\frac{\Delta V^F}{V_o^F} = \frac{\Delta V_m^F - \Delta V_o^F}{V_o^F}$$

where:  $V_m^F$  is the measured unreacted fuel volume, and

 $V_o^F$  is the as-fabricated fuel volume minus the fuel consumed in the formulation of interaction volume V'. The results for the six U-10Mo miniplates are given in Table I.

2) 
$$\frac{\Delta V^F}{V_o^F} = \frac{\Delta V^m - \Delta V^I + \Delta V_e^P}{V_o^F}$$

where:  $\Delta V^m$  is the measured meat swelling,

 $\Delta V^{\prime}$  is the meat swelling due to interaction phase formation, and

 $\Delta V_e^p$  is the difference between as-fabricated and measured residual porosity.

The data thus derived are also given in Table I.

Inspection of the resulting fuel swelling values indicates that fuel swelling is  $\sim 0.5\%/\%$ U-235 burnup and is independent of temperature. This conclusion is supported by the scanning electron micrographs shown in Fig. 4. The fission gas bubble morphology at various temperatures is very similar, with similar-size visible bubbles at the grain boundaries. It appears that the machined fuel has reached the initial stage of recrystallization; this was expected based on RERTR-1, -2, and -3 results.

#### 5. Miscellaneous

#### 5.1 Cladding Failure

During the postirradiation examination of the RERTR-5 experiment, a small lateral crack was observed in the fuel zone near the trailing edge of plate Q8003I. Q8003I is one of a series of plates fabricated with a uranium loading of 8 g cm<sup>-3</sup> using U-7Mo ground powder. A slight increase in coolant and stack activity was noted by ATR Operations on August 29, two days after irradiation start. Activity decreased to normal levels after approximately two weeks. Irradiation of RERTR-4 and RERTR-5 continued for 116 days, through the remainder of cycle 123B and cycles 123C and 124A with no further activity increase. Total irradiation time for RERTR-5 was 116 EFPD. It appears from the metallographic examinations that the failure of Q8003I was due to a thin area in the cladding that was introduced during fuel plate fabrication. The cladding measures 0.002-0.003 inches (50-75  $\mu$ m) thick in the region of the failure. Fuel pile-up in this region increases stresses from thermal expansion and fuel meat swelling (owing to reaction and fission products) beyond the ultimate strength of the





Atomized



Machined

Fig. 4 Microstructure of unreacted U-10Mo fuel particles at low and high temperatures irradiated to 40 to 50% U-235 Bu (SEM).

thin area in the cladding. This is exacerbated by the additional local heat loading from the highdensity 'dogbone' region of the fuel plate. More information is presented in a paper by A. Languille [7].

## 5.2 Comparison with U<sub>3</sub>Si<sub>2</sub> Fuel

Three  $U_3Si_2$  miniplates were included in test RERTR-5 to provide comparison with our previous experiments on this fuel type. As shown in Fig. 1, the swelling of the  $U_3Si_2$  miniplates was measured as 0%. Metallographic examinations showed that the original as-fabricated porosity of ~7% was reduced to ~1%. This behavior is consistent with our previous experience. The fuel swelling rate of the  $U_3Si_2$  is approximately 0.25%/%U-235 burnup. This is, considering the lower fission density at equivalent burnup when compared with U-10Mo, only fractionally lower than the latter. This is probably due to the absence of grain boundaries in  $U_3Si_2$  and, therefore, the absence of grain boundary bubbles (see Fig. 5). The main difference is in the meat swelling component represented by fuel-aluminum interaction. As is clear from Fig. 5, the extent of interaction is much greater in U-10Mo. Moreover, the (U-Mo)Al<sub>4</sub> compound contains much more aluminum than the U(Al<sub>0.75</sub>, Si<sub>0.25</sub>)<sub>3</sub> compound formed in  $U_3Si_2$  dispersions while the volume increase associated with the latter is only

Plate Number/ Position	BOL T, °C	Bu, %	Interaction Depth, y, µm	$\frac{\Delta V^F}{V_o^F}, * \%_0$	$\frac{\Delta V^m}{V_o^m}, \frac{0}{0}$	$\frac{\Delta V^F}{V_o F}, ** \%_0$	$\frac{\Delta V^{F}}{Bu}$
A8002L B00V C-4	195	47	15	20	9.5	22	0.43*/0.48**
V8005B <sup>α</sup> 600 AH D-8	180	41	14	17	11.8	20	0.42/0.48
V6019G <sup>α</sup> 600M B-4	149	49	13	20	12.4	28	0.42/0.57
V6018G <sup>α</sup> 600D A-4	116	38	8	23	~8	22	0.59/0.58
A6008H 600Y C-7	160	49	13	27	8.7	22	0.56/0.45
A6005H 600C A-3	109	38	5	17	~6	17	0.46/0.45

Table I. Results from fuel swelling analysis of RERTR-5 U-10Mo miniplates

- $\alpha$  atomized power
- ~ interpolated values
- \* by quantitative metallography
- \*\* by measured density

 $\sim$ 3% versus 16% for the former. The main difference between U-Mo and U<sub>3</sub>Si<sub>2</sub> fuel behavior is thus the kinetics and extent of fuel-aluminum interaction, making the U-Mo fuel swelling more sensitive to irradiation temperatures. The swelling of both unreacted fuel types is similar and athermal.

#### 6. Conclusions

The following conclusions may be drawn from the initial results of the postirradiation examination of high-density dispersion fuel test RERTR-5.

- The extent of fuel plate swelling is acceptable and stable.
- Temperature dependence of swelling is due to temperature-dependent U-Mo/Al interdiffusion up to burnup where the matrix aluminum is consumed by this interdiffusion process.
- The aluminide interaction product appears stable and contains no fission gas bubbles. It has, however, a low thermal conductivity, which results in an increased fuel temperature.
- The swelling behavior of the unreacted fuel appears to be athermal in the range of temperature and burnup tested.

U-10Mo





U3Si2

Optical





#### SEM

Fig. 5. Meat microstructure of U-10Mo and U<sub>3</sub>Si<sub>2</sub> miniplates from RERTR-5 irradiated at BOL temperature of 149°C to ~50% U-235 Bu.

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# MTR FUEL PLATE QUALIFICATION CAPABILITIES AT SCK+CEN

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# ABSTRACT

In order to enhance the capabilities of BR2 in the field of MTR fuel plate testing, a dedicated irradiation device has been designed. In its basic version this device allows the irradiation of 3 fuel plates. The central fuel plate may be replaced by a dummy plate or a plate carrying dosimeters. A first FUTURE device has been built. A benchmark irradiation has been executed with standard BR2 fuel plates in order to qualify this device. Detailed neutronic calculations were performed and the results compared to the results of the post-irradiation examinations of the plates. These comparisons demonstrate the capability to conduct a fuel plate irradiation program under requested and well-known irradiation conditions. Further improvements are presently being designed in order to extend the ranges of heat flux and surface temperature of the fuel plates that can be handled with the FUTURE device.

#### 1. Introduction

The BR2 research reactor operated by the Belgian Nuclear Research Center SCK•CEN at Mol is one of the most powerful material testing reactors in the world. The BR2 fuel elements are still manufactured on the basis of HEU, as the presently available LEU fuels don't have the required densities to preserve the irradiation characteristics of BR2. However with the developments on UMo fuels going on, some fuel plate qualification activities are to be foreseen in the future.

Recently BR2 performed some MTR fuel plate qualification tests for a customer. Those tests were performed by incorporating the fuel plates to be tested in standard fuel elements of BR2. In each case three plates to be tested formed the outer ring of the 6 concentric rings of the standard fuel element.

In order to enhance our capabilities in the field of MTR fuel testing and qualification, a dedicated irradiation device has been designed in 2000. The objective is to provide BR2 with a reusable device for the irradiation of fuel plate under representative conditions, that is geometry, neutron spectrum, heat flux and thermal-hydraulic conditions.

#### 2. The MTR Fuel Plate Testing Device FUTURE - Basic Design

The first version of this device, called FUTURE (FUel Test Utility for REsearch reactors) has been designed and built in 2000. The design of this device has been presented at RRFM-2000 [1]. A cross-section of the device is shown in figure 1.

In order to keep the design simple the only standard instrumentation foreseen are 4 peripheral holes which can hold dosimeters.

This device allows to irradiate up to 3 fuel plates. A fuel plate may be replaced by a dummy plate or a plate carrying dosimeters (in addition to the 4 dosimeter-carrying rods foreseen in the periphery of the device).



Fig.1: Cross Section of the FUTURE device

The major advantages of this device compared to the 'mixed-type' fuel elements used previously (i.e. by incorporating the fuel plates to be tested in standard fuel elements of BR2) are:

- the possibility for unloading the plates in between reactor cycles for inspections and measurements,
- the possibility to continue the irradiation of some plates after the removal of a failed plate,
- a larger choice of irradiation conditions for the plates to be tested,
- a reduced risk to BR2 in case of swelling or failure of a fuel plate.

# 3. Qualification of the FUTURE Device

In order to qualify this device as a meaningful testing installation, a qualification irradiation campaign has been carried out using 2 standard UAIx-Al plates loaded with HEU. The objectives of this program are:

- qualification of the irradiation device including the thermal-hydraulic conditions and the fuel plates unloading and reloading after an irradiation cycle,
- qualification of the methods to determine the irradiation conditions of the fuel plates to be tested with the FUTURE device.

The test plates were manufactured as a surplus during a fuel fabrication campaign of standard BR2 fuel elements. They are identical to the 5th format plates are our standard elements, except for the bending.

Previous to the irradiation campaign the hydraulic characteristics of the FUTURE irradiation device have been checked in a H4 hydraulic test loop of the Reactor Experiment Department. The H4 water loop facility simulates a channel of the BR2 reactor from the hydraulic viewpoint.

## 4. Post-irradiation Examinations

After the irradiation campaign the plates have been unloaded and reloaded several times after irradiation without difficulty, demonstrating that:

- the device is reusable,
- the irradiation of fuel plates can be interrupted for intermediate examinations.

A visual inspection of the plates have been performed and documented in the BR2 hot-cells. The dosimeters have been recovered and sent for evaluation. Gross gamma-spectrometry on the plates have been carried out at the LHMA laboratory in order to determine the relative distribution of the burn-up. Absolute determinations of
the burn-up will soon be carried out by TIMS (Thermal Ionisation Mass Spectrometry) at the Nuclear Chemistry Department on samples taken from the fuel plates.

#### 5. Neutronic Calculations

Detailed neutronic calculations have been executed to predict the irradiation conditions, in particular the heat flux distributions in the plates.

The newly developed three-dimensional full-scale Monte Carlo model of the BR2 reactor has been used for that purpose. This new reactor model includes a detailed geometrical description of the inclined reactor channels and of all irradiation devices with their experimental contents. The burn-up of the BR2 fuel elements and the poisoning of the beryllium matrix are also represented in the model. The calculations of neutron fluxes and heat load distributions in irradiated MTR fuel plates were performed taking into account the contents of all loaded experimental devices in the reactor channels. These MCNP calculations are reported at the poster session during RRFM 2002 [2].

The Monte Carlo model is brought in as a complement to the code used for the determination of the overall irradiation conditions by the standard core calculation method. This 2D nodal code, called GEXBR2-TRPT4, is based on the integral transport theory and will also be presented during the RRFM'02 conference [3].

The Monte Carlo model has been benchmarked on comparisons of calculated and measured axial distributions of thermal neutron fluxes in the various reactor channels, thermal and fast neutron fluxes in irradiation samples, fission activity in the irradiated fuel elements. These comparisons are being conducted in a continuous manner. Up to now the results are very satisfying.



Fig.2: Comparison of MCNP calculations with gross gamma spectroscopy

For the FUTURE benchmark irradiation the MCNP results have already been checked against the results of the gross gamma spectroscopy (see fig.2). Further checks are foreseen against the results of the evaluation of the dosimeters and the post-irradiation burn-up measurements. These comparisons will be carried out as soon as the experimental results become available. A comparison of the calculated and measured values of neutron fluxes and of heat loads in the BR2 reactor is presented in the poster session [2].

These comparisons serve to establish the capability to conduct a fuel plate irradiation program under requested and well-known irradiation conditions despite the rather limited instrumentation of the FUTURE irradiation device.

## 6. New Developments

Recently new designs efforts have been undertaken to enhance the capabilities of the FUTURE device. One objective is to broaden the ranges of heat-fluxes and plate surface temperatures that be achieved with this device. It is useful to recall here that some flexibility can be achieved by the choice of the most adequate BR2 channel and/or by adapting the content of the surrounding channels (this is a very common procedure at BR2 where the core configuration is regularly adjusted to satisfy the experimental requests). However with the basic version of the FUTURE device this procedure will only allow to choose the heat-flux within certain limits; the surface temperature of the plates will then automatically being fixed by the design of the water gap between plates (the primary water cooling flow through the basic FUTURE device being fixed by the BR2 operational conditions).

Another objective is to allow more flexibility in the choice of the number and the format of plates that the device can accommodate.

The design efforts are therefore directed to develop:

- a replaceable plate holder which can be connected to one reusable suspension system. If needed a new adequate plate holder is to be build (the parameters being the number and the format of the plates, the water gap between plates and the possible addition of a plate containing dosimeters). The design must be such that this can be achieved with a minimum effort;
- a flow reduction system which can be easily attached to the plate holder; this should allow to choose both characteristics (heat-flux, surface temperature) independently, within extended limits,
- a flow acceleration system in order to be able to tackle the higher end heat-flux values while still maintaining an acceptable surface temperature.

#### 7. Conclusions

BR2 is actively developing a family of irradiation devices for MTR fuel plate testing. These developments have already lead to a first benchmark irradiation. Further developments are underway in order to enhance the flexibility of the FUTURE iradiation device which should be able to help in the qualification of future MTR fuels. SCK-CEN stands open for collaborations in this field.

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# **Session 3**

Reactor operation, fuel safety and core conversion

# **PROGRESS OF THE RERTR PROGRAM IN 2001**

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# **PROGRESS OF THE RERTR PROGRAM IN 2001**

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# ABSTRACT

This paper describes the 2001 progress achieved by the Reduced Enrichment for Research and Test Reactors (RERTR) Program in collaboration with its many international partners.

Postirradiation examinations of microplates have continued to reveal excellent irradiation behavior of U-Mo dispersion fuels in a variety of compositions and irradiating conditions. Irradiation of two new batches of miniplates of greater sizes was completed in the ATR to investigate the swelling behavior of these fuels under prototypic conditions. These materials hold the promise of achieving the program goal of developing LEU research reactor fuels with uranium densities in the 8-9 g/cm<sup>3</sup> range.

Qualification of the U-Mo dispersion fuels has been delayed by a patent issue involving KAERI. Test fuel elements with uranium density of 6 g/cm<sup>3</sup> are being fabricated by BWXT and are expected to begin undergoing irradiation in the HFR-Petten reactor around March 2003, with a goal of qualifying this fuel by mid-2005. U-Mo fuel with uranium density of 8-9 g/cm<sup>3</sup> is expected to be qualified by mid-2007.

Final irradiation tests of LEU <sup>99</sup>Mo targets in the RAS-GAS reactor at BATAN, in Indonesia, had to be postponed because of the 9/11 attacks, but the results collected to date indicate that these targets will soon be ready for commercial production. Excellent cooperation is also in progress with the CNEA in Argentina, MDSN/AECL in Canada, and ANSTO in Australia.

Irradiation testing of five WWR-M2 tube-type fuel assemblies fabricated by the NZChK and containing LEU  $UO_2$  dispersion fuel was successfully completed within the Russian RERTR program. A new LEU U-Mo pin-type fuel that could be used to convert most Russian-designed research reactors has been developed by VNIINM and is ready for testing.

Four additional shipments containing 822 spent fuel assemblies from foreign research reactors were accepted by the U.S. by September 30, 2001. Altogether, 4,562 spent fuel assemblies from foreign research reactors had been received by that date by the U.S. under the FRR SNF acceptance policy.

The RERTR program is aggressively pursuing qualification of high-density LEU U-Mo dispersion fuels, with the dual goal of enabling further conversions and of developing a substitute for LEU silicide fuels that can be more easily disposed of after expiration of the U.S. FRR SNF Acceptance Program. As in the past, the success of the RERTR program will depend on the international friendship and cooperation that has always been its trademark.

#### 1. Introduction

The 2001 event that will undoubtedly stand longest in our memory is the terrorist attack that destroyed the World Trade Center in New York, and killed thousands of its occupants, on September 11, 2001. The RERTR program was immediately and directly affected by that event, because the resulting international reaction forced the cancellation of the 2001 International RERTR Meeting that had been planned to occur in Bali, Indonesia, on October 21-25, 2001. We are very grateful to the organizers of RRFM 2002 for providing us with a forum at which several of the papers originally meant for Bali can be presented.

More important for the RERTR program than the cancellation of the meeting, the September 11 attack has caused a paradigm shift in the way we view our work and our goals. In the past, our main concern was that rogue nations or terrorist groups would develop nuclear weapons and that, by threatening to

use those weapons, they would secure for themselves political and economic advantages that could drastically alter the world balance of power. September 11 changed this view of the threat facing us. Today we know that if a nuclear weapon were to fall in the hands of those who organized the September 11 attacks there would be no threats and no negotiations. Millions of innocent victims would die in a flash, without warning, killed by people driven by a twisted ideology and devoid of any respect for human life, including their own.

It is with this terrible vision in mind that we must face the task ahead of us: how to remove from civilian traffic any amount of highly enriched uranium that a terrorist could use to manufacture an explosive device. Achieving this goal will eliminate one of the most dangerous pathways that a terrorist could follow. Much progress has been accomplished towards this end since 1978, when the RERTR program began. This progress has been the result of a concerted effort by many international organizations involved with research reactors, including fuel and target developers, fabricators, regulators, shippers and, especially, reactor organizations. In close collaboration with this international community, the RERTR program will pursue with increased urgency the goal of eliminating civilian use of highly enriched uranium.

#### 2. Status and progress of the RERTR program

The main events, findings, and activities of the RERTR Program are summarized below, with special emphasis on those that have occurred since the Las Vegas International RERTR Meeting [1].

 Reprocessing studies at the Savannah River Site (SRS) concluded in 1983 that RERTR silicide fuels could be reprocessed there. These results were rendered moot, however, by DOE's decision to phase out reprocessing at SRS and by the expiration of the Off-Site Fuel Policy at the end of 1988. A new DOE policy was issued in 1996 [2] allowing, until May 2009, the return of spent research reactor fuel elements of U.S. origin irradiated before 13 May 2006.

Implementation of this policy through the U.S. Foreign Research Reactor Spent Nuclear Fuel (FRR SNF) Acceptance Program has been very successful [3]. In particular, between October 1, 2000, and September 30, 2001, four additional shipments of spent research reactor fuel containing 822 MTR-type elements were received at the SRS. With these additional shipments, 3,727 MTR elements had been received at SRS and 835 TRIGA elements had been received at INEEL by September 30, 2001, under the FRR SNF Acceptance Program, for a total of 4,562 elements.

Many research reactors intend to send their spent fuel to the COGEMA plant in La Hague, France after expiration of the FRR SNF Acceptance program, but COGEMA has indicated that it cannot accept significant amounts of silicide fuel in its present plant configuration. Some options for coping with this issue are addressed later in this paper.

2. A fundamental activity of the RERTR program has always been the development and qualification of safe LEU fuels that can replace HEU fuels without significant performance or economic penalties. The qualified uranium densities of the three main fuels which were in operation with HEU in research reactors when the program began were first increased significantly with LEU (UAl<sub>x</sub>-Al, from 1.7 g/cm<sup>3</sup> to 2.3 g/cm<sup>3</sup>; U<sub>3</sub>O<sub>8</sub>-Al from 1.3g/cm<sup>3</sup> to 3.2 g/cm<sup>3</sup>; and UZrH<sub>x</sub>, from 0.5 g/cm<sup>3</sup> to 3.7 g/cm<sup>3</sup>). A new LEU fuel type, based on U<sub>3</sub>Si<sub>2</sub>-Al was also developed and qualified with uranium densities up to 4.8 g/cm<sup>3</sup>. This fuel type has been internationally accepted and is fabricated routinely for more than twenty research reactors by several international fuel fabricators.

The effort to develop new advanced LEU fuels with higher effective uranium loadings was restarted in 1996 after a pause of about six years. Three batches of 32 microplates each, containing a variety of promising fuel materials, were irradiated between 1997 and 1999 in the Advanced Test Reactor (ATR) in Idaho. Postirradiation examinations indicated very promising behavior of U-Mo alloy particles dispersed in an aluminum matrix, with Mo content between 6% and 10% and uranium densities up to 8-9 g/cm<sup>3</sup>. Irradiation of two new nearly identical batches of plates, RERTR-4 and RERTR-5, containing 32 positions each and planned to reach 80% and 50% burnup, respectively, began in 2000 and was completed in September 2001. The plates have larger dimensions than those used in prior experiments, have uranium densities of either 6 g/cm<sup>3</sup> or 8 g/cm<sup>3</sup>, and are intended to investigate the swelling behavior of U-Mo dispersion fuels under a variety of realistic operating conditions. Two of the plates have solid U-Mo [4] meat, to investigate the feasibility of using this concept in research reactor plates. Results to date of the postirradiation examinations of RERTR-5 (50% burnup) are reported at this meeting [5], while examinations of RERTR-4 (80% burnup) have recently begun. The activities related to U-Mo dispersion fuels are conducted in close collaboration with a parallel French fuel development program [6,7]. In general, all results collected to date are consistent with the view that low-enriched U-Mo dispersion fuels can be used successfully in research reactor fuels with very high uranium densities.

3. The effort to qualify U-Mo dispersion fuel with high uranium densities has been divided into two parts. An initial effort to qualify U-Mo fuel with uranium density of up to 6 g/cm<sup>3</sup> is aimed at preventing potential disruptions in the operation of research reactors currently operating with LEU silicide fuel due to lack of a method to dispose of their spent fuel after termination of the FRR SNF Acceptance policy. A later effort is intended to qualify LEU U-Mo fuel with uranium densities of 8-9 g/cm<sup>3</sup>, for reactors requiring such high densities to convert.

The initial schedule, which envisioned qualification of the LEU U-Mo fuel with 6 gU/cm<sup>3</sup> by the end of 2003, and with 8-9 g/cm<sup>3</sup> by the end of 2005, has suffered a slippage of more than one year. The cause of this slippage is directly linked to patents obtained by KAERI in 1999-2000 covering the use of spherical U-Mo particles in research reactor fuels. After some initial discussions on the subject by KAERI and ANL personnel, at the beginning of August 2001 the DOE General Counsel (DOE/GC) assumed total responsibility for resolving this issue between DOE and KAERI. BWXT was initially reluctant to produce a fuel covered by the KAERI patent but, responding to a letter by DOE/GC, it has recently agreed to fabricate two fuel elements needed to qualify LEU U-Mo dispersion fuels with a uranium content of 6 g/cm<sup>3</sup>. To provide insurance against other problems that may be caused by the KAERI patent, approximately half of the plates in the two BWXT elements will be fabricated using non-spherical powder. Similarly, one or two fuel elements will be fabricated by the CNEA, in Argentina, using both spherical and non-spherical powders to qualify LEU U-Mo fuel with uranium density of 7 g/cm<sup>3</sup>. Irradiation of these elements in the HFR-Petten reactor is expected to begin around March 2003. The good behavior of low enriched U-Mo fuel samples during irradiation provides convincing evidence that LEU U-Mo fuels with high uranium densities can be successfully qualified.

- 4. Cooperation with the Russian RERTR program has continued. The purpose of this program is to conduct the conversion studies, safety analyses, fuel development, and fuel tests needed to establish the technical and economic feasibility of converting Russian-supplied research and test reactors to the use of LEU fuels. Irradiation of five LEU UO<sub>2</sub>-Al tube-type assemblies, fabricated by the Novosibirsk Chemical Concentrates Plant (NZChK) with a uranium density of 2.5 g/cm<sup>3</sup>, was successfully concluded in the WWR-M reactor at the Petersburg Nuclear Physics Institute (PNPI), St. Petersburg. A new "universal" LEU U-Mo pin-type fuel [8], which could be used with minor modification to convert most Russian designed research and test reactors, has been developed by the A. Bochvar Institute for Inorganic Materials (VNIINM). 72 mini-pins have been fabricated with uranium densities of 4 g/cm<sup>3</sup> and 6 g/cm<sup>3</sup> for irradiation testing in the MIR reactor at the Russian Institute of Atomic Reactors (RIAR), Dimitrovgrad, and two full-size pin-type assemblies have been fabricated with uranium density close to 6 g/cm<sup>3</sup> for irradiation testing at PNPI. The irradiations are planned to begin in the spring of 2002.
- 5. Significant progress was achieved during the past year on several aspects of producing <sup>99</sup>Mo from fission targets utilizing LEU instead of HEU [9]. This activity is conducted in cooperation with several other laboratories including the Indonesian National Nuclear Energy Agency (BATAN), the Argentina Comisión Nacional de Energía Atómica (CNEA), MDS Nordion/AECL (Canada), and the Australian Nuclear Science and Technology Organization (ANSTO).

- 6. Improvements were made to the methods and codes that are used to study the design, performance and safety characteristics of research reactors. These improvements included upgrades of the WIMS-ANL cross section generation code, the REBUS diffusion theory burnup code, and the codes used to determine thermal-hydraulic safety margins.
- Several joint studies are in progress to facilitate reactor conversions or to improve utilization of LEU fuel in converted reactors. In particular, conversion studies continued for the WWR-SM reactor in Uzbekistan, the HFR-Petten reactor in the Netherlands, and the SAFARI-1 reactor in South Africa.
- 8. With the recent conversions of the R2-0 reactor in Sweden and of the ULRR in the U.S., 20 research reactors have been fully converted to LEU fuels outside the United States, including ASTRA (Austria), BER-II (Germany), DR-3 (Denmark), FRG-1 (Germany), IAN-R1 (Colombia), IEA-R1 (Brazil), JMTR (Japan), JRR-4 (Japan), NRCRR (Iran), NRU (Canada), OSIRIS (France), PARR (Pakistan), PRR-1 (Philippines), RA-3 (Argentina), R2 (Sweden), R2-0 (Sweden), SAPHIR (Switzerland), SL-M (Canada), THOR (Taiwan), and TRIGA II Ljubljana (Slovenia). Eleven research reactors have been fully converted in the U.S., including FNR, GTRR, ISUR, MCZPR, OSUR, RINSC, RPI, ULRR, UMR-R, UVAR, and WPIR. Seven foreign reactors, GRR-1 (Greece), HOR (Netherlands), La Reina (Chile), MNR (Canada), SSR (Romania), TR-2 (Turkey), and TRIGA II Vienna (Austria), have been partially converted. (ASTRA, DR-3, GTRR, ISUR, MCZPR, SAPHIR, and UVAR were shut down after conversion).

#### 3. Planned Activities

The major activities that the RERTR Program plans to undertake during the coming year are listed below.

- 1. Continue postirradiation examination of all the U-Mo microplates and miniplates irradiated to date in the ATR, including plates containing monolithic LEU U-Mo fuel with a uranium density of 16 g/cm<sup>3</sup>.
- 2. Continue out-of-pile characterization tests on the main fuel materials of interest, to assess their properties and likely performance.
- 3. In cooperation with fuel manufacturers, complete fabrication of full-size LEU fuel assemblies containing U-Mo dispersion fuels with uranium densities of 6 g/cm<sup>3</sup> and 7 g/cm<sup>3</sup> for irradiation testing in the HFR-Petten reactor.
- 4. Begin irradiation of full-size LEU fuel assemblies containing U-Mo dispersion fuels with uranium densities of 6 g/cm<sup>3</sup> and 7 g/cm<sup>3</sup> in the HFR-Petten reactor under prototypic conditions.
- 5. Continue LEU conversion feasibility studies for U.S. and foreign research reactors. Continue calculations and evaluations about the technical and economic feasibility of utilizing reduced-enrichment fuels in reactors that require such assistance, and in reactors of special interest.
- 6. In collaboration with the Russian RERTR program, continue to implement the studies, analyses, fuel development, and fuel tests needed to establish the technical and economic feasibility of converting Russian-supplied research and test reactors to the use of LEU fuels. Begin irradiation testing of pin-type mini-elements at RIAR and pin-type fuel assemblies at PNPI. Fabricate two prototype full-size pin-type fuel assemblies for irradiation testing in the WWR-SM reactor in Uzbekistan.
- 7. Continue development of viable processes based on LEU for the production of fission <sup>99</sup>Mo in research reactors in cooperation with several current and future <sup>99</sup>Mo producers.

#### 3. Summary and Conclusion

- A patent issue with KAERI has caused a slippage in the schedule for qualification of LEU U-Mo dispersion fuels, but all other activities have proceeded as planned, with positive results. The RERTR program plans to continue to concentrate its fuel development effort on fuels based on U-Mo alloys with two major thrusts:
  - T o develop, demonstrate, and qualify LEU U-Mo fuels with uranium densities of up to 6 g/cm<sup>3</sup> that can replace silicide fuels in current use. Qualification of these fuels is scheduled for mid-2005.
  - (2) To develop, demonstrate, and qualify LEU U-Mo fuels with uranium densities in the 8-9 g/cm<sup>3</sup> range, to enable conversion of reactors that cannot be converted today. Qualification of these fuels is scheduled for mid-2007.
- Excellent progress was made in the development of suitable commercial processes to produce fission <sup>99</sup>Mo using LEU targets.
- The Russian RERTR program has made excellent progress. In particular, VNIINM has developed a "universal" LEU U-Mo fuel pin design that could be used, with minor modifications, to convert most Russian-designed research reactors.
- The events of September 11, 2001 have made the task of minimizing and eventually eliminating worldwide use of HEU for civilian applications more important and pressing than ever. We believe that this goal is achievable and intend to do our best, with the collaboration of the many participants in the international RERTR effort, to attain it in the shortest possible time.

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# CH0300013

# PROGRESS OF THE RUSSIAN RERTR PROGRAM: DEVELOPMENT OF NEW-TYPE FUEL ELEMENTS FOR RUSSIAN-BUILT RESEARCH REACTORS

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# ABSTRACT

The new design of pin-type fuel elements and fuel assembly on their basis for Russian research reactors has been developed. The number of following activities has been performed: computational and experimental substantiation of fuel element design; development of fabrication process of fuel elements; manufacturing of experimental assembly for lifetime in-pile tests. The relevant fuel assemblies are considered to be perspective for usage as low-enriched fuel for Russian research reactors.

#### Introduction

During last years, advanced, new-type fuel assembly (FA) for research reactors was being developed in Russia [1, 2, 3]. Selected conception of (FA) was supposed to be used in all types of Russian research reactors. Pin-type fuel elements (FE) have been proposed in this (FA) instead of tubular-type currently used at present time. The main advantage of pin-type FE is their simplicity in production process. Besides, it is very important that the design and fabrication process of pin-type FE make it possible to utilize dispersion meat with high uranium concentration.

In its turn, this promotes usage of low enriched uranium. The plan of R&D process involved the following activities:

- development of FA and FE design;
- development of FE fabrication process;
- development of plan for irradiation tests;
- manufacturing of experimental FA;
- conducting of irradiation tests.

This paper contains the results of pre-irradiation R&D activities. The works were being performed in the frames of RERTR Program with the support and creative participation of Argonne National Laboratory (ANL, USA).

#### 1. FE and FA design

Fuel element represents the rod (pin) with square cross section and twisted separating fins (spacers) (fig. 1b). The design of experimental FA consists of ending parts, top and bottom separating grids and hexahedral shroud with 37 fuel elements allocated in compact triangular arrangement (fig. 1a). Separating of fuel elements is realized by spiral fins and separating grids disposed in top and bottom parts of shroud. The main dimensional parameters of FE and experimental FA are given in table 1.



a) experimental fuel assemble

b) fuel element

Fig.1 Cross section of FA (a) and FE (b).

Fuel assembly		Fuel element	
Number of fuel elements in fuel assembly	37	Pitch of (FE) twisting (mm)	320
Area of Water Passage (cm <sup>2</sup> )	467,3	Altitude of (FE)/fuel column (mm)	570/500
Heat yield surface (m <sup>2</sup> )	0,288	Circumscribed Diameter (mm)	4,85
Wettable Perimeter (mm)	682,2	Cladding Thickness (mm)	Min 0,3
Hydraulic Diameter (mm)	2,74	FE area (mm <sup>2</sup> )	9,0
Volume of fuel (cm <sup>3</sup> )	94,8	FE perimeter (mm)	15,6
Volume fraction of Coolant	0,6	Area of fuel meat (mm <sup>2</sup> )	5,12

## Table 1. Dimensional parameters

Computational parameters of FA correspond to that for FA, which are currently operated in VVR-M-type reactor (Hungary, Uzbekistan).

To confirm computational parameters of FA, the hydraulic bench tests have been performed using dummy of fuel assembly (fig. 2).



Fig.2 Dummy of FA for hydraulic tests

Hydraulic parameters of FA have been elaborated on the results of tests. Optimal design of separating grids has been selected taking into account possible vibration of fuel element bundle etc.



Fig.3. Pressure fall vs. consumption

The results of tests showed, that even in the case of usage of only supporting grids, rotation and vibration of fuel elements hadn't been observed even at coolant consumption trough FA more than twice higher than projected one (fig. 3).

#### 2. Fabrication process of FE

Fabrication process of pin-type fuel elements was based on the method of joint extrusion of cylindrical cermet meat and cladding. The main tool for pin-type FE fabrication by this method was forming female die with body size hole of complicated form. Thus fabrication process of pin-type elements has been significantly simplified in comparison with that for tubular FE and has had number of advantages. Very important is that more convenient for extrusion process shape of pin-type fuel element promotes the increase of fuel concentration in the fuel meat up to 50 and higher volume % in comparison with maximum 25-30 vol. % for tubular fuel elements.

T	abl	e	2.	FE	parameters
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Parameter	First version	Second version
Fuel component	UO <sub>2</sub>	U-9 wt%Mo
U <sub>235</sub> loading per fuel element (g)	1.27	2.62
Enrichment on U <sub>235</sub> , %	19,7	19,7
U concentration (g\cm <sup>3</sup> )	2.47	5.1

Control of overall FE dimensions, thickness of cladding and dispersion meat has been performed during metallography researchers (fig 5). General view of experimental fuel assembly with pin-type fuel elements is given in figure 6.



Fig. 5. Cross-section of fuel element



Fig. 6. General view of FA

## 3. Parameters of in-pile tests

In-pile tests of experimental FA are being planed to be performed in VVR-M5 (Gatchina, RF). Main parameters of in-pile tests are the following:

- average power of FA 100KW;
- average burn-up an  $U_{235} 60\%$ ;
- duration of tests 300 effective days (UO<sub>2</sub>)
  - 600 effective days (U-Mo alloy).

## Conclusions

Spadework on arrangements for in-pile tests of experimental fuel assemblies with pin-type fuel elements on the basis of low-enrichment uranium has been carried out.

Developed fuel elements and fuel assemblies can be recommended for usage in Russian research reactors as perspective ones for conversion to LEU.

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# **USE OF HIGHLY ENRICHED URANIUM AT THE FRM-II**

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# ABSTRACT

The new FRM-II research reactor in Munich, Germany, provides a high flux of thermal neutrons outside of the core at only 20 MW power. This is achieved by using a single compact, cylindrical fuel element with highly enriched uranium (HEU) which is cooled by light water and placed in the center of a large heavy water tank. – The paper outlines the arguments which have led to this core concept and summarizes its performance. It also reports on alternative studies which have been performed for the case of low enriched uranium (LEU) and compares the data of the two concepts, with the conclusion that the FRM-II cannot be converted to LEU. A concept using medium enriched uranium (MEU) is described as well as plans to develop such a fuel element in the future. Finally, it is argued that the use of HEU fuel elements at the FRM-II does not – realistically - involve any risk of proliferation.

## **1. Introduction**

Far beyond their traditional fields of application in fundamental research in nuclear physics and in solid state physics, neutrons have become a most important "tool" in many disciplines such as physics, chemistry, molecular biology, archaeology, environmental science, medicine, and for the industry. The need of neutrons is so strong that many experts call it a "gap" which exists in the supply of neutrons. It is from these reasons that the Technische Universität München (TUM) has decided, many years ago, to design and build a high performance neutron source, the new research reactor FRM-II. This new FRM-II will replace the existing FRM which was the first nuclear reactor in Germany and a most successful research facility until it was shut down only very recently to allow the training of the operators for the new facility. The new FRM-II, the construction of which has been completed but which has not yet obtained its final nuclear license to operate, will be a high flux, multi-disciplinary national neutron source which is expected to be very attractive for international users, too.

## 2. The most efficient core concept

The work to design the new research reactor core started already in the late 70's. The goal was to design a new and most efficient core concept which should be optimized primarily for beam tube research but should also provide excellent conditions for sample irradiations and other applications (research with positrons, ultra cold neutrons, a fission products accelerator, medical cancer treatment). It was an important boundary condition to limit the reactor power to 20 MW only, which seems to be about the maximum which can economically be afforded by a university reactor and which leads to many additional benefits as will be shown later. On the other hand, a flux of thermal neutrons as high as possible (close to  $10^{15}$  n/cm<sup>2</sup>/sec) should be made available with a pure thermal spectrum in a large volume outside of the fission zone of the reactor in order to provide optimum accessibility to beam tubes (in some cases in combination with cold or hot neutron sources) and irradiation thimbles.

This target could be met by designing a particularly compact reactor core containing one cylindrical fuel element only, which is cooled by light water and placed in the center of a large heavy water tank. The smaller the core volume – at constant reactor power – the higher is the power density and, consequently, the source density of fast fission neutrons. Since the reaction cross sections are low at these high energies, about 70 % of the fission neutrons leak out of the fuel element into the heavy water tank where most of them are moderated. Since the life time of the neutrons is very long in the heavy water,

a high flux of thermal neutrons piles up there which is available for utilization, but some of these neutrons diffuse back into the fuel element to contribute to a high value of reactivity.

It was clear from the beginning – and was supported by all the reactor physics calculations we performed – that the highest performance would be obtained using highly enriched uranium (HEU). Although by definition "HEU" stands for any value of enrichment of the fissile isotope U235 above 20 %, in practice "HEU" is mostly understood to mean a U235-enrichment of the order of 93 %. It follows that the concentration of the non-fissile isotope U238 is only about 7 %, then; U238 is a "neutron poison" since it only absorbs neutrons (without fissioning) to produce plutonium.

Although we studied core configurations with low enriched uranium (LEU, with below 20 % U235), too, our target was to achieve a significant progress in the performance of research reactors in general. So we focussed our attention on the combination of medium or highly enriched uranium with the new type of fuel with high uranium density the development of which started at that time [1] [2]. In this way it became feasible to realize a particularly high packing density of the isotope U235 and a particularly small volume of the core. This approach was in full agreement with the conclusions of the "International Nuclear Fuel Cycle Evaluation (INFCE)", a conference of 59 countries and 6 organizations performed during 1977 - 1980. This conference recommended to convert research reactors, if possible, to the use of low enriched uranium but also "recognized that there are specific applications requiring high flux reactor operation which can only be met with high enrichment fuel" [3]. The investigations we started at that time were site-independent in a sense that they should also be relevant to other projects of high flux reactors in the world. So the first research grant which we obtained, in January 1984, from the German Federal Government was entitled "Project Study of the Feasibility of a Novel Compact Core for High Flux Research Reactors". Later, the US project of the "Advanced Neutron Source ANS" followed the same approach [4].



Fig. 1. Cross section of the fuel element of the FRM-II. 113 fuel plates of involute curvature are welded between 2 concentric aluminum tubes. The cooling channels between the plates are 2.2 mm wide and the fuel zone is 700 mm long.

The calculations showed that the best performance was achieved using the new high density uranium-silicide (U<sub>3</sub>Si<sub>2</sub>-Al) dispersion fuel not at the highest uranium density which would be technically feasible  $(4.8 \text{ gU/cm}^3)$  but at 3.0 gU/cm<sup>3</sup> only and in combination with the highest enrichment of 93 % (HEU). Again, the ANS concept was very much analogous to that. The final design of the compact fuel element of the FRM-II is shown in Fig. 1. The core of the FRM-II only contains this single element which is cylindrical in shape to reduce power peaking effects. It consists of two concentric tubes of aluminum, the outer one having a diameter of only 24.3 cm. A total of 113 fuel plates, 1.36 mm thick with a fuel meat zone of 0.60 mm and two claddings of 0.38 mm each, are welded to the tubes. The plates have involute cur-

vature so that the cooling channels (for light water) between the plates have a constant width of 2.2

mm. Such a "cylindrical, involute-type" core concept has been used – also with HEU fuel - by two high flux reactors only in the world so far: the HFIR (100 MW, with two concentric elements) at Oak Ridge, USA, and the RHF (nearly 60 MW, with one element) at Grenoble, France. The ANS (330 MW), planned at Oak Ridge, USA [4], also used this concept but was unfortunately terminated in 1995.

#### 3. Performance of the HEU compact core reactor FRM-II

With this HEU compact core reactor concept as just described the FRM-II features, at 20 MW, an unperturbed flux of thermal neutrons outside of the core (in the heavy water tank) of  $8 \times 10^{14}$  n/cm<sup>2</sup>/sec. The corresponding ratio of flux to power is by far the highest in the world. The realization of this high flux at a low power is not only most economic considering both the costs of construction and of operation. It is also best for the scientific utilization of the reactor by beam tube experiments since a low reactor power is inseparably connected with a low background of fast neutron and gamma radiation. Since the pool structure (diameter, wall thickness) can be made particularly compact, the scientific instruments can be placed particularly close to the neutron source (beam tube noses) to benefit from large solid angles of the neutron beams. A low power also leads to a relatively low heat production in cryogenic installations close to the core (in the heavy water tank) such as cold and ultracold neutron sources. With HEU fuel plates, the uranium converter installed at the FRM-II to produce fission neutrons for medical cancer therapy can be positioned relatively far away from the beam tube noses in order not to affect the thermal neutron spectrum there.

Further, the compact core is characterized by particularly pronounced safety features. The small fuel element would not even become critical if immersed in a sea of water – to become critical and allow a cycle of about 52 full power days requires the combination of light water inside and heavy water outside the fuel element (further, the central control rod has a beryllium follower). Considering a postulated severe accident it is favourable that the inventory of radioactive fission products in the fuel element ("risk potential") is small because of the low power. The full decay heat could be absorbed in the pool water, and even in case of a total core melt an evacuation of the population outside of the facility would not become necessary. Finally, the volume of high-active nuclear waste to be disposed of is smallest because of the compactness of the element. - The aspect of nonproliferation will be discussed in the last section of this paper.

