

Beryllium Use in the Advanced Test Reactor

8th IEA International Workshop on Beryllium Technology

Glen R. Longhurst
Rhonda D. Rohe

December 2007

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



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.

BERYLLIUM USE IN THE ADVANCED TEST REACTOR

Glen R. Longhurst and Rhonda D. Rohe

Idaho National Laboratory, P. O. Box 1625, Idaho Falls, Idaho 83415-3860, USA

ABSTRACT

The Advanced Test Reactor (ATR) at the Idaho National Laboratory (INL) began operation in 1967. It makes use of a unique serpentine fuel core design and a beryllium reflector. Reactor control is achieved with rotating beryllium cylinders to which have been fastened plates of hafnium. Over time, the beryllium develops rather high helium content because of nuclear transmutations and begins to swell. The beryllium must be replaced at nominally 10-year intervals. Determination of when the replacement is made is by visual observation using a periscope to examine the beryllium surface for cracking and swelling. Disposition of the irradiated beryllium was once accomplished in the INL's Radioactive Waste Management Complex, but that is no longer possible. Among contributing reasons are high levels of specific radioactive contaminants including transuranics. The INL is considering disposition pathways for this irradiated beryllium, but presently is storing it in the canal adjacent to the reactor. Numerous issues are associated with this situation including (1) is there a need for ultra-low uranium material? (2) Is there a need to recover tritium from irradiated beryllium either because this is a strategic material resource or in preparation for disposal? (3) Is there a need to remove activation and fission products from irradiated beryllium? (4) Will there be enough material available to meet requirements for research reactors (fission and fusion)? This paper discusses the present status of considerations on these issues.

I. INTRODUCTION

The Advanced Test Reactor (ATR) was built at the Idaho National Laboratory in response to needs for high-flux, large volume irradiation testing of advanced reactor fuels and materials. The reactor first went critical in July 1967. Since that time it has served as a premier test bed for investigating the effects of neutron irradiation on a wide variety of materials and the performance of a number of reactor fuel concepts.

The ATR, with its unique serpentine core design (Figure 1) is really 5 nearly independent reactors in one. Each of the 4 corner lobes can be controlled independently, and the center region operates with contributions from each of the corner lobes. Each of the 5 regions is capable of generating 50 MW_{th} for a total rated reactor power of 250 MW_{th}. In recent years, most of the operation has been at the lower power level of 110 – 120 MW_{th}.

There are 9 flux traps in the reactor core, one in each of the 4 corner lobes, one on each side, between each of the corner lobes, and another at the core

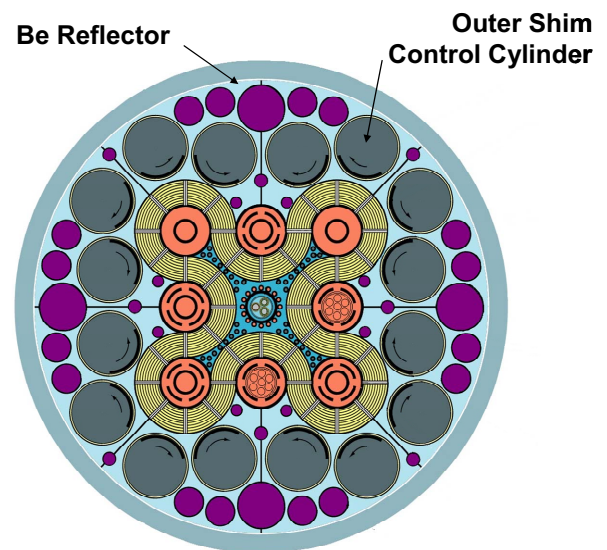


Figure 1. Cross section of the ATR core showing key beryllium components.

center. These flux traps have the same outer dimension but are of two sizes in the test space: 127 mm diameter in the northwest and northeast lobes

(top outside corners in Figure 1) and 76-mm diameter in the other flux traps. The smaller flux traps have hafnium safety rods (arc sectors) taking up space around the outer periphery of the flux traps. Five of the flux traps have pressurized water loops installed (Figure 2) where coolant conditions representative several different kinds of reactors can be replicated.

