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The status of beryllium research for fusion in the United States

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Use of beryllium in fusion reactors has been considered for neutron multiplication in breeding blankets and as an oxygen getter for plasma-facing surfaces. Previous beryllium research for fusion in the United States included issues of interest to fission (swelling and changes in mechanical and thermal properties) as well as interactions with plasmas and hydrogen isotopes and methods of fabrication. When the United States formally withdrew its participation in the International Thermonuclear Experimental Reactor (ITER) program, much of this effort was terminated. The focus in the U.S. has been mainly on toxic effects of beryllium and on industrial hygiene and health-related issues. Work continued at the INEEL and elsewhere on beryllium-containing molten salts. This activity is part of the JUPITER II Agreement. Plasma spray of ITER first wall samples at Los Alamos National Laboratory has been performed under the European Fusion Development Agreement. Effects of irradiation on beryllium structure are being studied at Oak Ridge National Laboratory. Numerical and phenomenological models are being developed and applied to better understand important processes and to assist with design. Presently, studies are underway at the University of California Los Angeles to investigate thermo-mechanical characteristics of beryllium pebble beds, similar to research being carried out at Forschungszentrum Karlsruhe and elsewhere. Additional work, not funded by the fusion program, has dealt with issues of disposal, and recycling.

1. INTRODUCTION

Beryllium has been used in nuclear applications for many years. Traditionally, beryllium has been chosen for its neutronic properties. The strong (n.2n) cross section combined with its low atomic mass has made beryllium an ideal material for neutron reflectors. Many fission test reactors have used beryllium reflectors, and it is an integral part of nuclear weapons. For magnetic-confinement fusion, beryllium has been included in many blanket designs, both as simple blocks and in pebble beds. It has also been considered and experimented with as a plasma-facing material. Its low atomic number means that surface atoms incorporated into the plasma will be fully stripped of electrons and thus have essentially no line radiation from the plasma. Its high affinity for oxygen makes beryllium an excellent oxygen getter, further improving the plasma effective Z and improving performance. It is also incorporated into some target designs for inertial-confinement fusion.

Notwithstanding these many admirable qualities, there are a number of issues associated with its use. A concern that has received considerable attention is the biological hazard associated with beryllium inspiration, chronic beryllium disease (CBD). Extensive efforts are underway to develop methods of identifying sensitivity to beryllium in at-risk individuals. There is a need for real-time beryllium air concentration monitoring at the low levels now recommended for worker exposure.

In fission reactor applications particularly, beryllium swells under neutron irradiation and must be replaced periodically. For magnetic fusion, securing of the beryllium to plasma-facing surfaces remains to be fully developed. Irradiated beryllium will require disposal, which is presently a problem.

Because of reduced research budgets generally within the U.S. and particularly within the U.S. fusion program, there has not been much programmatic support or activity in beryllium technology research for fusion in the U.S. in recent years. With the possible re-entry of the U.S. into the ITER Project, there may be a resurgence of interest and research in beryllium technology for fusion. In this paper, we summarize work that has been lately accomplished, focusing on that which is currently underway. This is intended only to be a summary. Further information can be found in references cited or by contacting the institutions where the work is being performed.

2. IRRADIATION EFFECTS

Investigations have been underway at Oak Ridge National Laboratory (ORNL) to investigate mechanical property changes for high quality beryllium materials subjected to low-temperature, low-dose neutron irradiation in water-moderated reactors. Materials chosen were the S-65C ITER candidate material produced by Brush Wellman, Kawecki Berylco Industries P0 beryllium, and a high-purity zone refined beryllium. Mini-sheet tensile and thermal diffusivity specimens were irradiated in the temperature range of ~100-300°C with a fast (E>0.1 MeV) neutron fluence of 0.05 to 1.0×10^{25} n/m² in the High Flux Isotope Reactor (HFIR) at the Oak Ridge National Laboratory and the High Flux Beam Reactor (HFBR) at the Brookhaven National Laboratory. Irradiation conditions are listed in Table 1.