## 4. Alternative studies for LEU reactor cores

Low enriched uranium (LEU), as defined by a concentration of less than 20 % of the fissile isotope U235, contains about 80 % of the non-fissile isotope U238 which acts as a "neutron poison" absorbing neutrons and producing plutonium. Many existing low and medium flux reactors, which typically use HEU fuel with uranium densities in the range of 0.4 - 0.8 gU/cm<sup>3</sup> [2], can be converted from HEU to LEU since by using the new silicide fuel the uranium density can be increased (up to 4.8 gU/cm<sup>3</sup>) by a factor of about 6 - 12, whence the loss in enrichment (factor 93/20 = 4.65) can be more than compensated by the gain in density (a small degree of overcompensation is required anyway to balance the influence of the "neutron poison"). This large overcompensation can be made use of to even improve the reactor performance by either reducing the core volume – leading to a higher neutron flux – or by increasing the inventory of U235 – leading to a longer reactor cycle.

Such a conversion, however, would not be feasible at the FRM-II. Since the uranium density is already as high as 3.0 gU/cm<sup>3</sup> the margin up to the present technical limit of 4.8 gU/cm<sup>3</sup> is very small – in any case far too small to allow a compensation as just described. So there would be no other choice as to increase the core volume significantly. This question has been studied by both the groups in Argonne [5], USA, and Munich [6]. To maintain the neutron flux and the cycle length of the FRM-II the power would have to be increased from the present 20 MW to about 32 or 40 MW, depending on whether the larger core could still consist of one element, as in Fig. 1, with wide plates or would have to be made up of two concentric elements with smaller plates in order to provide sufficient mechanical stability. The core volume would have to be larger by a factor of about 2.5.

It is clear that such a research reactor would have been very much different from the FRM-II. The larger power would require a larger cooling capacity of the reactor (water pumps, heat exchangers, air coolers, tubing system). The reactor pool would have to have a thicker wall structure, in order to improve the shielding of the stronger fast neutron and gamma radiation, and a larger volume of the water inventory, in order to maintain the favorable response of the facility in a postulated severe accident scenario (Section 3). All this, on an obligatory basis, would have led to a larger reactor building. The costs of both constructing and operating the reactor would have been significantly higher. It is clear, too, that such changes could not be performed in the reactor building of the FRM-II once this has been erected.

The higher reactor power would result in a higher background of fast neutron and gamma radiation for the experiments. This could, at least to some extent, be compensated by the thicker walls of the pool as just argued. Nevertheless, the larger dimension of the pool would affect the scientific instruments to be positioned at larger distances from the neutron source (beam tube noses) leading to smaller solid angles of the neutron beams. As a rule, the higher power will also induce a larger heat production in the cryogenic installations in the moderator tank, although very close to the core surface the flux density of the radiation might remain about the same if the core surface area increases by about the same factor as the reactor power. Also, with LEU fuel plates, the optimization of the uranium converter for the medical applications can not be made as good as in the present situation (Section 3).

Further, the safety features of the LEU reactor would generally be reduced. The absolute value of the subcriticality of the LEU fuel element (as designed for 32 MW operation) in plain water is only about one half of that of the compact fuel element of the FRM-II. With respect to a postulated severe accident the inventory of radioactive fission products in the fuel element ("risk potential") would be larger by about the same factor as the reactor power. Since the LEU element contains a much larger quantity of the non-fissile isotope U238 the production of plutonium would be larger by a factor of about 15 to 20. The full decay heat could only be absorbed in the pool water if the water volume had been made larger by the same factor as the reactor power (see above). To make sure that even in the case of a total core melt under water an evacuation of the population outside of the facility would not be necessary would require further studies and probably design changes. Finally, the volume of high-active nuclear waste to be disposed of would be larger as a consequence of the larger volume of the fuel elements.

## 5. Case of a MEU compact core

The only realistic possibility to reduce the enrichment of the uranium used in the FRM-II, if e.g. requested from the political side, could be to reduce the enrichment as far as this could be fully compensated by an increase in the uranium density of the fuel. In this case the outer dimensions of the fuel element would remain unchanged as well as the whole reactor facility including the reactor power (20MW) and the reactor cycle length (52 days).

Preliminary calculations have been performed by the TUM for this case for various types of fuel with medium enrichment (MEU). If the presently available uranium-silicide fuel  $U_3Si_2$ -Al can be used at the maximum qualified density of 4.8 gU/cm<sup>3</sup> the enrichment could be reduced to about 70 % and the penalty in the thermal neutron flux would be about 4 %. With the same fuel a density of 7.8 gU/cm<sup>3</sup> would be required, which is unrealistic, for 50 % enrichment leading to a flux penalty of about 7 %. The new uranium-molybdenum fuel UMo-Al which is presently under development internationally can be relevant if it can be qualified for use under FRM-II conditions. Since molybdenum is a stronger neutron absorber than silicon, the density required would be about 8.5 gU/cm<sup>3</sup> for 50 % enrichment and the flux penalty would be about 8 %.

Only very recently an agreement has been achieved between the present red-green German Federal Government and the Bavarian State Government. According to that the FRM-II should be allowed to start operation with its as-designed HEU fuel element. After a time period of less than about 10 years this type of fuel element should be replaced by an appropriate MEU fuel element which has to be developed during this time. When this agreement will be officially signed, which can be expected at the time of obtaining the nuclear license to operate the FRM-II, the TUM will establish a working group

to participate in the international development of the new fuel and to design, test, qualify and license the new MEU fuel element.

## 6. Use of HEU Fuel Elements and Nonproliferation

The old Munich research reactor FRM and many research reactors in Germany have been using HEU fuel (enriched to 93 %) for many decades. Even much more, HEU fuel elements have been used throughout the world in more than 130 research reactors for more than 40 years. In all these cases the HEU fuel has been supplied by the five Nuclear Power States. The "as declared" use of the HEU fuel elements for peaceful purposes only is safeguarded in all countries, which have signed the Nonproliferation Treaty, by the inspectors of the International Atomic Energy Agency IAEA and, in Europe, additionally of EURATOM. Any clandestine deviation of HEU fuel would have been detected immediately. There has never been a case of proliferation.

Those "Non Nuclear Power States" which have developed nuclear weapons on their own have all chosen an approach different from diverting HEU from the fuel element cycle. They have either built their own enrichment plants to produce HEU or they have used their own reactors to breed plutonium. In this way they could keep their activities as secret as they wanted. To ban the use of HEU fuel elements in research reactors would not have prevented any one of these developments.

In the case of the FRM-II, the HEU material is delivered from the supplier directly to the fuel element fabricator which is a company in France (a Nuclear Power State anyway). From there only the complete fuel elements (assemblies) are transported to the FRM-II in Germany. These elements are about 1.3 m high and very easy to control. There is absolutely no risk of proliferation in using these fuel elements at the FRM-II. This assessment of the TUM is shared by the Bavarian State Government and by the German Federal Ministry responsible for safeguards. It goes without saying that the use of HEU fuel elements at the FRM-II is also in agreement with all international contracts.

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# CHARACTERISTIC DIFFERENCES OF LEU AND HEU CORES AT THE GERMAN FRJ-2 RESEARCH REACTOR

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# ABSTRACT

As a sophisticated computational method for reactor physics analysis and fuel management an MCNP model in very high fidelity was developed and coupled with a depletion code and applied to the HEU-LEU core conversion study. The analysis show that as a consequence of the high amount of U-238, the amount of U-235 in the LEU core is about 14 % higher than in the HEU core. The reduction of the thermal flux varies between 16 % (core) and 5 % in the reflector zone. The rate of U-235 burnup in the LEU core is approx. 11.5 % lower which allows an extension of irradiation time. Due to the effect of neutron spectrum the worth of the absorber system decreases in an LEU core by 17 % resulting in a decrease of shutdown and excess reactivity. The kinetic parameters of the core are slightly reduced causing changes in the reactivity values and transient behavior of the core. The moderator coefficient is decreased by 18 % and the Doppler coefficient is increased by 63 %. Due to shortening of the absorption length of the fission neutrons the prompt neutron lifetime is reduced by 7 %.

## 1. Introduction

In view of the fuel supply for long-term operation and return of spent fuel, the operator of the German research reactor, FRJ-2, has participated from the beginning in the RERTR programme and intended to convert the core from HEU to LEU fuel. The development and the application of the high-density uranium silicide fuel required a modification of the fuel element design of FRJ-2 resulting in performance tests under operating conditions. For the conversion of the whole reactor core with the new LEU silicide fuel element design a long term programme was established in agreement with the German licensing authorities and representatives of the US RERTR program [1].

In the first phase of the conversion program, the reactor had to be shutdown for comprehensive inspection resulting in a postponement of the conversion activities [2]. In parallel, a sophisticated computational method was developed, which is capable of precisely simulating the physical behavior of HEU, LEU and mixed core as well as LEU fuel element performance. The method was also considered as a tool for replacing comprehensive in-core measurements and criticality tests being performed at the beginning of each operating cycle (BOC). For reactor physics analysis and conversion studies the Monte Carlo code MCNP was chosen, coupled with a depletion program and applied to the FRJ-2 [3-5]. The MCNP code was selected because of its comprehensive capability of modeling the complex 3D geometries of the fuel element assembly, shutdown system and core structures of FRJ-2.

The present paper describes the results of the first study with regard to characteristic differences in the neutronic behavior of typical HEU and LEU cores.

# 2. Description of FRJ-2

The FRJ-2 is a DIDO-class tank-type research reactor cooled and moderated by heavy water. The core consists of 25 tubular MTR fuel elements arranged in five rows of 4, 6, 5, 6 and 4 fuel elements (Fig. 1). It is accommodated within an aluminum tank 2 m in diameter and 3.2 m in height. The tank is surrounded by a graphite reflector of 0.6 m thickness enclosed within a double-walled steel tank.

The active part of the tubular fuel elements is formed by four concentric tubes having a wall thickness of 1.5 mm and a length of 0.61 m. Each tube is formed by three standard fuel plates containing fuel meat and aluminum cladding. The tubes are encased by a shroud tube of 103 mm diameter, to which they are attached by four combs at either end (electron beam welded tube for HEU-fuel elements; EB design).

According to the new design the pre-curved fuel plates are swaged into 3 plates provided with special grooves then forming the final fuel tube (roll swaged element; RS design). Fig. 1 illustrates the differences between the EB and RS element design.

The fuel elements with HEU fuel contains  $UAl_x$  in an aluminum-matrix with an U-235 mass of 170 g and 150 g, respectively. The meat of the fuel element with LEU fuel consists of  $U_3Si_2$ -Al with an uranium mass of 180 g and 200 g, respectively. The annular water gap between the tubes has in both cases a width of about 3 mm leaving a central hole of 50 mm diameter filled with a thimble for irradiation purposes.

The reactor is equipped with two independent and diverse shutdown systems, the coarse control arms (CCAs) and the rapid shutdown rods (RSRs). In case of demand, the six CCAs are released from their electromagnets and drop into the shutdown position by gravity, whereas the three RSRs are shot in by pneumatic actuators. The CCAs are lowered and raised manually around a pivot in order to control power levels during normal operation. A large number of horizontal and vertical channels give access to the neutron field in the reactor.



Fig. 1: HEU and LEU fuel element design and arrangement of fuel elements in the core

## 3. MCNP model of FRJ-2

The MCNP model of FRJ-2 is a complete 3-dimensional full-scale model with a very high level of geometric fidelity. It comprises the reactor core, CCAs, core structures, beam tubes, the graphite reflector and the biological shield. The core region consisting of 25 fuel elements was modeled as a cylinder containing a square lattice with an array of cells representing the individual fuel elements, part of the absorber arm and cooling gaps. For the detailed modeling each individual fuel element is divided into 15 axial, 35 radial and 6 azimuthal material zones. Due to the continuous change of the material composition in the fuel meat resulting from the fuel consumption, it was necessary to couple the MCNP code with a depletion code. In this way the variation of the neutronic states of the core could be simulated by multiple linked burnup and MCNP calculations. The detailed segementation of the core and the surroundings for the MCNP and depletion calculation resulted in a model with 11250 material zones. To achieve sufficient numbers of neutron tracks and events in all cells and consequently reduce the estimated error of the physical values (keff and local neutron flux) all simulations were run for a high number of particle history resulting in a standard deviation of 0.001 for the multiplication factor (i.e. 0.10 % dk/k) and 0.05 for the local n-flux. Due to the complexity of the geometrical model of the reactor and the large number of particle histories, the computing time was reduced significantly by the application of the PVM-version of MCNP and utilization of a massively parallel computer system, CRAY-1200, with 32 processors (available 520).

## 4. Results of HEU-LEU Simulations

As a first approach, two types of fuel elements with different fissile amount of U-235 were used. To set up the final LEU core, the HEU fuel elements with an initial mass of 150 g and 170 g (U-235) were successively replaced by 180 g and 200 g fuel elements with an enrichment of 20 %. After completing 5 operating cycles the whole core was set up as a LEU working core. The type and the characteristics of the HEU and LEU fuel elements are summarized in Table I.

Fuel type	HEU	LEU
Fuel specification	UAlx	U3Si2-Al
U-235 mass (g)	150/170	180/200
Density (g/cm <sup>3</sup> )	0.50/0.570	2.40/2.65
Enrichment (% w)	80	20

Table I: Main characteristics of the HEU and LEU fuel element

For the analysis of the characteristic differences of the LEU from the HEU core, different neutronic parameters were considered including the burnup behavior during an operating cycle. At first it was found out that the amount of U-235 in the LEU core is about 14 % higher than in the HEU core. This is a consequence of the high absorption rate in U-238 resulting from the high concentration in an LEU fuel element. According to the simulations, a change of the enrichment from 80 % to 20 % in one fuel element with the same amount of U-235 causes the fission rate to decrease by approx. 2.3 % due to the reduction of neutron flux in the fuel meat. In this case, the reactivity of the system is decreased by 0.4 % dk/k due to the increase of the content of U-238. As a consequence of the different neutronic state resulting from different material composition, the rate of the U-235 burnup in the LEU core amounts to 1.07 g/MWd and is approx. 11.5 % lower than in an HEU core which results in an extension of the transmutation of U-238 in an LEU core is significantly higher and contributes -to a considerable amount-to the total fission rate and power. In a core with an average burnup of 36 %, 252 f of the fissionable plutonium isotopes are generated (8.9 g in the HEU core). The contribution of fissile plutonium to the total fission rate is about 11 % which results in a compensation of the reduced fission rate in U-235.

From the utilization point of view, the neutron flux is changed to a considerable extent in an LEU core The change of the neutron flux depends on the location in the core and surrounding as well as on the core loading in particular. The simulation shows that its level in the moderator region of the core is reduced by 15 % on average. The distribution of the neutron flux in the core region is given in Fig. 2. It shows that the variation of the thermal flux is strongly influenced by the core configuration. In the peripheral zone (reflector region accommodating beam tubes and irradiation facilities) the change depends on the location of the experiments. At the reflector peak around the core (location of the vertical 2V-3 and 2V-8 channels) the reduction of the neutron flux remains limited to 5 % only. In the center of the cold neutron source (liquid hydrogen) located on the midplane at a distance of 0.6 m from the core axis, the change of the thermal flux is less than 4 % due to the thermal character of the reflector area. A compilation of the neutron flux at different positions is given in Table II.

Core position	Core average	Vert. Beam Tube (2V3/2V8)	Cold n-Source
HEU Core (cm <sup>2</sup> /s)	1.88E+14	1.35E+14	1.90E+14
LEU Core (cm <sup>2</sup> /s)	1.60E+14	1.28E+14	1.82E+14
Ratio	0.85	0.95	0.96

Table II: Compilation of the neutron flux at different positions

The effectiveness of the absorber systems is mainly influenced by the neutron spectrum. Because of the high amount of U-238 in LEU fuel, the neutron spectrum is slightly hardened causing a reduction of the effectiveness of the absorber systems. The calculation for the HEU and LEU cores shows that due to the spectral effect the absorber rate decreases by 17 % resulting in a decrease of reactivity from 16.3 %dk/k



Fig. 2: Distribution of the thermal flux in HEU and LEU cores

to 13.25 % dk/k at a CCA angle of 20°. The consequence of the reduction of effectiveness is a change of shutdown and excess reactivity. The transient behavior of the core is significantly influenced by the kinetics and safety characteristics of the LEU core. Due to the variation of the neutron spectrum, the temperature coefficient of reactivity at the average operating temperature is significantly changed. The results of the calculations are summarized in Table III. Accordingly, the contribution of the moderator to the total reactivity coefficient is decreased by 18 %. By comparison, the Doppler coefficient increases by 63 % in the case of conversion of the core from HEU to LEU fuel. However, it remains lower than the moderator contribution by one order of magnitude. Due to shortening of the absorption length of the fission neutrons resulting from the higher absorption rate in U-238, the prompt neutron lifetime is reduced by 7 %. In the event of transients with reactivity insertion the change of the neutronic variables becomes faster causing a fast transient behavior in comparison to the HEU core. To cope with this type of transients the limiting values for the max. amount of reactivity insertion by experiments are accordingly fixed and modified. For the control of the design basis accident (rupture of the most effective absorber arm) with a high amount of the reactivity insertion, the loading is managed in such a way that the shutdown reactivity is not reduced below a minimum level required for a stable and continuous subcriticality.

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Core parameter	Moderator Coefficient	Doppler Coefficient	Prompt neutron lifetime		
HEU Core	2.30E-2	1.14E-3	5.44E-4		
LEU Core	1.90E-2	1.86E-3	5.10E-4		
Ratio	0.82	1.63	0.94		

Table III: Safety-related parameters for HEU and LEU core

Reactivity coefficients in % dk/k °C, neutron lifetime in s

Due to the difference of fuel composition and neutronic states in the HEU and LEU fuel elements, the inventory of the fission products becomes different in the process of power generation and irradiation. As discussed before, the high amount of U-238 causes a reduction of the neutron flux resulting in a decrease of U-235 fission product activity in an LEU fuel element in the case of the same amount of fissile U-235. However, due to the high amount of U-235 in an LEU fuel element and the contribution of fission products from fissile plutonium isotopes, the total activity of the fission products becomes slightly higher. The results of the calculation performed for a fuel element of average thermal load ( 800 kW) and maximum burnup of 60 % (fifa) are summarized in Tab. IV. Accordingly, the total activity of the fission

products in an LEU fuel type (200 g U-235) is approx. 4 % higher than in a standard HEU fuel element with 170 g U-235.

Parameter	Init. U Mass	Burnup(%)/	Neutron	Actinides	Fission	Total
	U-235(g)	Irrad. Time(d)	flux	(Ci)	Products	Activity
					(Ci)	(Ci)
HEU	170	60/103,3	2.17E+14	4.77E+4	3.52E+6	3.56E+6
LEU	200	60/135,2	1.66E+14	5.04E+5	3.62E+6	4.12E+6
Ratio	1.17	0/1.31	0.76	10.6	1.028	1.16

Table IV: Activity of the actinides and fission products for HEU and LEU fuel element

#### 5. Conclusions

For the HEU-LEU conversion of the FRJ-2 core the design of the fuel elements had slightly to be changed but the principal dimensions remained unmodified. Using MCNP, first calculations of the neutronic differences between the HEU and the LEU cores revealed that the U-235 content of the LEU core must be increased by about 14 % due to the high inventory of U-238. Although the thermal neutron flux in the core decreases by nearly the same amount, the flux at the experimental positions located in the reflector zone including the cold neutron source is only slightly reduced. Thus the utilization quality of the reactor is preserved. In addition, the MCNP calculations showed that the effectiveness of the shut down system is reduced by 10%. Due to the existing high safety margins, the reduction of the effectiveness does not impair the safe operation of the FRJ-2 with a LEU core.

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# A PRELIMINARY STUDY on the SUITABILITY of HIGH DENSITY LEU FUEL ELEMENTS to MAINTAIN BR2 REACTOR OPERATIONAL CHARACTERISTICS

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# ABSTRACT

The present international efforts to develop high density fuel based on U-Mo (6-9%wt) triggered the present preliminary study to analyse the suitability of these high density LEU fuels to maintain the operational characteristics of the BR2 reactor.

The analyses concern the perturbation mode in a fuel cell for determining the available thermal neutron flux, the power generated, Pu-239 production and reactivity variation versus the burnup of fuel elements loaded in a cell where the statistical weight is unitary. The comparison with several types of fuel elements used at the BR2 reactor gives preliminary answers asked for using LEU fuel type in the BR2 reactor.

#### 1. Introduction.

The objective of the BR2 operation is to satisfy the irradiation conditions requested by the experimental load, and to do this by guarantying safe operation and by making optimal economical use of the available fuel elements.

The main nuclear characteristics of the BR2 reactor (a high flux MTR-type reactor) are the fast neutron flux available for experiments loaded in the axial position of a standard fuel element and the thermal neutron flux in the adjacent reflector channels. Those characteristics are limited by the maximum allowed heat flux at the hot spot. The safety criteria impose to keep a specified minimum negative reactivity worth available from shim rods at all times during a cycle (that may require a modification in the BR2 core arrangement).



fig. 1. model of fuel element

BR2 standard fuel elements type 6n contains 6 concentric Aluminium fuel plates 0.127 cm thick 97 cm long; the side plates are 0.59 cm wide; fuel meat is a dispersion in Al powder 0.051 cm thick and 76.2 cm long; the inner available space has a diameter of 2.6 cm and the Be channel has a diameter 10.82 cm. The water gap is  $0.30 \pm 0.03$  cm (see fig. 1).

This preliminary study was focused on a high density fuel: 8 gU/cm<sup>3</sup> (or ~35%vol dispersion) and on determination of suitable burnable poisons. The standard fuel geometry (meat and cladding thickness) was conserved. Uranium being enriched at 19.75% in <sup>235</sup>U, the fuel density becomes 0.0806 g<sup>235</sup>U/cm<sup>2</sup> (536 g<sup>235</sup>U for a standard fuel element type 6n); this may be the maximum acceptable.

This preliminary study has also considered dispersion fuel: 7.1 gU/cm<sup>3</sup> (or  $\sim$ 30%vol dispersion), where the fuel density becomes 0.0715 g<sup>235</sup>U/cm<sup>2</sup> (476 g<sup>235</sup>U for a standard fuel element type 6n). These fuel elements are referenced as types L1 and L2 respectively in this paper.

#### 2. Integral transport in large cell

For calculating the neutron fluxes, the method used by the GEXBR2-TRPT4 code is the integral transport theory, that introduces the most significant parameters of the neutron transport between neighbouring cells, after the elimination of angular variables and the elimination of spatial inhomogeneities. To make the collision probability method applicable to large cells, the albedo between cells of different compositions is calculated simply with the first order correction according the generalised perturbation theory: the net current at the boundary is given by the difference of the leak ( $\Sigma_{L,i}$ ) between neighbouring cells taking into account the escape probability for absorption and the escape probability to be slow-down in a given interval of lethargy [1].

$$(\Sigma_{L,i} + \Sigma_{a,i} + \xi \Sigma_{s,i}) \cdot \Phi_i = S_i + \int_j \longrightarrow_i \Sigma_{L,j(i)} \cdot \Phi_j / 6$$

$$\Sigma_{L,i} = \Sigma_{et,i} \cdot P_{esc} + \Sigma_{a,i} \cdot G_i + \xi \Sigma_{s,i} \cdot dQ_i/du$$

where:

Large cells  $(2.r_0 \Sigma_{tot.i} > 4)$ , good moderation  $(\Sigma_{et.i} >> \Sigma_{a.i})$  and several lethargy intervals generally permit to neglect the direct influence of second neighbouring cells. The modal approximations [3] is used to obtain average cross-sections, taking into account the axial burnup, the microscopic distribution of the neutron flux, effective resonance cross-sections and self-shielding having been previously calculated in each BR2 cell,... Axial neutron flux in a BR2 channel is obtained by composition of nodal neutron flux and of currents with neighbouring BR2 cells. But the main advantage of this model is the significance of the diagonal term in the matrix system describing the neutron flux distribution in the reactor, making easier the perturbation calculation; indeed the thermal adjoint flux can be expressed in a simple elliptic equation :

$$(1 - A_i) \cdot \Phi_i^+ = \int_{j \to i} \Phi_{j(i)}^+ / 6 \quad \text{where} \quad A_i = 2 \cdot r_0 \cdot (v \Sigma_{f,i} / k_{1group} - \Sigma_{a,i})$$

because in large cell 
$$P_{esc} \approx 1 / (r_0 \Sigma_{tot}) \rightarrow 2 r_0 \Sigma_S P_{esc} \approx 1$$
.

The parameter  $A_i$  actually represents the 'fraction of neutrons available per cell'.

A configuration of 6 fuel elements around the channel D\_0 [4] with fresh fuel elements type A 6n  $244g^{235}U$  (HEU=93% and no burnable poison) gives:

$$A_i = 2.r_0 . (v \Sigma_{f.i} / k_{1group} - \Sigma_{a.i}) = 1.0 / k - 0.5 = 0.2 \implies k_{1group} \approx 1.25$$

In configuration 10 (fig.2) with standard fuel elements type G, we found:  $k_{1group} = 1.30$  (see fig. 3).

The adjoint of thermal neutron flux is the general shape of a configuration when the BR2 core contains an homogeneous fuel loading.

The perturbation theory [5] applied on the system: **B**.  $\Phi = \lambda F \cdot \Phi$  gives the variation of the eigenvalue  $\lambda$  and the perturbed solution  $\Phi' = \Phi + \delta \Phi$ ; the variation of the eigenvalue is obtained with the bilinear product by weighting the neutron importance:

$$\delta \lambda = \lambda' - \lambda = \frac{\langle \Phi^+.(\delta B - \lambda.\delta F)\Phi \rangle}{\langle \Phi^+.F\dot{\Phi} \rangle} \quad \text{and} \quad (B' - \lambda'F')\delta \Phi = -(\delta B - \delta(\lambda F))\Phi$$

#### 3. Neutron cross-sections

The cross-sections for <sup>235</sup>U, <sup>236</sup>U, <sup>237</sup>Np, <sup>238</sup>U, <sup>239</sup>Pu, <sup>10</sup>B, <sup>149</sup>Sm, Mo,... have been determined by the standard library ENDF-6 used by the code MCNP-4B in the meat of fresh standard fuel elements type G 6n containing 400 g<sup>235</sup>U (HEU=90% +3.8 g B.nat + 1.4 g Sm.nat) loaded in channels C of the reference core loading of cycle 9007A. The resonance cross-section (above 0.5eV) of <sup>238</sup>U is found 264.4 barn for HEU and 65.2 barn for LEU. Thermal flux depression in fuel plate and epithermal self-shielding are calculated at each step of the activation of transuranians by reciprocal escape probabilities. The radial distribution of neutron flux is calculated by transport of thermal neutrons in concentric annular cells with diffusion boundary condition (which determines the epithermal ratio).

#### 4. Economical use.

In the present BR2 reference core arrangement (see fig. 2), fresh fuel elements are loaded in channels C (2.0 MW), and partially used fuel elements are successively loaded in the channels A (2.4 MW), B (2.0 MW), D and F at the  $2^{nd}$  until 5<sup>th</sup> batch (D and F channels receive low power in order to optimize neighbouring irradiation conditions); at the present reference power of ~ 60 MW the power generated in channels A,B and C is 37 MW.

The economical use of the fuel cycle is determined by the energy produced by a fuel element (fig. 3) until it reaches the burnup value for elimination. The residual fuel mass has been observed to be about 200  $g^{235}U$  in an external fuel channel during the last operation cycle.



The cycle length is determined by burnable poisons to be consumed at the end of first operation cycle. The variation of the reactivity excess of a core loading at the end-of-cycle in repetitive loading of N=n.k fuel cells (*n* fuel elements of *k* batches of burnup) looks as follow:

$$\delta \rho = \delta \left[ n \sum_{i=1}^{i=k} \rho(\beta_i) \right] \quad \text{or} \quad \delta \rho = \delta n \left[ \sum_{i=1}^{i=k} \rho(\beta_i) \right] + n \left[ \delta \rho(\beta_{el}) - \delta \rho(\beta_{el} / k) \right]$$

Because burnable poisons have to be burnt at the end of the first batch,  $\delta \rho(\beta_{el}/k) = 0$ , the burnup at elimination  $\beta_{el}$  determines the energy to be produced in a fuel element. The present core loading contains N=30 fuel elements type G 6n of 400 g.<sup>235</sup>U in k=5 batches, n=6 fresh fuel elements

loaded while also **n=6** fuel elements are eliminated. We found the reactivity of a fuel element by the gradient at end of life :  $\beta_{el} = 50\%$ .

$$\rho_{fuel.el} = \beta_{el} \cdot \frac{\partial \rho}{\partial \beta}\Big|_{el} = 1.4 \text{ } \text{fuel.HEU} \text{ and } \approx 1.0 \text{ } \text{fuel.LEU}$$

Near the burnup at elimination, we obtain after averaging axial isotopic composition:

$$\beta_{\max} = 1 - \exp(-\sigma_{a.U5} \cdot \Phi_{\max} \cdot \tau = 1) = 63\%$$
 and  $\beta_{mean} \approx 1 - (1 - \beta_{\max})^{0.70} = 50\%$ 

The mean axial isotopic density retated to initial <sup>235</sup>U mass is given at table 1.

			table 1				
type of fuel	U-235	U-236	Np-237	U-238	Pu-239	Pu-240	Pu-241
A 244gU5 HEU	0.5066	0.0744	0.0012	0.1065	0.0014	0.0003	0.0001
G 400gU5 HEU	0.5066	0.0758	0.0017	0.1050	0.0021	0.0004	0.0001
L1 536gU5 LEU	0.5066	0.0769	0.0022	3.9423	0.0296	0.0050	0.0022
L2 476gU5 LEU	0.5066	0.0765	0.0020	3.9425	0.0294	0.0051	0.0021

According the composition of fuel elements of types L1 and L2 (U-Mo has 7wt% Mo density), the reactivity variation at BOC and fluxes versus the equivalent burnup  $(^{235}U+^{239}Pu)$  for the energy produced (1.24 g<sub>fissile</sub>/MWd) in a BR2 channel with an unitary weight (in a core loading of 30 fuel element of the same type A,G, or L and a mean burnup of 25%) is presented at figures 4 and 5. The perturbation of the thermal neutron flux and of power generated in a fuel cell is presented at figures 6 and 7.



fig. 4 reactivity versus burnup at Wi=1



fig. 6 relative thermal neutron flux



fig. 5 reactivity versus equivalent burnup



fig 7 relative power

The loss of reactivity at the  $2^{nd}$  and  $3^{th}$  batch (to be multiplied by the weight of 6 central ring channels A and B : Wi=1.5) is respectively 0.07\$ and 0.05\$ for type L1 ( $\beta$ el=50% for  $\rho$ = -0.18\$) and 0.04\$ and 0.11\$ for type L2 ( $\beta$ el=40% for  $\rho$ = -0.12\$,). With reference to a HEU core arrangement, the LEU antireactivity is quoted 0.72\$ and 0.9\$ respectively for types L1 and L2. The antireactivity of the experimental load reduces the burnup at elimination proportionally to the gradient of reactivity of the fuel element at elimination.

		table 2			
	type 6n :	A	G	L1	L2
fuel mass	U-235 [g]	244	400	536	476
	enrichment	0.90	0.90	0.1975	0.1975
poison	Boron [g] (B₄C)	0	3.8	3.8	3.2
·	Sm [g] (Sm <sub>2</sub> O <sub>3</sub> )	0	1.4	1.4	1.4
density	g <sup>235</sup> U/cm2	0.037	0.060	0.0806	0.0715
·	g U/cm3	0.80	1.31	8.0	7.1
	form of U-X	UA14	UAlx	U-Mo	U-Mo
	%vol U-X	24%	28%	35%	30%
elimination	$\beta \ (\rho =36\$)$	33%	50%	59.4%	56.6%
	$\beta_{e1}$ ( <sup>235</sup> U+ <sup>239</sup> Pu)	30%	50%	50%	40%
	$\beta_{a1} = (^{235}U + ^{239}Pu)$	40%	63%	63%	52%
reactivity	ρ <sub>fuel al</sub> max [\$]	0.95	1.39	0.98	0.74
fission/cm3	max [1e+21]	0.625	1.60	2.14	1.57
	fuel.el /1000 MWd	~13	6.2	4.63	6.5
	batches	2	4 - 3	4	4
	cycle length [day]	14	21-28	28	20

# 5. Conclusion

The reactivity curves of LEU fuel elements with at least 480 g  $^{235}$ U indicates the feasibility of repetitive cycle length of 21 days with a consumption rate of 6.2 fuel elements for 1000 MWd [1].

The fissile load being increased when using LEU fuel elements the reactive effect of devices and of shim rods is decreasing accordingly. The worth of shim rods is presently minimum  $\geq 12$  \$ in the configuration 10 and therefore the core arrangement has to be modified for a whole core conversion.

The BR2 reactor is operated at constant power. The thermal neutron flux is strongly reduced in the axial position of fuel elements and by influence the thermal neutron flux is also reduced in neighbouring reflector channels. More detailed whole core calculations will have to be performed to check the irradiation conditions of the experimental devices and the hot spot factors in driver fuel elements.

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# BEHAVIOUR OF UMO WITH CLADDING FAILURE UMUS TEST IN THE HFR

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## ABSTRACT

The High Flux Reactor, located at Petten (The Netherlands), is often used to investigate various types of MTR fuel. During the year 2000 an irradiation test called UMUS was made on four UMo plates with a dedicated device.

After two cycles, a cladding failure occurred on one of the plates.

This paper explains how this cladding failure was detected, the response of measurements systems and the necessary time before to stop the plant.

The detection procedure to confirm the cladding failure is also explained.

From non-destructive and destructive examinations performed on the failed plate, it is also possible to give geometric indications on the failure itself and on the quantity of fission products released from the failed plate.

In conclusion, the information presented demonstrate the possibility of promptly detecting UMo cladding failure, of measuring fission product release and of assessing the consequences of such an occurrence. This experiment is intended as a part of the validation report of the UMo fuel.

## **1. Introduction**

UMo Project is a working group with the objective to develop new high-density low enrichment Uranium (LEU) fuel for research and testing reactors. This French UMo group is composed of five partners, namely CERCA to test the manufacture feasibility, CEA for neutronic calculations and incore performance studies, COGEMA for investigating the reprocessing feasibility, Technicatome and Framatome. Their investigation is concentrating on the development of Uranium-Molybdenum (UMo) alloys with 8-10 gU/cm<sup>3</sup> [1].

The irradiation programme, supervised by CEA, started with two series of irradiation of few full-sized plates: one at the OSIRIS reactor with on-line measurements of the plate thickness during the reactor stops and one at the HFR Petten. The objective of the irradiation experiment in the HFR, called

UMUS was to irradiate these experimental plates to a  $^{235}$ U burn-up of at least 50%. A release of fission product in primary water was observed during the irradiation and has conduced to shut down the reactor. The experiment has been unloaded from the core due to a failure detected in core and confirmed afterward from the post irradiation examinations.

This paper analyses the behavior of UMo fuel, in case of cladding failure: release of fission products, detection, consequences and comparison with other bibliographic results.

## 2. Irradiation history

Four UMo plates with 7 & 9% Mo and with 19.75 & 35% enrichment in  $^{235}$ U have been irradiated in a dedicated device, in the HFR, in the core position D2, [2]. The UMUS experiment irradiation has been stopped after 48.4 <u>Full Power Days</u> (FPD), due to a failure of plate. At the end of the irradiation, the average burn-up for the 4 plates has been estimated less than 15%.

#### 3. Chronology of events/Detection of failure

First important peak of activation products activity in primary water was noted on June, 3<sup>rd</sup> 2000, after 47 FPD. The evolution of primary water and fission products activity from the 2<sup>nd</sup> June to the 4<sup>th</sup> June is shown in Fig 1. The first increase of primary water activity and following fission gas monitor indications occurred at 02:00 AM. Afterwards, the activity became stabilized but still at elevated level. Several small peaks of activation products activity in primary water have been observed.



Fig 1. Evolution of primary water activity and fission products activity during the UMUS failure

A water sample taken the morning after showed that gaseous fission products were present. The results of the water analysis from primary water show that the activity is due to the fission products (Xenon, Krypton, Iodine, etc.). The results of radioactivity measurement in the primary loop water are typical of a nuclear fuel failure are given in Table I. In spite of an increase of primary activity and increase of fission products we had no fuel failure monitoring indications. Since the water activity was still decreasing and there were not signs of complete fuel failure the preliminary conclusion was that a fuel blister has released its activity. The discussions on the observations and preliminary conclusions continued during the following 24 hours with the shift supervisors involved.

On the day after a second peak of activity occurred. Since this peak was more profound and did not diminished immediately it was decided to end cycle 2000.05 one day in advance i.e. on Sunday 4<sup>th</sup> on June at 16.00. That means an operation with cladding failure of 38 hours. The KFD (Dutch safety authorities) has been informed the following day. A decision was made to keep the primary system closed for about 24 hours to enable decay of short-lived isotopes. The initial activities with the primary system were done by operators weaving protective clothing and air masks after additional measurements these measures were abounded.

A leakage from UMUS experiment was suspected. UMUS has been unloaded for inspection. Water was taken above UMUS experiment and above standard fuel elements. Measurements of water activity in the pool were made for reference. The gamma-spectrometry results show an important activity above UMUS, 50 times higher than above standard elements. This confirms the failure of UMUS experiment.

A control of activity in water has been carried out by gamma-spectrometry from the 3<sup>rd</sup> June 2000 first peak of activity to the 13 June 2000 reactor re-start, see Table II. An activity of I-131 of about 7050 MBq was released in the primary water. Due to the release of the fission products in the pool, we decided to isolate UMUS experiment from the pool water. The UMUS sample holder has been inserted in a closed holder to avoid further contamination of the pool. The stainless steel container was stored in the HFR hot cell pool.

Radionuclide	T 1/2	Activity (MBq/m <sup>3</sup> )	Activity (MBq/m <sup>3</sup> )
		on June, 3 <sup>rd</sup> at 14:43	on June, 5 <sup>th</sup> at 9:42
Kr-85m	4.5 h	158	31
Kr-88	2.8 h	203	9
Xe-131m	12 d		140
Xe-133	5.3 d	3648	4360
Xe-135	9.1 h	266	
Na-24	15	3720	382
Mn-54	312 d	54	168
Co-58	71 d		
Mo-99 +	66 h		7
Cd-109	453 d	155	61
Cd-115 +	45 d	234	
Sr-91	9.5 h		141
Ru-106	368 d		14
Te-132	76 h		45
I-131	8 d	30	237
I-132	2.3 h	40	68
I-133	20.8 h	84	316
I-134	52 m	117	59
I-135	6.6 h		
Cs-138	32 m	440	30

Table I. Primary water analy	ses
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Based on the calculated decay heat power some heat transfer calculations have been performed to estimate the cooling conditions for the UMUS fuel plates in water and in air. A calculation was made to assess the temperature rise that will occur after stopping the natural convection. The results showed that due to the decay heat the UMUS plates had to remain in the reactor pool, to avoid high temperature of the plates itself, for 3 months after the unloading. After the time cooling UMUS sample holder has been transported from the pool reactor to the hot cell laboratory of NRG for examination.

Date	Location	Activity (MBq/m <sup>3</sup> )
03-06-2000	Primary water	30
05-06-2000	Primary water	68
09-06-2000	Pool above UMUS experiment	199
09-06-2000	Pool	2
11-06-2000 to 13-06-2000	Pool	1.40

Table II. Activity measurement of I-131

#### 4. Post-irradiation examination results

The post-irradiation examination, confirms that the cause of the activity in the preliminary circuit is due to the failure of one of the plate U9Mo 35 %. The post-irradiation examination of plates has been performed in the hot cell laboratory of NRG, Petten [3], [4]. Large spots with decolorized zones have been observed on the two 35 % enriched plates. The high enriched U-9% Mo plate (No4) failed at the maximum flux location where a 7 mm long transversal crack can be observed. It was observed that a significant swelling of the plate U7Mo 35% had occurred. The investigation of UMo/AI interaction from electron probe micro-analysis (EPMA) on a sample of this plate shows that the UMoAl3 compound has been formed. **Fig 2** shows a cross-section of the radial gamma spectrometric measurement performed on the failed plate. From the gamma spectrometric analysis, the loss of fuel is evaluated at about 0.085 cm<sup>3</sup> of meat, that means about 0.75 ± 0.15g of alloy with 0.23 ± 0.02g of U235. The most quantity of UMo fuel lost has been released in the water and some of radionuclides have perhaps migrated around the failure. This could explain the high activity near the failure see **Fig** 2.

An aluminum oxide layer has been observed at the outside of the aluminum cladding. Considering the behavior of this layer in the SEM and optical microscopy it is rather certain that this layer consists of aluminum oxide. The thickness of this layer has been measured and ranges from 10-80 micron, with the highest rated positions having the thickest oxide layer. This aluminum oxide could be a boehmite layer ( $Al_2O_3$ , H2O), the details of the post-irradiation examination are given in [5].



#### Fig 2. Gamma spectrometric measurement of Ce-144 in the failed plate (U9Mo35%)

#### 5. Discussion on UMo behaviour under cladding failure

In the irradiation conditions of HFR (water temperature about 40°C, pressure 4.0 bar), we have lost in 38 h of operation about 0.75 g of meat, essentially in two separates peaks. A correlation of the estimation of fissile material volume lost with the increase of activity measured in primary water, or with the filters extra-activity is very difficult and has not been tried because these activities are created mainly by specific fission products.

The behavior of UMo in water observed during the incident is in good agreement with the existing bibliography.

In first some tests were made in [6], where corrosion of UMo samples in water was tested for different Mo concentration in function of time. The order of values obtained was a weight loss of about 10 mg/cm2 in one day in water at 315°C. These values show already a good behavior of the UMo fuel in water.

The good behavior of the UMo fuel irradiated in a reactor cooled by primary coolant water is confirmed by a similar failure reported by the Argone National Laboratory, ANL [7]. In 2001, a series of U-7Mo dedicated plates have been irradiated in the ATR (Advanced Test Reactor). 10 days after irradiation start a slight increase activity afterwards an activity decreased to normal levels has been

observed during the irradiation. The reactor has not be shutdown, the experiment remain in the reactor with no further activity change. After 116 EFPD'S (Effective Full Power Days) a small lateral crack was observed on a plate loaded at 8 gU/cm<sup>-3</sup>. From the post irradiation examination, it has been concluded that the failure was due to a fabrication defect. The quantity of fuel lost was estimated at 0.024 cm<sup>3</sup> of fuel alloy that means 0.42 g including 0.19 g of U235. It seams, with the fact that the fuel plate operated for more than 100 days after initial release that the transport through the defect after initial release on the defect, slowed down substantially or ceased.

## 6. Conclusion and next future

The UMUS test has confirmed the good behavior of the UMo fuel in case of cladding failure .The quantities released allow quick detection, but remain very low and without problems for the operator. Even in this test where too high temperatures were reached for the UMo fuel with the plates enriched at 35%, the release was bigger than in other bibliographic test, but without problems or consequences. This test can be used as a further validation of UMo behavior in case of cladding failure.

