INL/CON-07-13516 PREPRINT

The Idaho National Laboratory Beryllium **Technology Update**

8th IEA International Workshop on **Beryllium Technology**

Glen R. Longhurst

December 2007

This is a preprint of a paper intended for publication in a journal or proceedings. Since changes may be made before publication, this preprint should not be cited or reproduced without permission of the

author. This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, or any of their employees, makes any warranty, expressed or implied, or assumes any legal liability or responsibility for any third party's use, or the results of such use, of any information, apparatus, product or process disclosed in this report, or represents that its use by such third party would not infringe privately owned rights. The views expressed in this paper are not necessarily those of the United States Government or the sponsoring agency.

The INL is a U.S. Department of Energy National Laboratory operated by **Battelle Energy Alliance**



THE IDAHO NATIONAL LABORATORY BERYLLIUM TECHNOLOGY UPDATE

Glen R. Longhurst

Idaho National Laboratory, P.O. Box 1625, Idaho Falls, Idaho, USA

ABSTRACT

A Beryllium Technology Update meeting was held at the Idaho National Laboratory on July 18, 2007. Participants came from the U.S., Japan, and Russia. There were two main objectives of this meeting. One was a discussion of current technologies for beryllium in fission reactors, particularly the Advanced Test Reactor and the Japan Materials Test Reactor, and prospects for material availability in the coming years. The second objective of the meeting was a discussion of a project of the International Science and Technology Center regarding treatment of irradiated beryllium for disposal. This paper highlights discussions held during that meeting and major conclusions reached.

I. INTRODUCTION

Beryllium is used in fission research reactors as a means of enhancing the neutron flux by reflection and by (n, 2n) reactions. There are a number of issues relevant to the use of beryllium in reactors where there are uncertainties. One of these has been the available supply of metallic beryllium. Another relates to the composition and impurities in the beryllium. Mechanical properties are also of concern.

Early in 2007, discussions were held between the Idaho National Laboratory (INL) and the Japan Atomic Energy Authority (JAEA) regarding possible collaboration in providing irradiation services to the world community. One of the concerns for the restart of the Japan Materials Testing Reactor (JMTR) is the beryllium reflector. In particular, issues of cost, lifetime, and availability of material were important to resolve. The Advanced Test Reactor (ATR) at the INL also makes use of beryllium, as do several other research reactors around the world. When it was decided that there should be a meeting of JAEA and INL persons to discuss collaboration, it was suggested that perhaps persons with specific knowledge of beryllium technology should be invited to come to the INL for discussions particularly relevant to the concerns regarding beryllium technology.

A major research activity of interest to both INL and JAEA is supported by the International Science and Technology Center (ISTC), Project Number 3381, entitled, "Elaboration of Safe Methods for Radioactive Beryllium Waste Decontamination after Operation in a Nuclear Reactor." This work is being performed at the State Scientific Center of Russian Federation - Research Institute of Atomic Reactors (NIIAR) in Dimitrovgrad, Russia. Because the INL is one of the collaborators in this work, it was worthwhile and important to have a meeting of participants to discuss the scope and plans for the work that will be done under this project. The meeting proposed by JAEA and the INL would provide an excellent opportunity for the ISTC exchange to take place.

Further participation was graciously provided by Brush Wellman, provider of the beryllium currently used in the ATR and the JMTR. Representatives of the U.S. Department of Energy also participated in the meeting. Invitations were extended to other interested parties, including but not limited to SCK.CEN, the principal collator on the ISTC project mentioned, and the Ulba Metallurgical Plant in Kazakhstan, but for various reasons it was not possible for these other parties to participate.

II. MEETING PROGRAM

Five presentations were made at the meeting.

- Glen Longhurst of the INL discussed beryllium usage in the ATR and concerns for its disposal.
- Hiroshi Kawamura of JAEA discussed beryllium problems and concerns in the JMTR.

- Christopher Dorn of Brush Wellman presented a beryllium technology and business development update for ITER and other nuclear applications.
- Vladimir Chakin of NIIAR made an excellent presentation on beryllium research activity in the FSUE "SSC RF Research Institute of Atomic Reactors and plans for the ISTC Project No. 3381.
- Kay Adler-Flitton of the INL reviewed work that has been ongoing there to characterize the corrosion characteristics of buried beryllium.