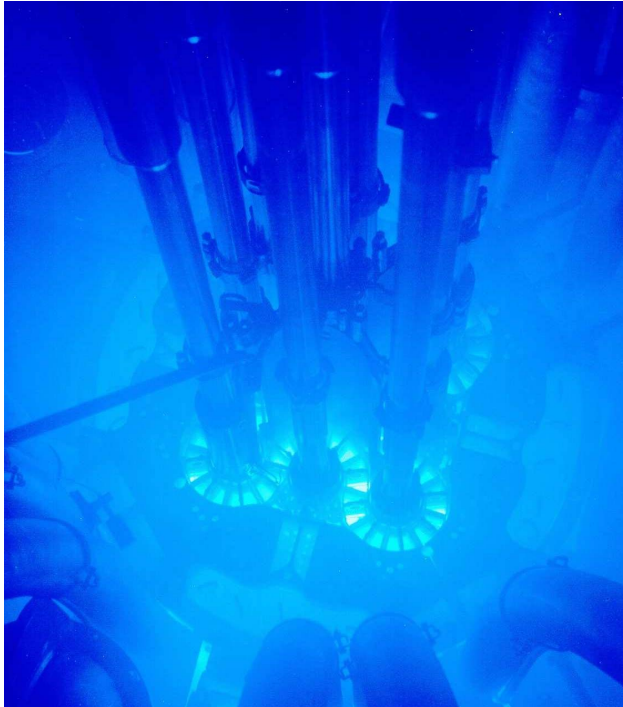


Figure 2. Operating ATR core showing pressurized water loops in the flux traps.

II. BERYLLIUM COMPONENTS

Beryllium is essential to the operation of the ATR. Surrounding the serpentine fuel elements (Figure 1) is the beryllium reflector. This structure enhances neutron flux in the core through neutron reflection and (n, 2n) reactions. Within the reflector are 16 outer shim control cylinders (OSCCs). Configured as shown in Figure 3, these are 18.2-cm diameter beryllium cylinders with 63-mm thick hafnium plates covering 120 degree arc sectors at the surface. The hafnium is a strong neutron absorber. When the OSCCs are rotated such that the hafnium plates are near the fuel, they absorb neutrons, maintaining core criticality with fresh fuel whose neutron production is higher than needed for operation. As the fuel is burned and specific neutron production falls off, the OSCCs are rotated away from the fuel, absorbing fewer neutrons and maintaining the core power at the desired levels.

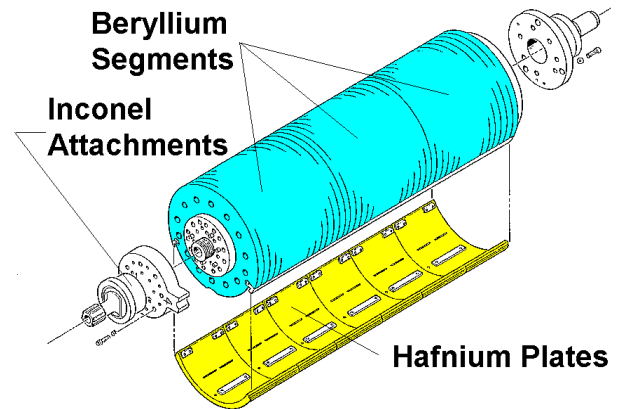


Figure 3. Outer shim control cylinders are comprised of three beryllium segments, hafnium plates for neutron absorption, and other pieces.

Also in the beryllium reflector are a number of test positions (see Figure 1). These range in size from 22.2-mm diameter in locations next to the fuel elements to 127-mm diameter at the four cardinal directions from core center. All of these test positions are 121 mm in length, running the full height of the core.