In the near future, a new irradiation of 4 UMo plates with 20% enrichment in LEU HFR standard device, named TULIP is planned for 2002. The design is ongoing. The new UMo experiment should consist of 4 UMo plates with inert plates loaded in a HFR standard device.

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# **Best Safety Practices for the Operation of Research Reactors**

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# Abstract

A survey on administrative, organisational and technical aspects for the safe and efficient operation of a 250 kW TRIGA Mark II research reactor is given. The replacement of the I&C system is discussed, maintenance procedures are presented and the fuel management is described.

#### 1. Introduction

Of the presently operating 278 research reactors 160 reactors are older than 30 years and have been constructed in a period where the licensing or safety requirements were far less stringent as of today. Further each facility has particular characteristics due to it's location (near city at a university campus or larger research centre away from the city), due to its utilisation (students training and education or material research, isotope production etc.) and due to its position in the country's infrastructure (country with a large nuclear program, country on the way to a nuclear program or country without any nuclear program) existing system.

The TRIGA Mark II reactor Vienna is a good example of an old (40 years), low power (250 kW) research reactor facility operating uniquely for students education and training in a country which has no nuclear power program and who in fact aims to convince the neighbouring countries to abandon nuclear power.

In the past 40 years many efforts have been invested to keep the reactor facility in an excellent organisational and technical state. On the organisational side the reactor group is responsible for the routine reactor operation, for all repair-, maintenance- and modification work, for training and retraining of reactor operators and for documentation of operational-, maintenance- and repair data. Maintenance-, training- and failure data collection programs were established which, in some cases, have been also implemented at research reactors with similar infrastructure.

Covering the more technical aspects of research reactors the reactor instrumentation has been modernised recently to be the only TRIGA instrumentation outside the USA operating a digital I&C system. Special optical inspection methods for the reactor tank and its internals were developed which have been also applied at several other research reactors in Europe and overseas. Fuel management has been carried out successfully in such a way that about 2/3 of the original core loading is still in use even with a high utilisation factor and totally only 8 fuel elements had to be eliminated from operation in 40 years.

This paper concentrates on the organisational and technical topics mentioned in the above paragraphs and gives a survey on safety practices for a typical low power research reactor.

## 2. Organisational and administrative safety practices

When the TRIGA reactor was initially started up in 1962 no Atomic Law was established in Austria and the reactor was licensed on an interim basis according to an environmental law, only in 1975 the reactor was finally licensed under the Austrian Law for Radiation Protection. This required a number of new

administrative and organisational procedures. One of them was to establish a reinspection and maintenance plan covering all reactor related systems. This plan has been several times upgraded and modified and is now in a well developed form [1]. It requires monthly, quarterly, semi-annually and annual inspections of all safety related components. Some of the inspections have to be carried out in presence of a governmental appointed external expert, other are carried out by external certified companies. Special inspection forms for each component have been developed which are completed during the inspection process. These forms are collected and submitted to the regulatory body once a year for evaluation and are the basis for the annual renewal of operation permission. As an example it is required that all nuclear channels are recalibrated once a year with an external signal generator, all temperature channels (water and fuel temperature) have to be recalibrated with signal generators, all alarm and scram settings have to be tested for its proper function. This work takes about 4 man-weeks.

A failure data collection system has been established where all component failures are recorded and analysed for the failure cause, the failure mode, for the impact on reactor safety etc. A special event record system has been developed [1], which allows further evaluation of the data for other purposes, i.e. for probabilistic safety analysis. The TRIGA reactor Vienna also participates in the Incident Reporting System for Research Reactors (IRSRR) established by the IAEA. In the past 40 years about 6 safety related incidents had been reported to the IRSRR most of them caused by experimental facilities around the reactor. In summary the incidents were as follows:

- Overnight flooding of the rotary specimen rack (1979)
- Sudden break-off of the absorber part of the transient rod from the guide rod (1980)
- Inlet tube of an aerosol monitor drops into the pool and draining 30 litres of water (1984)
- Leakage of pneumatic transfer system near core terminal and consecutive spraying of contaminated water when operated (1985)
- Leakage of pneumatic transfer system joints above pool water level and consecutive dust contamination when system was operated (1989)
- Rupture of a pneumatic system coupling under the pool water level (2001)

More detailed information is available through the IRSRR system.

#### 3. Technical safety practises

In order to keep the TRIGA reactor Vienna in an excellent technical state the I&C system has been replaced three times in this period. Originally, the reactor started with an electronic tube type instrumentation which operated until 1968. It was replaced by an analogue transistor type instrumentation operating until 1992. Finally, in 1992 a digital computerised instrumentation was installed with some upgrading (Y2K) which is in operation now (Fig 1.). Of course, this upgrading was only possible due to the financial support by the responsible Ministry of Science and Research and by the Technical University Vienna. This documents also the interest of the Austrian Government in nuclear sciences in spite of strong anti-nuclear tendencies.

It has to be mentioned, however, that the standard console offered by the supplier (General Atomics) had to be modified considerably both in order to fulfil the Austrian safety requirements following German standards and to allow efficient utilisation for training and education.

Another requirement is that all fuel elements have to be controlled for their elongation and bowing once a year with a special underwater measuring tool. Elongation limits are given by the supplier (6.5 mm for Al clad fuel and 2.54 mm for SST clad fuel) and the measured values are compared with previous years [2,3]. If a fuel element exceeds the maximum permissible elongation value it must be removed from the core. Since 1962 only 9 fuel elements had to be removed from the core, 5 of them due to excess
elongation and 3 of them for fission product release. This measurement procedure accounts for about 2 man-weeks (Tab. 2).



Fig 1. Diagram of the digital computerised instrumentation

Overall elongation since 1962 in mm	Number of fuel elements
< 0	3
0 - 1	17
1 – 2	18
2 – 3	20
3 – 4	11
4 – 5	7
5 - 6	3
	79

Table 2. Summary of fuel elongation measurements

Limits for fuel elongation set by fuel supplier: Al-clad fuel: 6.3 mm or 1/4 in SST-clad fuel: 2.54 mm or 1/10 in Besides the fuel inspections mentioned above, core management and fuel acquisition is an important fact for safe and efficient reactor operation. Fuel purchase started in 1965 in intervals of 2 to 3 years depending on the fuel consumption due to reactor operation. The last acquisition of fuel was actually triggered by the decommissioning of the TRIGA reactor Heidelberg, where it was possible to obtain 8 standard TRIGA fuel elements for symbolically 1 DEM each. Therefore, the total number of fuel elements at the facility are 104 while 9 of them have been removed permanently (Tab. 3).

Number of FE	Location	Clad	ding	Enrichment	Remarks
		Al	SST		
80 +	core	57	25	70 FE 20%	2 instr. FE
2 stored				9 FE 70%	
13	fresh fuel storage	-	13	20%	2 instr. FE
8	spent fuel storage	8	-	20%	1 instr. FE
1	hot storage facility	1	-	20%	cut into 3 pieces
total: 104	ý	66	38		

Table 3. Fuel element situation as per 1. 1. 2001

Other important procedures directly related to the safety of the facility are the development of inspection and maintenance methods especially for the reactor core and core internals. These methods have been published at several other occasions [4,5] and are repeated here only in summary form. They consist of

- Optical underwater endoscope with different viewing angles
- High pressure water jet to clean pool areas not directly accessible
- Tank cleaning pump with coarse and fine filters
- Pick up tool for small objects
- Miniature high density underwater lights
- Replica method

These methods have been developed and improved throughout the years and they were applied upon request from the IAEA or from individual research reactor operators successfully at four European research reactors [6] and at one overseas research reactor.

Off course, some of these maintenance methods described above are routinely applied at the TRIGA reactor Vienna. For example every three months the reactor tank internals are cleaned with the high pressure water jet and deposits and dust from the tank bottom and below the core are stirred up. At the same time the tank cleaning pump is operated to filter the water and to remove particles which had been stirred up with the water jet. Objects visible with binoculars are picked up with the special pick up tool. In addition to the annual fuel elongation measurement, each fuel element is removed from the core, turned and reinserted to check the free movement of fuel elements in their core position every 3 months.

Further important aspects of efficient and safe utilisation of research reactors is the constant information exchange between other operators. Therefore, the Atominstitute Vienna participates regularly at the following working groups:

- Arbeitsgemeinschaft Forschungsreaktoren (AFR = German speaking reactor club) meets two-times a year
- Research Reactor Operation Group (RROG), a subgroup of the European Atomic Energy Society (EAES) meets once a year
- TRIGA Conference meets every two years

- Nuclear Society of Slovenia meets once a year
- Research Reactor Fuel Management Conference, meets once a year
- IGORR meets every 18 months

The participation is the basis of a very valuable information exchange of current technical and administrative developments at an early stage and the personal contacts between the members are of utmost importance.

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# **Session 4**

Spent fuel management, corrosion and degradation



#### INFLUENCE OF THE FUEL OPERATIONAL PARAMETERS ON THE ALUMINIUM CLADDING QUALITY OF DISCHARGED FUEL

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#### ABSTRACT

In the last two years, the new MR6 type fuel containing 1550 g of U with 36% enrichment has been loaded into MARIA reactor core. Its aluminium cladding thickness is 0,6 mm and typical burnup - about 4 080 MWh (as compared to 2 880 MWh for the 80% enriched fuel used). However, increased fission product release from these assemblies was observed near the end of its operational time.

The results presented earlier [1] show that the corrosion behaviour of aluminium cladding depends on the conditions of fuel operation in the reactor. The corrosion process in the aluminium of fuel cladding proceeds faster then in the aluminium of constructional elements. This tendency was also observed in MR-6/80% and in WWR-SM fuel assemblies.

Therefore the visual tests of discharged MR-6/36% fuel elements were performed. Some change of appearance of aluminium cladding was observed, especially in the regions with large energy generation i.e. in the centre of reactor core and in the strong horizontal gradient of neutron flux. In the present paper, the results of visual investigation of discharged fuel assemblies are presented. The results of the investigation are correlated with the operational parameters.

#### **Reactor MARIA and MR fuel.**

The multipurpose high flux reactor MARIA is water and beryllium moderated, graphite reflector, water cooled reactor of a pool type with pressurized fuel channels containing concentric six tube assemblies of MR type fuel elements. The fuel channels are situated in a matrix made of beryllium blocks and surrounded by lateral reflector made of graphite blocks in aluminium cans. In original fuel assemblies of MR type, the 80% enriched uranium was used.

The new version of MR fuel assemblies has been designed and produced by USSR organisation especially to supply the MARIA reactor. In such assembly the fuel meat is a mixture of aluminium with  $UO_2$ , where U-235 is of 36% enrichment. The design principles and overall dimensions are the same as for fuel assemblies used before with 80% enrichment. MR6 fuel assembly was designed as a system of 6 concentric tubes. The fuel mixture in each tube was situated between aluminium clad. The thickness of each tube was 2 mm. The technical parameters of new and old fuel assemblies are shown in the table bellow. The construction of fuel assembly is shown in fig 1.

	MR6 (36%)	MR6 (80%)
Fuel composition	UO <sub>2</sub> in Al	UAl <sub>x</sub> in Al
U mass (g)	1530	428,9
U-235 mass (g)	550	345
Fuel thickness (mm)	0,64 ÷0,76	0,4
Clad thickness (mm)	$0,62 \div 0,68$	0,8
Burn up limit (MWh)	4080	2880
Max. power of fuel ass. (MW)*	1,6	1,6

\* Limited by the Polish Regulatory Body.

First new fuel assemblies were loaded in MARIA reactor in April 1999 and their number in the core was increasing in 2000 and 2001. Due to greater content of U-235 in MR6(36%) fuel assembly,

the number of fuel assemblies residing in the MARIA reactor was diminished from 17 do 14. In this period, the increase of the radioactivity in primary cooling system was observed



Fig. 1. Fuel assembly MR6(36%)

Reactor MARIA was equipped with a system for identification of fission product release from separate fuel assemblies. It is based on the detection of delayed neutrons from fission products released to the cooling water. This system can be connected to a particular fuel cooling channel and the delayed neutrons can be detected. The results of measurements show, that the fission product release from MR6/36% assemblies is greater by one order of magnitude then from MR6/80% assemblies[2]. In Fig. 2, the typical results of measurements of fission product release from MR6/36% assembly as compared to MR6/80% assembly in the course of operation are shown. The similar character of results was observed for other fuel assemblies.



Fig. 2. The dependence of fission product release during operation from MR6/80% assembly and MR6/36% assembly.

#### Visual inspection of MR6(36%) fuel assemblies.

Taking into account the results of the fission product release measurements, the visual inspection of few MR6(36%) fuel assemblies discharged from MARIA reactor in 2000 and 2001 was performed. Underwater pictures of the spent fuel cladding surface, registered by underwater video camera, were recorded on the video SVHS tape recorder and converted to bmp file using PC with MUTECH video card. The system was described in [1]. Of course, the intense gamma radiation of fuel assembly with short "cooling" time have the influence on the picture i.e. some flash points have been observed. But general picture is clear.

It can be stated, that we have observed the dependence the condition of the aluminium clad of on the type of uranium fuel used. Because of manufacturing technology of MR6 fuel, the boundary of the uranium meat was not uniform at the tip of fuel assembly. Such non uniform distribution of uranium at the tip of fuel assembly was confirm by the radiographic picture of fresh MR6 fuel assembly – Fig 3.



Fig. 3. Gamma radiography of the tip of the MR6 fresh fuel assembly.

The results of visual investigation of MR6/36% fuel assembly shows, that the aluminium clad surface – just after discharging from reactor core - has different state in the area with the fuel meat beneath. The photo of the tip of MR6/36% fuel element is presented in fig 4.



Fig 3. The video picture of MR6/36% fuel assembly tip with visible line of uranium distribution.

The similar results were observed in the corrosion processes – but only after ~15 years of storage in wet conditions – for MR6/80% fuel assemblies. The video picture of such fuel assembly tip is presented in fig 4. The programme of analysing of these phenomena is in elaboration.



Fig. 4. The video picture of MR6/80% fuel assembly tip with visible shape of uranium distribution (storage time - 17 years).

As the second result, it was observed, that the cladding of MR6/36% spent fuel assemblies discharged from the reactor core has areas with significant surface changes. On the surface the fuel black blemishes were observed in all investigated fuel assemblies. As an example, the picture of close to centre part of clad surface of MR6/36% fuel assembly with 4514 MWh burn up and discharged from reactor core at November 2000 is presented in Fig 5. This fuel assembly was exploited in the core region with significant gradient of neutron flux (about 20% in the horizontal area of fuel assembly) and therefore the clad state changes have the shape similar to the power flux distribution on the fuel surface.





Fig. 5. Photo of the MR6/36% fuel assembly cladding and its position in vertical flux distribution along the reactor core.

#### **Conclusions.**

The results of measurements of fission product leakage from MR6/36% fuel assemblies and results of visual investigation of clad condition lead to the conclusion, that during the storage time they should be regularly supervised. The sipping test and video control should be performed periodically for all MR6/36% spent fuel assemblies.

These results should be taken into account by the manufacturer of the MR fuel to improve production technology of such fuel assemblies in the future.

The research project for investigation of the observed phenomena is in preparation.

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#### MIR REACTOR FUEL ASSEMBLIES OPERATING EXPERIENCE

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#### ABSTRACT

The report summarizes the results of the MIR reactor operating FAs usage within the period from the start-up of the reactor in 1967 till present. The principle characteristics of various FA modifications operation are cited in it. Listed here are the same results of the post-irradiation examinations of fuel elements with burn-up of 30% and 87% under the standard operating conditions in the MIR reactor the FAs were operated with high reliability and serviceability. Prior to the transition to the ribless design there were no cases of FAs failure. On usage of the ribless FAs at the close to maximum heat power level 12 cases of FAs failure were detected. On the whole there were  $\sim 2350$  FAs used. This paper considers the possible causes of FAs failure and the trends in activity to improve the MIR reactor fuel serviceability.

#### Introduction

The MIR reactor was brought to power in mid-1967 and has been under operation till nowadays [1]. The tube-type FAs are used as standard nuclear fuel for this reactor. The UAl<sub>x</sub> or UO<sub>2</sub> particles dispersed in the aluminum matrix have been used as fuel particles within the fuel elements. The fuel matrix has a thickness of  $\sim 0.56$  mm and the height of  $\sim 1000$  mm, the fuel element claddings have a thickness of  $\sim 0.72$  mm. The material used for the fuel element claddings is SAV-6 aluminum alloy. As from the reactor commissioning till 1975 FAs consisted of 6 tube-type fuel elements with coaxial arrangement. Since 1976, after the reactor redesign the six-tube FAs were replaced by fourtube ones. To change the design two inner tubes were removed and the displacer dimensions were increased. The aim of the FA design modernization was to reduce the radial non-uniformity of energy release and to increase the mean fuel burn-up value, accordingly, as well as to increase the volume of the displacer used to position an ampoule for materials irradiation. Both in six-tube and four-tube FAs each fuel element had three spacing ribs located on the outer surface along the generating line, each 120°. In 1984 the UA1<sub>x</sub> fuel particles were replaced by uranium dioxide particles. In the late 80-s the next fuel element design change was made which consisted in transition from the rib-type outer cladding to the plain-cylindrical one. The main target of the fuel elements design change was to reduce the power distribution non-uniformity along the fuel elements perimeter, since there emerged an effect of fuel layer thinning in the ribs locations. Prior to mass production application 4 ribless FAs were successfully tested in the MIR reactor. However, it should be noted that the modified FAs testing was performed at the power of 2 MW maximum. From 1991 only ribless FAs were used in the MIR reactor. As of December 1999 ~ 650 ribless-design FAs were operated in the reactor. On the whole, under the rated operating conditions in the MIR reactor the FAs displayed high reliability and serviceability. Prior to the transition to the ribless design no cases of FA failure were recorded. On usage of the ribless FAs 12 cases of FA failure were detected. This paper presents the results of the FAs operating in the MIR reactor, the same results of the postirradiation examinations of fuel elements, the trends in activity to improve the MIR reactor fuel serviceability.

#### 1. Results of the MIR reactor operative FA Usage

As from the MIR reactor commissioning till present ~2350 FAs have operated in the MIR reactor.

The total period of the reactor operation at the set power makes up  $\sim$  19,5 years. To summarize the results of the FAs operation the whole operating period could be conventionally divided into 3 phases.



FIG.1. The MIR reactor FAs modifications.

Item	Characteristic	Dimension	Phase I	Phase II	Phase III
No			(1967-75)	(1976-90)	(1991-2001)
1.	FA type		rib type;	rib type;	ribless type;
			6 tubes	4 tubes	4 tubes
2.	Mean reactor power	MW	36÷50	25÷40	25÷50
3.	Max reactor power	MW	65	55	65
4.	Mean FA power	MW	0,75÷1,05	0,52÷0,83	0,52÷1,05
5.	Max FA power	MW	2,5	1,8	3,3
6.	Mean U-235 burn-up	%	~ 28	~35	~40
	in the FA				(60)*
7.	Time of the FA	days	~(40÷250)	~(60÷250)	~(30÷250)
	operation at the set				
	power				
8.	Axial non-uniformity		1,38÷1,74	1,38÷1,74	1,38÷1,74
	of power release along				
	the FA height				
9.	Coolant velocity	m/s	≤10	≤10	≤10
	between fuel tubes				
10.	Maximum heat flux on	MW/m <sup>2</sup>	3,85	2,80	5,10
	the fuel tubes surface				
11.	Number of spent FA		~500	~1090	~760
12.	Number of failed FA		-	-	12

Table 1. Basic characteristics of the reactor and the FAs operation in different operating phases.

\*Note : uranium-235 burn-up in the FAs during the last years of operation has increased as a result of beryllium blocks replacement and optimization of the active zone loading as well as the cycle duration.

On examining table 1 the following FA operation specific features in different phases could be noted. In phase II of the reactor operation if compared to phase I, the reactor and the FA power has dropped by ~20%, the burn-up has increased by ~30%. The given facts result directly from the reactor physical properties improvement due to the redesign (loop channels placement in the  $2^{nd}$  and  $3^{rd}$  reactor cells, opposite the  $2^{nd}$  and the  $4^{th}$  row before the redesign, beryllium blocks replacement, etc.). It was the modernization of the FA design that in a certain way also influenced the increase of fuel burn-up in the FA (going from 6 fuel elements to 4 fuel elements). In phase I and II if compared to phase III of the reactor operation no cases of fuel elements cladding failure in the FA have been detected. One more difference of phase III from the first two ones is that the maximum FA power has grown up to 3,3 MW, in phase I it has not exceeded 2,5 MW, in phase II - 1,8 MW. The given circumstance is in the first place connected with the specific character of the experiments being carried out in phase III ( providing of necessary heat loads in the experimental fuel elements with the

great burn-up depth, experiments with power leap, neutron flux high density creating during the experiments with structural and absorbent materials). In table 2 the FA coolant characteristics are given. The coolant parameters in all the reactor operation phases were identical.

Item No	Characteristic	Dimension	Value
1.	Inlet channel pressure:	MPa	1,1÷1,3
	Outlet channel pressure:	-"-	0,6÷0,8
2.	Inlet channel Temperature:	°C	40÷60
	Outlet channel temperature:	_''_	50÷95
3.	Flow rate	kg/s	8,3÷22,2
4.	pH		5,6÷6,0
5.	Conductivity	μS / cm	<1,5
6.	Solids	mg / kg	<1,5
7.	CI	_"_	<0,02
11.	N	n cm <sup>3</sup> /kg	≤50
12.	Ō		≤0,02

Table 2. Characteristics of F	A coolant
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Prior to the 90s when the FAs were operated under normal conditions no fuel elements failures were detected. Only when the ribless type FAs were used at the close to maximum power level there were detected the cases of the FA failure. 12 out of ~760 FAs failed. It should be noted that there were no claims in regard to the bulk FAs serviceability. Approximately 60 out of that bulk were operated at high power levels up to ~ (2,0...3,0) MW with maximum heat fluxes from the FAs surface up to ~ (3,4...5,1) MW / m<sup>2</sup>. The mean fuel burn-up depth in some FAs attained ~60%. To perform fuel elements irradiation stability investigation, there were carried out material structure investigations of the FA opearted at power level of <2 MW and having the mean fuel burn-up depth of 62,7%. In figure 2 macro and microsection metallographic specimen of fuel elements with 30% and 87% are presented. The results of the FAs material structure investigations affirm that the fuel elements' dimensions comply with the specifications. Both in the initial state and the state when the deep fuel burn-up is attained (~87%) there's a close contact of the fuel particles with the aluminum matrix and that of the matrix itself with the fuel elements cladding. The corrosion film does not exceed (10...15) µm.



FIG.2. Macro and microsection metallographic specimen of fuel elements a) burn up 30%b) burn up 87%.

In fig.3a one of the failed FAs' appearance is presented. It is typical for the majority of failed the FAs to have on the external FA surface an elliptical spot, that is different in colour from the rest of the surface. As a rule such spots were located on one side of the outer surface and covered from 1/3 to  $\frac{1}{2}$  of the perimeter. These spots were situated at the FA height sections from ~350 to ~900 mm from the active zone top. In the middle of these spots there is the cladding material corrosive damage that is indicative of the allowable temperature exceeding on the fuel elements surface. On one of the 12 failed FAs there occurred the "burn-out" of the external fuel element, followed by melting down and damaging of the area up to ~ 160 high and up to ~20 mm wide at ~2 MW, the allowable value being 2,6 MW. In fig.3b the picture of the damaged area is presented. The damaged area was situated at the elevation of  $\sim$ (650-800) mm from the active zone top. The results of the visual inspection of all failed FAs are indicative of the overheat of certain areas of external fuel elements. It is noteworthy, that these areas are not located along the whole fuel element perimeter and they are rather extent, up to ~550 mm along the active zone height. It should be also noted that the flow rate, the temperature and the power in each FA of the MIR reactor are controlled separately and no parameter deviation from the operating range was detected. Thus, within these areas either the heat loads exceeded the allowable ones, or there was an insufficient flow rate of the coolant, flowing about the given areas of the fuel elements surface.







b)

FIG.3. Appearance of failed FAs.

#### 2. Trends in activity to improve the FAs reliability

In order to determine and to exclude the causes of the reactor FAs failures during the last years the following work has been done :

- (1) Control system for coolant thermal parameters and channel power of FAs has been modernized. At present, operating parameters of each FA are visualized as reactor cartogram mnemonic on the operator's displays: coolant temperature and flow rate, as well as power.
- (2) There has been revised the procedure of experimental determination of FA power which considers the transfer of energy between FAs due to neutron and gamma-ray emissions, as well as the heat loss, resulting from the reactor pool during the coolant travel towards the location of the detectors.
- (3) On the physical reactor model there have been performed experimental investigations on refinement of energy release distribution along the FA height, cross-section and perimeter, depending on the high-altitude position of control rods and mobile FAs combined with absorbers, as well as on some fuel loading configurations. Following the results of the investigation, to determine the maximum allowable FAs power values, the following indices of energy release non-uniformity have been fixed: along the active zone height 1,74, along the FA perimeter 1,15; instead of 1,45 and 1,10, correspondingly, previously used in calculating procedures.
- (4) Calculating procedures for thermohydraulic and physical reactor calculations have been improved and updated. There has been made a comparison of critical heat fluxes calculation results for the MIR reactor FA, with the values resulted from the empirical corrections, recommended by IAEA for research reactors [3]. It has been determined that the values of the heat flux critical density, used in the MIR reactor, agree with a series of empirical corrections. However, there have been considerable calculation deviations on certain correlations, in both directions. That is why, there has been made a comparison of the calculation results with the experimental data, including Polish investigations [4]. It has been shown that within the range of mass velocities (6000-10000) kg/m<sup>2</sup> s, typical for the MIR reactor FAs, the experimental values of critical heat fluxes exceed or comply with the rated data, i.e. the rated correlation of the critical heat flux, applied in the MIR reactor, is conservative. To analyse the reactor safety there has been made a rated model based on the thermohydraulic code RELAP5 / MOD3.2, having a special block to calculate the maximum allowable FA power. At present, the verification of this model is being carried out, that considers the revised experimental and rated reactor characteristics, obtained during the last years [5].
- (5) The calculating procedure of the FA maximum allowable power has been updated. According to this procedure the maximum FA power is reduced from 4,0 MW to 3,2 MW at the maximum coolant velocity of ~10 m/s.
- (6) The material structure investigations of 2 failed FAs have been started, aimed at determination of the material structure in detail to find out the radical causes of the fuel elements failure.

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#### New Storage Mode for Spent Fuel at the Budapest Research Reactor

#### (Encapsulation of the fuel for semi-dry storage)

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#### Abstract

The Budapest Research Reactor is a light water cooled and moderated tank type reactor. The reactor was first commissioned in 1959; its principal functions at that time were to serve as a facility for basic research experiments in the frameworks of research programmes of the Hungarian Academy of Sciences and in industrial development projects. In the first years of operation (until 1966) the power was 2 MW; the type of the fuel used was EK-10.

In 1967 the reactor was upgraded, the power was increased to 5 MW, as a new fuel WWR-SM (36%) was introduced, the core grid was replaced and also beryllium reflector was put around the core. In this upgrading project a new wet spent fuel storage facility (AFR pond) was constructed near to the reactor hall.

In 1986 the second (full scale) reconstruction of the reactor started, the new reactor with unchanged fuel type was first critical in December 1992. After the reconstruction a new type of fuel was introduced, the so-called VVR-M2, with the same geometry and enrichment as VVR-SM, but with different material composition, i.e.  $UO_2$  granulate dispersed in aluminium matrix. Up to the present the reactor has been in operation for more than 43 years and all the spent fuel produced since the first commissioning of the reactor is stored in wet facilities on site. There is no real experience of wet storage for so long time worldwide, but it is well known that aluminium corrosion can accelerate rapidly once it has started. That is the reason why the operating organization of the BRR facility decided to encapsulate all the stored assemblies and keep them in inert gas atmosphere in order to slow down or to stop corrosion processes. The present paper describes the semi-dry storage transition option, i.e. the design and the operation of the canning device, as well as the canning procedure in detail.

#### Introduction

The unloaded burnt fuel is put into a temporary storage pool, situated close to the reactor tank (AR pool). Using high-density storage racks 786 single VVR type fuel elements can be stored in this pond. To allow filling one core load in case of emergency core unloading 230 positions in the AR pool have to be empty all the time, consequently 556 positions can be used only for storage. The failed (leaking) fuel assemblies can be placed into a special wet container and can be stored in the AR pond in sealed condition.

After a certain cooling time (approximately five years), the spent fuel elements can be transported to the other pool (AFR pond) for long-term storage. This pool is situated outside the reactor building; the distance from the reactor hall is about 100 m. The cask used for fuel handling in the reactor hall

can be used for in-site transportation too. The AFR pond is a cylindrical stainless steel vessel with racks without absorbers, sub-criticality is guaranteed by the geometry. At present all the spent fuel unloaded before 1986 is stored in the AFR pool, while the fuel assemblies irradiated after recommissioning of the reactor are stored in the AR pool.

The water in both of the pools is cleaned, time to time by a mobile ion exchanger filter. The corrosion of the aluminium fuel cladding did not cause a major problem yet, mainly due to the good water chemistry. The storage meets the requirements of the Hungarian authorities and a permanent surveillance program is implemented, but this type of wet storage is obviously not a final solution. Within two years, in accordance with the fuel consumption rate, fuel elements, irradiated after the reconstruction, should be transported from the AR pond to the AFR pond.

As there is no experience of long-term wet storage, an inspection programme was implemented. The visual inspection showed different levels of corrosion in case of some fuel assemblies. The heaviest corrosion was detected in the case of instrumented fuel having thermocouple in the cladding.

The encapsulated fuel assemblies will be stored in the same pool under water (providing the biological shield and cooling) unless some other solution will be available. The advantage of this solution is its simplicity, i.e. there is no need to change the pool structure, anyhow use can be made on the advantage of the dry storage The canning machine, was manufactured in 2001 by a Hungarian company (Ganz Energetic Ltd.). The canning procedure will be performed in the year of 2002.

#### The Canning Machine

The canning machine consists of four main components namely the canning cask with the transfer pipe, the car with the auxiliary equipment, the I&C cabinet and the cropping machine.

The Canning Cask



The main component of the cask is the stainless steel body containing the bottom shutter, the capsule holding device, the eddy current coil and the capsule-rotating equipment. The bottom shutter has two positions; open and close. In the open position the capsule can be transferred in and out, while in the close position a piston is placed under the capsule. This piston can lift and lower the capsule by demand. The transfer pipe is fixed to the bottom of the cask, while the pipe end lifting drive is mounted on the side of the body. A revolver head closes the upper part of the cask. This head has five positions, which means that every cycle of the canning procedure has a different head position.

Three stepping motors are installed on the top of the cask, for the revolver head, for the capsule rotation and for the bottom shutter. Water, nitrogen, air and vacuum pipes coming from and leading to the auxiliary equipment car are connected to the revolver head.



#### Auxiliary Equipment Car

The auxiliary equipment car has three levels. The main water pump, the transformer supplying the



the car a buffer tank can be found. This tank has three functions, namely keeps the main water pump filled by water, provides cooling water for the pre-cooler and serves as a buffer for the capsule transfer water. (In order to keep the required visibility under the water, the transport water doesn't go back directly to the pool). e main water pump, the transformer supplying the eddy current coil and the drying air pre-cooler can be found on the bottom level. The drying fan and the vacuum pump both with HEPA filters and transducers are mounted on the second level. The Peltier type chiller for final humidity removal is mounted on the second level too. On the top of



112

*I&C Cabinet* 



All of the equipment necessary to control the canning process are installed in the I&C cabinet. The canning procedure is controlled by a PLC; information about the cycles and steps of the process is displayed on a LCD display. In semi-automatic canning mode the operator uses only one push-button (CONTINUE), while in case of any malfunction or problem the operator can switch on manual mode and control the process manually.

#### The Cropping Machine

The cropping machine is an underwater circular saw used for

cutting off the leg of the VVR type fuel assemblies. It has two different cutting positions for the single and triple fuel assemblies. Another use of the

corpping machine is, that in case the welding is leaking, the capsule head can be cut off in the machine. In this case the canning process has to be repeated.

# CROPPING MACHINE

#### The Canning Procedure

The capsules are capable to accommodate either one EK-10 fuel assembly or three single (one triple) VVR type assemblies. The capsules are made of aluminium alloy; the wall thickness is 3

mm. The welded head of the capsule provides the same handling possibility, as the fuel has, consequently the same manipulation tools can be used for handling them. The capsules are filled with 2.5 bar nitrogen gas. The overpressure is needed for leak detection purposes. To assure sinking of the capsule and sub-criticality, an iron disk with aluminium cladding is screwed on the bottom of the capsule. In case of dry storage or transportation this weight can be removed easily.

The spent fuel assemblies now are stored in buckets, which are placed in storage tubes (three levels of buckets per

storage tube). The bucket is removed from the tube and put into the position dedicated for manipulation. The new capsule is placed beside it and the fuel having its leg cut off is loaded into the new capsule manually, under the water. There is no need for cropping in case of EK-10 fuel. Then the capsule is placed under the canning cask, the movable end of the transfer pipe is lowered and the main water pump is started. The water flow lifts up the capsule into the container. Upon arrival a special mechanism fixes the capsule in the cask, the bottom valve of the cask closes and the pump switches off. During the encapsulation the movable pipe-end remains in the bottom end position. This means that in case of malfunction or emergency the capsule can be transferred back to the pool any time.

Then the water must be removed from the capsule and the remaining water should be dried out. In order to reach perfectly dry fuel condition the capsule and the fuel shall be heated up to 120 °C



using an eddy current heater. The evaporated water is removed from the cask by a close circuit hot air circulation. The humidity is removed from the air by a pre-cooler and a Peltier type chiller, while the dry air is cleaned by HEPA (High Efficiency Particle) filter. When the fuel is perfectly dry, the inner cavity of the container is vacuumed and filled with nitrogen gas. Keeping the capsule in hot condition the cold capsule head is pressed in. This allows better sealing as the OD of the head in room temperature is slightly bigger (0.2 mm) than the ID of the container.

Finally the capsule head is secured by welding and the capsule is transferred back into the water. Under the water the capsule manually can be put back into a storage tube. Every capsule has an identification number for inventory and safeguards purposes. The canning process shall be fully documented and video recorded as appropriate.

#### Operation

Naturally all fuel cropping and underwater manipulation including the capsule handling are performed manually, while the operation of the canning machine is semi automatic i.e. individual cycles of the procedure are controlled by the PLC, but after every cycle the programme stops and waits for the approval of the operator (CONTINUE pushbutton). If the operator detects any failure or malfunction, he can interrupt the program and return back to the previous step or go forward, depending on the stage of canning. If the problem arises before pressing in the capsule head, it is better to send back the capsule (with the fuel) into the pool water, while after closing the capsule, it is better to solve the actual problem and finish the process with the welding.

#### **Monitoring programme**

Monitoring the pool water in the AFR pool has always been done since the pool is operated. The only on-line monitored parameter is the water level. The water quality is checked by sample analysis every second week. The analyses consist of the measurement of conductivity, radioactivity and pH. In every quarter of the year a full chemical analysis (including the quantitative determination of chloride, sulphur, iron, copper etc. ions) is performed on the water sample.

After having encapsulated every fuel assembly an extended monitoring programme will be elaborated. This programme shall naturally include all the investigations, which were included in the earlier monitoring programme. The extension of the monitoring programme will include leak tests and annual probe tests. Probes of the capsule material (welded material too) should be put into the pool, which will be used for annual monitoring. The tests of these probes will include visual inspection, metallurgic analyses and mass measurements.

If a capsule leaks, it can be observed, as in this case tiny bubbles come out of the storage tube. Anyhow the weight of the tubes shall be measured periodically, to have a quantitative measure of leak (corrosion), i.e. the increase of weight (the mass of a sealed capsule is  $\sim 1$ kg i.e. it is almost weightless in water). If the capsule is failed in time of canning or after it, then the capsule head can be removed in the cropping machine and the encapsulation process can be repeated.

#### Summary

The in-site inactive tests of the canning machine were started in November 2001. During the tests all "teething troubles" have been solved and in December 2001 the canning procedure was successfully demonstrated to the licensing authority. Based upon the tests the regular canning procedure can start as soon as the authority issues the license for the canning.



#### STORAGE, INSPECTION AND SIP TESTING OF SPENT NUCLEAR FUEL FROM THE HIFAR MATERIALS TEST REACTOR

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#### ABSTRACT

Aluminium clad U-Al fuel used within the HIFAR MTR has been stored both in dry (underground) and wet (pond) storage facilities at the Lucas Heights site since the 1960's. As part of ANSTO's current program to send this fuel for long term storage or reprocessing, a significant level of visual inspection and water sip testing has been performed. This data has been used to demonstrate the integrity and suitability of the fuel for transport and receipt at the reprocessors interim storage ponds. This paper presents the key technical background / history of HIFAR fuel and its storage at Lucas Heights, presents the data obtained to date regarding its condition and discusses some observations regarding visual corrosion indicators and actual sip test results.

#### 1. Introduction

The Australian Nuclear Science and Technology Organisation (ANSTO) has operated the 10MW DIDO class MTR reactor HIFAR (HIgh Flux Australian Reactor) since 1958.

The HIFAR core contains 25 fuel assemblies, 2 metres long and 100 mm in diameter (the fuelled section being only 600 mm long). The fuel plates are aluminium clad, Al-U matrix HEU which during the 43 year period of HIFAR operation have evolved through three key design variations (Mk II parallel plates, Mk III spiral plates and Mk IV concentric tubes).

Following unload from the reactor core, the fuel is stored in fuel ponds for a period of approximately 3 years, during which time it is cropped to retain only the fuelled length. ANSTO operates two fuel ponds which are used for a) cropping and interim / long term storage of fuel and b) preparation of fuel for shipment and loading of fuel shipment casks. Both ponds have demineralised water supplies and are maintained within the same closely monitored parameters (pH, Conductivity, Gross Beta, Alpha and Tritium). Water quality is maintained by re-circulation of the water through skimmer boxes, particulate filters and an ion-exchange system to remove radioactive contaminants arising from the stored fuel.

ANSTO's underground dry storage facility comprises 50, 15 metre deep, 140 mm diameter stainless steel tubes set into the rock. These storage tubes are capped with shield plugs seated on a rubber seal and each tube contains up to 22 cropped fuel elements, providing a storage capacity of 1100 elements. When loaded with fuel, these tubes are evacuated and then back-filled to atmospheric pressure with dry nitrogen to inhibit corrosion. Vacuum rise tests and atmospheric sampling of these fuel tubes (for relative humidity, temp, $O_2$ , NO<sub>x</sub> and Kr85) is facilitated via an in-situ quick-connect valving system and is performed biennially.

#### 2. Acceptance Criteria for Transport & Storage or Reprocessing

Contracts for the receipt of research reactor spent fuel for further storage or reprocessing considers that in the normal case spent fuel will be "physically sound". Spent fuel that does not satisfy this condition may require additional measures or restrictions governing its transport, receipt and handling arrangements. The term "physically sound" has been interpreted to mean fuel which is structurally sound (i.e. there must be no material dissemination during handling or transport) and that can therefore be transported within casks and received at fuel storage and reprocessing facilities without special provisions. Full visual inspections to identify any plate deformities, pitting in the cladding or other unusual markings are performed prior to loading of the shipment casks and reporting of this forms part of the basis for formal acceptance of the fuel.

In addition to visual indicators of fuel condition or degradation, quantitative criteria for radioactivity released from the assemblies are utilised. For acceptance of "sound" fuel being shipped for reprocessing, activity indicators such as the following are utilised:

- 1. Sip testing of any individual elements which have been classified as "questionable" based on visual inspection reports;
- 2. Reporting of pond gross beta activity following transfer of fuel elements for each individual cask;
- 3. Kr-85 concentration of the loaded cask following cask air drying;
- 4. Cs-137 activity concentration of demineralised water flushed through the loaded cask on receipt at the reprocessing or storage facility.

Compliance with Criteria 1 to 3 above must be demonstrated before fuel is accepted for transport. On receipt of the cask, failure to meet criteria 4 would lead to bottling of each individual fuel element prior to transfer to the storage ponds.

Through work performed at ANSTO to demonstrate compliance with the above acceptance criteria, it has been determined, that HIFAR spent fuel assemblies will fall into one of three possible categories:

Category	Definition
A	Mechanically sound, non-leaker assemblies. No special arrangements are needed for transport or acceptance of these assemblies.
В	Mechanically sound assemblies, having minor defects which lead to leakage of fission products when immersed in water (sip test > 10KBq/hr/assembly). Numbers of such assemblies in a given cask may be limited.
С	Unsound assemblies, being those with physical deformations or visible corrosion which could possibly cause flaking or shedding of material during transport or handling. These assemblies are expected to require 'bottling' prior to transport. These unsound assemblies may be in either the leaker or non-leaker category, and if leakers, they would be subject to the same limits on the number of assemblies per cask as Category B.

#### 3. Background on Spent MTR Type Fuels

It is important to recognise the considerable differences between HIFAR MTR type fuel and power reactor fuels. MTR type fuel plates consist of an inner fuel 'meat' made from a uranium-aluminium alloy dispersed in aluminium metal. This 'meat' is more than 95% by volume aluminium and is metallurgically bonded to the high purity aluminium cladding by hot rolling. To assure the integrity of the bond between the fuel core and the aluminium cladding, each fuel plate is subjected to a blister test where it is heated to 600°C for 20 minutes [1]. Once cooled, each plate is examined for blisters, and if found the plate is rejected.

Sindelar [2] explains that during irradiation, small isolated voids form in the fuel meat and contain fission product gases which are uniformly distributed throughout the fuel. Since these voids are not interconnected, Sindelar concludes, that if a minor cladding breach should occur, the only part of the fuel that could be exposed for release of fission products, or for water leaching would be the part immediately under the breach. Hence, with minor cladding breach, the great bulk of an MTR fuel plate continues to be effectively contained by the remaining sound cladding. Furthermore, the metallurgical similarity of the meat and cladding means that in general there is no preferential galvanic attack on the exposed meat even when the cladding preferentially corrodes and is protective with respect to any exposed fuel meat. This is illustrated in Figures 1 and 2. In fact, the fuel assembly shown in Figure 1 returned a radioactive leak rate of  $1.4kBq/hr \beta$  during sip testing. Despite the small breach illustrated, the leak rate was sufficiently low for the assembly to be classified as Category A (sound).

The metallurgically bonded, integral nature of MTR fuel also means that the chances of any pieces of fuel meat flaking or detaching during handling in storage or transport are remote. Even in the case of

severely corroded fuel, the flaking would be restricted to the localised area of corrosion. This would be immediately obvious to visual inspection, and such fuel would be considered as Category C unsound.



Figure 1. Pitting corrosion of the cladding has exposed the corner of the U-Al 'fuel meat'.

**Figure 2.** Pitting corrosion of the heataffected zone of the electron beam weld has exposed the edge of the U-A1 'fuel meat'.