As expected from earlier work on beryllium, both materials underwent significant tensile embrittlement with corresponding reduction in ductility and increased strength. Both thermal diffusivity and volumetric expansion were measured and found to have negligible changes in this temperature and fluence range.

A significant result from this work is that while both materials rapidly embrittle at these ITER relevant irradiation conditions, some ductility (1-2%) remains, which contrasts with a body of earlier work including recent work on the Brush-Wellman S-65C material irradiated to slightly higher neutron fluence.

The overall results on swelling, thermal conductivity, and tensile properties are in good agreement with present work on similar materials and are in qualitative agreement with the older literature data on less pure beryllium forms. Comparison of hardening and annealing behavior for the zone refined and powder-processed materials indicate that hardening is not dominated by helium stabilized defects forming along grain boundaries. Figure 1 compares fracture surfaces for irradiated and un-irradiated samples.

For the two materials that underwent tensile testing, the lower BeO content Brush Wellman S-65C material showed less severe embrittlement, retaining a small, but meaningful level of ductility following irradiation. This result is in contrast to other recent work on S-65B [1-8], which found that similar irradiation conditions yielded extremely low ductility or complete embrittlement.

Further information on this work is in publication and will be available soon [9].

3. PLASMA SPRAY DEPOSITION

The Los Alamos National Laboratory is continuing their work on plasma spraying of beryllium (Figure 2). The project currently underway is for the European Fusion Development Agreement (EFDA). The specimens to be produced are 5-mm and 10-mm thick beryllium coatings on CuCrZr heat sinks, which are mock-ups of first wall components. The intent of the program is to produce low cost components (no beryllium machining) and test their response to first wall

Irradiation conditions for beryllium specimen exposure.									
	Thermal Fluence x10 ²⁵ n/m ²	Fast Fluence x10 ²⁵ n/m ² (E>0.1MeV)*	Fast Fluence x10 ²⁵ n/m ² (E>1.0MeV)	dpa	He conc. (appm)	He/dpa			
HFIR	0.14	0.05	0.026	0.04	10				
(300°C)	0.6	0.20	0.11	0.16	42	254			
	2.8	1.0	0.53	0.82	208				
HFBR (95-278°C)	0.23	0.5 ± 0.02	0.21 ± 0.02	0.34	250	824			

Table 1					
Irradiation of	conditions	for ber	yllium s	pecimen	exposure.

* Energy cut-off is listed as E>0.11 MeV for HFBR results [10]



Figure 1. Fracture surfaces of non-irradiated and $0.2 \times 10^{25} \text{ n/m}^2$ HFIR irradiated S-65C specimens.

relevant heat fluxes. The recent work has fabricated one test specimen for the EFDA project (Figure 3), and another will soon be prepared. The first specimen had a sprayed beryllium coating 5-mm thick, while the one soon to be prepared will be 10 mm thick on CuCrZr. These specimens will be subjected to intense thermal cycling tests in the JUDITH facility at KFA Jülich.

4. PLASMA INTERACTIONS

Three U.S. facilities are presently capable of studying plasma interactions with beryllium. One of these is the PISCES-B facility at the University of California San Diego. Another is the INEEL Ion Implantation Facility, which has been used for



Figure 2. Beryllium plasma-spray facility at Los Alamos National Laboratory.



Figure 3. Plasma sprayed beryllium 5 mm thick on test specimen for the EFDA Project.

many years to investigate tritium permeation and retention issues in a variety of fusion materials. The third is the Tritium Plasma Experiment.

4.1 PISCES-B

The Center for Energy Research (CER) at the University of California San Diego is the home of PISCES-B, shown in Figure 4 [11]. This facility provides steady-state simulation of the boundary regions in a burning fusion reactor, with a maximum plasma density of 2.5×10^{19} ions/m³ in deuterium and temperatures in the range of 5 - 30 million kelvins. It features a fully-qualified safety enclosure for performing tests of plasma interactions with beryllium.