The beryllium reflector is fabricated in 8 separate sectors (Figure 4), 4 each in left hand and right hand configurations. Each of these sectors is originally pressed as a 740-kg billet that is machined down to 84.5 kg. The material for these pieces is Brush Wellman S200-F beryllium that has been hot pressed and sintered. There are specific limits on impurities for 13 different elements and the catch-all requirement that all other impurities must be less than 400 appm. There is no specific requirement for uranium impurity, but this naturally occurring contaminant has had substantial consequences for the ATR.

The beryllium in the ATR, as in most beryllium-reflected reactors, requires periodic replacement. To understand why, consider the neutron reactions for beryllium shown in Figure 5.

The He and H produced in the various reactions generates swelling. The amount of swelling depends on the temperature at which the irradiation takes place, as is illustrated in Figure 6 from Wolfer and McCarville.¹ Swelling of about 0.5% (5000 appm He) is typical at replacement in the ATR, although the specific time of replacement is determined by visual inspection for cracks and dimensional changes.

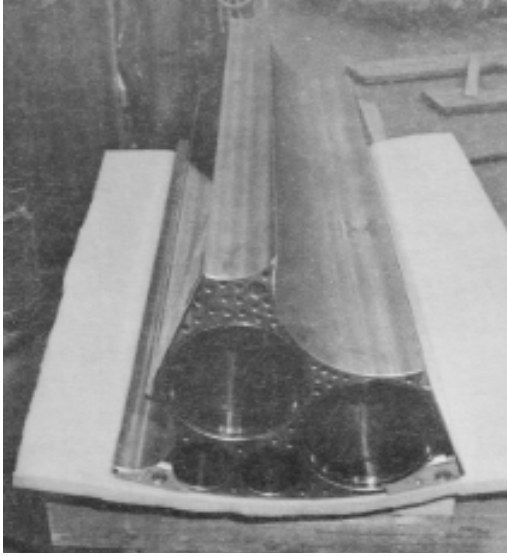


Figure 4. ATR beryllium reflector block as machined.

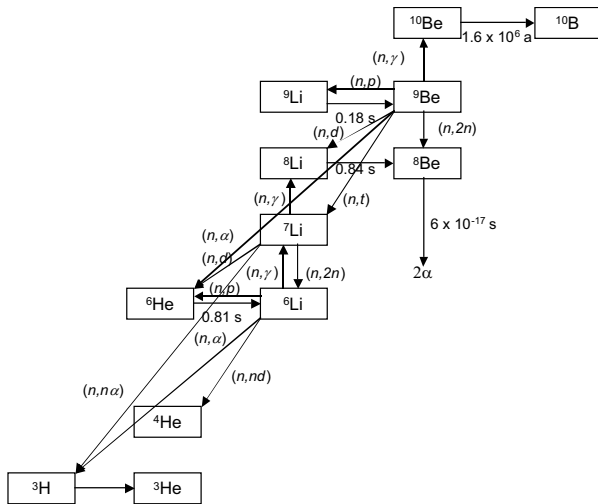


Figure 5. Nuclear reactions resulting from neutron irradiation of beryllium.

Since the ATR began operation in 1967, there have been four planned core internals change-outs (CICs), when the beryllium reflector blocks were replaced. These took place in 1977, 1986, 1994, and 2004. The first CIC, in the early 1970s, was not planned but was necessary when the reflector blocks developed major cracks. As a consequence, blocks since then have deep stress relieving saw cuts perpendicular to the core axis. The OSCCs installed during original plant construction were reused for reflector II, and were changed out in 1977. After that, OSCCs were replaced with the reflector blocks. Including the original reflector, five beryllium reflectors (40 blocks) have been removed from the ATR. The sixth beryllium reflector was recently