The other point that must be recognised with MTR fuel is that all spent fuel assemblies, no matter what their condition, will release radioactivity in a sip test. This radioactivity arises from a number of sources:

- Tiny traces of 'tramp' uranium are inevitably left embedded in the outer surface of the cladding during the manufacturing process. When the fuel is irradiated in the reactor this material also fissions and results in fission products in and on the surface of the assembly [3].
- Activation products and fission products are absorbed from the reactor coolant during operation and may be subsequently released [4].
- Fission products will be transferred onto non-leaker assemblies from leaker assemblies in storage facilities [4].
- Some trace elements left on fuel assemblies during the manufacturing process, such as from brazing or welding fluxes, and trace impurities in the aluminium cladding will become activated when the assemblies are irradiated in the reactor [5].
- Sub-Microscopic cladding defects (such as might remain from manufacturing or might occur with incipient pitting corrosion) may be so small as to not be detectable or significant. However, in operation or during periods of extended dry storage these defects can provide pathways for fission products to migrate from the fuel meat onto the cladding surface. (See for example, Kirsanov, 1999 [6]).

As a clear demonstration of these effects, it is always found by ANSTO that when spent fuel assemblies are first transferred to a pond after several years in dry storage there is inevitably a flush of elevated activity that washes off the assembly. This occurs with assemblies that are known to be perfectly sound and to have never exceeded recommended storage environmental conditions. After a period, this flush of activity ceases and ongoing activity release is then of the same order as that from other sound assemblies that have spent all their storage time in ponds and is generally indistinguishable from the pond water background.

Finally, it must be stressed that the use of sip testing to determine the condition of a fuel assembly is an extremely conservative approach. Quantitative assessments of data for fuel shipments of spent aluminium based fuel to Savannah River in wet environments resulted in sip values 2 to 3 orders of magnitude greater than for dry shipments (Vinson, Sindelar et al 1998 [7]). Massey et al [8] state that 'because transportation of the SNF to Savannah River will take place in a dry condition, it is expected that fission product release rates during transportation will be virtually zero, and essentially, immeasurable'.

#### 4. ANSTO'S Sip Test Procedure

The basic procedure developed at ANSTO for undertaking sip tests on assemblies as a basis for assessing their suitability for shipment is outlined below:

- The fuel assembly is immersed in demineralised water overnight to remove readily removable surface based contamination, prior to the commencement of each test.
- 10 litres of demineralised water is continuously circulated through the covered sip test tank.
- Samples are taken before the test commences and regularly over a 100 hour cycle.
- Samples are analysed for Cs-137, Cs-134, Co-60, and Co-58; plus gross alpha and gross beta.

ANSTO have undertaken sip tests both at ambient temperature and at 50°C to replicate potential temperature increases during transport) on the same fuel assemblies and no discernible difference in either leak rate or total activity release over the duration of the tests were yielded.

#### 5. HIFAR Fuel Condition and Sip Test Results

From ANSTO's operating records, routine monitoring of its spent fuel storage facilities (including pond activity levels), previous detailed examinations of spent fuel, and inspections performed prior to recent shipments, ANSTO is readily able to certify the soundness of the majority of the fuel it has in storage. There are a small number of assemblies which have been classified as unsound (those which were sectioned as part of a post irradiation examination campaign) and a small number which were designated as 'questionable' and required further investigation to allocate them to one of the categories.

A range of recent sip test results are presented in Figure 3.



Figure 3 Sip test results from "questionable" Mk II and Mk IV assemblies.

The Mk II assemblies showed signs of corrosion evident by the brown staining on the base of some assemblies. As a precaution, these six assemblies were retained for further characterisation. The Mk IV assemblies, were known to have minor breaches of the aluminium cladding, identified during detailed visual examination. Since that time they had been stored in sealed stainless steel canisters. As a consequence of the history of these fifteen assemblies, all were deemed to be questionable and submitted for sip testing.

There are two sets of results for each Mk II assemblies shown in Figure 3. The assemblies were sip tested initially in 2000 after having been transferred from dry storage and then retested in 2001 after a period of 12 months soaking in a pond. The results of the sip tests conducted in 2001 exhibit a significant reduction in the leak rate compared to the results from September 2000. Closer examination of individual results indicate that the total activity released during each test dropped to less than 10% of that recorded during the testing of 2000. When tested in 2000 the assemblies had only spent 5 months in the ponds after being transferred from dry storage. Clearly, the time spent soaking in the pond prior to a second sip test supports ANSTO's argument that when spent fuel assemblies are first transferred to a pond after extended periods of dry storage there is inevitably a flush of elevated activity that washes off the assembly.

The Mk IV assemblies were known to have minor breaches of cladding, which penetrate to the fuel meat (see Figure 1). However, the results of each assembly were less than 4kBq/hr gross  $\beta$ . This provides further evidence of the soundness of MTR fuel assemblies and reinforces the theory that only the fission products immediately under any breach are able to be leached from the fuel. The highest two MkIV leach rate results are considered to be associated with active contamination embedded on the outer surface of each assembly. A semi-adherent white powdery deposit was observed on the surface of each assembly identified as aluminium hydroxide. The deposit was largely removed by scrubbing, which exposed some minor breaches of the cladding. It was known that the assemblies had been exposed to water during their period of storage.

#### 6. Conclusion

Data and experience gained through ANSTO's preparation and testing of HIFAR spent fuel for shipment has reinforced a number of key observations regarding radioactivity release mechanisms from spent MTR fuel. It is concluded that the leakage of radioactivity from small visible cladding deformities is limited to the immediate area associated with the deformation and that all HIFAR assemblies, which have been in long term dry storage, release accumulated surface radioactivity when transferred to wet storage. For the purpose of classifying HIFAR spent fuel for shipment, an indicative soundness threshold of 10KBq/hr per assembly, gross beta activity, has been derived.

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# **Session 5**

**Back-end options and transportation** 



#### PROGRESS OF THE UNITED STATES FOREIGN RESEARCH REACTOR SPENT NUCLEAR FUEL ACCEPTANCE PROGRAM

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#### ABSTRACT

The United States Department of Energy (DOE), in consultation with the Department of State (DOS), adopted the Nuclear Weapons Nonproliferation Policy Concerning Foreign Research Reactor Spent Nuclear Fuel in May 1996. To date, the Foreign Research Reactor (FRR) Spent Nuclear Fuel (SNF) Acceptance Program has completed 23 shipments. Almost 5,000 spent fuel assemblies from eligible research reactors throughout the world have been accepted into the United States under this program. Over the past year, another cross-country shipment of fuel was accomplished, as well as two additional shipments in the fourth quarter of calendar year 2001. These shipments attracted considerable safeguards oversight since they occurred post September 11. Recent guidance from the Nuclear Regulatory Commission (NRC) pertaining to security and safeguards issues deals directly with the transport of nuclear material. Acceptance Program has consistently applied above regulatory safety Since the enhancements in transport of spent nuclear fuel, this guidance did not adversely effect the Program. As the Program draws closer to its termination date, an increased number of requests for program extension are received. Currently, there are no plans to extend the policy beyond its current expiration date; therefore, eligible reactor operators interested in participating in this program are strongly encouraged to evaluate their inventory and plan for future shipments as soon as possible.

#### Introduction

The Foreign Research Reactor (FRR) Spent Nuclear Fuel (SNF) Acceptance Program, in the sixth year of implementation, has completed 23 shipments safely and successfully to date. 27 countries have participated so far, returning a total of 4,957 spent nuclear fuel elements to the United States for management at Department of Energy (DOE) sites in South Carolina and Idaho. 19 of the 23 shipments, containing aluminum-based spent nuclear fuel from research reactors, went to the Savannah River Site (SRS) in South Carolina. The other four, containing Training, Research, Isotope, General Atomic (TRIGA) spent nuclear fuel, were transported to the Idaho National Engineering and Environmental Laboratory (INEEL). The FRR SNF Acceptance Program focuses on the planning and implementation of these shipments of research reactor spent fuel to the United States in support of worldwide nuclear nonproliferation efforts. Along with shipment logistics, the Department continues to address many other issues of importance to the program. Resolution of these issues is important in helping to improve our implementation activities.

#### Discussion

Several implementation successes were achieved over the past year in the Acceptance Program. Between January 2001-2002, a total of five shipments were safely completed under the program. Spent fuel from Argentina, Chile, Austria, Sweden, the Netherlands, Germany, Japan and Denmark entered the United States at the Charleston Naval Weapons Station (NWS) and was shipped by rail or truck to the SRS. In July of 2001, the third cross-country shipment of TRIGA spent fuel, German fuel arriving at the Charleston NWS, was safely completed. The cross-country shipment of TRIGA fuel, consisting of 126 spent fuel elements in 3 casks, was accomplished by truck shipment from SRS to the INEEL. This shipment encountered the usual high level of public and media interest in response to the movement of nuclear material, and extensive discussions were held among the DOE and State government officials along the planned transportation route. By working together closely with federal, State and international contacts during the planning stages, we were able to ensure that when the time came for shipment, the transport would occur smoothly. Approximately one cross country shipment per year is expected over the remaining eight years of the Acceptance Program, and DOE will build upon lessons learned from this and other cross-country shipments in making future plans.

This year also saw larger numbers of casks entering the United States as part of a single shipment. On July 19, 2000, a revision to the Record of Decision (ROD) on a Nuclear Weapons Nonproliferation Policy Concerning Foreign Research Reactor Spent Nuclear Fuel was published in the U.S. Federal Register. This revision increased the maximum number of casks on a single ocean-going vessel to 16. The May 1996 ROD limited the number of casks containing spent fuel on a single ocean-going vessel to eight, based upon the analysis in the Final Environmental Impact Statement on a Nuclear Weapons Nonproliferation Policy Concerning Foreign Research Reactor Spent Nuclear Fuel. At the time the ROD was issued, the worldwide supply of spent fuel casks available for use in transporting FRR SNF was very limited, and thus DOE forecasted that no more than eight casks could be made available to support any one shipment. However, since 1996, the worldwide supply of spent fuel casks has increased to the point where it is possible to transport up to sixteen casks on a single ocean-going vessel, thus potentially helping reduce the number of ocean transits and saving money for both high-income economy shippers and DOE. This revision to the ROD was first put to use in October 2001, when a total of 12 casks, containing a total of 438 fuel elements, were received at the Charleston NWS in a single shipment and then transported to the SRS.

The FRR SNF program continues to be successful in large part due to the aggressive planning efforts between the United States and those countries eligible to ship their research reactor fuel. It is imperative that cask licensing be initiated as soon as possible in the planning process. The 2001 cross-country shipment came dangerously close to being rescheduled because of significant delays regarding receipt of cask application materials and responses to requests for additional information from the Nuclear Regulatory Commission (NRC). The shipment timeline was saved due to extended efforts of many involved, including the German licensing authority and the NRC. The Acceptance Program enjoys a very professional working relationship with NRC staff and as such, wishes to take every measure possible to respect this relationship by ensuring that cask applications are timely and complete. In the past the Acceptance Program may have been able to rely on NRC to readjust its workload to accommodate a special request for package review and certification under less than optimum deadlines. However, the post-September 11 environment now has U.S. federal staff weighted down with evaluations into safeguards practices and preventive measures. Timelines for cask application submittals must be closely adhered to, and allowing more time for review is always a welcome move. In addition, it is important that we continue to schedule shipments as soon as possible in order to allow for as much spent fuel to be shipped to the United States over the remaining seven years of the program. It is also important to note that countries interested in participating in the Program should express their interest as soon as possible so that fuel and facility assessments can be scheduled and shipments may be entered in the long-term shipment forecast. The Program may not be able to accommodate a large number of last minute requests, particularly from geographically isolated regions. With more casks available for shipment and more casks allowed on a single ocean-going vessel, and with transportation plans into and across the United States established and implemented, shipments can be made more efficiently for all parties involved. Although some reactor operators and contractors have voiced support for extension of the program expiration date, it should be understood by all involved parties that the DOE has no plans, at this time, to seek extension of the FRR SNF Acceptance Policy. We remain committed to our program goals and hope to work with all remaining eligible research reactors to plan for shipments of their eligible spent fuel. The DOE continues to support research reactor operators' needs and would be happy to meet any interested parties to discuss the program.

A primary goal of the Acceptance Program is to support worldwide nonproliferation efforts by shipping high enriched uranium (HEU) of U.S.-origin to the United States for management and disposition. Integral to this process is the U.S. assistance offered in helping reactor operators convert their cores to low enriched uranium (LEU) as the reduced enrichment fuels become qualified and available. In addition, DOE plays a strategic role in ensuring a supply of enriched uranium for fuel fabrication. In the Acceptance Program, we realize our primary goal is intertwined with the missions of the Reduced Enrichment for Research and Test Reactors Program and the Enriched Uranium Operations group out of DOE's Y-12 plant in Oak Ridge, TN. We remain committed to working with staff in these other program offices within DOE to do whatever we can to assist in smooth transitions of core enrichment level and a steady supply of fuel.

The first research reactor to benefit from these policy changes within DOE was the High Flux Reactor (HFR) at the Joint Research Center in Petten, the Netherlands. On January 10, 2000 a diplomatic note was signed between the Government of the U.S. and the European Commission concerning supply of HEU for the HFR reactor and subsequent shipments of spent research reactor fuel from this facility to the United States. Petten has agreed to convert the core to LEU by May 2006. The U.S. has agreed to export HEU to France, for fabrication into fuel assemblies for Petten, while Petten awaits license approval for the converted core. This will allow Petten to continue operation pending regulatory approval and conversion. The Department has a contract with Euratom for the sale of material for Petten. Shipment of the first batch of fresh HEU occurred in 2001.

As alluded to earlier, security issues are now occupying a central focus as a result of the September 11 terrorist attack. The DOE, working in conjunction with international, federal, State, Tribal and local authorities, is re-examining procedures and requirements for transport of radioactive material, particularly commodities such as spent fuel. A temporary halt on all DOE-owned shipments of radioactive material in the U.S. was ordered by senior DOE management immediately after the September 11 attack, and again in October after commencement of the air campaign over Afghanistan. This action was taken in conjunction with other security measures throughout the DOE weapons complex and the nation at large. DOE was, and is, in constant contact with the Federal Bureau of Investigation, the Department of Defense, the Nuclear Regulatory Commission and other federal agencies responsible for transportation and infrastructure safety.

Historically spent nuclear fuel shipments have not been considered attractive targets for terrorist attack or sabotage, and threat assessments undertaken by law enforcement since last September seem to corroborate this view. However, across the globe spent fuel shipments are a matter of high concern for public officials due to the perceived perception that spent fuel transportation presents a heightened risk as compared to transport of other hazardous materials (e.g. propane and liquid natural gas). In addition, inspection, escort and other enforcement duties related to safe, routine transport can tax law enforcement and emergency response assets that could otherwise be deployed elsewhere. Within the United States, discussions and advances concerning the Yucca Mountain permanent geologic repository have renewed and invigorated ardent support, both pro- and anti-nuclear. Major steps have recently been taken in the U.S. with respect to identifying and backing Yucca Mountain as the proposed permanent geologic repository. The public has voiced opinions on both ends of the anti-nuclear spectrum; they are not comfortable with transport of nuclear material across interstate roadways, nor are they comfortable with having spent fuel and other high level radioactive waste stored at the 131 temporary storage facilities across the United States. Like others interested in permanent disposition of spent fuel, the Program continues to monitor closely developments in this issue.

#### Conclusion

We will have many challenges in 2002 as we resume planning for cross-country shipments of spent nuclear fuel for the first time since September 11. The United States, and likely as well for the international nuclear transport community, will have a more watchful public. Some of the issues DOE and other agencies are examining now include impact for State, Tribal and local resources should shipments be halted again, or should the additional temporary measures - such as those recommended in recent NRC guidance - become permanent. Advance notification and information are emerging as important related issues given the stricter climate of security; electronic mail, conference calls and fax machines have done much to improve how shipment planning and operations are accomplished, but the need for safeguarding sensitive information may change how these functions are performed. In addition, we can expect increased international coordination amongst security officials in order to ensure safety of the ship, its crew and cargo. In short, while DOE does not anticipate that security concerns will have a long-term impact on the schedules of the FRR SNF Acceptance Program, there may be short-term delays as a new concept of operations emerges. DOE expects to continue to work closely with reactor operators and others to ensure spent fuel is shipped safely.



### **RRFM 2002**

6th International Topical Meeting Research Reactor Fuel Management

## MANAGEMENT OF THE ACCEPTANCE PROCESS OF RTR ALUMINIDE TYPE SPENT FUEL

P. AUZIERE, J. THOMASSON COGEMA



Ghent, Belgium March 2002

#### ABSTRACT

A wide range of Research Test Reactor aluminide type spent fuel is already received for treatment conditioning at the La Hague reprocessing complex. Such a diversity calls for an utmost attention to be paid to all safety-related systems and technical aspects, to all regulatory and administrative constraints.

Despite of such multiple data inputs and rigid constraints, a close cooperation between the Research Reactor operator and COGEMA enables to reach adequate and cost effective solutions also relevant to spent fuel having had an uneven history.

The acceptance process is primarily based on the client descriptive data and status declaration issued by the Research Reactor (RR) operator under QA. This acceptance process is a key step, to be keenly scheduled as it is directly interactive with the RR evacuation plans and the La Hague industrial plant program. It is also governed by the reviews conducted by the French Safety Authority and generally translated into operational authorisations.

Concerned by maintaining high safety standards, reliable and proven operational levels of its nuclear services performed in the La Hague facilities COGEMA includes, all through this acceptance process, the operating, regulatory and administrative requirements.

This paper sets forth an overview of the approach implemented in the COGEMA organisation for the management of the acceptance process of RTR aluminide type spent fuel.

## MANAGEMENT OF THE ACCEPTANCE PROCESS OF RTR ALUMINIDE TYPE SPENT FUEL

#### P. AUZIERE, J. THOMASSON - COGEMA

#### 1. INTRODUCTION

COGEMA is applying its extensive experience with the many U-Al type RTR spent fuel assemblies already received at the reprocessing plant in La Hague, France, to gradually expand the range of services it offers research reactor operators. Its extended offering includes reliable, safe services concerning in particular the acceptability of fuel assemblies.

Using the precise pre- and post-irradiation characteristics that the research reactor operator provides for each U-Al type RTR spent fuel assembly, COGEMA has to verify that each assembly with its particular characteristics can be managed in full compliance with all safety rules at every step in a complex process including transportation, reception, unloading, storage and treatment in the La Hague plant.

In addition, long before fuel is shipped for reception, treatment and conditioning in the La Hague plant, COGEMA must first ensure that it falls within the scope of existing licenses and operating permits, which are subject to strict supervision by the competent French safety authority.

A special procedure has been established to determine whether a spent fuel assembly is acceptable, in particular within the framework of permits issued by the safety authority. COGEMA, the La Hague plant operator, applies this procedure in accordance with stringent quality assurance criteria.

Since COGEMA has the required shipping casks and manages fuel transportation, fuel transportability in a certified cask is usually verified at the same time. This aspect is not considered in this paper.

#### 2. FUEL CHARACTERISTICS

COGEMA has experience in managing fuel with a wide variety of characteristics, as illustrated in Figure 1.

Cross sections of fuel assemblies



The research reactor operator requesting COGEMA services must provide all the required data, including precise pre- and post-irradiation characteristics for each U-Al RTR spent fuel assembly, as indicated in Appendix 1.

The research reactor operator knows the history and condition of the spent fuel assemblies being stored before treatment in accordance with a predetermined schedule.

Certain spent fuel assemblies may be considered unsound, i.e. unsuitable for reuse in a reactor, due to traces of corrosion, perforation or other defects. In addition, they may have undergone testing to assess the potential rate of radioactivity release into the reactor pool water.

In some cases, the operator has installed special in-reactor instrumentation and introduced measures to separately manage spent fuel assemblies that have been declared unsound.

126
# 3. ACCEPTANCE PROCEDURE

### 3.1. Sound Fuel

COGEMA can generally accept RTR spent fuel that a research reactor operator has declared sound, provided that the fuel has been cooled for at least one year before loading into a shipping cask.

COGEMA has certified casks required for shipping the spent fuel and manages transportation from the reactor to its plant in La Hague. It therefore usually takes delivery of the fuel on the client's site.

The standard fuel acceptance procedure comprises the following steps:

(i) The research reactor operator provides COGEMA with the pre- and postirradiation characteristics as specified in Appendix 1, for each fuel assembly, in a document prepared in accordance with quality assurance requirements.

The characteristics are submitted in a single file covering a group of deliveries for a multi-phase disposal program at least 9 (nine) months before loading of the first cask specified under the program.

COGEMA reviews the documents and then notifies the operator of the provisional acceptance of the fuel concerned, and of any reservations, at least 3 (three) months before loading of the first cask in accordance with the scheduled delivery program.

(ii) The research reactor operator provides COGEMA with a plan for loading the assemblies into the assigned shipping cask at least 21 (twenty one) calendar days before the start of loading operations for each cask.

COGEMA reviews the plan and then notifies the operator of its approval of the proposed plan, and of any reservations, at least 14 (fourteen) calendar days before the start of loading operations.

(iii) If the fuel has been handled after the operator has transmitted the pre- and postirradiation characteristics to COGEMA as per step (i) above, the operator must re-confirm the integrity of the fuel and submit pool water inspection characteristics and any other documents with supporting information concerning conditions in the last place of storage.

This information is provided by the operator and commented on or accepted by COGEMA in compliance with the time requirements specified in step (ii) above.

(iv) One last verification is performed and COGEMA pronounces final acceptance of the fuel in accordance with steps (i) to (iii) above when its representative assigned to the reactor to monitor the fuel during cask loading completes the final inspection. COGEMA accepts the fuel following receipt of the operator's statement of conformity to the specifications indicated in Appendix 2 and review of the results of the scheduled pre-shipment inspections.

A Fuel Acceptance Certificate is drawn up by COGEMA and signed jointly with the research reactor operator prior to shipment.

Upon completion of this process, COGEMA takes delivery of the sound fuel.

### **3.2** Fuel Declared Unsound by the Reactor Operator

COGEMA can manage nearly all fuel declared unsound by the reactor operator, provided that a careful specific examination is conducted in each case. In the most difficult cases, a series of complementary precautions may be required primarily to guarantee protection against the risk of radioactivity releases and dissemination of materials.

### 3.2.1. Heavily Damaged Fuel

This category includes fuel that has been involved in an accident and fuel that has undergone significant corrosion that exposes extensive areas of the fissile material and/or that has destroyed the geometry or mechanical integrity of the assembly or its elements. It also includes dismantled fuel.

The precautions taken for extensively damaged fuel usually involve inserting the fuel assemblies or elements into containers. The containerization process is defined jointly by the reactor operator and COGEMA in order to apply the best possible solution allowing as precisely as possible for reactor and La Hague plant requirements.

The containerization process must also allow for constraints related to transportation and the cask employed. It is important to note that the use of organic sealing agents, such as epoxy resins, is totally prohibited. No organic material may be used in the treatment process implemented in the La Hague plant.

Sound elements from dismantled fuel assemblies may easily be managed by reconstructing a pseudo-assembly that meets the La Hague transportation and receivability criteria.

### 3.2.2. Slightly Damaged Fuel

This category covers fuel whose geometric and mechanical integrity the research reactor operator has ensured and guaranteed. It usually includes assemblies that have been subjected to limited corrosion attacks in the reactor or, in most instances, during storage in pools, dry shafts, vaults or casks.

Acceptance of this fuel for reception, unloading and storage is subject to testing to assess the radioactivity release rate for fuel that has been declared unsound by the research reactor operator. This testing is to be performed by the operator in accordance with the procedure proposed or approved by COGEMA.

Depending on the results of the radioactivity release test, the fuel declared unsound by the operator may be accepted without any additional measures. Otherwise, as a minimum, it will have to be placed in cylinders upon reception in La Hague.

This case will be detailed in a future paper.

### 4. CONCLUSIONS

The importance COGEMA assigns to the preparatory work designed to determine whether RTR spent fuel assemblies are acceptable for reception, unloading, storage and treatment in the La Hague reprocessing plant guarantees trouble-free management despite the intrinsic diversity of this type of fuel assembly. This approach also enables full compliance with safety requirements and limits specified by the competent safety authority.

Research reactor operators can benefit from COGEMA's experience in determining RTR spent fuel assembly acceptability to provide reliable and proven solutions for managing their fuel assemblies, regardless of whether they have been declared sound or unsound.

# **APPENDIX 1**

#### **Pre- and Post-Irradiation Fuel Characteristics**

The research reactor operator must provide COGEMA with the required spent fuel element data:

Fuel element identification number

Total mass of U and mass of U-235 and U-238 before irradiation (in g)

Total mass of U and mass of U-235, U-236 and U-238 after irradiation (in g)

If possible, the mass of U-234 and initial and residual masses of U should be indicated in grams with 2 or 3 digits after the decimal point.

Total mass of Pu and mass of Pu-238, Pu-239, Pu-240, Pu-241, Pu-242 (in mg)

Mass of Np, Cm, Am (in g)

Residence time in operating reactor (Effective Full Power Days - EFPD)

Date of unloading

Cooling time in days at assumed shipment date

Energy generated (MWd)

Burnup in MWd/t (total pre-irradiation U)

Total radioactivity (TBq)

Residual heat (W) at assumed shipment date

Inspection commitment codes: Euratom U and Pu code

Confirmation of fuel integrity and demonstration of integrity for core and pool storage, including such information as: inspection system, equipment, types of radioactivity monitored, leaktightness fault trip threshold, mean and maximum in-service radioactivity and current radioactivity.

# **APPENDIX 2**

# **Fuel Specifications**

GEOMETRY (in mm)	
Length	
Width	
Total thickness	
Fuel material thickness	
Cladding thickness	
Active length	
Max. active width	
COMPOSITION	
Fissile material alloy	
Cladding material	
Initial U-235 enrichment (%)	
Utotal concentration in (g/cm <sup>3</sup> )	
Initial Utotal mass (g)	
Initial U-235 mass (g)	
OTHER	
Drawings	
Specifications	



# DEVELOPMENT OF MELT DILUTE TECHNOLOGY FOR DISPOSITION OF ALUMINUM BASED SPENT NUCLEAR FUEL

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# ABSTRACT

The US Department of Energy (DOE) has for many years had a program for receipt and disposition of spent nuclear fuels of US origin from research reactors around the world. The research reactor spent nuclear fuel that consists of aluminum alloy composition has historically been returned to the Savannah River Site (SRS) and dispositioned via chemical reprocessing. In 1995, the DOE evaluated a number of alternatives to chemical reprocessing. In 2000, the DOE selected the melt-dilute alternative as the primary disposition path and direct disposal as the backup path. The melt-dilute technology has been developed from lab-scale demonstration up through the construction of a pilot-scale facility. The pilot-scale L-Area Experimental Facility (LEF) has been constructed and is ready for operation. The LEF will be used primarily, to confirm laboratory research on zeolite media for offgas trapping and remote operability. Favorable results from the LEF are expected to lead to final design of the production melt-dilute facility identified as the Treatment and Storage Facility (TSF). This paper will describe the melt-dilute process and provide a status of the program development.

### **INTRODUCTION**

Since the early 1960's the United States Department of Energy (DOE) has received spent nuclear fuel for disposition by processing at the Savannah River Site (SRS). The program stopped in the late 1980's. When it was restarted in 1996, the DOE also decided to pursue alternatives to processing the fuel. Research and development was begun to develop technology for two strong alternatives. One alternative was direct disposal of the fuel in an underground repository. The other involved a melt treatment for volume reduction and U235 dilution followed by disposal in an underground repository. By 2000, the DOE decided to pursue the melt and dilute technology as a primary disposition path and direct disposal as a backup. This paper will describe the melt dilute process as it has been developed at the pilot scale. This paper will also discuss some of the challenges remaining.

#### BACKGROUND

Historically aluminum based spent nuclear fuel has been chemically reprocessed at the SRS to separate fissile species. This process results in generation of aqueous waste streams. In the mid-1990's the DOE decided to pursue an alternative to chemical reprocessing. Today, only fuels designated as 'at risk' are being reprocessed at SRS. All other aluminum based fuel such as the research reactor spent nuclear fuel are being stored in a pool until a final disposition path is implemented.

Direct disposal of the SNF was developed as a primary path for several years before being declared the backup path. Direct disposal entails placing the SNF in a canister, drying the canister, sealing it and ultimately transporting it the underground repository. Major issues include how dry the fuel needs to be before storage, characterization of the fuel, addition of any neutron poisons required for repository disposal and degradation considering reconfiguration processes.

The melt dilute technology has also been developed. It was initially developed as an alternative to direct disposal. Many questions were resolved during the early development, and by 2000 the technology was sufficiently developed to declare this the primary disposition technology for aluminum based fuels.

#### **MELT-DILUTE PROCESS**

The melt-dilute process is conceptually simple, and flexible in the treatment process to produce a desired waste form for disposal. An intact fuel element is placed in a furnace and melted. Depleted uranium is added to dilute the uranium-235 enrichment to a desired level. In the process being developed at SRS, the enrichment is being targeted to below 20% to eliminate proliferation concerns during the storage and transportation of the waste forms prior to ultimate disposal. Aluminum is also added to the melt in order to keep the mixture near the eutectic (13.8 wt% uranium – aluminum) for the uranium/aluminum alloy and consequently near it's lowest melting point (646C). This minimizes release of volatile radionuclides from the spent fuel.

The melting and casting processes for production of aluminum fuel materials are well established at the site. The melt-dilute process for spent fuel necessitates contamination control and the need to work remotely. The L-Area Experimental Facility (LEF) furnace is an induction furnace that eliminates the need to have a mechanical stirrer in the operation. All stirring is accomplished inductively.

All fuel movements are done remotely or with the fuel in a shielded container such as a cask. For the demonstration facility, an existing crane is used to move the fuel. The fuel is picked up from a cask using a specially designed fuel handling tool. The tool is designed to operate when it is placed on a special power port. The power port provides electricity and/or air to operate the tool. As a safety feature, when the tool is not on a power port, none of its functions will operate. Power ports are provided on tool stands and on a datum plate above the furnace. The cask with fuel to be melted is placed under a tool stand. The tool is placed on the stand and used to pick up the fuel. The tool is a rectangular shaped box with an opening in one end. A gripper that travels the length of the box is used to draw the fuel up into the box through the opening. When the tool is removed from the tool stand a cover plate will swing under the opening. The box and cover plate are designed to contain any loose contamination on the fuel. The cover plate is also capable of holding the fuel if it is dropped. The tool has a camera inside that can be used to monitor the fuel.



Fig. I Fuel Handling Tool

The fuel handling tool is moved from the tool stand to the induction furnace. A similar power port is present over the furnace, which allows the tool to be operated to lower the fuel into the furnace. The furnace contains a graphite crucible, a carbon steel crucible liner, and a quartz liner outside of the crucible. After fuel loading, an equipment plate is loaded on top of the crucible. The equipment plate contains the primary zeolite absorber bed, a camera, and two samplers. The samplers allow samples of the molten metal to be obtained. After the system is closed and ventilation initiated, power is applied to the furnace to heat the fuel. For the demonstration facility the temperature of the fuel will be held at about 350C for one hour to drive off all residual free water. For the production scale facility the fuel will be dried before being charged to the furnace. The temperature will be increased to 850C after the one hour hold-point to ensure that the alloy melt is fully liquid. The melt will be inductively stirred, two samples retrieved, and the furnace shutdown. The molten metal will be allowed to cool in the crucible. Once cooled, the system can be opened remotely, the metal ingot removed and stored in a shielded cask and the system prepared for another melt.

The metal ingot is removed with the carbon steel crucible liner. A separate tool similar to the fuel handling tool but with a gripper designed to pick up the crucible liner is used to move the ingot to the



Fig. 2. Furnace in trailer space

The last major component of the melt and dilute demonstration facility is the control building. The control building is a fifteen by twenty-foot prefabricated building that houses the furnace controls, alarm panel and data acquisition system. The furnace controls are standard cask. This tool is also equipped with a camera and bottom cover plate capable of holding a dropped ingot.

A third, similar tool is used to move a crucible if that becomes necessary. Because of the crucible liner, the crucible is designed to be used for all the demonstration facility melts.



Fig. 3. Furnace control

induction furnace controls with the addition of two interlocks to shutdown the furnace on high furnace temperature and on the presence of moisture in the furnace refractory material surrounding the crucible. The alarm panel is also a standard alarm panel design that indicates which sensor is in alarm condition. The data acquisition system is uniquely designed for this system. The data acquisition system records all process system measurements for the duration of the melt. Any of the twelve cameras used in the process can be monitored and recorded at the data acquisition. Finally, control of the fuel handling tool, ingot handling tool, and crucible handling tool functions is at the data acquisition system.

Appropriate administrative and engineering controls have been included in the L-Area Experimental Facility (LEF) design to protect against postulated adverse events. Engineered controls include multiple barriers between water and molten metal, redundant zeolite absorber beds and several interlocks to shutdown the furnace upon loss of ventilation control, high temperature, and loss of cooling water. Administrative controls include controlled access, limit on number of fuel elements allowed in furnace area, limit on total number of melts allowed and limits on amount of combustible material allowed near the furnace.

Concurrent with developing a process for disposition of research reactor spent nuclear fuel, the waste form must be shown to be able to meet disposal system requirements or "qualified" for disposal in the repository. Qualification of the waste form is accomplished by evaluating the waste form on repository performance during its disposal lifetime. Studies have been in progress to determine the waste form thermal loads and degradation modes and rates. The waste form has also been evaluated for criticality potential under various degradation configurations. The results show that either gadolinium or hafnium up to 3 wt% will be included in the melt to ensure that the melt-dilute product remains subcritical under all postulated conditions. It has also been shown that the gadolinium and hafnium will remain with the uranium during decomposition and will not be preferentially separated.

## **TECHNOLOGY DEVELOPMENT**

The construction and installation of the LEF for demonstrating the melt-dilute treatment on a single spent nuclear fuel assembly scale is complete. Startup checks have been completed including integrated system tests using aluminum mock fuel. The overall system and operating procedures have been reviewed for readiness. The completion of an operational readiness review will allow operations with spent fuel to begin. The program has been placed on hold awaiting additional funding.

The major issue for the melt-dilute treatment technology development is the ability to continue funding the development. While the disposition path appears favorable, it requires a significant upfront expenditure to construct the TSF. Today's funding climate does not support this expenditure. The LEF demonstration has been suspended to make funds available for other DOE priorities. It is now likely that a disposition alternative with a more level funding profile will be executed.

The objectives of the pilot-scale experiments remain to be achieved. It should be shown on this scale that contamination can be controlled and that volatile radionuclides can be captured. Bench scale work to date continues to prove that the technology is viable. The melt-dilute treatment process will be demonstrated at a scaled up level before construction of a production scale facility to reduce the risks.

# CONCLUSIONS

The melt-dilute technology is a viable technology to treat highly-enriched spent nuclear fuels for ultimate disposition. It provides for spent SNF to be consolidated with flexibility and control of the composition of the waste form. In contrast, direct disposal of the fuel requires that analysis consider many types of fuel. Converting the fuel to a melt dilute waste form allows the enrichment and uranium composition to be adjusted to a specified level. Use of a melt dilute waste form also allows easier recovery of samples for confirmation of composition.



# SPENT FUEL MANAGEMENT PLANS FOR THE FIR 1 REACTOR

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# ABSTRACT

The FiR 1 –reactor, a 250 kW Triga reactor, has been in operation since 1962. The main purpose to run the reactor is now the Boron Neutron Capture Therapy (BNCT). The BNCT work dominates the current utilization of the reactor: three days per week for BNCT purposes and only two days per week for other purposes such as the neutron activation analysis and isotope production.

The final disposal site is situated in Olkiluoto, on the western coast of Finland. Olkiluoto is also one of the two nuclear power plant sites in Finland. In the new operating license of our reactor there is a special condition. We have to achieve a binding agreement between our Research Centre and either the domestic Nuclear Power Companies about the possibility to use the Olkiluoto final disposal facility for our spent fuel or USDOE about the return of our spent fuel back to USA. If we want to continue the reactor operation beyond the year 2006, the domestic final disposal is the only possibility.

At the moment it seems to be reasonable to prepare to both possibilities: the domestic final disposal and the return to the USA offered by USDOE. Because the cost estimates of the both possibilities are on the same order of magnitude, the future of the reactor itself will decide, which of the spent fuel policies will be obeyed. In a couple of years' time it will be seen, if the funding of the reactor and the incomes from the BNCT treatments will cover the costs. If the BNCT and other irradiations develop satisfactorily, the reactor can be kept in operation beyond the year 2006 and the domestic final disposal will be implemented. If, however, there is still lack of money, there is no reason to continue the operation of the reactor and the choice of USDOE alternative is natural.

#### 1. Introduction

The FiR 1 –reactor, a 250 kW Triga reactor, has been in operation since 1962. The main purpose to run the reactor is now the Boron Neutron Capture Therapy (BNCT). The epithermal neutrons (1 eV – 10 keV) needed for the irradiation of brain tumor patients are produced from the fast fission neutrons by a moderator block consisting of Al+AlF<sub>3</sub> (FLUENTAL<sup>TM</sup>) developed and produced by VTT. The material gives excellent beam values both in intensity and quality and enables the use of a small research reactor as a neutron source for BNCT purposes. Twenty-one patients have been treated since May 1999, when the license for patient treatment was granted to the responsible BNCT treatment organization, which has a close connection to the Helsinki University Central Hospital. The BNCT work dominates the current utilization of the reactor: three days per week for BNCT purposes and only two days per week for other purposes such as the neutron activation analysis and isotope production. Figure 1 describes the general layout of the BNCT facility at the FiR 1 –reactor. The facility gives a high epithermal neutron field,  $1.1 \times 10^9$  n/cm<sup>2</sup>s with a very low fast neutron and gamma component.

In the near future the back end solutions of the spent fuel management will have a very important role in our activities. There is a special condition in our operating license. We have now about three years' time to achieve a binding agreement between VTT and the domestic Nuclear Power Plant Companies about the possibility to use the final disposal facility of the Nuclear Power Plants for our spent fuel. If this will not happen, we have to make the agreement with the USDOE with the well-known time limits.



Figure 1. BNCT Facility at the FiR 1 -reactor

# 2. Current situation

In Finland also the research reactor must have a nuclear waste management plan, which contains among others a part for spent fuel management. The plan describes the methods, the schedule and the cost estimate of the whole spent fuel management procedure starting from the removal of the fuel from the reactor core and ending to the final disposal. The plan has been based on the assumption that the final disposal site will be somewhere in Finland. The Finnish Parliament ratified in May 2001 the decision in principle on the final disposal facility to be situated in Olkiluoto, on the western coast of Finland, for the spent fuel of the nuclear power plants. Olkiluoto is also one of the two nuclear power plant sites in Finland. The final disposal facility is supposed to be in operation in 2020.

There is a special condition in the current operation license of our reactor. We have to achieve a binding agreement between our Research Centre and the domestic Nuclear Power Plant Companies about the possibility to use the joint final disposal facility of the Nuclear Power Plants for our spent fuel, if we want to continue the reactor operation beyond the year 2006. We have had already for twelve years an agreement in principle with one of the Nuclear Power Companies about the final disposal matter, but it does not satisfy the requirements any more. If an acceptable agreement will not be accomplished in about three years' time, we have to use the USDOE alternative with the well-known time limits. The Ministry of Trade and Industry has the responsibility to decide, if the agreement is acceptable or not. Before we can start the real negotiations about the final disposal of our spent fuel with the Nuclear Power Companies, we have to prepare a safety study about the behaviour of the Triga fuel in the final disposal surroundings.

The operation license of our reactor will expire in 2011. It is very probable that there will be certain waiting time from the shut down of the reactor to the opening of the final disposal facility. Therefore there have to be a sufficient interim storage for the spent fuel before the transportation to the final disposal facility. After enlargement work in 1997 we have sufficiently storage capacity for the fuel in the reactor building. In addition to the domestic final disposal solution there is still the USDOE alternative available until 2006.

# 3. Finnish final disposal solution

The Finnish Nuclear Power Companies founded in 1995 a separate company Posiva to develop the technology and carry out safety analysis and site investigations for implementing spent fuel final disposal. In 1999 Posiva submitted an application for a decision in principle for a final repository to be built at Olkiluoto, the site of two nuclear power plant units. At the end of the year 2000 the Finnish government approved the application and sent it to the parliament for ratification. The ratification took place in May 2001. Separate licenses still will be needed for the construction of the facility, scheduled

to start in 2010, and also for the operation, 10 years later. The government alone will decide, if these licenses are granted or not.

For the final repository the spent fuel will be encapsulated in airtight copper canisters and situated in the bedrock at a depth of 500 m. The safety of this deep underground repository is based on multiple natural and engineered barriers. Each canister contains 12 normal fuel assemblies from nuclear power plants. The present concept for Triga fuel elements is that the elements will be loaded in containers, which have the same outer dimensions as the nuclear power plant fuel assemblies. This ensures that the Triga fuel will be easily handled in the final disposal facility and also loaded in the heavy copper canisters.

# 4. Long term safety of the Triga fuel in the final disposal surroundings

In order to start the negotiations with the Nuclear Power Companies or their representative Posiva we have to prepare a safety study about the long-term behaviour of the Triga fuel in the final disposal surroundings. As was already mentioned the Triga fuel elements will be loaded in containers, which have the same outer dimensions as the nuclear power plant fuel assemblies. We need from 3 to 5 such containers for all the Triga fuel elements. The containers can easily be loaded into the heavy copper canisters, which have 12 positions to be loaded. For the criticality reason the Triga containers will be situated in the outer zone of the canister and the inner zone will be left empty. In practice the empty positions will be loaded with dummy assemblies made of cast iron. It can be shown that the system is critical safe. This is important, because if the criticality safety would demand the fuel to be divided to two or more canisters, the expenses would also be about twice or more compared to the one canister alternative.

### 5. Decisions in the near future

If we want to utilize the USDOE policy and return our fuel back to USA, it means in practice, for economical reasons, that the whole inventory of the irradiated fuel should be sent to USA at the same time. Thus the return of the fuel back to USA means in other words the shutting down of the reactor. In our case the important question will be: Are we willing to shut down the reactor within the time limits declared by the USDOE?

The BNCT work is today the main purpose to run the reactor. The amount of BNCT irradiations is still rather low: twenty-one irradiations during the first thirty months. At the moment we assume that we are getting funding for the reactor and the BNCT for the next two or three years. During that time we together with the treatment organization have the opportunity to show that the BNCT irradiations will be needed also in the future and that the annual number of patients will be satisfactory to run the reactor. If the turnover of the BNCT and other irradiations will be satisfactory and the positive trend seems to continue, there is no reason to use the USDOE alternative. Instead it is reasonable to continue the reactor operation beyond the year 2006, which means inevitable also the choosing of the domestic alternative for the treatment of the spent fuel. If, however, the frequency of the treatments is not growing up to a acceptable level, the funding of the reactor will be stopped after the said period, which leads inevitably to the permanent shut down of the reactor. In that case the USDOE alternative seems to be the right one.