Brief summaries of these presentations will be presented below. More detailed information on most of these topics will be available at this conference from the respective authors.

II. BERYLLIUM IN THE ATR

The ATR began operation in 1967 and has been used since as one of the world's most capable test reactors. The fuel is in a serpentine arrangement forming flux traps in each of the four corners and in the center. Surrounding the fuel is a beryllium reflector.

The reflector is fabricated in 8 separate blocks, each of which contains holes for the outer shim control cylinders, which are also made of beryllium with hafnium plate sleeves for reactor flux control. The reflector also contains a number of test positions where irradiations are performed in addition to the positions in the main flux traps.

Because of swelling and embrittlement, the beryllium reflector blocks and the outer shim control cylinders must be replaced at nominally 10-year intervals. In addition to the continuing need for large pieces of beryllium metal, issues of recent concern include disposal of the irradiated material. It has been found that typical impurity levels of uranium result in material that is classified as transuranic waste. This is material with alpha-emitters having atomic numbers greater than 92 and half-lives greater than 20 years in concentrations greater than 100 nCi/g. Transuranic wastes are not permitted in most radioactive waste disposal facilities.

After several investigations into processes for removing the transuranic contaminants, it appears that this waste beryllium can be classified as Greater-Than-Class-C waste and held for disposal with other radioactive material of like characteristics. A recent proposal that future reflector blocks be fabricated from beryllium with ultra-low levels of uranium was considered. It was decided that this option was not needed. A major contributor to that decision is that there is a large quantity of legacy material awaiting disposal in which the transuranic contamination will need to have a specific pathway defined. Additional beryllium with the same radiological characteristics can be dealt with at minimal cost using the same procedures.

The ATR should be in use until about 2050. That will result in the need to purchase two complete beryllium reflectors in addition to one that is now being procured. If a new fast-flux test capability is installed, the needs for beryllium in ATR would increase by about 25%.

III. JMTR BERYLLIUM ISSUES

The JMTR operation was stopped in 2005. A plan is now in place to refurbish the reactor and begin operation again in 2011 for an expected period of about 20 years. Part of that refurbishment is the replacement of the beryllium reflector. While conventional hot-pressed beryllium powder is planned for many components, another concept in the design modification planned for the reflector is the replacement of solid beryllium blocks with beryllium pebbles. These pebbles can be produced by the rotating electrode process and have the advantage that swelling and embrittlement will not be major problems.

Reflector lifetime is a significant concern in the JMTR life extension planning primarily because of cost. The planners are very interested finding ways of increasing the lifetime of beryllium. In addition, they are investigating the possibility of recycling the irradiated beryllium, not only to mitigate waste disposal concerns but as a means of cost savings. A cooperative research project has been arranged with the Ulba Metallurgical Plant in Kazakhstan starting in Oct., 2007. Additionally, there is an ISTC project sponsored by the Japanese Government starting in October, 2007 and running for two years. The work will be performed by the National Nuclear Center of Kazakhstan with Kazatomprom.

Waste issues for irradiated beryllium in Japan are similar to those found elsewhere.

Factors strongly influencing beryllium lifetime in reactors include irradiation temperature, fabrication methods and material purity. Beryllium in the JMTR will be irradiated at temperatures of 50 to 150°C. Fabrication techniques include hot pressing powder and, for pebbles, rotating electrode production. Purity of 98.5% beryllium is sought.

Research is being conducted to determine mechanical properties of irradiated beryllium and compare results with similar tests on unirradiated beryllium. Before the JMTR stopped operation, beryllium specimens were irradiated. Tensile and impact strength tests are now being performed on those specimens and on corresponding specimens of unirradiated vacuum hot pressed S-200F and S-65C. Bending tests are being performed on vacuum hot pressed S-65C. No radiation induced bending or swelling were observed on a specimen irradiated to 1×10^{25} n/m² at a nominal temperature of 200°C.