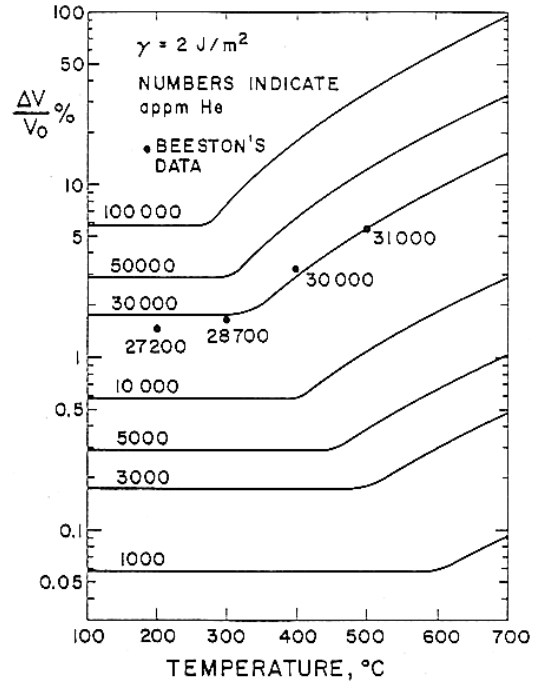


Figure 6. Irradiation induced swelling in beryllium.

installed in the ATR. A seventh reflector is currently in process of being procured for a probable 2014 CIC.

III DISPOSAL ISSUES

These beryllium components have great advantages for operation, but their disposal has recently become a problem. Some reflector blocks and OSCCs from the first few ATR cores were disposed of at the INEEL Radioactive Waste Management Complex (RWMC) in earthen vaults. The blocks from the 1986 CIC (core III) were kept in the canal until 1993 when six of the blocks were sent to the RWMC and placed in retrievable storage.² At the present time, there remain in the canal 20 reflector blocks (8 from core V, 8 from core IV, and 4 from earlier cores) and 55 OSCCs, pending removal to a more permanent storage facility. The reason disposal is difficult is the content of radioactive contaminants compared with acceptance limits for the disposal site.

III.A. Uranium

A major concern has been the presence of transuranics, produced from uranium impurity in irradiated beryllium. The presence of the uranium impurity in the beryllium metal was not identified nor incorporated into INL computer characterization modeling before 2002. Uranium had not been reported as present in previous beryllium reflector

blocks. Uranium was not specifically limited in the ATR beryllium procurement specification nor was it indicated on the vendor supplied chemical assay data sheets that are routinely provided to the INL. The ATR procurement specification limits all metallic impurities in the beryllium to less than 400 weight parts per million (wppm).

The presence of uranium in the new beryllium blocks procured for the ATR Core VI was determined from detailed chemical assay sheets from Brush-Wellman. Those concentrations ranged from 23 wppm to 105 wppm with an average concentration of approximately 71 wppm. Calculations³ indicated that even at 30 wppm, the concentrations of alpha-emitting radioactive isotopes with atomic numbers greater than 92 and half-lives greater than 20 years would exceed 100nCi/g in the ATR beryllium, which is the threshold for transuranic (TRU) waste. Some of these activation processes are shown in Figure 7.

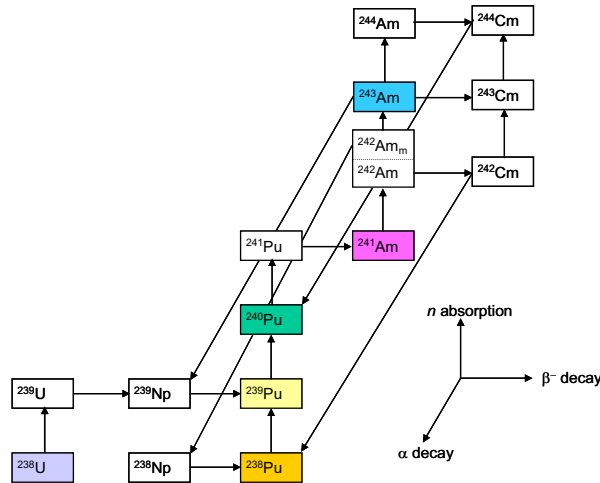


Figure 7. Activation processes for uranium impurity in beryllium.