### 6. Conclusions

We have now ahead of us a restricted period of time, during which we have the possibility to show that the BNCT method works successfully and that the annual amount of patients is satisfactory. After about two years, in 2004 we are in the situation, when we have to decide in every case, if the reactor will continue the operation or will it be shut down. As the criterion there will be the possible success of the BNCT. After the decision of the reactor operation the choosing between the USDOE and the domestic back end solution will be rather easy, because the expenses of both of the spent fuel management alternatives seem to be of the same magnitude.



# THE RUSSIAN FEDERATION LEGISLATION. THE NEW LAWS. PROSPECTS FOR INTERNATIONAL COOPERATION.

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# ABSTRACT

- Survey of the regulatory basis for the international cooperation of the Russian Federation in the area of foreign commercial and research spent fuel management. Analysis of the latest legislative amendments. Complex approach and environmental priorities of the new legislative initiatives (three Federal laws):
- Amendments to Articles 1, 47 and 64 of the Federal Law on "Utilization of atomic energy"
- Amendments to Articles 50 of the Federal Law on "Environmental protection"
- The new Federal Law "On Special ecological programs for the clean-up of areas, contaminated by radiation".

The main regulatory documents being the legal basis in the 90s for foreign NPP irradiated nuclear fuel to be returned to the Russian Federation are the following:

• Intergovernmental Agreements on cooperation in the field of atomic energy;

• The Law of the Russian Federation "On the Use of Atomic Energy" as of October 20, 1995;

• The Law of the RSFSR "On Environmental Protection" as of December 19, 1991;

• Decree of the Government of the Russian Federation No. 773 "Procedure for acceptance of foreign NPP irradiated nuclear fuel for further reprocessing at Russian enterprises and return of radioactive waste and materials resulted from reprocessing" as of July 29, 1995;

• Decree of the Government of the Russian Federation No. 745 "Amendments to and modifications of the procedure for acceptance of foreign NPP irradiated nuclear fuel for further reprocessing at Russian enterprises and return of radioactive waste and materials resulted from reprocessing" as of July 29, 1995

In accordance with the above list of regulatory documents the following terms and conditions for acceptance of irradiated nuclear fuel have been applied down to recent times:

• Irradiated nuclear fuel (INF) should be reprocessed provided that radioactive waste is to be returned;

• Return of uranium and plutonium is subject to the consent of the Parties.

As you see the Russian legislation being in force limited the potential of the Russian Federation at irradiated nuclear fuel markets. However, it is necessary to note the positive changes made in 1998. The Decree of the Government of the Russian Federation No. 745 as of July 10, 1998, introduced amendments to the existing procedure for acceptance of foreign NPP irradiated nuclear fuel which allowed to extend the potential of the Russian suppliers of irradiated nuclear fuel reprocessing services for the Eastern European countries in the framework of intergovernmental agreements concluded by the USSR before 1991.

The activity to improve the legislation was carried on in 2000-2001 and as a result, as you are probably aware, in July 2001 the President of the Russian Federation signed the Laws aimed to extend the range of services of the Russian enterprises in this area:

• On Amendments to Article 50 of the Law of the RSFSR "On Environmental Protection"

• On Amendments to the Federal Law "On the Use of Atomic Energy"

• On special ecological programs for the recovery of the radioactively contaminated areas of the Russian Federation

All the laws had passed state ecological expertise, had been adopted by the State Duma in three readings and had been approved by Federation Council prior to signing.

With regard to the modifications of the existing Russian legislation it is necessary to note that the new laws introduce certain changes and amendments to the previous terms and conditions of international cooperation of the Russian Federation in the field of nuclear fuel cycle.

This Law:

• clarifies the meaning of INF and RW;

• provides the possibility to store INF as a specific technological operation;

• enables to set the procedure (terms and conditions) to import foreign INF as well as to solve the issue of waste return by the Government of the Russian Federation.

Import terms and conditions:

- to conduct ecological and other expertise;
- to reduce the risk of radiation impact;
- to improve the level of environmental safety;
- to consider international agreement as legal basis;
- to ensure non-proliferation principle;
- to have priority right to return waste

This Law:

• clarifies the terminology being previously in use (fuel assemblies, irradiated fuel assemblies);

• provides the opportunity to make foreign trade deals involving nuclear fuel on the basis of contracts (leasing);

• provides the opportunity to store irradiated fuel assemblies as a specific technological operation.

This Law:

• regulates the activity related to the implementation of the programs for recovery of radioactively contaminated areas financed from INF import funds.

We would like to consider this Law in detail as it is a key point for understanding of our approach.

The Law introduces the term "special ecological program". "Special ecological program for recovery of radioactively contaminated areas " (hereinafter referred to as "special ecological program") means the program for rehabilitation of radioactively contaminated sites to be financed from the earnings received from foreign trade transactions with irradiated fuel assemblies of nuclear reactors.

"Specific ecological programs are aimed to ensure public radiation safety, to generally reduce radiation impact risk and to improve environmental situation at radioactively contaminated sites by means of rehabilitation measures, disposition or elimination of radiation hazardous facilities being decommissioned. Special ecological programs include:

• goals, measures, phases and timing to implement major measures;

• information regarding environmental condition and public health;

• assessment of environmental impact resulted from economic and other activities;

list of measures to recover contaminated areas;

• availability or necessity to develop environment control system as well as ecological and sanitation monitoring systems;

• results of the implementation of special ecological programs involving expected changes of the environment and public health;

• financing level for the above programs in general and for specific periods"

"The procedure and priority for special ecological program financing shall be specified by the Government of the Russian Federation as agreed up on with state authorities of the entities of the Russian Federation. The priority is given to the special ecological programs of the entities of the Russian Federation on the territory of which the facilities dealing with reprocessing of imported foreign irradiated fuel assemblies of nuclear reactors and temporary storage are located."

"The amount of currency funds received from foreign trade transactions with irradiated fuel assemblies of nuclear reactors and transferred to special account of budget fund of Federal Executive Body exercising state management of the use of atomic energy as well as the expenditures of the above fund are specified by the Federal Law on Federal Budget for corresponding year indicating funds breakdown with regard to areas and entities of the Russian Federation. In accordance with the Federal Law on Federal Budget for corresponding year 75% of currency funds received from foreign trade transactions with irradiated fuel assemblies of nuclear reactors and transferred to special account of budget fund of Federal Executive Body exercising state management of the use of atomic energy shall be allocated to specific ecological programs excluding the costs of management of irradiated fuel assemblies of nuclear reactors and products of their reprocessing approved by the Government of the Russian Federation as appropriate."

In accordance with legislation of the Russian Federation Accounting Chamber shall handle control over the specific budget funds of the Federal Executive Body exercising state management of the use of atomic energy.

"The entire project involving foreign trade deal related to the import of foreign irradiated fuel assemblies of nuclear reactors and implementation of specific ecological program or programs financed from the funds of the above deal should be subject to the State Ecological Expertise. Foreign trade deals involving irradiated fuel assemblies of nuclear reactors shall be made by the organization specially authorized by the Government of the Russian Federation provided positive decision of the State Ecological Expertise."

However, the legal basis shall become the most comprehensive when appropriate modifications are incorporated in the bylaws (primarily, to the INF import procedure mentioned in the list of regulatory documents). Now we can say with rather high level of probability that the new bylaws shall meet today's requirements. Additionally it is worth to mention that it is planned to provide the possibility not to return the products of reprocessing for the countries operating the reactors of Russian (Soviet) type and (or) for those countries to which fresh nuclear fuel of Russian origin or fuel fabricated from Russian nuclear materials is supplied. For instance, fresh fuel transfer schemes based on leasing operations when irradiated nuclear fuel being used at foreign NPP is returned to the country is under development in Russia. As it is mentioned above, the new Russian legislation meets modern goals and opens new perspectives for international cooperation at the final stage of nuclear fuel cycle. We are confident that such cooperation may be mutually beneficial and effective.

Thank you.

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# **Posters**

# IN-REACTOR BEHAVIOUR OF CENTRIFUGALLY ATOMIZED U<sub>3</sub>Si DISPERSION FUEL IRRADIATED UP TO HIGH BURN-UP FOR HANARO

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# ABSTRACT

The irradiation test on atomized and comminuted  $U_3Si$  dispersion mini-element fuels has been performed up to about 69 at.% U-235 burn-up under normal linear power condition in OR-4 hole position of HANARO reactor, in order to examine the irradiation performance of the atomized  $U_3Si$  for the driver fuels of HANARO. Irrespective of the powdering method, the in-reactor interaction of  $U_3Si$ dispersion fuel meats is generally assumed to be acceptable with the range of ~9  $\mu$ m in average thickness. The  $U_3Si$  particles have relatively fine and uniform bubble size distribution, irrespective of the powdering method. Both the atomized and comminuted  $U_3Si$  dispersion mini-element fuels exhibit sound swelling behaviours with the swelling range of ~5 % in  $\triangle V/V_m$ , which meets with the safety criterion of the fuel rod, 20vol.% for HANARO.

#### **1. Introduction**

 $U_3Si$  fuel dispersed in aluminium matrix was developed over the world from 1970s, and has been used as nuclear fuel for research reactors with verification of the in-reactor performance. The rod-type  $U_3Si$ fuel assembly has also been applied in HANARO as a driver fuel after confirmation of fuel safety in AECL [1]. KAERI has performed the localization development of the  $U_3Si$  driver fuel since 1987.

During the development process of the nuclear fuel for HANARO, new fuel fabrication technology was applied, preparing the spherical fuel powders directly from the alloy melt by centrifugal atomization method instead of conventional comminution method, was applied and eventually registered as an invention patent in USA, Canada, Germany and Korea. The centrifugal atomization technology, without the homogenization treatment and the mechanical comminution of an as-cast uranium alloy billet, has brought the advantages of the process simplification, the minimization of the fabrication space, the improvement of the production yield, the fuel productivity, the powder purity, the fuel formability and the thermal conductivity along the transverse direction, and the decrease of the as-fabricated porosity and the thermal swelling (Fig. 1) [2-4].

In this study, the  $U_3Si$  fuel mini-elements with a fuel loading of 24vol.% were irradiated to characterize the in-reactor behaviours of atomized  $U_3Si$  dispersion fuels at approximately 69at.% U-235 burn-up at OR-4 hole position in the HANORO reactor. Thereafter, the in-reactor performance of atomized  $U_3Si$  dispersion fuels was examined primarily using an optical microscope (OM) and a scanning electron microscope (SEM), compared with that of comminuted fuels.

#### 2. Experimental procedure

The  $U_3Si$  fuel powders were prepared by rotating-disk centrifugal atomization and mechanical comminution, using low enriched uranium lumps (99.9wt.% pure) and silicon chips (99.9999wt.% pure) [5]. The reduced fuel cores having  $\phi 6.35$ mm in diameter and L200mm in length were dispersed by the atomized or the comminuted  $U_3Si$  particles of 24vol.% in pure aluminum matrix having uranium loading of 3.15 g-U/cm<sup>3</sup>. The type and the dimension of the fuel assembly for the irradiation test were

the same as that of the 18 rod type driver fuel assembly for HANARO reactor. The test rod was composed of an extruded  $U_3$ Si-Al dispersion fuel meat, an aluminum cladding having eight cooling fins, and two aluminum end plugs. The fuel assembly for the irradiation test was fabricated with six test fuel mini-elements (three atomized fuel mini-elements and three comminuted fuel mini-elements) and twelve dummy fuel elements.

The mini-elements from the atomized U<sub>3</sub>Si fuel and the comminuted fuel, were loaded at OR-4 hole position in the HANARO reactor and irradiated for 220 F-P-Day residence time at about 70 % operating capacity from December 1997 to June 1999. Table 1 summarizes typical irradiation conditions for mini-elements in the HANARO reactor. The maximum linear power was evaluated to be about 89 kW/m, in which the peak temperature of mini-elements was calculated to have been 262°C during irradiation. The average burn-up and the fission density in the fuel meats after irradiation test were estimated to be about 69 at.% U-235 and 1.26 x 10<sup>21</sup> fissions/cm<sup>3</sup>, respectively. Thereafter, in order to prepare the samples for OM observation from irradiated mini-elements, the mini-elements were cut down into three pieces, and then the optical observation was focused at the sample positions from the fuel meat center to the edge region in the middle section of mini-element. In order to prepare SEM sample, a special punching tool was used to make the samples for SEM observation. Two small pieces of irradiated samples (atomized and comminuted) with having a diameter of 1.57 mm using punching jig in hot cell was taken from the center region of fuel meat. Then the fuel meat in the punched sample was cut down by hand in a glove box to observe the fractured surface of irradiated fuel particles. SEM observation on the polished fuel samples was also carried out to investigate the bubble size distribution and the fuel/Al reaction layer.

#### 3. Experimental Results and Discussion

Fig. 2 shows the typical optical micrographs of polished surfaces in the atomized and the comminuted  $U_3Si$  mini-elements at about 69 at.% burn-up. The fuel-matrix interaction layer of the atomized spherical particles is relatively uniform and generally acceptable in the range of ~9 µm in average thickness. Irrespective of the powdering method, the  $U_3Si$  mini-elements do not have a prominent reaction layer between  $U_3Si$  fuel and aluminium matrix; however, the comminuted irregular particles have less uniform and even thicker reaction layer relative to the atomized particles (Fig. 3). The possible reason is supposed as follows. The atomized particles do not have a prominent deformation damage formed during the powdering process. Whereas, the comminuted particles have severe deformation damage with a high dislocation density formed during the powdering process. The local deformation zone formed during the comminution process increases the diffusion rate of aluminium atoms to the irregular particles, which leads to less uniform and thicker fuel/Al reaction layer relative to the atomized particles.

The scanning electron micrographs of fractured surface and the apparent fission gas bubble size distribution in the atomized and the comminuted  $U_1$ Si mini-elements at about 69 at.% burn-up are shown in Figs. 4 and 5. There are no fission gas bubble-free zones in the U<sub>3</sub>Si particles, in contrast to the atomized U-Mo particles. There are apparent similarities in comparing the bubble formation as well as bubble distribution between the atomized and the comminuted fuel samples. The bubbles formed to be visible have covered entirely the fuel particles, nucleating and growing at the whole regions. The  $U_3Si$ particles have relatively fine and uniform bubble size distribution, and relatively thin and uniform fuel/Al layer thickness, irrespective of the powdering method. The bubble population in the atomized fuel appears to be more homogeneous with the characteristics of narrower bubble size distribution than that in the comminuted fuel. The maximum and the average bubble diameter of the U<sub>3</sub>Si mini-elements are approximately 1.2  $\mu$ m and 0.4  $\mu$ m. The U<sub>3</sub>Si dispersion fuels show no indication of breakaway swelling, similar to that of U<sub>3</sub>Si<sub>2</sub> and U-Mo [6-8]. There is no evidence of interlinking as the relatively uniformly and randomly distributed fission gas bubbles in places in particles. The atomized U<sub>3</sub>Si dispersion fuels do not show any indication of breakaway swelling, but stable irradiation behaviour, similar to that of U<sub>3</sub>Si<sub>2</sub> and U-Mo. The mini-elements have swollen by about 0.8 vol.% per 10 atomic percent burn-up to a final burn-up of about 69 atomic percent, which meets with the safety criterion of the fuel rod, 20vol.% for the HANARO. It is thought that stable irradiation performance resulted from the in-reactor irradiation at relatively low temperature of  $\sim 262$  °C up to high burn-up.

According to the previous studies, the stoichiometry of the interaction layer formed in uranium silicide dispersion fuel during irradiation, has been known to be  $U(Si_3,AI)_3$  [8]. In this study, the more quantitative analysis for compositional changes was carried out across the fuel/aluminum interface by using Electron Prove Micro Analysis (EPMA). Fig. 5 shows the typical compositional changes near the fuel particle surface in U<sub>3</sub>Si sample. In an unreacted fuel particle region, the result is fairly consistent with fuel composition of U<sub>3</sub>Si. Moreover, it is also observed that the compositions uranium, silicon and aluminum in the inter-diffusion layer changes gradually and continuously across the reacted layer, not being in unique composition such as  $U(Si_3,AI)_3$ , indicating that the formation of interfacial layer in U<sub>3</sub>Si/AI during irradiation is a diffusion controlled reaction.

#### 4. Conclusions

In order to localize the driver fuels for HANARO, centrifugally atomized U<sub>3</sub>Si mini-element fuels, have been fabricated and irradiated up to high burn-up under normal power condition in HANARO reactor, compared with mechanically comminuted fuels. The fuel-matrix interaction layer of the atomized spherical particles is relatively uniform and generally acceptable in the range of ~9  $\mu$ m in average thickness; however, the comminuted irregular particles have less uniform and even thicker reaction layer relative to the atomized particles. The U<sub>3</sub>Si particles have relatively fine and uniform bubble size distribution, irrespective of the powdering method. The bubble population in the atomized fuel appears to be more homogeneous with the characteristics of narrower bubble size distribution than that of the comminuted fuel. The atomized U<sub>3</sub>Si dispersion fuels do not show any indication of breakaway swelling, but stable irradiation behaviour with the irradiation swelling of ~5 % in  $\Delta V/V_m$ , similar to that of U<sub>3</sub>Si<sub>2</sub> and U-Mo, which meets with the safety criterion of the fuel rod, 20vol.% for the HANARO.

#### Acknowledgements

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Fig.1. The fabrication process flow sheet of for HANARO fuel.

Table 1. Irradiation Conditions in the HANARO Reactor.

Fig. 2 The Optical micrographs of polished surfaces in the atomized (a) and the comminuted (b) U<sub>3</sub>Si mini-elements at about 69 at.% burn-up.



Fig. 3. The Fuel/Al reaction layer thickness of the atomized and the comminuted U<sub>3</sub>Si mini-elements at about 69 at.% burn-up.



Fig. 4. The scanning electron micrographs of fractured surface in the atomized (a) and the comminuted (b) U<sub>3</sub>Si mini-elements at about 69 at.% burn-up.



Fig. 5. The apparent fission gas bubble size distribution in the atomized (a) and the comminuted (b) U<sub>3</sub>Si mini-elements at about 69 at.% burn-up.



Fig. 6. The typical compositional changes at near the fuel particle surface in the atomized(a) and the comminuted (b) U<sub>3</sub>Si mini-elements at about 69 at.% burn-up.



# BORO SILICATE GLASS : THE PROVEN CONDITIONING OF RTR ULTIMATE WASTE

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# 1. INTRODUCTION

Long term management of the Research and Test Reactor (RTR) spent fuels is one of the main concerns expressed by the research reactor operators.

In the past, a number of RTR operators have chosen to send their spent fuels to the US in the framework of the US take back program. It was then a major advantage since the US in counterpart returns no waste. However, this possibility ends as of May  $12^{th}$ , 2006.

The RTR operators will have then to choose a way to manage safely, on the long term, their spent fuels.

As promising solutions, currently under study, have been found for a final *direct disposal* of the spent fuels coming from power reactors, the methods are not directly applicable to the RTR spent fuels. In both cases, the spent fuel is placed into canister acting as the first barrier in the final disposal concept. Due to their metallic structure and Al cladding, RTR spent fuel of both HEU (High Enriched Uranium) and LEU (Low Enriched Uranium) types face degradation phenomena during a long-term storage. The uncertainties are mainly on the corrosion rate of the cladding and subsequent leaching rate of the fission products and the transuranium elements in case of groundwater intrusion into the canister. As it has already been detailed in others instances, neither nuclear safety is ensured due to uranium enrichment linked criticality risks nor repository mechanical stability due to hydrogen production through corrosion processes.

Therefore, direct disposal requires watertight and durable conditioning on a geological time scale, for which no satisfactory solution has yet been found. As a consequence no direct disposal of RTR spent fuels without prior treatment is worldwide neither existing nor planned.

As it is said here above treatment of the RTR spent fuels is necessary but, the foreseen treatment must ensure the production of raw waste which can be conditioned under a compatible form with a final disposal. Although no final disposal exists, all the concepts developed around the world are very demanding on the various properties of the conditioned waste foreseen to be placed in.

In fact, a key safety criterion of all the final disposal designs and their associated requirements for the waste aims at protecting future generations. As a consequence, the radioactive waste are not dumped, even carefully selected, into repositories for geological time scale without proper encapsulation.

To meet this safety requirement, the conditioned waste must have the following properties :

- A good chemical, mechanical and thermal stability
- The matrix, if any, must have a good resistance to the radiations,
- A low leachability.

The treatment – conditioning by reprocessing offers a proven management solution by the conditioning of the ultimate waste in the boro silicate glass.

# 2. BORO SILICATE GLASS

Glass, in general, have a long term durability. The best proof is given by the obsidian, by the glass accessories made several thousand years ago in Egypt which still exist, or by the glass ornaments with an history of more than a thousand years remain attractive in Japan.

Vitrification of high-level liquid waste makes use of glass's characteristics, such as strong mechanical, thermal, chemicals and radioactive resistance.

After extensive research in several countries, glass has proven to be the optimal solution for immobilising the chemicals elements present in the highly radioactive liquid waste solution.

An international consensus (France, Japan, UK, USA, Germany, Belgium, Russia, Switzerland, Netherlands, Italy ...) exists on the choice of borosilicate glass as being the matrix for immobilising fission products and long lived actinides in solid form.

For more than 40 years now, French scientists and nuclear experts have been dealing with vitrification in order to give a proper and comprehensive understanding of the glass properties. These long-term studies performed by the CEA (French Atomic Energy Commission) resulted in the formulation of the so-called R7 and T7 glasses.

The principal criteria for the formulations were : mechanical and thermal stability, homogeneity, radiation resistance, high containment capacity, low volume, corrosion resistance, low leachability, easy fabrication (mastered technology), and flexibility with regard to the composition of the waste to be conditioned.

It was found rapidly that boro silicate glasses were the best adapted to the immobilization of fission products solutions resulting from the reprocessing of LWR fuel using a PUREX-type process.

The studies, which lasted for several years, were based on several hypotheses regarding the composition of the feed and evolving sets of criteria. As a result, a large number of compositions were tested in the silica-boron-soda system, including glasses with very high waste loadings. The influence of several constituents has been investigated (iron, gadolinium, lithium, zinc, aluminum, calcium, molybdenum, phosphate, magnesium, titanium, zirconium, tin...). These studies, which involved the fabrication of hundreds of non-radioactive and radioactive melts, enabled the accumulation of a very large knowledge database.

The glass development was conducted in three steps :

- Demonstration,
- Technology development,
- Long-term containment characterisation.

The demonstration phase was started in 1962, by experimentation on the active test called VULCAIN. Based on the result of this experimentation, a longer phase starts in order to develop the right technology to implement on an industrial scale this type of conditioning. A first pilot was designed, named GULLIVER, in 1964, the capacity of which was 5 litres per batch ; a second one, named PIVER, in 1969, implemented on Marcoule site.

Based on this experience, a first facility, AVM (Atelier de Vitrification de Marcoule) was implemented in Marcoule in 1978. This facility is still working at that time.

Today, French vitrification process clearly appears as one of the best solution both in terms of industrial maturity and efficiency. Commercial-scale high level waste vitrification is on line at La Hague reprocessing plant (R7 and T7 facilities).

These two units were commissioned, respectively in 1989 and 1992. Each unit was designed to produce around 600 glass canisters per year.

The main records for these facility are :

	Type of facility	Date of commissioning	Number of containers produced until end 2001
PIVER	Pilot	1957	N/A
AVM	First generation of production	1978	~2800
R7	State of the out	1989	~5100
T7	State of the art	1992	~3600

In addition, for the R7 and T7 facilities, radioactive samples were taken after the hot start-up of the facilities, and characterised to confirm that the product displayed the expected characteristics. These analyses were conducted in two independent laboratories CEA (France) and JAERI (Japan). The results found by the laboratories were in accordance with the values declared by COGEMA.

The last stage of the glass characterisation, required for its acceptance, is the evaluation of its longterm performance. This stage includes irradiation stability, thermal stability, and long-term leach testing. This stage is necessary to provide information to the organisation in charge of disposal (ANDRA in France), for incorporation into the Safety Analysis of the future disposal site.

Operational models are supported by a scientific "state of art" dealing with the containment capacity of high active and long lived radioactive waste package and can be used in numerical simulation tools. They take into account a step by step phenomenology approach, necessary to answer in time the radionuclides release question. The limits of model validity have to be specified and their input parameters have to be accessible through industrial characterisation.

Mathematical models which describe at millennium scale the containment function of high active and long lived radioactive waste packages, under aqueous alteration conditions, have been defined for nuclear glass.

Because of its high thermal power and high activity content, the long-term behaviour of the French glass has been deeply investigated for many years and is now well known. Many studies of the kinetics for aqueous alteration have been performed all over the world, and particularly in France.

Operational model, simply associated with the initial kinetics alteration of the glass is now available and taken into account by repository designers. In the extremist approach, where the water in contact with the glass is continuously removed, the glass release is around  $10^{-5}$  of the initial content per year.

#### **Specification**

Characterisation studies have verified the main properties of the final products and have established the glass specification. The vitrified waste specification was subjected to review by an independent commission of nuclear specialists.

The here above studies allow identifying an optimum glass composition for High Liquid Waste, along with an operating range of process parameter for the "AVM type" technology.

This was formalised in the "Specification of Vitrified Residues produced from the reprocessing at UP2 / UP3 La Hague plants". The specification includes "Guaranteed parameters" i.e. those parameters identified as key parameters in the process to ensure the glass canister's quality.

The specification was subjected to review by a commission of scientists and nuclear experts acting on behalf of the French regulatory authority, DSIN. It was also provided to ANDRA (French National Radioactive Waste Management Agency) for comments. The French safety authority approved the vitrified residue specification in 1986.

This specification was then submitted and approved by the safety authorities of the COGEMA's foreign customers :

Japan	August	1988
Germany	August	1988
Belgium	January	1995
Switzerland	August	1988
Netherlands	November	1989

#### 3. RTR TREATMENT CONDITIONING

RTR spent fuels treatment – conditioning by reprocessing features two advantages. First, it benefits from the long experience of existing flexible industrial facilities from countries like France. Secondly, it offers a dramatic volume reduction of the ultimate waste to be stored under well-characterised, stable and durable forms.

It exists two types of RTR fuels, which have the following typical content per assembly :

	Fuel type	Weight	U <sub>tot</sub> (g)	Al (kg)	Si (g)	Initial U235 enrichment (%)
HEU fuels	UAl	from 2,7	from 150 to 900	from 2.5	/	90 %
LEU fuels	U <sub>3</sub> Si <sub>2</sub>	to 8,5	from 2000 to 2500	to 7,5 kg	200	20 %

These two types of typical RTR fuels, have the following typical content after irradiation in the reactor :

	Al (W %)	Si (W %)	U (W %)	Pu (W %)	Fission Products and Actinides
HEU fuels	~ 92.5	/	5	0.05	2.5
LEU fuels	64.7	3	29.7	0.3	2.3

After a dissolution operation of the RTR spent fuel, the resulting liquors are diluted into the power reactors UO2 spent fuels to blend down the U-235 content to a maximum of 2% before the separation facility. This blending insure the non-proliferation.

The following step consists in the separation of the Uranium, the Plutonium and the Fission products. The aluminium, the silicium and in the future the molybdenium follow the fission products flux in in order to be conditioned with them.

The treatment –conditioning allows then :

- In one hand, the recovery of the uranium which can be recycled,
- On an other hand, the recovery of the plutonium which can also be recycled,
- On an other hand, the immobilisation in boro silicate glass of the fission products, the actinides and the aluminium.

# 4. CONCLUSION

Any spent fuel management methods must comply with the final disposal key objective to store safely the waste. A consequence is that a lot of very stringent criteria for the waste to be stored appear in all the studies performed to design a final disposal.

The possibility to dispose of the RTR spent fuel in a geological repository is neither internationally forecast nor seem realistic, due to first, degradation phenomena and possible criticality incident, the treatment – conditioning by reprocessing is then the solution for back end management of RTR spent fuels.

The main advantage of the treatment –conditioning by reprocessing stands in the fact that the incorporation of the ultimate waste in a boro silicate glass matrix is proven :

- An international consensus exist on this method of conditioning the fission products and the actinides,
- The formulation of the boro silicate glass is the result of many years of R&D programs,
- The boro silicate glass matrix allows sufficient flexibility to condition the ultimate waste coming with various type of spent fuels (NPPs spent fuel, UA1 RTR spent fuel, UM0 RTR spent fuel ...),
- The operational scheme get many years of experience with the reprocessing of NPPs and RTR spent fuel,
- As for the R7 T7 vitrified residues, it exists a specification, which lists a number of parameters deriving from the R&D programs. This specification is approved by several countries around the world,
- The boro silicate glass is one of the waste taken into account to design the geological repository in many countries around the world like France, the USA, Japan, Germany, Belgium, Switzerland, Netherlands, the United Kingdom,...



# CONSORTIUM NCS/GNS: DISPOSAL OF SPENT NUCLEAR FUEL FROM THE DKFZ (HEIDELBERG, GERMANY)

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# ABSTRACT

Before the decommissioning of the TRIGA HD II reactor of the "Deutsches Krebsforschungszentrum" in Heidelberg (DKFZ) the spent nuclear fuel (126 TRIGA-F/A) had to be discharged from the reactor and shipped in 2001 to the United States.

The Consortium NCS/GNS, together with the DKFZ, carried out this task including the following essential details:

- Implementation of structural measures inside the reactor building.
- Development and fabrication of loading units for the F/A.
- Development and fabrication of a transfer system for the loading units.
- Obtaining new German and US certifications for the casks.
- Loading of the casks.
- Shipment of the casks from DKFZ to the United States by road and sea.

The successful completion of this task once again proved the operational experience of the Consortium NCS/GNS, which will be useful for further worldwide shipments of spent nuclear fuel from research reactors, e.g. shipments from Europe, South America and Australia to the United States and France.

#### 1. Introduction

The Consortium NCS/GNS is involved in the management of the worldwide shipment of spent nuclear fuel assemblies (F/A) from research reactors, e.g. shipments from Europe, South America and Australia to the United States and France.

The operational experience of NCS/GNS was shown during the shipment of TRIGA-Fuel from the TRIGA HD II reactor of the <u>Deutsches Krebsforschungszentrum Heidelberg</u> (DKFZ, German Cancer Research Center) in 2001. Before the decommissioning of the DKFZ-Reactor, the spent nuclear fuel (126 TRIGA-F/A) had to be discharged from the reactor and shipped to the United States in three casks (two GNS 16 and one TN 6/3).

The Consortium NCS/GNS was engaged for this task by DKFZ. NCS/GNS, together with DKFZ, carried out the complete task including the following details:

- Planning and implementation of structural measures inside the reactor building.
- Development and fabrication of special loading units for F/A including those for bent F/A.
- Development and fabrication of a transfer system for transferring F/A from the reactor pool to the casks (dry loading procedure).
- Drawing up of application documents (safety analysis reports) for the casks.
- Obtaining new German and US certifications for the casks according to the inventories.
- Accompanying of all reviews by site authorities and independent experts.
- Cold tests of casks and equipment at Idaho Falls and DKFZ.

- Loading of the casks.
- Planning and preparation of the shipment.
- Shipment of the casks from DKFZ to the United States by road and sea.
- Supervising the unloading of the casks at Idaho Falls, United States.

In the following sections several of the steps concerning the cask loading inside the reactor hall are described.

#### 2. Fuel Assemblies

126 spent TRIGA-F/A had to be discharged from the reactor. The F/A were divided into the following groups:

- 65 Standard F/A with aluminum cladding (length 720.6 mm),
- 56 Standard F/A with steel cladding (length 732.6 mm) and
- 5 Fuel Follower Control Rods (FFCR) with steel cladding (length 1175 mm).

The max. diameter of all F/A was < 38 mm, 11 F/A with aluminum cladding were bent, three of them by more than 12 mm. The total U-mass of all 126 F/A was appr. 23.2 kg with appr. 4.6 kg U-235 (appr. 20 % enrichment). The maximum heat decay per F/A was appr. 0,9 W.

### 3. Casks

Due to their length of 1175 mm the five FFCR had to be loaded into the cask type TN 6/3 (usable cavity length 1500 mm). Additionally the two mostly bent Standard F/A were also loaded into this cask. The remaining 119 Standard F/A were loaded into the cask type GNS16 (usable cavity length 980 mm).

### 4. Loading Units

The F/A intended for the cask type TN 6/3 could be loaded directly into the cask basket. The basket of the cask type GNS16 is equipped with 15 cavities, which means that it can contain up to 15 loading units. Twenty-nine loading units were equipped with steel tubes for the reception of four F/A each. One loading unit, especially designed for bent F/A, was equipped with three steel tubes for the reception of three F/A (two normal tubes for non-bent F/A and one long slot tube for F/A, bent by more than 12 mm). At the beginning of the task, damaged fuel could not be excluded. So all loading units were equipped at the upper and lower ends with filter plugs to avoid contamination of the cask cavity. Figure 1 shows a loading unit for the reception of the F/A.



Fig. 1 Loading Unit (with dismounted upper Filter Plug)

# 5. Reactor Hall and Crane Capacity

An important limiting factor for the fuel handling inside the reactor hall was the available crane capacity. The crane inside the DKFZ reactor hall is licensed for nuclear loads with a maximum capacity of only 3 t and for non-nuclear handlings with a maximum capacity of only 5 t. Due to these limitations no handling of the two cask types inside the reactor hall was allowed with the crane, as the maximum weight of each of the casks (loaded, without impact limiters) is 13.2 t for the cask type GNS 16 and 6 t for the cask type TN 6/3. As a consequence, no loading of the casks inside the reactor pool was possible and a transfer system had to be designed for the fuel element transfer from the reactor pool into the casks.

Figure 2 and 3 schematically show the reactor hall with the reactor pool, the 3 positions of the casks in the truck entrance area and the crane.



Fig. 2 DKFZ Reactor Hall (Ground Floor)

Fig. 3 DKFZ Reactor Hall (Section A-A)

# 6. Transfer System

The transfer system designed for the fuel element transfer from the reactor pool into the casks was equipped with the following components:

- the <u>transfer flask</u> for the loading of a loading unit, filled with up to 4 F/A, inside the reactor pool and for the shielded transfer of the loading unit to the cask,
- the <u>mechanical grab</u> (with six elongation rods) as part of the transfer flask, especially fitted for the pintle of the loading units (grabbing and pulling the loading unit into the transfer flask),
- the loading lock, mounted onto the open cask, for shielding purposes during the loading, as the loading of the loading units is performed ,,dry",
- the <u>grid plate</u>, mounted onto the loading lock, for the defined positioning of the transfer flask and the specific lowering of a loading unit into a basket cavity and
- the <u>mobile water pool</u>, mounted onto the cask and filled with water after completion of the cask loading. The mobile water pool serves as shielding during the lowering of the primary lid onto the cask. After fixing of the primary lid the water is removed and the mobile water pool is removed from the cask.

Figure 4 and 5 schematically show the handling of the loading lock and the pulling out of the mobile water pool.



Fig. 4 Handling of Loading Lock

Fig. 5 Pulling out Loading Lock through mobile Water Pool

# 7. Loading Process

Due to the tight timeline of the loading situation, the possibility of first removing the five FFCR (absorber rods) from the reactor into the cask TN 6/3 was discussed in conversations with independent experts (TUEV) and the supervising authority as well as the companies involved. After intensive examination and calculations as to the criticality behavior of the reactor, it was agreed upon to accept the proposal of the DKFZ, i. e. to set a large part of the fuel rods into the storage shelves located on the side of the reactor pool, and to leave the remaining rods arranged in a ring shape on the outside of the reactor core. It was certified that the reactor was unambiguously and safely non-critical with a criticality factor of  $k_{eff} = 0.64$ .

# 8. Handling of F/A

In the following sections the main steps of the F/A handling are summarized (exemplary for the cask type GNS 16):

# 8.1 Acceptance of Unloaded Cask

- Removal of container roof and cask top impact limiter.
- Lifting out the cask with a mobile crane.
- Bringing in the cask and the equipment into the reactor hall via truck gate.

# 8.2 Cask Preparation

- Transport of the cask to the loading position and disassembly of protection plate and primary lid.
- Assembly of the loading lock onto the cask and function control of the loading lock.

# 8.3 Loading of Loading Units

- Loading of the F/A under water into the corresponding loading unit once immediately after each other. The F/A are manually handled with a F/A-grab from the reactor bridge by DKFZ-personnel.
- Closing of the loading unit with the filter plug. The filter plug is manually handled with a special grab from the reactor bridge by DKFZ-personnel.

# 8.4 Cask Loading

• Lowering the transfer flask into the water pool directly over the loaded loading unit.

- Pulling the loading unit into the transfer flask by means of the mechanical grab.
- Lifting the transfer flask over the water level of the reactor pool.
- Closing of the lower opening of the transfer flask by means of a special closure mechanism and performance of dose rate measurements at the transfer flask (see figure 6).
- Transport of the filled transfer flask by means of the hall crane to the loading lock on the cask .
- Coupling of the transfer flask to the loading lock, opening of the corresponding position of the grid plate and lowering the loading unit into the corresponding cavity of the basket by means of the mechanical grab.
- Pulling out the grab into the transfer flask, closing of the loading lock and lifting up the transfer flask (figure 7 shows the loading units inside the cask type GNS 16 after the cask loading).

The handling steps 8.2 to 8.4 are repeated until the cask is completely loaded.



Fig. 6 Dose Rate Measurement at Transfer Flask



Fig. 7 Loading Units inside the Cask GNS 16

# 8.5 Disassembly of the Loading Lock and Insertation of the Primary Lid

- Lifting of the mobile water pool by means of the reactor crane and assembly onto the cask.
- Filling of cask and mobile water pool with water.
- Lifting out the loading lock by means of the reactor crane (see figure 8)
- Insertion of the primary lid into the cask through the water by means of the reactor crane.
- Sucking out the water from the cask and the mobile water pool (see figure 9), disassembly of the mobile water pool and closing of the cask.



Fig. 8 Lifting out Loading Lock



Fig. 9 Water Sucking of Cask and mobile Water Pool

# 9. Conclusion

After loading of the casks, shipment of the casks from the DKFZ to the United States by road and sea. and supervising the unloading of the casks at Idaho Falls it can be concluded that the Consortium NCS/GNS has once again proven its operational experience, which will be useful for further worldwide shipments of spent nuclear fuel from research reactors, e.g. shipments from Europe, South America and Australia to the United States and France.



# U.S. SPENT NUCLEAR FUEL ACCEPTANCE POLICY 2006 AND BEYOND

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# ABSTRACT

In 1992, a large group of research reactor operators joined with Edlow International Company (EIC) forming the **Edlow Group** and promote the interests of members before the U.S. government and to enhance the negotiating leverage of each member. The goal was to convince the U.S. Department of Energy (DOE) to resume acceptance of foreign research reactor spent fuel after a three-year suspension. Approximately four years later in May of 1996, largely due to the efforts of the Edlow Group, DOE, in consultation with the Department of State (DOS), issued a new research reactor spent nuclear fuel acceptance policy. After five years of successful spent fuel shipments, new issues have surfaced which led of the Edlow Group to represent the interests of the foreign research reactor community before the DOE. This paper discusses the new spent research reactor fuel issues, and proposes utilizing a reactivated Edlow Group to address these concerns.

#### **INTRODUCTION**

Since the 1950's, as part of the "Atoms for Peace" program, the United States has provided research reactor technology and the reactor fuel necessary to fuel research reactors around the world. In the past, after irradiation in the research reactor, the used or "spent" fuel was returned to the United States, where it was reprocessed to extract the uranium still remaining in the spent fuel. The U.S. accepted foreign research reactor spent nuclear fuel until the program expired (1988 for HEU fuels, and 1992 for LEU fuels). At that time, the DOE suspended its acceptance of spent fuel primarily based on environmental concerns raised by Sierra Club and other groups pending a review of the environmental impacts of extending the program. Because of these concerns, the DOE decided to complete an Environmental Assessment (EA) pursuant to the National Environmental Policy Act (NEPA) and regulations implementing NEPA before issuing a renewal of the program.

In 1991, after nearly three years during which shipments were suspended under the program, the EA was finally completed and issued for public comment. A proposed Finding of No Significant Impact (FONSI) was published in the Federal Register. Since the public comment received by DOE in response to the EA and the proposed FONSI generally opposed the DOE findings, they decided to prepare a full-blown Environmental Impact Statement (EIS).
Suspension of the program forced many foreign research reactors to store their spent nuclear fuel inventories for a considerable period of time. This resulted in some reactor owners being faced with the prospect of substantially increased storage costs or the risk of shutting down operations altogether until their spent fuel could be shipped to the DOE.

In 1992, a large group of research reactor operators and other interested parties formed the "Edlow Group" to promote the interests of members of the group before the U.S. government and to enhance their negotiating leverage. This would also include litigation, if required, to convince the DOE to resume receipt of foreign research reactor spent fuel. The Edlow Group membership included research reactor operators representing 18 reactors from 15 countries. The goal of the Edlow Group was to obtain a resolution of the spent fuel problem in a manner that was cost effective, efficient, and responsive diplomatically to research reactor owners' urgent needs. To accomplish this, the following course of action was taken:

#### A. Executive Branch Action Plan

EIC and the Edlow Group established high-level contacts and meetings with the DOE and other government agencies to:

- 1) Present the views of research reactor owners to the highest level of the Executive Branch and to present legal and policy arguments directly to those officials best positioned to make favorable decisions.
- 2) To convince the U.S. that the balance of its legal and policy obligations rested with research reactor owners, and that circumstances permit and/or demand the prompt receipt of research reactor fuel.
- 3) To work with agency officials to develop and implement a solution to the problem.
- 4) To develop a mutually acceptable legislative strategy to solve the problem.

#### B. Mediation with Environmental Groups

After outlining the rights and remedies of the research reactor owners and exploring the nonproliferation aspects of the problem facing them, EIC, as the research reactor operator's designated representative, approached key environmental and political groups opposed to the shipment of spent research reactor fuel to the U.S. and attempted to work out agreements with them that would avoid litigation against the DOE for receiving the fuel.

C. Litigation

As a last resort, consideration was being given for one or more of the research reactor owners to attempt litigation against the DOE to compel the agency to receive their spent fuel. Such litigation was not pursued. However, the Edlow Group did prepare testimony and assisted the DOE in defending against litigation by other groups attempting to block acceptance of the research reactor spent fuel.

Approximately four years later, in May of 1996, largely due to the efforts of the Edlow Group, the DOE in consultation with the DOS, issued a new research reactor spent nuclear fuel acceptance policy and began receiving shipments of spent fuel.