IV. BRUSH WELLMAN TECHNOLOGY AND BUSINESS DEVELOPMENT UPDATE FOR ITER AND OTHER NUCLEAR APPLICATIONS

Brush Wellman has undertaken some organizational changes to better serve the needs of the nuclear applications community. There is a new Chief Executive Officer in the Brush Wellman Engineered Materials branch of the company, a new President of Beryllium Products, and a new Vice President of Sales & Marketing. The company has developed a new long-range strategic direction.

An important modification to the company structure is the creation of SPADE (Special Products and Applications Development Engineering). SPADE offers concurrent engineering, detailed part design, analysis support, prime contracting, and access to new technologies. With the retirement of Don Kaczynski, the company has launched a search for a new Technical Director.

Among projects recently completed is the ISIS Neutron Reflector at the Rutherford Appleton Laboratory. This is an integrated assembly involving beryllium, aluminum and cadmium. The company has also been active in an ITER-like wall project, working with the European Fusion Development Agreement (EFDA) and the Joint European Torus (JET).

With regard to beryllium supply, Brush Wellman has been in contact with all the ITER participants, seeking to establish needs and develop planning to provide the required material. Orders have been placed for their S-65C grade material. Brush Wellman is pursuing the acquisition of a material export license to China. There are several applications for beryllium in ITER including plasma-facing structures, tritium breeding modules, and vacuum windows. Studies are being conducted to determine the preferred orientation of beryllium crystals in the plasma-facing material.

Brush Wellman suspended reducing ore to metallic beryllium early in this decade because of more stringent environmental requirements. Since then, orders for metallic beryllium have been filled from various available stockpiles. A new reduction facility is now under construction at their Elmore, Ohio plant, one that will allow the company to once again produce the primary beryllium metal.

V. BERYLLIUM RESEARCH AT THE FSUE SSC RF RESEARCH INSTITUTE OF ATOMIC REACTORS

The Research Institute of Atomic Reactors (RIAR) is located at Dimitrovgrad, in the Ulyanovsk region of Russia. Among the nuclear reactors there are the SM reactor, the BOR-60 reactor, and the MIR. The SM has one of the highest neutron fluxes of all research reactors in the world. This allows accelerated tests of materials up to large neutron fluences and production of transuranium elements and isotopes with a very high activity. There are support facilities for studying corrosion under pressurized water reactor conditions, thermal cycling effects. neutron activation, and certifying radionuclide compositions of test samples. Bor-60 is a sodium-cooled fast reactor where research is performed on many different materials at a wide range of temperatures, up to 1.000°C. Fast reactor safety is also a main topic of BOR-60 research. The MIR is a channel type research reactor with water coolant and a beryllium moderator and reflector. The operating and loop channels are located in the central holes of the beryllium moderator blocks. The main purpose of the reactor is to perform long-term tests and special experiments with fuel rods and assemblies, both for operating and advanced reactors of different types.

In addition to the three large reactors just mentioned, there is a complex of three pool-type research reactors in the Institute. RBT-6 was commissioned in 1975, RBT-10/1 commenced operating in 1983, and in 1984 RBT 10/2 was commissioned. The RBT type reactors are pool-type reactors, which use spent fuel assemblies of the SM reactor as fuel. These reactors feature maximum design simplicity and flexibility, high accessibility of experimental devices during reactor operation, and simplicity of transport operations. They provide an effective after-burning of the SM reactor fuel. There are specialized devices for in-pile mechanical tests, in-pile investigations of thermo- and electro-physical properties of materials, testing fuel elements and structural materials, producing short-lived radioactive nuclides 99Mo and 133I, and for nuclear silicon doping.

There are extensive laboratory research facilities at the RIAR Materials Testing Complex. These support a very wide spectrum of destructive and nondestructive analyses for examining radiological, mechanical, structural, and chemical aspects of materials. Many of these instruments are in large hot cells where the work can be performed on highly radioactive materials.