No existing waste repositories could be found that would accept ATR's transuranic waste beryllium. Even the Waste Isolation Pilot Plant (WIPP) in New Mexico, which accepts transuranic waste from defense programs, could not accept ATR beryllium, in part because of its large ^3H inventory.

III.B. Tritium

An additional complicating factor for beryllium disposition is tritium (^3H). The inventory of ^3H in a beryllium reflector when it comes out of the reactor is about 2.2 MCi. Measurements of ^3H were made at the INL (then Argonne National Laboratory-West) on

samples taken from the reflector blocks in the ATR canal. Reported values range from 60–350 $\mu\text{Ci/g}$.

These samples were from low-flux positions at the outer edge of the reflector blocks, so ^3H concentrations should have been low. However, several measurements have been made on samples of beryllium irradiated in ATR to higher fluences. An early one was extraction by dissolution of a beryllium sample irradiated as part of the Phillips Atomic Energy Division Surveillance (PAEDS) program.⁴ These samples were of the same S-200E beryllium used for the first ATR reflectors and were cylinders 1 cm in diameter and 3 cm long. Neutron fluence was $5.5 \times 10^{22} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$). Expected ^3H inventory at the time of the experimental measurement was 32.6 Ci in the 4.7-g sample. ^3H actually measured was 32.8 Ci (6.9 Ci/g) in the specimen. Of this, 32 Ci was found to be in the form of elemental gas, probably resident in gas bubbles in the beryllium. The remaining 0.8 Ci went into solution in the dissolving acid, indicating it was probably resident as a hydroxide, probably at oxide inclusions, or as a hydride in the beryllium lattice.

Another measurement of ^3H in PAEDS samples was in a demonstration of heat-only-extraction of ^3H from beryllium.⁵ Gamma spectrometry showed that each sample contained 1.2 mCi of ^{60}Co , 33 μCi of ^{137}Cs , and a smaller quantity of ^{134}Cs . Two samples were tested. Quantitative ^3H release was not reported for the first, but it seemed to agree well with the 25–30 Ci of ^3H thought to be in the sample. The second was heated to melting and released 24.6 Ci of ^3H (5.2 Ci/g), in good agreement with expectations. It is interesting that none of the tritium escaped at temperatures below 650°C. Melting caused 87 percent of the ^{137}Cs to be released, but none of the ^{60}Co came out. One reason for the difference was probably the very high vapor pressure of Cs at that temperature.

An additional thermal desorption measurement of ^3H in a PAEDS sample was conducted at Pacific Northwest Laboratory where the tritium content was found to be 2,500 appm (8 Ci/g).⁶ There, too, tritium was not released until temperatures were in excess of 600°C.

While the ^3H in irradiated beryllium is tightly bound in the beryllium lattice, it is released as the beryllium corrodes. Corrosion is common in ordinary radioactive waste soil depositories. The result is a plume of ^3H observable in ground water and in the atmosphere over the burial site.²

III.C. Radioactive Carbon

One further complication is ^{14}C . Approximately 19% of the total ^{14}C in the INL Subsurface Disposal Area at the RWMC is associated with buried irradiated beryllium.⁷ That 19% constitutes about 90% of the mobile ^{14}C , which is a problem for containment of radionuclides in the disposal facility. The ^{14}C is an activation product from ^{14}N , which is virtually impossible to keep out of the beryllium because the powder from which it is formed is processed in air. The amount present is lower in some beryllium grades than others because of differences in processing procedures. Measurements of nitrogen and ^{14}C inventories in samples taken from the reflector blocks presently stored in the canal, combined with detailed computer simulations of their operating history in the reactor, have demonstrated that the ^{14}C levels in the reflector blocks are lower than the 80 Ci/m³ limit for Class-C designation when ^{14}N concentrations in the beryllium are less than 300 appm.

III.D Niobium

^{94}Nb is formed by neutron absorption in ^{93}Nb impurity. The concern for ^{94}Nb arose because of previously unsuspected levels of ^{93}Nb measured in the beryllium of the ATR reflector blocks. Measurements of ^{93}Nb were in the range 1–23 wppm, up to two orders of magnitude greater than the previously accepted value of 0.2 wppm.