After five years of successful spent fuel shipments from foreign research reactors under this program, new issues have surfaced which require the Edlow Group to be reactivated and again represent the interests of the foreign research reactor community: namely, extension of the return

policy beyond May of 2006, and expansion of the return policy to include other miscellaneous nuclear material containing uranium enriched in the U.S. EXTENSION OF THE POLICY BEYOND MAY, 2006

To further reduce the danger of nuclear weapons proliferation, in 1978 the U.S. initiated the RERTR program, which was aimed at reducing the use of HEU in civilian programs by promoting the conversion of research reactors from HEU fuel to LEU fuel. From the beginning of the RERTR program, foreign research reactor operators have made it clear that their willingness to convert their research reactors to LEU fuel was contingent upon the continued acceptance by the DOE of their spent nuclear fuel for disposition in the U.S. The current program for the return of research reactor spent nuclear fuel is based upon the Record of Decision (ROD) published by the DOE in May 1996.

The ROD describes the technical and legal details that must be met by the fuel and the operating organization returning the fuel, as well as the schedule requirements for the return of the fuel. To be eligible for return to the U.S., the spent fuel must be removed from the reactor by May of 2006. The Foreign Research Reactor Spent Nuclear Fuel Acceptance Program established with the issuance of the EIS in May 1996, restricts the take-back policy to the following types of reactor fuels:

- Aluminum-clad reactor fuels where the uranium-235 content is equal to or greater than 20 percent, by weight, of the total uranium content (i.e., HEU fuel). The active fuel region of these fuels may be configured as uranium-aluminum alloy, uranium-oxide, or uranium-aluminide. Spent nuclear fuels containing significant quantities of uranium-233 are excluded from receipt.
- 2) Aluminum-clad reactor fuels, where the uranium-235 content is less than 20 percent by weight of the total uranium content (i.e., LEU fuel). The active fuel regions of these fuels may be configured as uranium-silicide, uranium-aluminide, or uranium-oxide. Fuels containing significant quantities of uranium-233 are excluded from receipt.
- 3) Aluminum-, incoloy-, or stainless steel-clad, uranium-zirconium hydride (other than uranium-233) TRIGA fuel types.

Most of the HEU to LEU conversions under the RERTR program to date have been accomplished using silicide fuels. Since there is no operating facility for the reprocessing of silicide fuel, there is no alternative disposition option in many countries when the program draws to a close in 2006.

When the DOE initiated this program, the development of a new high-density reprocessable fuel that could be used for the conversion of all research reactors to LEU was planned. Although this development (U-MO fuel) has begun slowly and is moving slower than the earlier LEU fuel development program, it is currently underway. By May of 2006, the new U-Mo LEU fuel currently under development must replace all currently used silicide fuel in order to continue reactor operation with a fuel for which reprocessing is a disposition option.

Since the qualification for this fuel is anticipated somewhere between 2003 and 2005, the date by which the silicide fuel has to be replaced by the new fuel is far too short. Efforts must therefore be made to extend the timeframe for returning U.S. origin spent nuclear fuel beyond the May 2006 date. An extension of the date will allow the reactor operators to safely replace the current fuel without disrupting reactor operation during the development, licensing, and procurement phase of the new fuel.

During the period from the early 1960's continuing into the late 1970's, the U.S. encouraged other nations to buy and use various forms of radioactive materials. This included many forms of uranium (of varving assavs), thorium, plutonium, and other miscellaneous nuclear materials.

Typically these materials were used at research facilities. Of course, they were not consumed and residual amounts remain scattered around the world, both in developed and developing nations.

For the most part, these materials do not represent a proliferation risk. They are nonetheless hazardous materials including both radioactive and chemical hazards. Many of these materials do not meet standards for disposal within the host countries either because they have no capacity or their existing capacity cannot accommodate these materials.

Many reactor operators possess these miscellaneous nuclear materials containing uranium that was enriched in the U.S. There is currently no disposition option for these fuels since they were not included in the current spent nuclear fuel return program. The U.S. should expand the above acceptance criteria to repatriate these miscellaneous nuclear materials.

The first steps necessary to create this program include a review of the types and quantities of materials held around the world so that the DOE could determine its ability to receive and manage the various forms of nuclear material. It seems likely that whatever is held by foreign research reactors must also be similar to materials that the U.S. must also manage for itself. The incremental amounts are most likely small and could be easily accommodated within the existing disposal programs. After some initial exploratory discussions with several research reactors, EIC believes there is interest in the research reactor community to pursue this idea further. This service would be widely welcomed by those countries that have miscellaneous nuclear materials.

### **CONCLUSION**

We at Edlow International (EIC) believe that in order to achieve an extension of the return program of U.S. origin spent nuclear fuel and expand the existing take-back policy to include miscellaneous (orphaned) nuclear materials, we should progress along similar lines established by the current program. Our present success was due to the different research reactor operator's ability to coordinate their common interests within a group setting. Within the group, each member gave a detailed description of the reactor's situation and special needs. This information was compiled and coordinated by EIC, and the DOE was then apprised of the existing situation and made aware of the international interest in having corrective action taken. The predominant reason for the Edlow Group's success was due to the large number of research reactor operators who joined the group and shared their common interests. It is within this type of group setting that we are able to gain the voice and the required momentum to assure that our needs are not only recognized, but also met by the U.S. Government.

#### REFERENCE

[1] U.S. Department of Energy, Assistant Secretary for Environmental Management, "Final Environmental Impact Statement on a Proposed Nuclear Weapons Nonproliferation Policy Concerning Foreign Research Reactor Spent Nuclear Fuel," DOE/EIS-0218F, February of 1996.



Historical First Shipment To Savannah River Under The Renewed

U.S. Department of Energy Spent Fuel Policy Program



## MR-6 TYPE FUEL ELEMENTS COOLING IN NATURAL CONVECTION CONDITIONS AFTER THE REACTOR SHUT DOWN

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#### Abstract

Natural cooling conditions of the nuclear fuel in the channel type reactor after its shut down are commonly determined with relatively high uncertainty. This is not only to he lack of adequate measurements of thermal parameters i.e. the residual power generation, the coolant flow and temperatures, but also due to indeterminate model of convection mechanism. The numerical simulation of natural convection in multitube fuel assembly in the fuel channel leads to various convection modes including evidently chaotic behaviour.

To determine the real cooling conditions in the MARIA research reactor a series of experiments has been performed with fuel assembly equipped with a set of thermocouples. After some forced cooling period (the shortest was half an hour after the reactor shut down) the reactor was left with the only natural convection. Two completely different cooling modes have been observed.

The MARIA core consists of series of individual fuel channel and so called bypasses, maintaining the hydraulic properties of the fuel channel, connected in parallel. Initially, the convection cells were established trough few so-called bypasses providing a very effective mode of cooling. In this mode the flow charts were identical to those existing in forced cooling mode. After certain period the system switched on the second cooling mode with natural circulation within the individual fuel cells. Higher temperatures and temperature fluctuations were characteristic for this mode approaching 30 deg in amplitude.

In almost all the cases the system was switching few times between modes, but eventually remained in the second mode. The switching times were not regular and the process has a chaotic behaviour.

#### 1. Introduction

Following up the International Atomic Energy Agency recommendation on transition from HEU to LEU the Institute of Atomic Energy switches the operation of MARIA research reactor from 80% enrichment fuel into 36% U-235 enriched tube type fuel elements of MR-6 /36% type. The geometry of the new fuel elements is the same as the previous one but the contents of U-235 is  $(540 \pm 2)$  g instead of  $(350\pm2)$  g to be in the former elements. To retain the same outer dimensions of fuel tubes the thickness of aluminium cladding has been decreased from 0.8 mm to 0.61 mm (outer clad) and 0.641 mm (inner clad). In this connection new thermal-hydraulic calculations of the core were performed and some thermal hydraulic examination of an instrumented MR-6/36% fuel element was accomplished. During the experiments position of the fuel element examined was changed in the reactor core to provide various physical and thermal conditions. In the course of experiments after scramming the reactor two modes of cooling the fuel element under natural convection conditions were discovered. Both of them provided safe cooling of the fuel element even after disactivating all reactor cooling pumps at 0,5 h after reactor shutdown.

#### 2. Objective of experiments

Experiments were aimed to examine the thermal behaviour of the MR-6/36% fuel elements under various operational conditions [1] and on that basis to verity thermal hydraulic calculations in

the Safety Analysis Report. Due to decrease in enrichment and substantial increase in  $U^{235}$  contents in the fuel the thermal neutron flux densities are significantly lower. Changes in neutron fluxes and spectra in the fuel are additionally intensified by dramatic increase in  $U^{238}$  contents (over 10 times). Though lower the neutron flux densities are also deteriorated in beryllium and graphite in which most of irradiation processes took place. Recovery of thermal neutron fluxes in beryllium can be achieved only by increasing the fuel channel thermal power from the actual value of 1,6 MW to ca. 1,8 ÷ 1,9 MW. This might be done by proving that under transient conditions including natural convection cooling mode, the cladding "hot spot" temperature doesn't exceed 180°C and the maximum heat flux is less than 350 W/cm<sup>2</sup>. The calculated thermal parameters exhibited much lower values but for the Regulatory Body the theoretical values should be proven by experimental data [2].

To perform the experiment one of the MR-6/36% fuel channel was instrumented with nine chromel-alumel sheath type couples of 1 mm diameter.

The thermocouples measured coolant temperatures at the fuel part inlet (3 thermocouples with an angular spacing 120° in channel cross section) at the mixing cell (3 thermocouples) and at the fuel part outlet (3 thermocouples). Other parameters including water temperatures at the inlet header and channel outlet as well as reactor pool temperature and coolant flow in the fuel channel were recorded by a computerized system GTREMA.

Schematic of the fuel channel is depicted in Fig. 1. Attention has been focused on the fuel element behaviour after reactor shut down with accounting for the delay time of the circuits cooling pumps switching off after reactor scram.

An analysis of the initial experimental results showed that without violating the safety requirements after reactor scram the cooling time by means of forced convection can be reduced up to 0.5 h. Then the after-heat generated by fuel elements can reliably be removed by mechanism of natural convection. In the period from 30.04.1999 to 25.02.2000 eighteen experiments with various thermal hydraulic parameters in the instrumented fuel channel were performed.



Fig. 1. Schematic drawing of water flow in the fuel channel for two modes of natural convection

#### 2. Course of experiments.

In the course of experiments the range of parameters was as fol	lows:
reactor thermal power before scram	15-20 MW
fuel element thermal power before scram	0.92÷1.68 MW
fuel burn up in the instrumented channel	6.6÷152 MWd
cooling circuits pumps switching off time after reactor scram	4÷0.5 h

The major parameters to be recorded were coolant temperature at the inlet and outlet of the fuel part and in the coolant mixing cell at the lowest part of the fuel channel. Fig. 2 shows the general trend of coolant temperature changes in the channel after switching off the cooling pumps with 0,5 h delay time after reactor scram.

In Fig. 3 one can distinguish quite different modes of fuel element cooling under natural convection conditions.



Fig 2. Coolant temperature course in measurement of 19.11.99

#### 4. Two modes of natural convection in the fuel [2].

The study of the residual heat removal from MR-6/36% fuel at the absence of forced flow showed that the mechanism of natural convection in the tubular fuel geometry is more complicated than it was predicted in the analyses so far [3].

After disactivating of cooling circuits the water temperature oscillations are growing in several dozens second but then after a few damping oscillations they are getting established. Typical temperature run is shown in Fig. 3.

Relation between individual temperatures indicates that the water convection movement spreads from the inlet through the mixing cell and further to the channel outlet.

However, in several or dozens of hours since the circuits pump have been disengaged the system passes into another mode of natural convection. It is associated with water temperature increase in 20-30 deg and also much greater the temperature fluctuations at channel inlet and outlet are to be observed.

Temperatures at the inlet and outlet are definitely higher that the water temperature at the mixing cell which evidences that the two convection cells - inside and outside of the separation tube have been developed. Up to now it was impossible to identify the factors to be decisive for the transition from mode I to mode II. In all cases after switching off the pumps at first mode I is being activated and mode II comes at the final stage.



Fig. 3. Coolant temperature course in measurement of 10.12.99

One can assume that the channel basic thermal parameters are responsible for the transition onset, i.e. fuel element residual power and channel water temperature.

In general the transition periods from mode I to mode II cover a broad time range:  $1.3 \div 26h$  and the return times from mode II to I:  $1.4 \div 25h$ . Such a large time scatter with rather small fuel element power scatter evidences on a weak correlation between those times and fuel element after - heat.

Similar situation appears when correlating the water temperatures with the modes transition onset. There is also noticeable temperature scatter, i.e. 35÷45°C at the inlet and 45-60°C at the outlet.

Using the ORIGEN code the relation between residual power and cooling time for one of the experimental runs has been calculations (Fig. 4). It is a monotonous relations so it allows to substitute the time scaling with residual power scaling.



Fig. 4. Relation between water temperature and fuel element residual power

## 5. Conclusion

- Ten measurements results indicate that after cooling pumps switching off the convection mechanism defined as mode I is always being activated. The water temperatures are characterized by substantially lower values than the estimated temperatures in [3].
- The circulation direction of coolant in mode I has not been fully explained; it seems that bypasses are responsible for the reverse flow direction.
- To determine the impact factor for the transition phenomenon it is foreseen to conduct further study after installing measurement rigs over and under the reactor core.
- Due to the chaotic character of the process it will be necessary to conduct some experiments in out-of-pile stand to find out the nature of transition between the two modes.
- There will be an attempt to develop a new convection model for mode II and later evaluate it on experimental way.

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#### ENCAPSULATION TECHNOLOGY OF MR6 SPENT FUEL AND QUALITY ANALYSIS OF THE EK-10 AND WWR-SM SPENT FUEL STORED MORE THAN 30 YEARS IN WET CONDITIONS.

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#### Abstract.

The research reactor MARIA has been in operation for more than twenty years and all the spent fuel assemblies used since the first commissioning of the reactor are stored in wet facility on site. The present paper deals with the spent fuel MR-6 encapsulation technology in MARIA reactor.

The encapsulated spent MR-6 fuel will be stored under water in the same pool unless some other solution is available.

The capsules made of stainless steel are capable to accommodate one MR-6 fuel assembly. The encapsulation process is performed in the hot cell by the MARIA reactor.

The spent fuel having its leg cut off is loaded to the transport cylinder manually and next transferred to a trolley. The trolley is moving to a position directly below the entrance to the hot cell and the spent fuel is entering the hot cell. The spent fuel assembly is then put into the drying cell. Dried out spent fuel is moved into the capsule mounted on the grip of the machine. Next, the capsule lid is pressed in and welded. After the leak test and filling up with helium the capsule returns from the hot cell to the pool. The hermetic capsule is sunk back into the water and positioned in the separator.

The results presented earlier show, that the limiting time of WWR-SM and Ek-10 type spent fuel residence in wet storage is about 40 - 45 years. Therefore, the systematic quality investigation of all Ek-10 fuel elements and WWR-SM fuel assemblies discharged from EWA reactor in the period of 1959 - 1969 was performed. All together, about 2 500 Ek-10 fuel elements and 47 WWR-SM fuel assemblies were investigated. The results of these investigations are presented in the present work.

The sipping test, visual investigation and ultrasonic techniques were used for that purpose. The radioactive isotope Cs-137 was used as the indicator of fission product release from the fuel assembly. Taking into account the value of Cs-137 release from damaged WWR-SM fuel assembly the criteria of damaged fuel assembly were proposed. It was established, that part of analysed fuel assemblies is close to the state specified as damaged one.

The visual investigations of outer clad surface of WWR-SM fuel elements were combined with their ultrasonic scanning. The values of the depth of the pit corrosion holes was analysed.

#### 1 Encapsulation technology of MR-6 spent fuel.

#### 1.1. Introduction

During the operation of the MARIA research reactor 297 fuel subassemblies of the MR type were burned up. This number includes: MR-6 (80% U-235) - 266pcs., MR-5 – 18pcs. and 11 fuel elements of the MR-6 (36% U235). Beyond that there are 16 partly burned up fuel elements.

In connection to the anticipated construction of the dry storage facility for nuclear spent fuel which presumably will be located in the EWA reactor building the necessity occured to develop the technology for encapsulation of the spent fuel accumulated in the IAE at Świerk for its further storing.

It has been decided to encapsulate in the first order the MR-type fuel, to develop the technology relevant to this process and exploiting the MARIA reactor storage pool and the hot cell in which the handling operations with spent fuel will be accomplished.

#### 1.2. Main stages of encapsulation of the MR-type fuel

In the process of encapsulation of the MR-type spent fuel it is necessary to distinguish the two locations of operations to be accomplished with the fuel, i.e. the storage pool and the hot cell of MARIA reactor. The most important part of the MR-type spent fuel encapsulation process, namely the

drying out, leak-tight closure in capsules and checking their tightness, is carried out in the hot cell of MARIA reactor.

The hot cell is located at the end of the storage pool adjoining the external shell of the building. The cell has the following horizontal dimensions: length 6.6 m., width 2.5 m. The minimum height is 4.5 m. Internal walls and the floor are lined with stainless steel sheet of 1H18N9T. The cell shielded wall from the operational side and the side walls are made of the barytes concrete and the basalt concrete. Operational gamma radiation limit for the hot cell is 100 kCi – Co-60. The cell is equipped with two lead sight-glasses.

The basic equipment of the cell comprises the following devices: machine tool, transport device, overhead crane, lifting magnet, the two manipulators, bolting machine; ventilation, waste and decontamination system, radiological monitoring system, television camera and five culverts. For the encapsulation needs of the MR-type spent fuel, the cell has been additionally equipped with drying chamber and vacuum chamber located in the sockets on the cell floor and chucks fixed on the face, milling cutter to be used for: welding head, helium probe, milling cutter for possible cutting off the lid.

The peripheral equipment is placed on the platform in front of the cell and both the crane and lifting magnet are operated from this platform.

The spent fuel assemblies are located in the storage pool. Before removing the spent fuel element from the sheath the spent fuel element is cut off from the channel construction and the water contained inside is monitored.

If radiological measurements results exhibit a failure (leakage) of cladding of the fuel element the element is being treated by an individual procedure. In most cases the fuel channel with leaking fuel will be separately cut off in water-lock. Then it will be closed in a leak-tight thimble in which water is filtered.

To displace safely the fuel element into the hot cell, it is necessary to install on it a gripping head accommodated to the lifting magnet, which belongs to one of the cell accessories. This operation is accomplished in the storage pool, below the water layer. The fuel element equipped in such a manner will be brought to the transport sleeve, which is placed in a special rack on the floor of the storage pool, and then along with the transport sleeve on the carriage belonging to the transport equipment.

After approaching the spot right below the inlet hatch of the dismantling cell, the spent fuel element will be pulled in to the transport sleeve, dried out and prepared to be placed inside the dry storage facility. To reach this goal it is necessary to apply the technology of the encapsulation process that is shortly described below.

In compliance to the developed technology, the spent fuel element is transferred to the drying chamber. The drying chamber is connected to the outlet of the hot cell ventilation system by means of a flexible duct. The head is equipped with an absolute filter aimed to capture the possible loose solid parts, which could have broken away from the cladding of the dried out fuel element. The head is located at the ventilation system inlet. After placing the spent fuel element in the drying chamber it is closed and the drying system is activated. The electric air- heater with temperature regulation within the range of  $50 \div 110^{\circ}$ C at the inlet to the culvert is located at the outlet of this system. The heated air is delivered over the bottom of the drying chamber by means of a steel pipe and when passing at the direction of the drying chamber head it washes the fuel element surface to dry it out. The absolute filter is located at the head outlet. After passing through the filter, the air is sucked into the hot cell ventilation system. The drying process is conducted until the air relative humidity at the outlet of the system is around 5%. On achieving an appropriate relative humidity degree, the dried out fuel element is ready to be closed in the capsule.

The most important moment of the encapsulation operation is the leak-tight closing of the dried out spent fuel element in the capsule. The capsule and its cover have been earlier manufactured of certified materials and they are verified as regards to the welding tightness. The dried out fuel element is being transferred into the capsule thimble. The operation of loading the spent fuel element into the capsule is being recorded on video cassette by means of a film camera to identify the number on the capsule and the number of fuel element. In the next operation the welding head is positioned onto the welding spot and rotation of the chuck and the welding machine TIG are activated. Having finished the full process of welding the checking of the tightness of the capsule with the fuel element closed inside is performed by the operator. The check of welding tightness on the capsule perimeter is being carried out by helium method by means of the universal helium leak detector of the ASM-type.

The first operation to be done is creating the vacuum inside the capsule. At the next step the capsule is being connected to the helium leak detector. Later, the probe supplying helium is being installed over the examined girth weld. On activating the detector, the weld tightness on the entire perimeter by means of the method of washing its surface with helium is being examined. If the threshold limit of the signal of the detector to be  $10^{-7}$  Pa m<sup>3</sup>/s is surpassed, the operator determines the leaking spot of the weld and the welding operation is repeated. If the capsule is not leaking, the capsule can be filled with helium to the value of pressure of 0.2 MPa.

Next, operation of mounting the gripping head on the capsule is started. It is the same like described below during the leak-tight closing of the capsule.

After the gripping head has been mounted, the capsule is transported to the vacuum chamber. A sucking in head is being installed on the chamber and the helium leak detector, connected to it, is activated. When the absolute value of the overall leakage rate of the capsule has been measured and recorded the leak-tight capsule is removed from the vacuum chamber to be ready for transportation to the storage pool.

The capsule containing a spent fuel element is shifted by means of the lifting magnet over the inlet hatch of the hot cell. Then it is let down into the transport sleeve. The sleeve is being transported from below the hot cell using the carriage of the device P-109 to the storage pool. By means of the crane, the capsule along with transport sleeve is transferred to the rack. The last operation is based on taking out, by means of grapple fork, the capsule from the sleeve and shifting it to the separator for the capsules that is located in the storage pool of the MARIA reactor.

The review of the developed encapsulation technology of the spent fuel elements has confirmed its full compatibility with the required safety and strength of the elaborated procedure relevant to store the spent fuel in the dry storage facility for at least 50 years period and more. The presented technology refers only to the spent fuel elements with undamaged cladding. By implementing some modifications this technology can also be applied to the leaking spent fuel elements. According to our assessment the presented technology seems to be cheaper than the American technology based on the combined vacuum and heating drying out process.

To implement the verified encapsulation technology of the MR-type fuel the encapsulation of the first three MR-type fuel elements was accomplished in MARIA reactor in June 2001.



Fig.1. Leak-tight capsule after the operation of mounting the gripping head in the hot cell.

# 2. Quality analysis of the Ek-10 and WWR-SM spent fuel stored more 30 years in wet conditions.

As the result of examination of the status of the spent fuel stored at Institute of Atomic Energy the time limits of its storage in wet conditions were estimated [1]. For the Ek-10 fuel rods, the limit time was 46 years, for WWR-SM assemblies - 41 years, for WWR-M2 assemblies - 36 years and MR-6 assemblies - 36 years. These limits should be treated as approximate indicators. The corrosion processes is varying in a wide range due to differences in conditions of storage, manufacture,

operation and material. Therefore, the state sponsored program of inspection of the fuel rods and assemblies was executed. The program required, that the quality inspection will be performed for all fuel elements stored in wet condition for more than 30 years. The analyses covered fuel elements which were discharged from EWA reactor in the period from 1958 to 1969 i.e. the all Ek-10 fuel elements and WWR-SM fuel assemblies discharged from EWA reactor in the period of  $1967 \div 1969$ .

The status of separate fuel elements was examined using the sipping test of Cs-137 isotope from investigated element. The Cs-137 isotope was chosen as a marker of fission product release from the fuel element tested. Great efficiency of its production in the fission processes, long decay time and easily detected decay radiation are the reasons for accepting this isotope as a marker.

During storage time in a water pool the aluminium clad fails due to degradation in the processes of corrosion. The effective thickness of clad material was decreasing with storage time, and consequently, the release of fission products from fuel - among them Cs-137 isotope - was increasing. For the storage time close to limit time, the value of Cs-137 release grows up rapidly. In [1], the relation between storage time and the rate of Cs-137 release was shown. On the basis of such relation, the remaining time for safe storage in water, for the analysed fuel elements was determined.

The tested fuel element was placed into a container filled with fresh water. After a specified time - usually after 24 hours - the radioactivity of water sample from container was measured by NaJ(Tl) scintillation gamma spectrometer. The daily activity of Cs-137 isotope in the sample is chosen as the magnitude characterising the state of fuel element. In the case of Ek-10 type, the sipping procedure was applied for fuel rods stored in the storage container (from 43 to 48 fuel rods in container).

The results of measurements presented as the number of assemblies or containers in a function of daily Cs-137 activity release are presented in Table 1 - for WWR-SM fuel assemblies and in Table 2 for containers with Ek-10 fuel rods. It is obvious, that three WWR-SM assemblies with Cs-137 activity daily release greater than 200 Bq should be removed from the water, placed in dry conditions and put in dry containers. The three containers with Ek-10 fuel rods with daily release greater then 200 Bq also should be removed from water and put into an inert gas.

ruble 1. Results of 11 million rue		
Daily release of Cs-137 (Bq)	No of fuel assemblies	
0 - 20	8	
20 - 40	16	
40 - 60	11	
60 - 80	1	
80 - 100	2	
100 - 120	3	
120 - 140	1	
140 - 160	1	
160 - 180	1	
260	1	
420	1	
912	1	

Table	1. Results of	f WWR-SM fuel	sipping test
ruore	T. Results of	i ii ii ii onii idei	orpping cost

Table 2. Results of Ek-10 fuel sipping te				
Day release of Cs-137 (Bq)	No of containers			
20 - 40	9			
40 - 60	15			
60 - 80	8			
80 - 100	9			
100 -120	2			
120 - 140	4			
140 - 160	2			
160 - 180	3			
180 - 200	0			
200 - 220	1			
220 - 240	1			
240 - 260	0			
260 - 280	1			
280 - 300	1			
1770	1			

Using ultrasonic equipment - its principle is presented in fig 2 - the underwater surface scanning of spent fuel was performed. The result of pit corrosion hole scan is presented in fig 3



Fig. 2 The principle of ultrasonic equipment use for scanning spent fuel clad surface



Fig 3. The result of WWR-SM fuel clad surface scan of pit corrosion hole (above - the picture of the fuel clad surface).

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## **STUDY ON HANARO CORE CONVERSION USING U-MO FUEL**

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## ABSTRACT

Two types of fuel rods with different fuel meat diameter and uranium density are considered for HANARO core conversion with high density U-Mo fuel. Arranging standard fuels of 5.0 gU/cc and 6.35 mm in diameter at the inner ring of an assembly and reduced fuels of 4.3 gU/cc and 5.49 mm in diameter at the outer ring of an assembly flattens the assembly power distribution and avoids the increase of linear heat generation rate due to using higher uranium density and less number of fuel rods. The maximum linear heat generation rate is similar with the current reference core and four fuel sites at the outer core in the reflector tank is converted to the irradiation sites to suit more demand on fuel tests and radioisotope production at outer core sites. This new core has 32% longer fuel cycle than the current reference core.

#### 1. Introduction

HANARO has thirty-two fuel channels and seven irradiation sites in the core. The inner core is cooled and moderated by light water and has 20 hexagonal fuel channels, 8 circular fuel channels, and 3 hexagonal irradiation sites. The outer core, located in the reflector tank, is cooled by light water but moderated by heavy water and has 4 circular fuel channels and 4 irradiation sites. Outer core irradiation sites are appropriate for fuel irradiation tests or radioisotope production. High power fuel irradiation tests are carried out in the inner core irradiation sites. HANARO driver fuel is 19.75 w/o 3.15 gU/cc  $U_3Si$  in rod type.

High density atomized U-Mo fuel is considered to replace current driver fuels. There are a few merits that can be obtained by replacing current silicide fuel with high density U-Mo fuel. Four fuel channels in the outer core can be used as irradiation sites. These sites are very much in demand and sometimes a high burnup fuel test continues for up to five years. Secondly, the increase of uranium loading per fuel assembly extends the fuel cycle and reduces the number of spent fuels discharged per year. It may be possible that current spent fuel storage pool is used through the lifetime of HANARO without transporting spent fuels to the interim storage site. Lastly, the fact that U-Mo is reprocessable allows another backend option to HANARO.

HANARO started its operation in 1995 and does not have to confront ageing problem yet. Hence the core conversion that can be considered in the near future is the one without major system modification. Only the fuel change is under consideration even without shape change. Any change affected by this fuel change should be minimized. Two major concerns are to ensure the thermal margin and the endurance of the fuel. Thermal margin can be ensured either by upgrading cooling system or by achieving uniform power distribution within a fuel assembly. A marginal reduction of the existing margin could be accepted. Operation experience and more accurate and proven analysis method would be helpful. Upgrading cooling system will be excluded to minimize the effect of fuel change. Another concern is the

soundness of the fuel integrity against the flow-induced vibration under the condition of prolonged fuel cycle and heavier fuel assembly.

#### 2. Preliminary study

The parameters that can be considered in fuel change are mass fraction of molybdenum, fuel density, fuel meat diameter, uranium loading, excess reactivity, linear heat generation rate, and mass fraction of poison. Fuel irradiation performance is confirmed through fuel qualification test. New fuel specification should be selected by analysis to fulfil a maximum linear heat generation rate ensuring the minimum thermal margin and neutron flux required by users at various locations.

It is assumed that a new core has 28 fuel channels. All of 8 channels in the outer core, called OR, are irradiation sites. U-9Mo is selected in this study since more stable fuel is preferable than the slight handicap of parasitic absorption of Mo. Fuel meat density and diameter determines the uranium loading and directly affects linear heat generation rate (LHGR). The uranium loading in the new core is 32% more than the existing 32 fuel assembly core. The thermal conductivity is measured along the fuel volume fraction and temperature. As the fuel volume fraction increases, thermal conductivity decreases very much. In manufacturing fuel rods, more unacceptable fuel rods are produced when the diameter of the fuel rod becomes lower than 5.49 mm. With a given fuel density, larger fuel meat diameter produces higher linear heat generation rate.

MCNP [1], VENTURE [2], and HELIOS [3] are used for this analysis with fresh fuels at 30 MW. The reference core and a new core with control rods at the middle of the core height are compared in Table 1. In the reference core, all fuel meat has the same density while the reduced fuels, which are located in the outer ring of hexagonal fuel assembly, have 5.49 mm diameter and the standard fuels have 6.35 mm. The reduced fuels are introduced to reduce the power peaking within a fuel assembly. The reduced fuels in the new core, however, has 4.3 gU/cc and 5.49 mm diameter and are located in the outer ring of both hexagonal and circular fuel assemblies. The standard fuel has 5.0 gU/cc and 6.35 mm diameter. When four OR fuel channels are replaced with dummy fuels, the average LHGR is basically increased by 8.3%.

The LHGR ratio of outer ring average to inner ring average is, however, greatly reduced for circular assemblies by more than 20%. In MCNP calculation, the excess reactivity is increased by only 1.9 mk although uranium is loaded 32% more. This is because the neutron loss becomes larger due to the reduction of the core size excluding 4 fuel sites and the parasitic absorption of Mo. Another reason is the increased self-shielding due to higher uranium density. The control rod worth of the new core is 105.0 mk that is 5.1% smaller than that of the reference core.

The cross section library for VENTURE is provided by WIMS [4]. VENTURE is underestimating LHGR compared to that from MCNP by 1.5% in average and 7.0% in maximum. The excess reactivity of the new core is even decreased by 1.8 mk.

HELIOS is a two-dimensional transport code and two core states such as all rod out (ARO) and all rod in (ARI) are analysed. LHGR shown in Table 1 is for ARO and the average LHGR's are underestimated within 4% except CAR site compared with those from MCNP.  $k_{eff}$  is the average of ARO and ARI. When the maximum LHGR's of HELIOS are compared with those of MCNP and VENTURE, the axial peaking factors deduced from MCNP and VENTURE are 1.61 and 1.50, respectively. The control rod worth in the new core is 105.1 mk that is 5.7% less than that of reference core. The excess reactivity of the new core is increased by only 0.5 mk.

In MCNP calculation, the thermal-to-total flux in the fuel region is decreased by a few percent from the reference core to the new core. This hardening effect is also shown in assembly burnup calculation using HELIOS but the extent of hardening does not change much in burnup. While the uranium density increases, the spectrum becomes harder due to self-shielding effect.

When the fuels at the inner ring of the hexagonal fuel assemblies increase their density from 5.0 gU/cc to 5.5 gU/cc, the improvements in LHGR and LHGR ratio are only a few percent.

Core	Fuel specification	Uranium	Code	LHGR	LHGR ratio	k <sub>eff</sub>
	(enrichment,	Loading		max/avg	(outer/inner	CAR worth
	density, diameter)	(kg)		(kW/m)**	ring)***	(mk)
Reference	19.75 w/o U <sub>3</sub> Si	58.92	MCNP	88.2/40.6	1.03	1.17499
core	3.15gU/cc			110.9/37.1	1.17	
32 fuel	6.35/5.49 mm*			108.1/54.3	1.24	110.7
assemblies				83.3/48.7	1.36	
			VENTURE	80.6/40.8	1.00	1.16642
				98.8/36.8	1.17	
				98.7/53.2	1.21	119.7
				78.5/47.5	1.29	
			HELIOS	55.9/39.5	1.03	1.18477
				64.6/50.5	1.22	
				64.1/51.2	1.22	111.4
				57.5/48.5	1.30	
New core	19.75 w/o U-9Mo	77.72	MCNP	103.3/45.2	1.02	1.17762
28 fuel	5.0/4.3 gU/cc*			107.2/38.5	0.89	
assemblies	6.35/5.49 mm*			106.4/55.3	0.95	105.0
			VENTURE	96.1/45.4	0.95	1.16397
				101.6/37.5	0.85	
				102.0/53.7	0.88	114.2
			HELIOS	66.3/44.2	1.02	1.18544
				65.3/51.7	0.93	
				64.9/52.3	0.94	105.1

Table 1. Comparison of reference and new core with fresh fuels at 30 MW

\* standard/reduced

\*\* R: hexagonal fuel assembly (36 rods),

CAR: circular fuel assembly (18 rods) in the control absorber rod (shroud)

SOR: circular fuel assembly (18 rods) in the shut-off rod (shroud)

OR: circular fuel assembly (18 rods) in the outer core

\*\*\* Hexagonal fuel assembly has 3 rings. Two inner rings have 6 and 12 fuel rods and an outer ring has 18 fuel rods. Circular fuel assembly has 2 rings. An inner ring has 6 fuel rods and an outer ring has 12 fuel rods.

## 3. Burnup analysis

The burnup analyses for the reference and new cores without any fuel shuffling are conducted using HELIOS. Assuming the discharge burnup of 55%U235 as HANARO, the irradiation day is 192.9 days and  $k_{eff}$  is 0.8891 at 90 GWD/tHM (54.04 %U235) in the reference core. For the new core, the irradiation day is 254.4 days and  $k_{eff}$  is 0.8894 at 90 GWD/tHM (53.79 %U235). The irradiation length of new core is 32% longer as uranium loading is 32% more. When the uranium enrichment is different, the same U235 loading does not mean the same cycle length due to the different quantities of U238 [5]. For reference and new cores,  $k_{eff}$ 's and their slopes are about same as up to 55%U235 burnup.

## 4. Conclusions and further study

The feasibility of core conversion with high density U-Mo fuel is analysed. In addition to the advantages of high density U-Mo such as longer fuel cycle, less number of spent fuel produced per year and reprocessability, smaller core size is pursued to have more irradiation sites. It is assumed that the reactor cooling system should not be modified to hold this core conversion. This means that the maximum linear heat generation rate should be maintained as it is in the current core. The reduced fuels with smaller fuel meat diameter and lower uranium density at the outer ring of the fuel assembly are proposed in order to reduce the assembly power peaking. This new core has 32% more uranium with the

same enrichment of 19.75w/o and four more irradiation sites than the current core. The maximum linear heat generation rate is about same as the current reference core while the linear heat generation rate ratio of outer to inner ring fuel is greatly reduced making an assembly power distribution more uniform. The characteristics of the new core will be analysed more for flux level, spectrum and reactivity coefficients. The transition and equilibrium cores will be searched and the characteristics of the equilibrium core will be analysed.

#### Acknowledgements

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## STUDY ON POSSIBLY USAGE OF 36% OR 20% ENRICHED UO<sub>2</sub> TWR-S FUEL TYPE AT RB HEAVY WATER CRITICAL ASSEMBLY

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## ABSTRACT

In view of possible including of Yugoslavia to the RERTR program, an idea for possibly replacement of current HEU (80%) fuel at RB heavy water critical assembly with a similar Russian TWR-S fuel type of lower enrichment (36% or 20%) is studied. Initial criticality analyses, carried out in this paper, are the first ones related to study of proposed fuel replacement. These criticality analyses are done for selected HEU benchmark cores of the RB reactor. Current characteristics of heavy water of the RB reactor and available material data for Russian 36% or 20% enriched TWR-S uranium fuel of oxide type, dispersed in aluminium matrix, are used. Initial results, obtained by using the Monte Carlo based reference code MCNP for criticality safety, confirm possibility of RB reactor operation with proposed LEU fuel.

### 1. Introduction

RB research reactor is heavy water critical assembly operated by Nuclear Engineering Laboratory of the "Vinča" Institute of Nuclear Sciences, since 1958 [1]. The reactor uses metal natural uranium, 2%-enriched metal uranium (LEU) and 80%-enriched UO<sub>2</sub> (HEU) fuel dispersed in aluminium matrix. Natural uranium fuel is rod type (diameter 2.5 cm, length 210 cm, 1 mm Al cladding). The LEU and HEU fuel elements are Russian TWR-S type, bought in ex-USSR in 1959 and 1977/1982, respectively. These elements are slug type, 11.3 cm total length, total diameter 3.7 cm; annular fuel layer has outer/inner diameter 3.5/3.1 cm and 10.0 cm total length, covered by inner and outer side with 1 mm aluminium cladding.

Different lattices, with regular basic square lattice pitch from 7 cm, 8 cm, 9 cm, 12, cm, 13 cm, and their various multiplies, can be formed within bare aluminium tank (diameter 200 cm, height 230 cm) filled by heavy water. More than 600 different reactor core configurations are examined by end of 2001, using all three types of fuel elements, including several coupled fast-thermal cores [2].  $D_2O$  moderator, used in the same time as core reflector and fuel coolant, is not isolated from the air during reactor operation. Due to that circumstance, the contents of light water in heavy water was increasing during past years, as consequence of absorption of moisture from the air. This 'degradation' of heavy water isotopic composition has an approximate average rate of 0.05% molar per year and influences criticality parameters of the reactor. Current isotopic composition of heavy water of the RB reactor is 97.24  $\pm$  0.01 molar percent, determined in August 2001.

## 2. Nuclide composition of the HEU and LEU fuel of TWR-S type

Three-dimensional cross section view of the TWR-S fuel type is given in Figure 1. According to available fuel manufacturer data [3], it is assumed that uranium dioxide is dispersed in aluminium (PA-2 type, [4]) matrix within fuel layer. The fuel layer has annular shape with ID/OD 3.1/3.5 cm and total length of 10.0 cm. Nominal mass of nuclide <sup>235</sup>U in each fuel slug is 7.7 g  $\pm$  0.35 g for all cases of uranium enrichment (20%, 36%). Certificate mass of 7.67 g of nuclide <sup>235</sup>U in HEU (80%) fuel, utilised at RB reactor, is used in calculations. Impurities in UO<sub>2</sub>, are not known, and were not

included in the calculations. Ratio of U to O in uranium dioxide is assumed exactly 1:2. A content of nuclide <sup>234</sup>U in the fuel material is not reported by manufacturer. It is estimated by calculations under assumption given in ref. [5]. Nuclide <sup>236</sup>U in the fuel is neglected under assumption that uranium used as fuel material was obtained by enrichment process in the factory that has used only natural uranium at inputs of cascades, but not the uranium obtained from reprocessed fuel. Also, fuel layer cladding (1 mm thick at both sides of fuel layer), bottom and top stars of the slug and slug expeller are made from AMSN2 aluminium alloy [4]. Nuclide composition of the fuel layer, used in the calculations, is shown in Table 1.



Table 1. Calculated nuclide composition  $(10^{24} \text{ cm}^{-3})$  of TWR-S type UO<sub>2</sub> in PA-4 fuel layer for various <sup>235</sup>U enrichment

Nuclide	U(80%)O <sub>2</sub>	U(36%)O <sub>2</sub>	U(20%)O <sub>2</sub>
	in PA-4	in PA-4	in PA-4
0	$2.3591 \cdot 10^{-3}$	5.2436·10 <sup>-3</sup>	9.4189·10 <sup>-3</sup>
Al	4.9948·10 <sup>-2</sup>	5.3915·10 <sup>-2</sup>	3.3181·10 <sup>-2</sup>
Si	1.9343.10-4	2.0879.10-4	1.2850.10-4
Fe	8.5116·10 <sup>-5</sup>	9.1876·10 <sup>-5</sup>	5.6544·10 <sup>-5</sup>
Cu	4.2745.10-6	4.6140.10-6	2.8396.10-6
<sup>234</sup> U	1.0262.10-5	1.0086.10-5	9.6797·10 <sup>-6</sup>
<sup>235</sup> U	9.4510·10 <sup>-4</sup>	9.5147·10 <sup>-4</sup>	9.5147·10 <sup>-4</sup>
<sup>238</sup> U	2.2420.10-4	1.6602.10-3	3.7483·10 <sup>-3</sup>
Slug mass [g]	162	175	175

Figure 1. TWR-S fuel slug

## 3. Reactor RB HEU fuel benchmark cores

Basic characteristics of the HEU fuel benchmark cores of the RB reactor [5] are given in Table 2. Few assumptions, compared to data of the benchmark cores given in ref. [5], are made in model and data used in calculations carried out in this study. The SAV-1 alloy of fuel slug cladding is replaced by AMSN2 aluminium [4]. The assumed SAV-1 matrix of the fuel layer is replaced by PA-4 aluminium. Heavy water with 2.76 molar percent of light water is used for moderator. Temperatures of fuel and moderator are equal to 20 °C in all cases. Influence of part of fuel elements in air, above heavy water level, at criticality is included in calculations, as well.