There are a number of problems with use of beryllium as a neutron reflector and moderator in fission reactors. Among these are the absence of beryllium production in Russia, the high price of beryllium, the degradation of physical-mechanical properties of beryllium under irradiation, cracking, and problems with disposal of radioactive beryllium waste. The SM reactor has beryllium reflector blocks arranged along the core perimeter and beryllium neutron traps located in its center. In the MIR reactor beryllium hexagonal blocks make up the core. This beryllium block material has a life-limiting neutron fluence of 6×10^{22} n/cm² (E>0.1 MeV). This results in high induced activity of beryllium. When discharged from the reactor, the beryllium blocks require disposal in a high-level storage facility of limited capacity. These facts necessitate investigations on optimization of high-level beryllium waste decontamination and disposal, e.g. categorization into low-level waste.

Extensive research has been conducted on the response of beryllium to irradiation in reactors. Swelling, hardness, tensile strength, thermal conductivity, corrosion, and microstructure have all been shown to be markedly influenced by irradiation.

The ISTC project now underway at RIAR has the main objective of investigating and optimizing the processing and disposal of irradiated beryllium waste. Its main tasks are

• Theoretical definition of the final elementary composition and contribution of radioactive elements to the resulting radioactivity of irradiated beryllium blocks

- Study the effect of high-dose neutron irradiation on physical-chemical properties and microstructure of irradiated beryllium
- Experimental evaluation of the surface decontamination effectiveness of irradiated beryllium blocks
- Development of decontamination methods for removal of radioactive metal transmutant-impurities from irradiated beryllium
- Investigation of the effect of high-temperature annealing on volume decontamination of irradiated beryllium blocks
- Choice and study of getter materials to provide efficient sorption of tritium releasing on high-temperature annealing of irradiated beryllium
- Estimation of the radioactivity level and physical -chemical state of irradiated beryllium blocks after various types of treatment

Implementation of this project will contribute to more optimized use of costly radioactive waste storage facilities and environmental improvements. The experience gained from the research will also be useful for American, European and Japanese specialists in this field.

VI. UNDERGROUND CORROSION OF BERYLLIUM

Corrosion tests have been underway for more than 6 years in a location very near the Subsurface Disposal Area (SDA) where beryllium components from INL reactors have been buried. This site has identical soil and weather characteristics with the actual burial site. The objective is to evaluate the rate at which corrosion takes place on a variety of materials under these conditions.

The corrosion coupons are 21 x 21-cm plates of material, typically 3-mm thick. Each of these has a nominally 2-cm hole in the center which allows it to be positioned on an electrically non-conducting rod. The beryllium specimens were made from Brush Wellman S-200F rolled plate.

Test specimens were installed, vertically oriented (mounting rods horizontal) at depths of 1.22 m and 3.05 m below the surface. Soil was returned to nearly its original location and packed to achieve densities similar to soil around the actual buried wastes in the SDA. Samples have been retrieved from these installations at times of 1-year, 3-years, and 6-years after the specimens were placed in the test location. The retrieved specimens underwent surface cleaning, and the extent of corrosion has been measured, noting corrosion rate and typical pit depth. Observations are tabulated in Table 1.

Exposure	Test Depth	Average	Pit Depth
	(m)	Corrosion	(µm)
		Rate	
		(mm/y)	
l-yr	1.22	2.007E-3	113
	3.05	4.540E-3	180
3-yr	1.22	4.635E-4	80
	3.05	7.248E-3	115
6-yr	1.22	2.160E-4	80
	3.05	6.633E-3	115

Table 1. Beryllium corrosion data.

VII. SUMMARY

The Beryllium Technology Update meeting held at the INL provided an excellent opportunity for exchange of information relative to the use of beryllium in fission reactors. The prospects for beryllium availability for future use in reactors, both fission and fusion are good. The principal hinderances of material cost and problems with disposal are being addressed. Several methods have been proposed to reduce or manage the radioactive contamination from uranium impurity. Research is proceeding both commercially and by government laboratories in the U.S. and Russia to facilitate disposal and find ways of reducing the cost of beryllium usage by lifetime extension.

ACKNOWLEDGMENT

This work was performed for the U.S. Department of Energy, Assistant Secretary for Nuclear Energy under DOE-NE-ID Operations Office Contract DE-AC07-05ID14517.