It is believed that the reflector blocks will be greater than 10 CFR 61 Class C (GTCC) for disposal purposes because of high ^{14}C and ^{94}Nb levels. Even though they meet the Class-C requirement for ^{14}C and ^{94}Nb specific activity individually, the total activity, from modeling based on measurements taken, is great enough (sum-of-fractions greater than 1) to make them GTCC. Though no measurements have yet been made on the OSCCs, available information suggests that some, if not most, of these will also be GTCC.

IV. RECOMMENDATIONS FOR BERYLLIUM DISPOSITION

Early considerations addressed the possibility of removing the transuranics, the ^3H , and possibly even the other beta-emitters from the irradiated beryllium. Several possibilities were considered. The process for getting the ^3H out by thermal desorption is quite well understood.⁸ An INL research project explored removal of heavy metals using chemical dissolution.⁹ Alternative means of purification including gaseous

chlorine reaction, distillation, and zone purification have also been suggested.

An INL internal ATR beryllium disposition alternative study was conducted which strongly supports the following steps for beryllium management and disposal.

- Continue storage of ATR beryllium in the ATR canal until a repository is available.
- Classify the ATR beryllium as GTCC-like waste and include it in DOE's complex-wide inventory of waste with similar characteristics, and co-disposition with such waste.
- Develop a current radiological inventory of beryllium waste presently stored in the ATR canal for comparison with potential storage and transportation container requirements.
- Establish clearly defined requirements with respect to the National Environmental Protection Act (NEPA) and ensure ATR beryllium-specific NEPA-relevant issues are considered and appropriately addressed for past and future CICs.
- Participate in and support development of national R&D strategies directed toward beryllium processing/recycling.

The only presently defined requirements for DOE waste to be included with the GTCC waste is that it has similar characteristics and would not otherwise have a path to disposal. The ATR beryllium meets these requirements.

Efforts to develop processes for transuranic contaminant removal have been deferred pending further determination of disposal pathways. If it is found that the ultimate disposition pathway requires removal of the radioactive contaminants from irradiated beryllium, those efforts will be resumed. The available beryllium resource is sufficient to meet anticipated needs such that recovery of irradiated beryllium for the beryllium's sake is unnecessary. Consideration was given several years ago to ^3H removal from irradiated beryllium for ^3H resource recovery, but it was decided that the costs for such recovery were greater than the value of the ^3H that would be recovered.

V. FUTURE BERYLLIUM NEEDS

The ATR is presently beginning procurement of the 7th reflector set. That beryllium will probably be

installed in about 2014. Anticipating ATR operation to the year 2050, it is likely that two and possibly three additional reflectors beyond the 7th will be purchased.

A concept for conducting fast flux testing in the ATR is presently being considered. It would require one lobe of the ATR to be operated at nominally twice its usual power level. That would mean the reflector blocks surrounding that lobe would require replacement at 5-year intervals rather than the typical 10 years. It would create additional demand for ATR beryllium. It should be emphasized that this change in ATR operation is only at the conceptual stage with no determination to proceed.

Regarding the composition of future beryllium purchases, one suggestion for reducing the production of irradiated beryllium waste with high TRU inventories is to remove the uranium from beryllium before it is fabricated into components and irradiated in the reactors. Brush Wellman has identified the possibility of supplying beryllium with uranium impurity levels in the vicinity of 1 appm. This material would be furnished at a necessary cost premium, but its other properties would be essentially the same as those of the normal S-200F grade.