Table 2. Criticality data for benchmark cores with HEU fuel of the RB reactor [5]

Case no.	Core no. /Year	Lattice pitch [cm]	No. fuel elements in the core & (no. slugs inside)	D <sub>2</sub> O isotopic composition [molar %]	Critical level [cm]	T (D <sub>2</sub> O) [°C]
1	32/1977	13	37 (12)	99.43 ± 0.05	$94.46 \pm 0.01$	$23.0 \pm 0.1$
2	95/1999	7	32 (13)	$97.60 \pm 0.05$	$104.14 \pm 0.01$	$14.0 \pm 0.5$
3	96/1999	7√2	24 (13)	$97.60 \pm 0.05$	$120.00 \pm 0.01$	$14.0 \pm 0.5$
4	97/1999	14	32 (13)	$97.60 \pm 0.05$	$114.15 \pm 0.01$	$14.0 \pm 0.5$
5	98/1999	16	32 (13)	$97.60 \pm 0.05$	131.87 ± 0.01	$13.0 \pm 0.5$
6	99/1999	8√2	24 (13)	97.60 ± 0.05	$123.63 \pm 0.01$	$14.0 \pm 0.5$
7	100/199	8	24 (13)	$97.60 \pm 0.05$	$125.36 \pm 0.01$	$13.0 \pm 0.5$
8	101/199	$12\sqrt{2}$	32 (13)	97.60 ± 0.05	$145.66 \pm 0.01$	$13.0 \pm 0.5$
9	102/199	12	32 (13)	$97.60\pm0.05$	$103.38 \pm 0.01$	$13.0 \pm 0.5$

## 4. Results of calculation and discussion

The criticality studies are carried out using the Monte Carlo based reference, well known, code for criticality safety, MCNP version 4B2 [6], with continuous energy neutron data library based on ENDF/B-VI data. This code is verified in many applications, including RB benchmark cores with natural uranium, 2% enriched and 80% enriched uranium fuel, published in the OECD/NSC Handbook of International Criticality Safety Evaluated Experiments, editions 2000 and 2001. The 3D model of the fuel slug described in ref. [5] is applied. Calculations are carried out for 750 cycles with 1000 neutron histories each, after 25 initial cycles run to determine neutron initial space distribution. Initial results for calculated heavy water critical level are obtained and shown in Table 3. These results confirm possibility of RB reactor operation with TWR-S fuel based on 36% or 20% enriched UO<sub>2</sub> dispersed in aluminium. Number of fuel elements was not changed in the core during criticality calculations, i.e., the core radius was assumed constant. The critical height of heavy water is estimated, based on  $k_{eff}$  calculations for at least two different D<sub>2</sub>O levels, supposing that necessary number of fuel slugs (10-15 in all cases) within the fuel elements was available and assuming constant gradient of reactivity between the two water levels.

Case no.	Heavy Water Critical Height $H_c \pm 1\sigma$ [cm]			
	U(80%)O <sub>2</sub> in PA-4	U(36%)O <sub>2</sub> in PA-4	U(20%)O <sub>2</sub> in PA-4	
1	$107.86 \pm 2.98$	$116.41 \pm 3.04$	$120.69 \pm 3.08$	
2	$127.95 \pm 7.96$	$136.28 \pm 8.33$	$141.05 \pm 8.71$	
3	$102.25 \pm 2.46$	$105.84 \pm 2.56$	$107.73 \pm 2.51$	
4	$104.77 \pm 1.75$	$107.07 \pm 1.75$	$109.01 \pm 1.75$	
5	$116.16 \pm 3.40$	$119.08 \pm 3.45$	$119.60 \pm 3.74$	
6	$135.01 \pm 9.38$	$138.06 \pm 10.17$	$138.99 \pm 10.87$	
7	$121.52 \pm 5.75$	$127.65 \pm 5.83$	$131.18 \pm 6.11$	
8	$125.08 \pm 6.06$	$130.08 \pm 6.39$	$132.76 \pm 7.16$	
9	$146.14 \pm 9.59$	$152.89 \pm 9.25$	$149.74 \pm 12.34$	

 Table 3. Heavy water critical levels for benchmark cores with new RB reactor fuel calculated by the MCNP code

Some of the calculated critical heights of heavy water show very high uncertainty ( $1\sigma$  error). It could not be explained at this initial level of calculations. Verification of experimental and code input data, and better statistical error (more neutron histories) could help to understand these discrepancies.

## 5. Conclusion

Initial results for calculated heavy water critical levels for new 36% and 20% enriched UO<sub>2</sub> fuel, used instead HEU fuel in the 'HEU benchmark cores' of the RB reactor, are obtained by MCNP code. They confirm possibility of RB reactor operation with TWR-S fuel based on 36% or 20% enriched  $UO_2$  dispersed in aluminium and current content of light water (2.76% molar) in heavy water moderator and/or reflector.

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## POWDER PRODUCTION OF U-Mo ALLOY, HMD PROCESS. (Hydriding-Milling-Dehydriding)

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## ABSTRACT

Uranium-molybdenum (U-Mo) alloys can be hydrided massively in metastable  $\gamma$  (gamma) phase. The brittle hydride can be milled and dehydrided to aquire the desired size distributions needed for dispersion nuclear fuels. The developments of the different steps of this process called hydriding-milling-dehydriding (HMD Process) are described. Powder production scales for industrial fabrication is easily achieved with conventional equipment, small man-power and low investment.

## **1. Introduction**

Powder metallurgy technology offers different alternatives for obtaining uranium-molybdenum (U-Mo) alloy particles in the size range (< 150  $\mu$ ) needed as raw material for the fabrication of material testing reactors (MTR) nuclear fuel elements. Since as cast U-Mo is a ductile alloy, conventional milling alternatives used with brittle materials are forbidden.

Centrifugal or gas atomization are the two more generalized high yield methods for metallic powder production [1]. The major drawback of these alternatives is that since particles solidify in an inert gas atmosphere flight, big chambers are needed. High initial equipment investment is required for a process where small batches have to be used, limited to a few pounds because of safety conditions required for the management of an alloy with enriched uranium. A cyclonic centrifugal atomization (CCA) process that curves the cooling trajectory of particles, would allow the use of smaller chambers.

Hand and lathe filing, and wheel grinding, have been considered as possible processes. These alternatives are usually associated with additional treatments of classification, purification from introduced debris or rounding of filings by milling and scrap reprocessing. The recently developed process in which  $\gamma$  phase is decomposed in two phases, one of which is  $\alpha$  uranium that when hydrided comminutes the alloy [2, 3], needs high control of thermo-mechanical treatments for obtaining desired particle size distribution.

The recent discovery of the massive hydriding [4] of uranium-molybdenum alloys in the  $\gamma$  phase structure allows to obtain a brittle interstitial compound that can be readily milled. This is the starting point for the hydriding-milling-dehydriding -HMD- process of powder production described in this work.

## 2. Hydriding

Hydriding is a well known process of comminuting materials that form brittle hydrides. Evidence of localized hydride formation in U-Mo was reported in cathodic charge of hydrogen in a uranium 10% weight molybdenum alloy (U-10Mo) [5]. Also it is known that this alloy has low hydrogen [7] solubility that for values over 5 ppm slightly embrittles the material [6]. A massive hydriding of U-Mo

alloy in metastable  $\gamma$  body centered cubic (bcc) phase can be obtained in samples where hydrogen has been solubilized previously.

The equipment used consisted in a vacuum chamber -evacuated with mechanical and diffusion pumpswith a total volume of 6.85 liters (V) with a quartz tube inside a resistance furnace. The heating zone is of approximately 2 liters and a needle valve allows the incorporation of hydrogen or argon gases. Batches of more than one kilogram can be processed measuring temperature and pressure. Testing was performed fundamentally using U-7Mo alloy. The alloy was melted in an inert gas induction furnace using high purity molybdenum and natural uranium of different purities. Plates of 5 mm thickness were casted in graphite molds.

Fragments of the alloy are introduced in the quartz tube and heated during one hour in pure hydrogen (H<sub>2</sub>) at one atmosphere pressure and 700 °C. Absorption of hydrogen is usually greater than 10 ppm in weight. Sample gas absorption can be detected in a closed chamber by the evolution of pressure and temperature while cooling or heating. Simple calculations can be performed with the suppositions that the chamber has two zones at different temperatures [7] and the ideal gas law:

$$n(t) = \frac{V}{R} P(t) \left( \frac{1}{T(t)} + \frac{V_o}{VT_o} \right) (1); \qquad \frac{dn(t)}{dt} = \frac{V}{R} \left[ \left( \frac{1}{T(t)} + \frac{V_o}{VT_o} \right) \frac{\delta P(t)}{\delta t} - \frac{P(t)}{T^2(t)} \frac{\delta T(t)}{\delta t} \right] (2)$$

where *n* is the quantity of hydrogen moles in the chamber of volume  $V' = V + V_q$ , V is the volume of the heated zone, R is the ideal gas constant and P and T are the pressure and temperature respectively. Variables depending on time (t) are explicitly identified and sub index indicates the volume  $(V_0)$  of the part of the chamber that is at room temperature  $(T_o)$ . The values of  $V_o/V.T_o$  and V/R are obtained by calculation using a variable temperature and pressure range in which no absorption takes place (n(t) = constant ). In figure 1 are represented the absorption velocity during heating and cooling runs of batches of approximately 700 g. Massive hydriding of the material takes place in a temperature range between 50/190 °C and is maximum at around 120 °C. Normally the hydrogen pressure was maintained at 1 bar. Runs performed at 2 bars and 0.5 bars practically doubled and made half respectively the hydrogen absorption rate. Hydrogen flow can reach values of 0.5 lt/min and normally is around 1400 ppm/hr. It is usual to notice during changing conditions of the experiment that absorption velocity is greater during cooling than when heating. Anyway, hydriding of the material is done at constant temperature and pressure. Pressure is maintained constant with a hydrogen gas flow continually incorporated to the chamber and temperature control is needed because the hydriding reaction is exothermic. The process is stopped after less than 15 hours when the rate of gas absorption is lower than 0.02 lt/min and more than 85% of the material has been hydrided. The hydrided alloy is fractured in millimeter size fragments as it is formed.



Fig 1. Hydrogen absorption rates during heating and cooling of U-7Mo alloy. Notice the different absorption rates in each case. Error bars are indicated for all data points.

The U-Mo hydride is a dark gray brittle compound of low hardness (Vickers < 300) and when oxidized is dark brown. It is pyrophoric and burns with flame because of hydrogen liberation with temperature. The stoichiometry is  $MH_x$ , where M stands for the U-7Mo alloy and X > 2.8. X-ray diffraction (XRD) and Rietveld refinement (Figure 2) show that the structure is of the prototype Cr<sub>3</sub>Si (A-15), the same as  $\beta$ -UH<sub>3</sub> [8]. The density (DRX) is 10.39 g/cm<sup>3</sup>, much smaller than the U-7Mo which is 17.5 g/cm<sup>3</sup>. The hydriding of the U-Mo in  $\gamma$  phase is controlled by hydrogen diffusion but, because of the high density difference between the alloy and the hydride, internal cracks and fractures are formed parallel to the diffusion front and new surface is formed. The consequence is that the rate of hydride formation is greater than the expected 0.5 power law usual in diffusion processes.



Fig 2. XRD and Rietveld Refinement of U-7Mo hydride A-15 structure. The width of peaks is probably ought to high distortion of the crystal lattice. Some  $\gamma$  phase alloy (bcc) is still present.

#### 3. Milling

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Before exposing the hydride to air it is needed to do a controlled passivation to avoid burning the material. The general appearance of the interstitial compound is of small platelets 0.5 mm thick with inside small cracks. The milling of the material can be done gently in air by hand using a mortar or in any conventional milling machine in a low oxygen atmosphere. Size distribution control will depend on the choice of the mill to be used. Hydride milled and sieved particles are shown in Figure 3. A decrease in size distribution is expected in the dehydriding of the material because of density differences between the hydride and the final U-7Mo powder. The usual powder specification of less than 50% fines is easily achieved.



Fig 3. U-7Mo hydride powder. Mean particle size is 80 µ.



Fig 4. U-7Mo powder obtained by HMD process. Scale bar divisions correspond to 100 µ.

## 4. Dehydriding

In vacuum conditions hydrogen liberation from the hydride powder begins at 125 °C. As hydrogen evacuation takes place, the particles surface densify and diffusion rate decrease. For a complete removal of hydrogen the powder is submitted during one hour at 700 °C in a diffusion pump vacuum. After cooling, controlled passivation must be done before air exposure of the material. Measured hydrogen content is less than 50 ppm, probably attributed to water adsorption at the particles surface after air exposure than to remnant hydrogen. Figure 4 is a general view of the final U-Mo powder particles retained in a 44  $\mu$  sieve. XRD of the powder is shown in Figure 5 indicating the major presence of the U-Mo metastable  $\gamma$  phase. Some traces of uranium oxide are detected and are attributed to excessive oxidation in air during the hand milling of this laboratory sample.



Fig 5. X-Ray Diffraction of U-Mo alloy obtained by hydriding, milling and dehydiding.

## 5. Conclusions

The development of the HMD process has been done simultaneously with the initiation of basic research to study the up to now unknown properties of the massive U-Mo hydride. Additional thermodynamic data, equilibrium stoichiometry and interstitial hydrogen positions still have to be elaborated. Hydrogen in solution, crystallographic orientation and macro and micro stress field alloy conditions are fundamental in the kinetics and massive hydriding of  $\gamma$  phase U-Mo, as in the formation of other hydrides.

One important aspect of the hydriding of U-7Mo alloy is that fractures are not as fine as in  $\alpha$ -U hydride which produces nanometric size powder. In the case of the U-Mo hydride fractures are macroscopic with micron size internal cracks. Brittle transgranular fractures are produced during comminution giving as result polyhedrical shaped particles.

The developed process (Figure 6) is susceptible of improvements. Higher pressures can be used to increase hydrogen rate absorption; temperature and pressure controls can be automated; etc. The process is fully scalable to maximum U-Mo powder production using enriched uranium compatible with security standards. Low manpower and equipment investment are required.



Fig 6. Flow diagram of the HMD process for U-Mo industrial powder production.

## 6. Acknowledgements

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## IMPROVEMENTS IN IRRADIATED FUEL HANDLING AT DHRUVA FOR ENHANCED SAFETY

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## ABSTRACT

Dhruva is a 100 MWt research reactor located at the Bhabha Atomic Research Center, Trombay, Mumbai, India. The reactor achieved its first criticality in August 1985 and full power operation in the year 1988. Heavy water is used as moderator, primary coolant and reflector in the reactor. The fuel assemblies are made of natural metallic uranium pins clad in aluminium. The reactor provides facilities for basic and applied research, material testing, production of radioisotopes and training of manpower. Operation of Dhruva for over sixteen years has an excellent safety record. Continued efforts are made towards enhancing operational safety of the reactor by way of reviewing incidences and implementing necessary modifications including changes in operating procedures. This paper highlights some of the improvements carried out in irradiated fuel handling at Dhruva for enhanced safety.

## 1. INTRODUCTION

India's fifth research reactor DHRUVA which became critical on August 8,1985, is a natural uranium fuelled, heavy water moderated and cooled thermal neutron research reactor with an operating power level of 100 MWt and a maximum thermal neutron flux of 1.8 X 10<sup>14</sup> n/cm<sup>2</sup>/sec. The indigenously designed and built reactor is located on the east Coast of Trombay Island at the Bhabha Atomic Research Centre (B.A.R.C.), and is about 10 km north-east of Mumbai city. The reactor has a vertical core and employs natural metallic uranium seven-pin cluster fuel assemblies installed in Zircalloy guide tubes in stainless steel reactor vessel. Heavy water is used as the moderator, primary coolant and reflector. Helium is used as cover gas. Reactor power regulation is achieved by moderator level control. Fast shut down of the reactor is effected by nine Cadmium Shut off rods with simultaneous dumping of heavy water moderator. Heat from primary coolant is transferred to a secondary closed loop system recirculating demineralised light water in a set of heat exchangers. The sea water coolant is drawn from the Mumbai harbour bay and flows through the heat exchangers on once-through basis.

## 2. THE REACTOR FUEL CYCLE

The fuel cycle for Dhruva Reactor is represented in fig.1. The basic source material is uranium ore. The concentrated ore is shipped to Uranium Metal Plant (UMP). At UMP uranium ore is converted into uranium metal. The uranium metal is transported to Atomic Fuel Division (AFD) for fabrication of fuel. Dhruva fuel pin is three meter in length and 12.7 mm in diameter. These pins are clad with 1mm thick aluminium. Seven pins are assembled inside an aluminium flow tube to form one cluster sub assembly. Aluminium spacers are fixed to the central rod of the fuel cluster and distributed over the length. Each fuel pin and clad tube is subjected to Eddy current testing, Radiography and Visual inspection to ensure integrity of fuel pin and its cladding. Fuel pins are subjected to Glycol testing before assembly. These fuel

# DHRUVA REACTOR FUEL CYCLE



## SPENT FUEL STORAGE

FIG 1. OUTLINE OF COMPLETE URANIUM FUEL CYCLE.

cluster sub assemblies are transported to Dhruva Reactor for final assembly and loading in to reactor.

At Dhruva each fuel cluster is subjected to visual inspection and dimensional checks. Fuel cluster sub assembly, aluminium shield subassembly and seal-and-shield plug subassemblies are assembled to form a fuel assembly. The fuel assembly is 9.2 meter in length. The aluminium shield sub assembly and seal-and-shield sub assembly form the extension of fuel cluster sub assembly up to the top of the reactor and provide shielding and means for locking the fuel assembly to its channel. The full-length assembly is flow tested, dried and tagged certifying its pile worthiness. All fuel assemblies are stored vertically.

Refueling is carried out by 300 Te fuelling machine having layers of lead and borated paraffin shielding to protect against primary gamma, photo neutrons and secondary gamma rays. The machine has two barrels, which are indexed to load and unload fresh and irradiated fuel in to and from the core. All operations of machines are carried out remotely by hydraulic drives from machine console. All drives are backed up by manual drives. Limit switches indicate actuations of various operations and these indications are used in interlocks for ensuring sequential operations of machine drives. The fuelling machine is designed to have its own arrangement for cooling fuel with heavy water. Provision also exists for cooling the fuel in machine with light water in exigencies. Necessary arrangement is provided in machine to minimise tritium exposure during fuel handling. A test channel is provided to qualify the machine prior to the fuelling operation. All drives are hydraulically operated and set suitably to ensure the limited force on fuel, coolant channel and machine components.

Refueling is carried in accordance with approved agenda. The fuelling engineer-in-charge prepares the agenda. The station reactor physicist certifies the load changes and Dhruva Reactor Superintendent approves the agenda. Each movement of a fuel assembly is recorded in Transfer Slips to ensure proper movement and inventory of fuel assemblies.

The fuel assemblies are removed from the core after completion of scheduled irradiation and transferred by the fuelling machine into fuel transfer buggy which carries it through an underwater fuel transfer trench to the adjoining spent fuel storage building (SFSB). In the SFSB the fuel assembly is bisected separating the reusable seal and shield sub assemblies from the fuel sub-assembly. The fuel sub-assemblies are stored vertically under water. Towards ensuring safety of fuel and operation staff following provisions exist

- Bays are built above ground level and extend up to pile elevation. This facilitates to minimise dry handling duration of irradiated fuel assembly while transferring from machine to SFSB.
- Bays are lined with Stainless Steel to provide leak tightness and ease in decontamination. This also obviates the problem of water table exerting pressure on the stainless steel lining.
- The fuel sub-assembly storage bay cannot be drained completely which ensures fuel submergence.
- SFSB is divided into cutting bay, storage bay and inspection bay. Provision exists to isolate each bay for any repair job.
- Under water tools cannot be lifted beyond specified height by design to ensure adequate water shielding.
- All under water equipments are subjected to regular preventive maintenance checks and surveillance testing to ensure the intended functioning.
- Bay water chemistry and specific activity is maintained by use of ion-exchanger resin.
- Ventilation is provided to ensure personal safety, air curtain is maintained above water level to minimise air activity in the SFSB.

### 3. IMPROVEMENTS IN FUEL HANDLING

The following are some of the improvements carried out in fuel handling systems and operating procedures based on experience:

- 3.1 During initial commissioning trials with dummy fuel, it was realized that possibility of fuel assembly remaining unlocked from a channel couldn't be ruled out. In Dhruva upward force exerted on the fuel by the coolant flow when main coolant pumps are in operation is more than the weight of the fuel and an unlocked fuel can get lifted up. In view of this apprehension, provisions were made to deny access to fuel channels when main coolant pumps are in operation in the form of physical barriers and interlocks. In addition special locking gadget was provided on top of each fuel assembly whose design does not permit the assembly to be unlocked while coolant pumps are in operation. [1]
- 3.2 During transfer of irradiated fuel from reactor to the underwater fuel transfer buggy, the fuel is cooled by heavy water cooling system of the fuelling machine. Just prior to discharge of fuel, heavy water from the machine is dumped in to storage tank and the fuel is quickly lowered into the buggy. Detailed procedural checks ensure that the irradiated fuel assembly is discharged within the "stipulated dry time". Provision also exists for dousing the fuel assembly with light water if there is a possibility of exceeding the "specified dry time". Based on experience further provisions have been made to make available additional critical information to fuelling engineer and also to enhance reliability of light water dousing system.
- 3.3 A pool site inspection facility is being provided to carry out post-irradiation examinations of routine nature at the reactor site itself avoiding the need for frequent transportation of irradiated fuel to hot-cells [2]
- 3.4 For canning of fuel elements with clad failure, the flow-tube itself is used as a can. For achieving this, significant development work had to be done towards working out the design of plugs and the plugging tools. [2]
- 3.5 Adequate storage capacity for irradiated fuel has been provided for storage of fuel for few years. The irradiated assemblies are stored vertically. Being metallic fuel, if fuel clad fails during storage the recovery of uranium powder becomes difficult task. To recover any debris/uranium powder, removable trays have been provided below the underwater fuel storage racks. With this the bay floors remain clean and the trays can be removed for cleaning when required. [2]

#### 4. CONCLUSION

Enhancement of safety is an on going process. From over 16 years of experience in the field of handling and storage of irradiated fuel from Dhruva we have incorporated several changes towards enhancing safety and improving system performance. These include design modification, procedural changes, provision of physical barrier, development of in house inspection facility and development of canning facility for fuel.

#### 5. ACKNOWLEDGMENT

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## EXAMINATION OF U-9% Mo ALLOY POWDER MICROSTRUCTURE IN ITS INITIAL CONDITION AND AFTER FUEL PIN FABRICATION

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## ABSTRACT

Within the frames of a program for reduction of research reactor fuel enrichment a powder of U-9%Mo (OM-9) alloy has been prepared by the method of the rotating electrode atomization. The analyses of its composition, distribution of its principal chemical elements over the cross-sections of individual particles have been carried out as well as the analysis of the initial OM-9 powder. To be able to conduct a high-grade process of fabricating pins whose dispersion type fuel is the OM-9 grit embedded in aluminum matrix, both the initial OM-9 grit and this fuel composition have been investigated at 500 and 620°C. A substantial interaction has been shown to occur between the OM-grit and the aluminum matrix at 620 °C.

### 1. Introduction

The works aimed at the conversion of the research reactors built-up in accordance with Russian (Soviet) projects to the lower enrichment fuels (up to 19,7% of  $U^{235}$ ) are being carried out in conformity with "The Russian Program of reducing the research reactor fuel enrichment" under the supervision of a joint-stock company "TVEL".

According to this program a novel technique for preparation of the U-9%Mo (OM-9) alloy powder by atomizing a rotating electrode has been developed in the SSC RF IPPE [1].

A fuel pin fabrication process [2] stipulates a number of processing steps proceeding at elevated temperatures. To evaluate the effects of these elevated temperatures on the fuel composition, both the initial grit of the OM-9 alloy and the fuel composition being the OM-9 grit embedded in the aluminum matrix were investigated after their isothermal testing at 500° and 620°C.

## 2. Investigation of the initial grit of the OM-9 alloy

The initial grit of the OM-9 alloy had the properties as follows:

- the γ-phase structure;
- the microhardness of 286 to 297 kg/mm<sup>2</sup>;
- it contained 90,7 % uranium, 8,9 % molybdenum, and (not taking oxygen into account) no more than 0,1 % contaminant elements.

The employed grit fraction had the particle size of  $100\div315 \,\mu\text{m}$ . The sampled grit was subjected to X-ray phase analysis. X-ray exposures were taken in the Co emission in the IRIS-3 apparatus with the chamber photo-recording.

An X-ray photograph presented in Fig.1 shows the presence of the Mo solid solution in  $\gamma$ -U as a basic phase; it has a body-centered cubic lattice whose parameter  $a \approx 3,470 \pm 0,005$  Å. The presence of faint uranium dioxide lines in the X-ray photograph is related to a slight OM-9 grit oxidation in the course of its preparation.



Fig. 1. A typical X-ray photograph of the OM-9 grit after its preparation  $\times$  -  $\gamma$ -phase solid solution of Mo in U  $\circ-UO_2$ 

For the purpose of the metallographic analysis a microsection was prepared that was subjected to electrolytic etching and photographed at a 200-fold enlargement. As one can see from Fig. 2, the grit has a fine-grain structure (its grain size number varies from 7 to 10 N). In certain cases individual dendrites characteristic for a cast structure could be visible in a rather distinct way.



Fig.2. The microstructure of the OM-9 alloy grit as prepared, ×200.

With the purpose of revealing the uranium and molybdenum distribution over the cross sections of the individual particles the X-ray spectral microanalysis of metallographic microsections has been carried out in the MAR-2 setup. The probe spot diameter was of 2 to 3  $\mu$ m which enabled to get the complete pattern of the element distribution. The analysis of the X-ray photographs has revealed a good homogeneity of the molybdenum distribution over the sections of individual particles without any significant deviations from the average values, see Fig. 3.





## 3. Investigation of the "OM-9 grit in aluminum matrix" fuel composition

The interaction between the OM-9 grit and aluminum matrix has been carried out at  $500^{\circ}$  and  $620^{\circ}$ C as characteristic temperatures for processing steps of fabrication fuel rods containing the fuel composition containing the UO<sub>2</sub> grit embedded in the aluminum matrix. Fuel composition samples were produced by cold pressing. Sintered density of these samples was of 95 to 97 % from the theoretical density.

The samples were tested in vacuum at 500° and 620°C.

The duration of testing was of 6 hours. After cooling the samples were taken out of the furnace and subjected to the metallographic analysis of the fuel composition (see Figs 4 and 5).



Fig. 4. The microstructure of a sample tested at  $620^{\circ}$ C (×200).



Fig. 5. The microstructure of a sample tested at  $620^{\circ}C$  (×200).

As can be seen from Fig. 4, at  $620^{\circ}$ C some interaction of the OM-9 grit with aluminum has occurred. The microhardness of the extant parts of the particles has not changed which points to the fact that the OM-9 alloy has preserved its  $\gamma$ -phase structure. The interaction region has a microhardness of 607-636 kg/mm<sup>2</sup> which is characteristic for the uranium-aluminum intermetallides.

The microstructure of the fuel composition tested at  $500^{\circ}$ C indicates (see Fig. 5) that the interaction between the OM-9 and aluminum has just begun (the interaction layer is yet 1 to 2  $\mu$ m thick).

A certain rise of the grit microhardness values from 321 to 372 kg/mm<sup>2</sup> can be observed.

For the purpose of revealing the distributions of uranium, molybdenum and aluminum over the crosssection an X-ray spectral microanalysis has been carried out on the metallographic microsections of the tested samples, see Figs 6, 7.



Fig.6. The patterns of aluminum, uranium and molybdenum distribution along the grit particle diameter after testing a sample at 620°C.



Fig.7. The patterns of aluminum, uranium and molybdenum distribution along the grit particle diameter after testing a sample at 500°C.

According to the results of the X-ray spectral microanalysis, the grit of the OM-9 alloy is sufficiently well compatible with the aluminum matrix at 500°C. The distributions of U, Mo  $\mu$  Al presented in Fig. 7 indicate that the thickness of the interaction region does not exceed ~5  $\mu$ m, and the interaction is of a diffusional character.

Thermal testing at  $620^{\circ}$ C leads to the formation of an interaction region up to  $\sim 30 \ \mu$ m thick at a boundary of contact between the grit and the matrix. The character of the element distribution over the interaction region presented in Fig. 5 corresponds to the character of the element distribution in case of chemical compound formation.

## Conclusions

The investigations carried out have indicated that no processing steps in the course of fabricating fuel pins with the fuel composition of the U-9 % Mo alloy in the aluminum matrix are allowed to be carried out at temperatures exceeding 500°C.

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# STUDY OF IN-REACTOR CREEP IN THE ALLOYS EMPLOYED AS STRUCTURAL MATERIALS FOR RESEARCH REACTOR CORE COMPONENTS AND FUEL PINS

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# ABSTRACT

In order to make a computational estimate of the stressed-strained state as well as the working capacity of research reactor fuel pins it is necessary to know mechanical properties of materials under irradiation (deformation curves, creep, long-term strength or deformability). With the purpose of investigating in-reactor creep, a series of in-reactor test devices have been developed in the SSC RF IPPE that ensure the execution of fully instrumented and controlled experiments intended for the "dry" channels of the BR-10 research reactor. A specially developed technique enabled to carry out in-reactor creep rate measurement for various structural materials (an austenitic steel, zirconium and aluminum-based alloys) employed as fuel pin cladding. A number of special features have been revealed that take place in the process of radiation creep of the materials investigated.

# 1. Introduction

In-reactor creep test was carried out by the method of tubular specimen torsion at a constant level of the torsional moment. With that end in view a series of in-channel "Sigma" type test facilities specially developed in the SSC RF IPPE have been used [1]. The temperature of specimens during the tests was adjusted at the expense of the test facility radiation-induced heating or by means of an electric heater. The loading of a specimen by the torsional moment of a specified value and the automated maintenance of this loading was carried out during a stable (both in power and in temperature) operation of the reactor. The strain of the specimen in the course of testing was being determined by the dynamometric method [2]. The temperature of the specimen in the process of irradiation was being measured by chromel-alumel thermocouples. The specimen was placed within a leak-proof cavity of the test facility. The working fluid was helium under standard pressure or an argon-helium mixture.

The research of steady-state radiation creep rates for austenitic steel, zirconium- and aluminum-based alloys resulted in the accumulation of a data array enabling to determine the form of a functional relationship between the radiation creep rate and stress in a wide range of stress variation. From the measured data related to shear strain,  $\gamma$ , and tangential stress,  $\tau$ , the values of creep rate intensity,  $\dot{\epsilon}$ , and stress intensity,  $\sigma$ , have been computed. These computations were made taking into consideration the relationships:  $\dot{\epsilon} = \dot{\gamma}/\sqrt{3}$  is  $\sigma = \tau\sqrt{3}$ .

# 2. In-reactor creep of steel

The in-reactor creep of the .09C16Cr15Ni3MoNb type austenitic steel has been investigated at  $T \le 0.4$  $T_{melt}$  [3,4] in the central channel of the BR-10 reactor, and the result is shown in Fig. 1.As can be seen from this figure, the evolution of the in-reactor creep of the steel is stress dependent. With stresses lower than the yield strength the creep rate is proportional to the stress  $\sigma$ . In case of exceeding the yield stress that is dependent on temperature and cumulative radiation dose, the nature of deformation changes and the dependence of  $\dot{\epsilon}$  on  $\sigma$  becomes close to the exponential one. The tests have shown that, notwithstanding the fact that creep is triggered by radiation, under stresses exceeding the yield strength the temperature rise slightly accelerates the deformation process. At the same levels of stresses and temperatures but in the absence of the neutron flow the creep rate of the steel has reduced by a factor of 100.



Fig. 1. Stress dependence of the in-reactor creep rate for the .09C16Cr15Ni3MoNb type steel: o - results of testing at 470°C up to  $K = 3.1 \cdot 10^{-7}$  dpa/s; •- those at 500°C up to  $K = 4.1 \cdot 10^{-7}$  dpa/s

#### 3. In-reactor creep of the zirconium-based alloy

The in-reactor creep of the zirconium-based alloy Zr+1% Nb has been tested in a periphery channel of the BR-10 reactor at the temperatures eliminating any appreciable contribution of the thermal creep into the overall strain. The results of measuring the creep rate at the stage of the steady-state creep depending on the stress applied are shown in Fig. 2.

It is to be noted that up to a stress value of  $\sigma \sim 350$  MPa (which corresponds to  $\sim 90\%$  of the yield strength of this material irradiated up to  $\Phi = 6 \cdot 10^{21}$  n/cm<sup>2</sup> at the irradiation temperatures, T, in the range of 150 to 350°C), the in-reactor creep rate,  $\dot{\epsilon}$ , like that of the steel, is proportional to the applied stress  $\sigma$ .

At stresses  $\sigma$  exceeding 90% of the yield strength the radiation creep of the Zr+1% Nb alloy has been evolving with substantially higher rates. This radiation creep acceleration is similar to that observed in case of testing the austenitic steel (see Section 2).



Fig. Fig. 2. Stress dependence of the (Zr+1% Nb) alloy in the temperature range of 150 to  $350^{\circ}$ C.

Irradiation temperature: •-230-350°C; o-150-160°C.

#### 4. In-reactor creep of the aluminum-based alloy

The in-reactor creep of the aluminum-based alloy (SAV-1) containing Mg, Si, Si (< 5% in a total) has been tested in a periphery channel of the BR-10 reactor. The irradiation temperature range was within 80 to 90°C. At these temperatures the thermal (out-of pile) creep of this alloy at stresses lower than  $\sigma_T$ is  $\leq 10^{-7}$  h<sup>-1</sup>.In-reactor creep measurement data for this alloy show (see Fig. 3) that its creep rate under irradiation has grown by several fold. But, just like for other materials, too, the creep rate,  $\dot{\epsilon}$ , under the stress values of  $\sigma \leq \sigma_T$  is proportional to  $\sigma$ , and under the stress values of  $\sigma \geq \sigma_T$  the creep rate is nonlinear in  $\sigma$ .



Fig. 3. Stress dependence of the in-reactor creep rate for the SAV-1 aluminum-based alloy (containing Mg, Si, Fe < 5% in a total) within the temperature range of 80 to  $90^{\circ}$ C

#### 5. Conclusion

The in-channel test facilities developed in the SSC RF IPPE have enabled to carry out the in-reactor creep testing (in the BR-10 reactor) of various structural materials employed as fuel pin cladding. The results of the tests have shown that for all the materials investigated (steel, zirconium-based and aluminum-based alloys) the in-reactor creep rate under the stresses below the corresponding yield strength is proportional to the stress. In case of exceeding the yield strength that is dependent on temperature and cumulative irradiation dose, the stress dependence of the in-reactor creep rate,  $\dot{\epsilon}$ , becomes close to the exponential one.

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# MANAGEMENT AND INSPECTION OF INTEGRITY OF SPENT FUEL FROM IRT MEPHI RESEARCH REACTOR

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# ABSTRACT

The information on wet storage and dry storage of the spent nuclear fuel (SNF) of the IRT MEPhI reactor and experience from SNF shipment for reprocessing are presented. The procedure and a facility for nondestructive inspection of local power density fields and the burnup of fuel assemblies based on studying the  $\Upsilon$ -activity of some fission products generated in U<sup>235</sup> and procedure for inspection of the fuel element cladding leaktightness are described.

#### **1.Introduction**

The IRT MEPhI research reactor is a water-cooled water-moderated pool reactor with the power of 2.5 MW. It has been successfully operated at Moscow State Engineering Physics Institute since 1967. The first criticality of the reactor was attained on May 26, 1967 and its power was 500 kW in October 1967, 1 MW in 1970, 1.7 MW in 1971 and 2.5 MW in 1972. A facility for inspection of the fuel cladding leaktightness was installed in the interim SNF storage in 1975.

In the course of reactor operation 77 fuel assemblies (FAs) with EK-10 fuel elements and 102 IRT-2M, -3M FAs were used: until 1975, FAs with EK-10 fuel rods with meat of  $UO_2$  in a magnesium matrix and with an aluminium alloy cladding, and afterwards IRT-2M and IRT-3M FAs with annular fuel elements having a square cross-section and containing the meat of U-Al (or  $UO_2$ ) in an aluminium matrix and the aluminium alloy (SAV-1) cladding (the FAs contain three, four or eight fuel elements of such type).

In 1989, 48 IRT-2M spent fuel assemblies (SFAs) were transported to reprocessing plant RT-1.

The transportation for reprocessing of IRT-2M SFAs rather than older EK-10 SFAs has been caused by the fact that RT-1 does not reprocess EK-10 fuel elements because of their small amount, low enrichment and the necessity of using a process somewhat different from the regular process.

The SNF transportation was later stopped because of economic difficulties.

- Currently the SNF of the IRT MEPhI reactor is stored in two different ways:
  - wet storage (in a storage pool filled with chemically desalinated water (CDW) 54 SFAs with annular-type fuel elements;
  - dry storage (in leaktight containers with an air environment) 77 rod-type SFAs.

The Russian spent nuclear fuel handling concept provides that the strategic direction of nuclear power development is the nuclear fuel cycle closing that is to ensure a more complete use of natural nuclear fuel and artificial fissile materials produced by nuclear reactors (plutonium, etc.), minimization of the radioactive waste amounts from SNF reprocessing and approximation to the radiation migration equivalence of the disposed waste and withdrawn natural fuel.

In this connection, the SNF of the IRT MEPhI reactor as well as the fuel of other Russian research reactors should be reprocessed based on the extraction technology used by RT-1 (PUREX-process).

# 2. Experimental and calculational research of fuel burnup

Short-term and long-term loads experienced by FAs in the core are monitored for efficient and safe operation of the IRT MEPhI reactor. The reactor power is monitored based on measuring the heatup of the coolant flowing through the core. The contributions of different FAs to the overall power are determined by calculations. The fuel burnup in each FA is determined based on results of these calculations proceeding from their total power generation. A neutronic code TIGR [1] is currently used to calculate neutron fields, power density and fuel burnup values. The TIGR code was verified by comparing calculation results with experimental operation data. Integral characteristics of the reactor (reactivity, critical position of the control rods) were compared for different core loads. It is known that such measurements characterise the reactor as a whole. So quite an accurate and detailed experimental information is required for a more detailed verification of the calculations.

As direct measurements of the  $U^{235}$  content in the irradiated FAs using  $U^{235}$  own gamma radiation are not feasible, then the measurement of the burnt-up  $U^{235}$  in the irradiated FA was based on measuring the gamma activity of a particular fission product formed as the result of the  $U^{235}$  burnup.

The characteristics of the widely used fission products for analysing the irradiated nuclear fuel burnup are presented in Table 1 [2, 3].

				1 aute
Fission	T <sub>1/2</sub>	Yield at $U^{235}$ fission	Gamma radiation energy	Quanta yield (%)
product		(%)	(keV)	
Zr <sup>95</sup>	64.0 days	6.50	724.2	43.1
			756.7	54.6
La <sup>140</sup>	1.68 days	6.27	1596.17	0.844
Nb <sup>95</sup>	35.0 days	6.50	765.8	99.8
Cs <sup>137</sup>	30.17 years	6.22	661.6	85.1

A measuring scanning type facility consisting of a regular transfer container, a scanner, a collimator system and a spectrometric system was constructed in the reactor hall for studying irradiated FAs. Spectrometric systems with germanium detectors of different types and designs were used for measurements.

The reactor was shut down, all FAs were withdrawn from the core and put into the interim storage by the start of the experiment. For carrying out measurements, the FAs were alternately withdrawn from the interim storage, put into the transfer vessel and moved over a vertical channel to a lead thick-wall transfer container.

The experimental values were compared with the respective calculated data. And the calculated value of the relative burnt-up U<sup>235</sup> amount was compared with the experimental value of the relative amount of the accumulated fission product. Experimental and calculated values of the power density distribution over the FA height were mostly close and major differences are observed only for FAs with control rods. The calculated fuel burnup distribution over the height of FAs mostly correlate with the <sup>137</sup>Cs measurement results. Differences beyond the experimental error boundaties are observed for some of the six-tube low-burnup FAs containing control rods. The experimental and calculated results correlate better for eight-tube high-burnup FAs.

The produced spectra were processed using modern Russian and foreign codes. The obtained data on the initial fuel distribution in FAs, neutron field in the reactor core, fuel power generation and burnup in FAs were compared with the FA certificate data and calculation results produced using TIGR and GETERA codes being regular IRT MEPhI reactor neutronic calculation codes. Differences between the experiments and calculations have been found out and analyzed. General conclusions have been made with respect to the correctness of the calculation results and judgements were made regarding the errors of the calculated values.

Some new procedural approaches that have been developed during the work (loading indicators into the operating core FAs and use of a special irradiation time mode for getting information on processes in the fuel during different periods) expand the capabilities of in-pile experiments and may be used in other reactors.

# 3.Spent fuel inventory and ways of SFAs storage

		SFA type						
No.	Description of characteristic	IRT-2M (90%)	IRT-2M (36%)	IRT-2M KS <sup>*)</sup> (36%)	IRT-3M (90%)			
1	Residual enrichment in terms of $U^{235}$ , %	40 - 50	20	20 – 22	40 - 50			
2	Estimated residual U <sup>235</sup> content, g	90	130	230	150			

Some of the IRT reactor SFAs characteristics are presented in Table 2.

\*) – IRT-2M-KS FAs are experimental FAs developed at the Kurchatov Atomic Energy Institute and they differ from the regular IRT-2M FAs is a higher  $U^{235}$  load (from 230 g to 390 g).

15 IRT-2M KS FAs were tested in IRT MEPhI reactor. The tests have shown that such FAs are not reliable enough – according to the results of fuel cladding integrity detection in 1984-1986, 4 FAs with the fuel burnup of 10-15% were withdrawn from the core with the initial gas non-leaktightness and removed to the storage pool for regular storaging and there was no degradation of the storage pool water radiation characteristics.

Spent fuel inventory is summarised in the Table 3.

Table 3

Table 2

Location	SFA type	SFA quantity
Dry storage facility	SFAs with EK-10 fuel elements	77
Wet storage facility	IRT-2M (90%)	22
	IRT-2M (36%)	3
	IRT-2M KS (36%)	15 (4 of them are leaky)
	IRT-3M (90%) – 8 tube ones	14

Configuration of dry storage and wet storage is presented in Fig.1. The dry storage facility represents



dry channels with the diameter of 180 mm made in a concrete body for storage of irradiated experimental devices. SFAs are located in leaktight canisters in the dry channels, 3 SFAs in each canister. The channels are closed with a shielding plug made of heavy concrete in a steel casing.

SFAs were transferred to the dry storage facility after a 5 year cooling period in the storage pool. SFAs were put into the canisters and the canisters were sealed in a hot cell. The environment inside the canisters is air. The SFAs were preliminarily dried with hot air blowout. From 5 to 6 canisters are installed in the channel. Dry channels with the SFAs are ventilated forcedly during the reactor operation. The canisters are cooled by natural convection during longterm reactor shutdown periods. A separate pool filled with CDW is used for wet storaging. The storage pool has a system of ionexchange filters responsible for water chemistry and a system of water quality and temperature control. The water temperature is  $30-40^{\circ}$ C.

54 SFAs are placed in a special fuel storage rack in two layers – there are 48 SFAs in the lower layer and 6 SFAs in the upper one. Prior to the installation for storage, SFAs were cooled in the reactor pool outside the core where they were monitored for leaktightness based on water sipping in an isolated canister using the Te-I procedure of detecting leaky SFAs.

#### 4. Procedure for detecting SFAs cladding leakage

The SFA cladding leakage extent is determined using the method of measuring the FA cladding surface contamination with fission-produced  $Te^{132}$ .

Due to high sorption capacity and short path length of  $Te^{132}$  nuclei in the cladding material, the appearance of tellurium on the cladding surface is caused by the fission-produced tellurium release from beneath the cladding via its defects (cracks, pits, etc.) and the amount of the absorbed Te is proportional to the fission-produced Te release rate (the proportionality factor depends on the reactor operation mode). With the period of  $T_{1/2} = 77$  hours,  $Te^{132}$  undergoes a  $\beta$ -decay with generation of daughter I<sup>132</sup> ( $T_{1/2} = 2.3$  hour). As iodine has lower sorption capacity, it is released from the cladding to the coolant that flows around the FAs. Thus, the  $Te^{132}$  amount on the FA cladding surface can be determined by installing the FAs from the reactor core 40-50 hours after the shutdown, when the absence of fission-produced I<sup>132</sup> on the FA cladding surface is guaranteed, in a leaktight accumulator with very pure water and by measuring the  $Te^{132}$  amount in the accumulator. The I<sup>132</sup> concentration is measured based on the area of  $E\gamma=670$  keV and 770 keV photopeaks in the  $\gamma$ -radiation spectrum of the water sample taken from the accumulator.

#### 5. Equipment and technology for SFA shipment for reprocessing

In 1989, 48 IRT-2M SFAs were transported to reprocessing plant RT-1. A TK-5M shipping cask intended for these purposes was used for transportation. The weight of one shipping cask is not more than 5500 kg. Casks (in batches of 4 pieces) were transported by a trailer truck to a railway track to the Kurchatov Institute, Moscow, and put onto a TK-5 container car.

One of the basic conditions of putting the SFA into the shipping casks for SFA shipment for reprocessing was a gamma radiation exposure dose rate that was not to exceed 100 R/h at a distance of 1 m off the FA. The SFAs shipped for reprocessing were cooled in the storage pool for 3.5 to 11 years and the gamma radiation exposure dose rate was 20 to 80 R/h. The SFAs were put into casks using regular transfer equipment – rods with collet grips and a transfer container. Prior to being loaded into casks, the SFAs were dried by hot air blowout.

Russian regulations prescribe [4] that spent FAs to be delivered for reprocessing should not have leaky fuel elements causing contacts of the fuel composition with the SNF storage facility water (microdefects of the fuel cladding corresponding to the «gas non-leaktightness» of the cladding are permitted).

The SFAs to be shipped were:

- checked for leaktightness;
- visually examined;
- checked for passability in a gage.

The (average) burnup of the SFAs in this batch was more than 40%.

The leaktightness of the fuel cladding was checked by each FA sampling in the storage pool based on detection of  $I^{132}$  isotope technique.

There were no comments with respect to the SFAs condition after the checking operations being completed.

The SFA quantity coming annually to the storage pool is small (some 4 pieces) and the storage facility volume will be enough for approximately another 10 years. But according to the available information [5], the condition of similar SFAs (the fuel composition of U-Al alloy in an aluminium matrix, the

thickness of the fuel cladding made of an aluminium alloy is 0.9 mm) stored in the storage pool water and having the burnup of around 15 MW day, was normal for the first 15 to 20 years of storage after which problems started caused by more fission fragments released to the water as the result of the fuel cladding corrosion. The SFAs of the IRT MEPhI reactor that have been accumulated to date still have a time reserve of 10 to 15 years till the start of the potential loss of the fuel cladding leaktightness after which it is not possible to store them in the existing conditions. In this connection, it is necessary to consider possible variants of the subsequent SFA handling – their transfer to the dry storage facility or for reprocessing. Most of the accumulated SFAs (52 pieces) have been cooled for more than 5 years after their withdrawal from the core, which enables their transportation to reprocessing plant RT-1. It is rational to simultaneously transport 77 SFAs with EK-10 fuel elements stored in the dry storage for accumulating the body of such assemblies on the RT-1 site with their further reprocessing.

#### **6.Conclusions**

The IRT MEPhI reactor has been the basis for research activities and training of specialists in the industry throughout the period of its service. Most of the developments have been practically implemented in the field of nuclear engineering and technology. Thus, for example, verification work has been completed based on obtained experimental data of the neutronic TIGR code and the regular Te-I procedure for monitoring the fuel cladding leaktightness has been introduced.

Long-term water storage of SFAs in the storage pool has not lead to a visible degradation in the condition of the SFA structural materials, but if storage is continued in these conditions, the integrity of the fuel cladding is not guaranteed. The only possible solution of this problem both for the IRT MEPhI reactor and other Russian research reactors is transportation of SNF for reprocessing in near future as it is more expensive to set up a dry storage facility on the reactor site and to transfer the fuel to a more safe dry storage facility.

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# MONTE CARLO SIMULATION OF IRRADIATION OF MTR FUEL PLATES IN THE BR2 REACTOR USING A FULL-SCALE 3-D MODEL WITH INCLINED CHANNELS

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# ABSTRACT

A three-dimensional full-scale Monte Carlo model of the BR2 reactor has been developed for simulation of irradiation conditions of materials and fuel loaded in various irradiation devices. This new reactor model includes a detailed geometrical description of the inclined reactor channels, the irradiation devices loaded in these channels including the materials to be tested/loaded in these devices, the burn-up of the BR2 fuel elements and the poisoning of the beryllium matrix. Recently a benchmark irradiation of new irradiation device for testing and qualification of MTR fuel plates has been performed. For this purpose the detailed irradiation conditions of fuel plates had to be predetermined. Monte Carlo calculations of neutron fluxes and heat load distributions in irradiated MTR fuel plates were performed taking into account the contents of all loaded experimental devices in the reactor channels. A comparison of the calculated and measured values of neutron fluxes and of heat loads in the BR2 reactor is presented in this paper. The comparison is part of the validation process of the new reactor model. It also serves to establish the capability to conduct a fuel plate irradiation program under requested and well-known irradiation conditions.

### 1. Introduction

The Belgian high flux material testing light water reactor BR2 has been operational since 1963. An intensive experimental program of irradiation of different materials and fuel elements in the reactor core needs an effective and reliable theoretical model of the reactor for the calculation of different neutronic parameters and irradiation conditions in investigating devices. A specific feature of the BR2 construction is that all channels containing fuel elements and irradiated devices in the reactor core are inclined at different angles. All 2-dimensional and 3-dimensional (Monte Carlo) computational models used until now for the calculation of irradiation conditions in the BR2 reactor did not take into account the actual inclined orientation of the channels[1]. This note describes a full-scale Monte Carlo model of the BR2 reactor with inclined channels for calculating the neutron fluxes and the nuclear heating in different irradiation devices loaded into the BR2 core

### 2. Full-scale 3-dimensional model of the BR2 rector

The Monte Carlo transport code MCNP [2] was used for simulating neutron transport in the BR2 reactor. Three-dimensional BR2 model includes:

- precise geometrical description of fuel elements, control rods, regulation rods, individual orientation of hexagonal Be matrix in each channel and all experimental devices;
- individual description of contents loaded into each inclined channel;
- individual composition of fuel elements;
- flexibility for preparing and changing data describing a reactor load;
- possibility to obtain detailed information about distribution of neutron and photon fluxes, fission heating, gamma heating in any part of the reactor.

 possibility to obtain detailed information about distribution of neutron and photon fluxes, fission heating, gamma heating in any part of the reactor.

The core of BR2 reactor contains 79 channels: 64 standard channels ( $\emptyset$ 84.2 mm), 10 reflector channels ( $\emptyset$ 50 mm) and 5 large channels ( $\emptyset$ 200 mm). Each channel has a hexagonal Be reflector and is inclined by it own angle so, that in each cross section of the core by Z-plane a triangular water gap exists between the channels formed by the hexagonal Be matrix. The further that the cross section is from the middle plane, the larger the triangular gaps appear between the channels. An example of the core cross section by the plane Z=+20 cm above the middle plane is shown in Fig.1. Irradiated MTR fuel plates are located in the irradiation devices as shown in Fig.2. This shows the triangular water gaps do not appear. The MTR fuel plates are located in water gaps 7.6 mm and have a thickness of 1.27 mm, and dimensions 68.4 mm × 970 mm. Standard fuel elements of the BR2 reactor have a different burn-up composition. Dependence of nuclide composition on the burn-up of BR2 fuel elements is similar to the work [3].



Fig.1 Cross section of the BR2 core load containing the irradiation device with MTR fuel plates.



Fig.2 Cross section of the channel G300 containing the device with MTR fuel plates (cross section at Z=+20 cm).

The effect of Be poisoning was taken into account for the following reactions:

$$\begin{array}{rcl} & 9 \operatorname{Be}(n,\alpha) & \longrightarrow & 6 \operatorname{He} \\ & 6 \operatorname{He} & & & \beta \\ & 6 \operatorname{Li}, & & T_{1/2} = 0.807 \text{ s} \\ & 6 \operatorname{Li}(n,\alpha) & \longrightarrow & 3T \\ & 3 \operatorname{T} & & & & \beta \\ & 3 \operatorname{T} & & & & \beta \\ & 3 \operatorname{He}(n,p) & \longrightarrow & 3T \end{array}$$

Production of <sup>6</sup>Li and <sup>3</sup>T are the main source of poisoning of Be matrix. Both <sup>6</sup>Li and <sup>3</sup>T are large absorbers of thermal neutrons and they have an influence on the flux distribution and on reactivity in the core. Be material is used as a reflector & moderator in several reactors. Analysis of the influence of Be poisoning on the reactivity of BR2 was presented in paper [4]. Calculations of the Be poisoning in the MARIA reactor was considered in papers [5-6]. The influence of Be poisoning on the reactivity in the WWR-M reactor was considered in paper [7].

The system of differential equations describing an evolution of concentrations of  $^{6}$ Li,  $^{3}$ T,  $^{3}$ He has a form:

$$\frac{dN_{Be}(r,t)}{dt} = -N_{Be}(r)\int \varphi(r,E,t)\sigma_{n,\alpha}^{Be}(E)dE ,$$

$$\frac{dN_{Li}(\mathbf{r},t)}{dt} = N_{Be}(\mathbf{r},t) \int \varphi(\mathbf{r},E,t) \sigma_{n,\alpha}^{Be}(E) dE - N_{Li}(\mathbf{r},t) \int \varphi(\mathbf{r},E,t) \sigma_{n,\alpha}^{Li}(E) dE ,$$

$$\frac{dN_{T}(\mathbf{r},t)}{dt} = N_{Li}(\mathbf{r},t) \int \varphi(\mathbf{r},E,t) \sigma_{n,\alpha}^{Li}(E) dE + N_{He}(\mathbf{r},t) \int \varphi(\mathbf{r},E,t) \sigma_{n,p}^{He}(E) dE - \lambda_{T} N_{T}(\mathbf{r},t) ,$$

$$\frac{dN_{He}(\mathbf{r},t)}{dt} = -N_{He}(\mathbf{r},t) \int \varphi(\mathbf{r},E,t) \sigma_{n,p}^{He}(\mathbf{r},E) dE + \lambda_{T} N_{T}(\mathbf{r},t) ,$$
(1)

here N<sub>Li</sub>, N<sub>T</sub>, N<sub>He</sub> are atomic concentrations of Li, T and He in Be matrix,  $\sigma_r^n$  is the microscopic cross section of nuclide *n* for reaction *r*,  $\varphi(r,E,t)$  is a neutron flux, and  $\lambda$  is a decay constant for T. Reaction rates in the equations considered are the function of Be matrix position in the core and the time of irradiation. However, for reducing the number of calculations, reaction rates were computed for several channels situated in different places in the core for irradiation cycle 2a/2001. Using equations (1) and information about the duration and cooling times for each irradiation cycle, concentrations of Li, T and He were computed for each set of reaction rates, corresponding to different channels. The average concentrations over the channels considered were used as homogeneous concentrations of Li, T and He in the whole Be matrix in the core. It seems to be difficult to calculate the reaction rates for each channel and each irradiation cycle starting from 1997. But for more accurate calculation it is possible to calculate spatial dependence of Li, He, T in different channels in the core taking into account reaction rates for one typical cycle.

#### 3. Comparison of Calculated and Measured Neutron Fluxes in Irradiation Channels

Calculations of thermal neutron fluxes were performed by Monte Carlo code MCNP for cycle 2A/2001 in the central Be channel H1 containing no irradiation samples and in channels G60, G300. The thermal neutron flux in channels G60 and G300 was calculated for the beginning and for the end of the fuel cycle for a nominal reactor power of 56 MW. The average value over the considered fuel cycle and over both channels was compared with neutron fluxes measured by 60Co integrated dosimeters (see Fig.4). The axial distribution of the thermal neutron flux calculated by MNCP in the central channel H1 and measured thermal flux is presented in Fig.3. Neutron fluxes for energy E<0.4eV and E<0.625eV were calculated for the H1 channel. The neutron flux in channels G60&G300 was calculated for  $1\sigma$ . The maximum value of neutron flux is located by -10 cm below the middle plane, that is explained by the position of absorbing elements of control rods above the middle plate.



Fig.3 Axial distribution of calculated and measured thermal neutron flux in the central



Fig.4 Axial distribution of calculated and measured thermal neutron flux in channels

G60&G300

#### 4. Irradiation of MTR fuel plates in channel G300

Two fuel plates were irradiated in channel G300 (see Figs.1-2) during fuel cycle in 2001. The axial distribution of integral  $\gamma$ -activity measured over the width of fuel plates is compared with axial distribution of fission rates. In Fig.5 both distributions are shown in arbitrary units for the plate located closer to the centre of the core. Comparison is performed in arbitrary units for both distributions. As can be seen, the calculated fission rate (histogram) and the measured  $\gamma$ -activity (points) have a similar axial distribution. More detailed distribution of fission rate was calculated over the heating surface of the fuel plate and presented in Fig.6.



Fig.5 Axial distribution of calculated fission power (histogram line) and measured  $\gamma$ -activity (dots) for the fuel plate located in channel G300. (nearest plate to the centre of the core).

Fig.6 Two dimensional distributions of fission rate in MTR fuel plates in channel G300. Fission power is normalised per cooling surface of fuel plate. The right distribution is calculated for the plate nearest to the centre of core.

#### 5. Conclusion

A full-scale 3-dimensional model of the BR2 reactor includes detailed description of the contents in all inclined channels and of all loaded irradiation devices. Effects of BR2 fuel burn-up and Be poisoning were taken into account.

Monte Carlo computations of neutron fluxes and fission power distribution performed by MCNP code using the present model demonstrated good agreement with experimental results.

#### 6. Acknowledgements

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# **NEUTRONIC DESIGN OF THE RSG-GAS' SILICIDE CORE**

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# ABSTRACT

The objective of core conversion program of the RSG-GAS multipurpose reactor is to convert the fuel from oxide,  $U_3O_8$ -Al to silicide,  $U_3Si_2$ -Al. The aim of the program is to gain longer operation cycle by having, which is technically possible for silicide fuel, a higher density. Upon constraints of the existing reactor system and utilization, an optimal fuel density in amount of 3.55 gU/cc was found. This paper describes the neutronic parameter design of the silicide equilibrium core and the design of its transition cores as well. From reactivity control point of view, a modification of control rod system is also discussed. All calculations are carried out by means of diffusion codes, Batan-EQUIL-2D, Batan-2DIFF and -3DIFF. The silicide core shows that longer operation cycle of 32 full power days can be achieved without decreasing the safety criteria and utilization capabilities.

#### 1. Introduction

The on-going BATAN program for the RSG-GAS multipurpose reactor is converting the core from oxide to silicide fuel. Based on the previous investigation conducted by Arbie et al. [1] and Liem et al. [2], a density of 3.55 gU/cm<sup>3</sup> of the silicide fuel was proposed for the RSG-GAS. The program was then fixed to conduct the conversion into two steps. Firstly, to convert the oxide fuel to silicide with same density of 2.96 gU/cm<sup>3</sup>. Secondly, it will then be converted from silicide 2.96 gU/cm<sup>3</sup> to silicide 3.55 gU/cm<sup>3</sup>. One amongst other reasons of this step is closely related to the BATAN R&D program in fuel fabrication. The former step was started in the year of 1999 and will achieve a full silicide core in about the end of the year 2002. The latter step is realized in 3 years.

In the mean time, calculations and analysis of all aspects for the silicide core of  $3.55 \text{ gU/cm}^3$  density are being conducted. These papers are then implemented in the preparation of the SAR (Safety Analysis Report) for licensing processes. In this occasion, neutronics design virtues of the RSG-GAS' silicide core are presented. Moreover, in order to keep the same shutdown margin as the original design, a change of control rod system is also proposed.

All calculations are done using the BATAN self developed diffusion code, Batan-EQUIL-2D for incore fuel management and Batan-2DIFF or -3DIFF for core calculations on 2 and 3 dimensions, respectively. For cell calculation, the WIMS/D4 code is applied. The same safety criteria and reactor performance for the utilization were imposed as the constraints of the calculations.

### 2. Design Constraints

Currently, the RSG-GAS is operated at maximum power of 30 MWth using 40 standard fuel elements (FE), 8 control fuel elements (CE) and 8 control rod absorbers (CR) on the  $10 \times 10$  core grid positions. The operation cycle length is 25 full power days (or 750 MWD) with the excess reactivity of 9.2 % $\Delta k/k$  and the shutdown margin reactivity of  $-2.2\%\Delta k/k$ . The objectives of the core conversion should be done without decreasing the safety criteria and utilization performance.

The safety criteria and constraints applied in the design are as follows:

- 1. The minimum shutdown margin reactivity ( $\rho_{OSR}$ ) is -0.5 % $\Delta k/k$ .
- 2. The maximum radial power peaking factor is 1.40.

- 3. The maximum discharged burn-up (BU) for the first and second steps of the core conversion at the end of cycle (EOC) are 56 and 70 (% loss of <sup>235</sup>U), respectively.
- 4. The number as well as the performance of irradiation positions and facilities should be maintained.

Furthermore, the burn-up classes and distribution as shown in Fig. 1 as well as the refuelling scheme as listed in Table 1 are considered as the strategy of in-core fuel management in generating all equilibrium and transition cores.



Note:BE = Beryllium Element;BS = Beryllium Element with plug;CIP/IP = Irradiation Position; PNRS/H);RS = Pneumatic/ Hydraulic Rabbit System;

Figure 1. Core configuration of RSG-GAS reactor with burn-up class in the second rows.

Table 1. Reshuffling and refueling strategy for the transition cores of RSG GAS reactor.

From	То	From	To	From	To
— H-9	F-10	F-5	F-8	C-7	B-8
H-8	C-4	F-4	F-6	C-6	G-5
H-7	F-7	F-3	C-10	C-5	D-4
H-6	D-10	E-10	B-4	C-4	D-5
H-5	E-5	E-9	G-6	C-3	H-8
H-4	F-9	E-8	D-3	B-9	C-9
G-9	E-8	E-5	A-8	B-8	out
G-8	Out	E-3	A-7	B-7	out
G-6	B-7	D-10	G-4	B-5	out
G-5	G-8	D-8	out	B-4	A-6
G-4	C-7	D-5	H-5	A-9	A-4
F-10	G-9	D-4	E-9	A-8	B-5
F-9	A-5	D-3	C-6	A-7	H-7
F-8	C-5	C-10	E-3	A-6	B-9
F-7	F-4	C-9	D-8	Δ-5	H-6
F-6	Out	C-8	F-5	A-4	E-10

The neutronic design parameters of the equilibrium and transition cores are performed by Batan-EQUIL-2D code [3] using the 4-group-energy neutron generated by WIMS/D4 code [4]. The Batan-3DIFF code [6] is used for calculating the 3-D core problem, such as, the axial power peaking factors and the reactivity worths of the partially inserted control rods of AgInCd. The kinetic parameters, such as, delayed neutron fraction and prompt neutron life time, are calculated by Batan-EQUIL-2D code for the 2.96 and 3.55 gU/cm<sup>3</sup> silicide equilibrium cores.

#### 3. Silicide core design

#### 3.1. Equilibrium cores

Table 2 summarizes the comparison of the calculated core parameters among the tree equilibrium cores. Significant increment of the operation cycle length in the core using silicide fuel of 3.55 g U/cm<sup>3</sup> is clearly shown in the Table. The cycle length of 32 full power days shows an increment of 30% or related to 7 full power days longer (210 MWD) compared the equilibrium oxide core one.

As seen in Fig. 2, the shutdown margin reactivity of the 3.55 g U/cm<sup>3</sup> equilibrium silicide core is 53% lower than that of the equilibrium oxide core due to harder neutron spectrum. Whereas, for the silicide of 2.96 g U/cm<sup>3</sup>, the bigger excess reactivity makes the shutdown reactivity 36% lower than that of the oxide core. An effort must be considered to increase the control rod capability which gives shutdown margin reactivity at least at a number of the initial design of  $-2.2\%\Delta k/k$ .

Core Parameters	Oxide 2.96 g U/cm <sup>3</sup>	Silicide 2.96 g U/cm <sup>3</sup>	Silicide 3.55 g U/cm <sup>3</sup>
Operation Cycle Length (MWD/days)	750/25.0	614.6/20.48	975/32.2
Max. discharged (% loss of <sup>235</sup> U)	56.0	56.0	69.1
Reactivity Balance (% $\Delta k/k$ ):			
- equilibrium Xenon poisoning	3.50	3.66	3.70
- burn-up	3.00	2.51	3.77
- Core excess reactivity	9.20	9.76	9.24
- Total control rod worth	-14.50	-13.60	-13.05
- Shutdown margin reactivity	-2.20	-1.32	-1.03
Max. Radial Power Peaking Factor	1.23	1.23	1.27
Max. Axial Power Peaking Factor	1.50	1.61	1.41
Delayed neutron fraction	7.204	7.186	7.024
Prompt neutron life time, µs	61.46	64.51	62.81

Table 2. Core parameter for the equilibrium cores





Fig. 2 Reactivity  $(\%\Delta k/k)$  vs. insertion position of eight AgInCd control rods

Fig. 3 Effect of the insertion of control rods to the axial PPF

The 3.55 g U/cm<sup>3</sup> equilibrium silicide core has smallest axial PPF because the maximum edge PPF takes place in F-10 core grid position. The axial PPF in F-10 is much lesser than that of in E-8 core grid position where the maximum axial PPF occurred in other two cores. This is because of F-10 grid core position far from the irradiation positions and the E-8 grid core position adjacent to the center irradiation position. However, the local power peaking factor of FE and CE adjacent to beryllium reflector and to irradiation position are slightly higher for the silicide of 3.55 gU/cm<sup>3</sup> core than for the oxide core.

#### 3.2. Modification of control rod system

As mentioned above, the designed silicide of 3.55 g U/cm<sup>3</sup> core has shutdown margin reactivity much lower than that of the initial value of -2.2.  $\%\Delta k/k$ . That's why, a modification of control rod system for the silicide core was proposed [7]. The objectives of the work is to increase the capability of control rod by adding the number of control rods acting as the safety rods in order to produce at least the same shut down margin reactivity as of for the oxide fuel one. The results show that, by adding 2 safety rods of AgInCd absorber in B-3 and G-10 grid core positions, the silicide core has shutdown margin reactivity of -3.32  $\%\Delta k/k$  that meets the safety criteria (-0,5 $\%\Delta k/k$ ) and even higher than that of the existing oxide fuel (-2,2 $\%\Delta k/k$ ).

#### 3.3. Transistion cores

In the first step of core conversion, from oxide to silicide fuel, the full silicide core (2.96 g U/cm<sup>3</sup>) will be achieved in the tenth transition core (TR-10), because the core was loaded using oxide and

silicide fresh fuels at BOC in the first (TR-1) and second (TR-2) transition cores. As seen in Table 3, all core parameters fulfil the design requirements. The detail neutronic design parameters of this first step can be seen in Ref. [5]. Table 4 shows that the levels of the thermal neutron flux ( $\leq 0.625$  eV) at selected irradiation positions do not change during the core conversion. For same operation cycle length and fuel burn-up distribution, the excess reactivity of the full silicide core is slightly smaller than that of the full oxide core one. This result is a consequence of the production to absorption ratio in the silicide core that is slightly smaller than that of the oxide core.

In the second step of core conversion, from silicide of 2.96 g U/cm<sup>3</sup> to 3.55 g U/cm<sup>3</sup>, the full higher silicide core (3.55 g U/cm<sup>3</sup>) will be achieved in the eighth transition core (TS-8). As seen in Fig. 2, all core parameters fulfil the design requirements, except in the TS-7, the core has maximum discharged burn-up of 70.4 % which is slightly greater than 70%. This condition can be avoided by reducing the operation cycle length. The detail neutronic design of this second step can be seen in Ref. [8].

Table	3.	Neutronic	calculated	parameters	for	the
		mixed oxid	de-silicide c	ores		

Carac	$\rho_{\rm OSR}$	Max. BU	Max.
Cores	(%)	(%)	Radial PPF
TR-I	-1.07	53.9	1.21
TR-2	-0.82	53.8	1.21
TR-3	-0.89	53.8	1.21
TR-4	-0.96	53.8	1.20
TR-5	-1.00	53.8	1.21
TR-6	-1.05	53.9	1.21
TR-7	-1.09	53.9	1.21
TR-8	-1.12	53.9	1.20
TR-9	-1.17	54.5	1.21
TR-10	-1.19	54.4	1.21

Table 4.	Average	Thermal	Flux	at	Selected
	Irradiatio	n Position	S		

Cores	$\times 10^{14}$ (neutrons cm <sup>-2</sup> sec <sup>-1</sup> )								
Cores	HYRS	(D-6)	PNRS	(G-7)					
Oxide	0.70	2.49	0.78	2.05					
TR-I	0.71	2.50	0.81	2.07					
TR-2	0.70	2.49	0.80	2.06					
TR-3	0.71	2.50	0.80	2.06					
TR-4	0.70	2.50	0.80	2.06					
TR-5	0.70	2.50	0.81	2.06					
TR-6	0.70	2.50	0.81	2.06					
TR-7	0.71	2.50	0.81	2.07					
TR-8	0.71	2.50	0.81	2.07					
TR-9	0.71	2.51	0.81	2.07					
TR-10	0.71	2.51	0.81	2.07					



Fig. 4 Calculated parameters for the transition cores in the second step

# 4. Conclusion

The silicide cores of the RSG-GAS were successfully designed which meets the safety criteria. It is unfortunately, produces lower shutdown margin reactivity compared to the original design one. To overcome the problem, a modification of control rod system was proposed by adding two safety rod absorbers.

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# SPENT FUEL MANAGEMENT AND SITE EXPLOITATION AT IFIN-HH, BUCHAREST-MAGURELE

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# ABSTRACT

This paper presents a discussion of the experience gained in the management of the spent nuclear fuel stored in water basis at the IFIN-HH, Bucharest-Magurele. At Magurele site a number of 223 burned fuel assemblies were stored in four water filled basins. Corrosion concerns on the spent fuel have been minimal over the years of fuel storage. The water in these basins is currently being maintained but no special issues were applied to improve water quality. Some pitting was reported on aluminum-clad of the fuel, but this was attributed to material defects and fabrication concern. An intensive effort was made at our department to understand the corrosion problems and to be able to improve the basin storage conditions for extended storage requirements. Because no significant improvements were accomplished in the last time, the ultimate solution is to remove the fuel from the ponds and after that to clean up the empty basins. The main goal of this strategy is to ensure that the fuels are maintained in a safe stable state until a final storage method for these fuels becomes available.

### 1. Introduction

The nuclear research reactor WWR-S from the National Institute of Research and Development for Physics and Nuclear Engineering "Horia Hulubei", Bucharest-Magurele, was first commissioned in July 1957 and it was shut down in December 1997. At the moment the reactor is in conservation state. The reactor is a Russian tank type model, using Russian fuel and it was the first nuclear reactor from his class build in a socialist country outside the soviet block.

This reactor was designed and constructed to serve general scientific research needs in different aspects of fundamental research (e. g. determination of cross sections, excitations level of nuclei and molecules, materials studies, etc.) and some applied research like isotope production and silicon doping. During its operation the reactor worked at an average power of 2MW, almost 3216 hours/year, producing a total energy of  $230 \times 10^3$  MW•h.

After 23 years of operation a large number of spent fuel elements became available for storage exceeding the stocking capacity of the small cooling pond near reactor.

Therefore, in 1980 was commissioned the nuclear spent fuel repository that contains at present all the fuel elements burnt in the reactor during years, minus 51 S-36 fuel ensembles, which are conserved in the cooling pond.

As a result of 40 years of intense utilization, 223 ensembles of burned fuel, were produced and stored in special ponds.

### 2. The spent fuel

The spent fuel ensembles used in the WWR-S reactor and stored in this repository contain two types of fuel: 153 pieces of EK-10 and 70 pieces of S-36.

The EK-10 and S-36 fuel assemblies have the same shape but distinct numbers of fuel roads and different percentage of U-235 enrichment.

So, the EK-10 contains 16 fuel roads with 10% enrichment and the S-36 contains 15 fuel roads with 36% U-235 enrichment. Total activity of the fuel assemblies is about  $10.69 \times 10^4$  Ci.

The most important fission products in fuel roads are Sr-90 and Cs-137. Depending on fuel assembly type the calculated values of the activity of these elements are:

1. EK-10:Sr-90  $\approx$  7,14  $\times$  103 Ci,Cs-137  $\approx$  7,67  $\times$  103 Ci2. C-36:Sr-90  $\approx$  8,21  $\times$  103 Ci,Cs-137  $\approx$  8,08  $\times$  103 Ci

#### 3. The repository

This repository contains 4 identical ponds, (Fig. 1) each of them having the depositing capacity of 60 fuel ensembles. Every pond having the outside sizes of 2750mm (length)  $\times$  900mm (breadth)  $\times$  5700mm (depth) is made from a special aluminum alloy (AlMg<sub>3</sub>), with the walls thickness of 10mm and bottom thickness of 15mm (Fig. 2). Pond's lids are made from cast iron having the thickness of 500mm; they provide only the biological protection for the maintenance personnel. A 1.5m concrete layer provides the lateral biological protection of the ponds.







Fig. 2. Spent Fuel Repository (Cross-Section View).

Over the fuel elements in every pond we can find a 4.5m water layer, which play the role of biological protection and coolant. Inside the ponds exist an aluminum rack, which contains 60 places for fuel storage. Serious safety problems concerning the condition of this fuel, particularly corrosion and leakage, must be avoid.

#### 4. Management and maintenance

The environment around the repository is permanently monitored; samples of soil and vegetation are periodically gathered and analyzed to see if some contamination with fission products is present. The status of every fuel ensembles is monitored and recorded in a database. Twice a year special inspection with a video camera is performed to observe structure modifications occurred from the last recording. The IAEA recommends water values of  $1\mu$ S/cm, but values up to  $2\mu$ S/cm might be accepted if the chlorine, copper and sulphate concentrations are very low. To maintain these parameters of the water in concordance with IAEA recommendations, it was designed and realized a filtration installation to improve the quality of the distilled water from the filling tanks (Fig.3).



Fig. 3. Distilled water filling system.

The manufacturer have specified the water conditions for the EK-10 and S-36 fuel assemblies, which they believe will avoid corrosion problems to be as follows:

Parameters	EK-10	C-36
- pH	5,5 - 7,5	5,5 - 6
- conductivity	5 µS/cm	2-3.3 μS/cm
<ul> <li>constant residuals</li> </ul>	8 mg/l	1 mg/l
- corrosion products	-	1 mg/l
- CF		0,02 mg/l
- O <sub>2</sub>		8 mg/l

Water chemistry is permanent monitored and if the quality is not good the entire quantity of bad water it will be replaced. An other problem is the detection of fuel assemblies with leakages. For this purpose was designed a special device for detecting and isolation of fuel assemblies with leakage.

Every fuel assembly will be isolated in a special vessel (Fig. 4) and the water around it will be measured (activity and nuclides components). After that we can draw conclusions about the fuel state and we can isolate the leaky fuel. We intend to elaborate a solution to confine the leaky fuel in a sealed aluminum eladding and after that to re-store it in the pond. Through this methods of detection, isolation and through a strictly water conditions we hope to extend the storage period till 30-40 years until the properly technology of final disposal will be choused and approved.



Fig. 4. Sipping test device.

### 5. Fuel Problems

Fuel is considered as spent nuclear fuel regardless of burnup when it is discharged from the reactor core for the final time. It is then normally placed in pools for cooling and interim storage until a final disposition is made. Some of this aluminum-clad spent fuel has been in water storage for more than 30 years and remains in pristine condition while other fuel has been some how degraded by pitting corrosion. Pitting corrosion of the fuel can lead to breach of the eladding material and release of radioactivity to the storage pond. Even most of the fuel assemblies are in good state there is some experimental one that is caring thermocouples. On this, the corrosion is much evident due to galvanic reactions (see Fig. 5). A special attention and often checks is given to the damaged fuel due to an improper manipulation in the reactor core (see Fig. 6). For this one, carefully examinations and maintenance procedures are done.



Fig. 5. Corrosion on experimental fuel assembly.



Fig. 6. Damaged fuel assembly.

Of course, the most important factor in controlling corrosion of aluminum-clad spent fuel assemblies is maintaining high quality water chemistry in the fuel storage pools. Until now, no special operational practices or techniques were applied to improve the water quality from the ponds. The distilled water produced at the site was used "as it is" without any serious concern regarding corrosion. In 1997 the first visual inspections were done to the reactor vessel and to the stored spent fuel revealing some corrosion traces. A number of 223 fuel assemblies were checked with an underwater camera and the information was recorded into a database. From that date all the data regarding the fuel were reviewed and a special attention was granted to water conditions and fuel state revaluation.

Table 1 showed the evolution of the main water parameters during the last three years, improved parameters are because of water changing.

Nr.	Date	Basin 2						Basin 3 E				Basin 4							
crt.		Cond	pН	Activity	Res.fix	CI	Ó	Cond.	pН	Activity	Res.fix	CI	0	Cond	pН	Activity	Res fix	CI	0
		[uS/cm]		Cs137[Bq/l]	[mg/i]	[mg/l]	[mg/l]	[uS/cm]		Cs137[Bq/l]	[mg/l]	[mg/l]	[mg/l]	[uS/cm]		Cs137[Bq/l]	[mg/l]	[mg/l]	[mg/l]
1	30.09.99	17.82	7.01	288	8.8	0		15.84	7.15	20636	7.9	0		19.8	7.05	1561	6	0	
2	28.10.99	17.16	7.44	838		0		15.84	7.21	22617		0		19.47	7.37	1370		0	
3	29.11.99	4.95	6.52	235		0		15.84	7,18	21750		0		19.8	7.34	1275		0	
4	16.12.99	4.95	6.47	185	1.6	0		15.83	7.08	22105	7.2	0		19.47	7.37	1108	6.9	0	
5	24.01.00	3.7	6.56	304		0		15.18	7.01	22120		0		14.52	6.67	1853		0	
6	28.02.00	4	6.58	295	_	0		7.92	7.1	22105		0		13.2	7.09	1847		0	
7	27.03.00	9.5	6.82	290	3.9	0.05		5.36	6.7	3769	3.8	0.05		19.43	7.09	1850	6.5	0.05	
8	24.04.00	6	6.59	226		0.05		7.03	6.68	3807		0.05		18.76	7.04	2132		0.05	
9	29.05.00	6.86	6.88	723		0.05		6.7	6.75	4721		0.05		17.2	7.19	1028		0.05	
10	20.06.00	8.04	6.74	153	7	0.05	8	7.52	6.63	4759	3.8	0.05	8	23.76	7.1	1218	6.3	0.05	8
11	25.07.00	11.88	6.84	418		0.05		9.24	6.74	4530		0.05		10.56	6.71	266		0.05	
12	27.08.00	9.83	6.68			0.05		8.53	6.63			0.05		9.24	6.27			0.05	
13	15.09.00	9.24	6.73	107	5.3	0.05		7.78	6.59	6065	4.5	0.05		8.58	6.75	179	4.8	0.05	
14	23.10.00	6.65	6.84			0.05		8.64	6.63	5914		0.05		8.25	6.71	314		0.05	
15	27.11.00	8.37	6.76	190		0.05		7.5	6.88	8224		0.05		9	6.76	457		0.05	
16	30.12.00	1.67	5.79			0.05		2.4				0.05		2.25	6.2			0.05	
17	22.01.01	11.3	4.81	0		0.02		13.86	4.55	1048		0.02		11.88	4.64	0		0.02	
18	19.02.01	19.43	4.09	0		0.015		21.44	4.46	1321		0.02		16.75	4.59	0		0.02	
										1635						1125			
19	19.03.01	21.12	4.93	920	18.1	0.015		26.4	4.38	Co60-1030	18.9	0.015		20.8	4.64	Co60-359	14.3	0.015	
20	23.04.01	23.1	4.86					27.72	4.25					22.11	4.5				
21	21.05.01	26.4	4.87					28.71	4.26					24.04	4.48				
										1675						278			
22	18.06.01	28.2	4.75			0.015		29.2	4.38	Co60-355		0.015		23.8	4.54	Co60-1422	0	0.015	
23	23 07 01			-		0.015		7.53	5.9	559		0.015				Co60-2964		0.015	
										/						103			
24	17.09.01	25	4.75	-	54.2	0.015		14.57	5.55	2102	9	0.015		26	4.5	Co60-3004	20	0.015	
24	21.10.01	27,7	4.69					15.16	5.72					28.3	4.56				

Table 1. Water parameters from spent fuel pools.

For such a small ponds with low water quality, the corrosion of aluminum-clad spent fuel is always a problem and can be solved only by operational practices using special filtration equipments and deionizers. A program to improve the water parameters was issued but the lack of funds for procurement of desired equipment make the achievement of this goal impossible yet.

At the moment the lonely viable solution to improve the water chemistry is water changing, but because no deionization can be done no high quality parameters can be obtain and maintained.

### 6. Conclusions

An optimum management of aluminum alloys in water environments can result in satisfactory durability of irradiated fuel cladding and the functionality of pool components for more than two decades. If pool operating parameters are systematically controlled the wet storage can be extended to more than 30 years.

All this operations mentioned above are important to keep the fuel cladding integrity as long as it is possible with this methods, and to prevent the irradiation of the personnel and environment contamination.

Mean time, new solutions for safe storage (e. g. dry storage) are studied and planed for the future use. During the 21 years of exploitation, was not encountered any major incident or accident or any other event regarding the health state of the operating personnel or environmental contaminations caused by the repository activity.

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# FUEL MANAGEMENT OPTIMIZATION FOR THE WWR-M RESEARCH REACTOR IN KIEV

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# ABSTRACT

Core loading patterns, number and types of fuel assemblies in the core as well as discharged fuel burnup are determined for the WWR-M research reactor in Kiev by the optimization procedure providing high neutron flux under the safety and fuel constraints. For neutronics calculation, the iterational hybrid method combining diffusion model with higher approximations of neutron transport equation is applied. The results of calculation are shown to be consistent with the results of measurement. To determine the best placement of fuel assemblies in the core, successive mixed-integer linear programming and backward diffusion calculation is used. An example of maximization of thermal neutron flux in large channels in the core is demonstrated.

# 1. Introduction

The WWR-M research reactor in Kiev is used for various purposes including neutron physics and materials research, radioisotope production and neutron transmutation doping of silicon. Thus, its fuel management should be flexible to correspond various objectives with different priorities and to provide the best possible result. Core loading patterns satisfying all the safety constraints and fuel requirements, fuel types used, number of fuel assemblies in the core and discharged fuel burnup should be determined by the optimization procedure using computer codes. To provide high reliability and safety of fuel, computer codes based on adequate models should be applied.

### 2. Neutronics Model

Diffusion approximation is invalid for neutronics modeling of the WWR-M reactor because of neutron streaming phenomenon in beam tubes. The Monte-Carlo method and other high-order approximations of neutron transport equation can be used for such calculations but they are very computationally intensive. The iterational hybrid method combining diffusion model with higher approximations of neutron transport equation has been developed for neutronics calculation of the WWR-M reactor [1]. This technique has been examined by comparing its results for 1-D and 2-D test problems to solutions obtained for the same problems using the Monte-Carlo and high-order discrete ordinate methods. High efficiency and accuracy of the iterational hybrid method and possibility of its application for neutronics calculation of the WWR-M research reactor using a personal computer has been proved [2].

The code VICA based on this method has been developed for 3-D neutronics calculation of the WWR-M reactor. The results of calculation using this code were compared to the results of measurement [3]. Some results are shown in Fig. 2 - 4 for the core configuration depicted in Fig.1. The total number of measurement data is about one hundred while average deviation of the calculated and measured values of the thermal neutron flux is about 7%. Errors in calculation of the effective multiplication factor for various core loading patterns are less than 0.5%.



Fig.1. WWR-M reactor core and reflector



Fig.2. Axial distribution of thermal neutron flux in cell 1



Fig.3. Axial distribution of thermal neutron flux in cell 2



Fig.4. Axial distribution of thermal neutron flux in cell 3

The code was examined also by the analysis of such research reactor accident as a damage of a large neutron beam tube located near the core with accompanying release of positive reactivity due to penetration of water. The penetration of water into the void tube was simulated by loading an ampoule filled up by water. The measured value of reactivity was 0.09  $\beta_{eff}$  while the calculated value was 0.12  $\beta_{eff}$ .

#### 3. Core Loading Pattern Optimization

The WWR-M reactor core loading pattern, fuel types used, number of fuel assemblies in the core and discharged fuel burnup are optimized using the code PORT [4]. Number and types of fuel assemblies as well as discharged fuel burnup are evaluated primarily for simplified models using an optimization algorithm based on backward diffusion calculation and linear programming [5]. As an example, solution of the problem  $p_n \Phi_{th}$ -C<sub>F</sub>  $\rightarrow$  max for 1-D model, where  $p_n$  is the "neutron price",  $\Phi_{th}$  is the thermal neutron flux in the channels of the reflector, and C<sub>F</sub> is the feed fuel cost, is demonstrated in Fig.5.



Fig.5. Optimal parameters for the problem  $p_n \Phi_{th}$ -C<sub>F</sub>  $\rightarrow$  max

To determine the best placement of real set of fuel assemblies in the core for realistic model, an algorithm based on the successive mixed-integer linear programming and backward diffusion calculation has been developed [6]. An example of application of the code PORT to maximize thermal neutron flux in large channels in the core is demonstrated in Table 1 and Fig.6.

Optimization problem	$\Phi_{51/58} \rightarrow \max$	$\Phi_{80/32} \rightarrow \max$	$\Phi_{51/58} + \Phi_{80/32} \rightarrow \max$
$\Phi_{51/58}$ , $10^{12}$ n/cm <sup>2</sup> /s/MW	16.7	12.5	15.8
$\Phi_{80/32}$ , $10^{12}$ n/cm <sup>2</sup> /s/MW	9.6	13.4	12.4

 Table 1. Thermal neutron flux in channels 51/58 and 80/32

 for different optimization problems



Fig.6. Optimal core loading pattern for the problem  $\Phi_{51/58} + \Phi_{80/32} \rightarrow \max$ 

To specify and control input data and review the results of neutronics and thermal-hydraulics calculation in convenient form, a graphical interface for automation and visualization of core design has been developed [4]. Information for all core reloads is recorded in database, thus whole history of each fuel assembly can be reviewed and analyzed.

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