There are two major reasons the low-uranium beryllium was not considered as necessary for ATR. One is that the current material specification provides material adequate for ATR operation. Administrative processes are such that making any changes to the technical specification for ATR beryllium would be a very time-consuming and expensive process. While there is no technical reason to believe that the mechanical and neutronic properties of the beryllium would change, except for the lesser transuranic contaminant production, the very conservative operational requirements for the ATR mean that mechanical properties of the new material would have to be demonstrated, and doing so would incur substantial additional cost. The second major consideration is that there is on hand, awaiting disposal or now in the reactor, contaminated beryllium in a quantity approximately equivalent to that which would be procured with low uranium concentration. A process or pathway for dealing with this legacy material will need to be developed regardless of what is done regarding future procurements. Any such process or pathway would be capable of dealing with the material yet to be irradiated in the ATR. A further consideration was the difficulty in measuring uranium in beryllium at levels low enough to assure that the transuranic issue would be fully resolved given present levels of

uncertainty in the predictive computer models. Verification would require the material to be produced, irradiated in the reactor, and then analyzed for transuranic content. That also would be expensive and time consuming. For these reasons, it was determined that it would not be in the ATR's best interest to pursue development of beryllium with very low levels of uranium at this time.

VI. CONCLUSIONS

Like many research reactors, the ATR benefits greatly by having beryllium as its neutron reflector. Use of beryllium has a number of challenges as well as benefits. Because it must be replaced at regular intervals, there is a need for a disposal pathway for material that has been irradiated. Disposal of irradiated beryllium is complicated by activation products, which include ³H from the beryllium itself, beta-emitting isotopes from common impurities such as ¹⁴N and ⁹³Nb, and alpha-emitting transuranic isotopes from uranium impurity.

While there is no immediately available repository for that disposal, it has been determined that beryllium irradiated in the ATR can be designated as Greater-Than-Class-C waste, for which a path is expected in the not-too-distant future.

Another interesting suggestion has been that by improving the quality of the beryllium, such as by reducing grain size, it may be possible to retard swelling of the beryllium for a given neutron dose and thus get longer life in the reactor. That is an interesting possibility that needs further exploration.

The ATR will continue to make use of beryllium for the next several decades, but decisions regarding changing the specification of the beryllium to reduce impurities or provide longer lifetime in the reactor have not yet been settled upon.

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.

REFERENCES

1. Wolfer, W. G., and, T. J. McCarville, July 1985, "An Assessment of Radiation Effects in Beryllium," *Fusion Technology*, 8, pp. 1157-1164.

2. Ritter, P. D. and D. L. McElroy, *Progress Report: Tritium and Carbon-14 Sampling at the Radioactive Waste Management Complex*, INEEL/EXT-98-000669, March 1999.
3. Mullen, C. K. et al., March 2003, *Beryllium Waste Transuranic Inventory in the Subsurface Disposal Area, Operable Unit 7-13/14*, INEEL/EXT-01-01678, Rev. 2, Idaho National Engineering and Environmental Laboratory.
4. Beeston, J. M., et al., June 1990, *Gas Retention in Irradiated Beryllium*, EGG-FSP-9125, Idaho National Engineering and Environmental Laboratory.
5. Anderl, R. A., and J. D. Baker, October 14, 1994, *Tritium Extraction from Irradiated Beryllium, Project Summary Report (SDRD-03)*, Idaho National Engineering and Environmental Laboratory.
6. Billone, M. C., et al., May 1991 "Tritium and Helium Behavior in Irradiated Beryllium," *Fusion Technology* 19, pp. 1707-1714.
7. S. L. Lopez et al., January 2005, *Summary Report for the OU 7-13/14 Early Actions Beryllium Encapsulation Project*, INL/EXT-04-00646, Rev. 1. Idaho Completion Project, BWXT Idaho, LLC.
8. Anderl, R. A., J. D. Baker, G. L. Bourne, and R. J. Pawelko, 1995, "Tritium and Helium Release from Irradiated Beryllium," Fifth Topical Meeting on Tritium Technology in Fission, Fusion, and Isotopic Calculations, May 28 to June 3, 1995, Belgirate, Italy.
9. Tranter, T., R. Tillotson, N. Mann, and G. Longhurst, 2006, "Processing Irradiated Beryllium for Disposal," *Proceedings of the 7th International Workshop on Beryllium Technology*, INL/EXT-06-01222, February 2006.