In 2002, UCSD commenced a collaborative program with the European Union Fusion Program to study plasma materials interactions with mixed materials including Beryllium, Carbon, and Tungsten. The purpose of this work is to systematically examine the interaction of a



Figure 4. The PISCES-B facility at the University of California San Diego's Center for Energy Research.

beryllium-seeded deuterium plasma with graphite and tungsten target plates in the PISCES-B facility. This work has application to the ITER FEAT device, currently being planned. That machine will have beryllium tiles as its plasma-facing surface. The divertor will be faced with tungsten. It is important to understand the interaction between beryllium in the plasma striking the divertor and the tungsten divertor face to properly assess the potential for tritium inventories in the machine [11].

4.2 INEEL Ion Implantation Facility

The Ion Implantation Facility at the INEEL (Figure 5) has been in service since the early 1980s. It also is used to study plasma-material interactions, including interactions with beryllium. It has the ability to generate ion fluxes up to 10^{20} ions/m² on



Figure 5. INEEL Ion Implantation Facility.

targets. Figure 6 shows the surface of one such beryllium target, showing the surface modification that such ion fluxes produce. Experiment modes include permeation transient analysis and thermal desorption spectroscopy.



Figure 6. Beryllium specimen exposed to deuterium plasma in the INEEL Ion Implantation Facility.

4.3 Tritium Plasma Experiment

Another system built to investigate plasma interactions with materials, including beryllium, is the Tritium Plasma Experiment (TPE) shown in Figure 7.



Figure 7. Tritium Plasma Experiment before shipment to Los Alamos National Laboratory.

This device was first installed in the Sandia National Laboratory Tritium Research Laboratory at Livermore, California. It was subsequently moved to the Tritium Systems Test Assembly at Los Alamos National Laboratory. It is now being installed in the Safety and Tritium Applied Research (STAR) facility at the INEEL. Because of external tritium contamination, it is presently housed in a special Permacon[™] enclosure (Figure 8) where it is awaiting reassembly. It will also be operated within that structure.

5. MOLTEN SALT PROPERTIES

Molten salts have long been considered as a coolant and heat transfer medium for nuclear reactors. Early fission-reactor designs made use of molten fluoride salts, in part because of their low vapor pressures at elevated temperatures. The Molten Salt Reactor Experiment at ORNL was designed to operate at 650°C. The secondary coolant in that reactor was 2•LiF-BeF₂, commonly referred to as Flibe. The Li in that salt was depleted in ⁶Li.

There has been recent interest in Flibe for fusion applications. In addition to its obvious heat transfer properties, it can serve as a tritium breeder.



Figure 8. PermaconTM enclosure in which the Tritium Plasma Experiment is housed.

Additional benefits are that its electrical conductivity, tritium solubility, and reactivity with water and oxygen are low. One key issue is the corrosion potential of fluorine/TF. TF can be formed as free tritium, transmuted from Li and even from Be, combines with free fluoride ions. TF is extremely corrosive. There is a need for a suitable REDOX control agent. Finding such an agent will have substantial impact on tritium control strategy in fusion reactors that use Flibe.

5.1 JUPITER-II

JUPITER-II is a joint Japan-US research program that is examining a number of issues including REDOX processes. The main objectives of the JUPITER-II program include

- Preparation, purification and characterization of Flibe
- Development and testing of REDOX control strategy
- Investigation of deuterium/tritium behavior in Flibe, including
 - Solubility
 - Diffusivity
- Investigation of mobilization behavior of molten Flibe exposed to inert and reactive gas streams

The Flibe Tritium/Chemistry task will demonstrate the REDOX approach using Be and determine its influence on tritium behavior and corrosion. The experimental apparatus in which these experiments are conducted is shown schematically in Figure 9. Figure 10 shows various experimental facilities used with JUPITER-II at the INEEL.



Figure 9. Schematic of experimental system for molten salt safety experiments. The glovebox is equipped with two furnace assemblies and interconnecting plumbing to enable experiments with two different salts.



Figure 10. Experimental apparatus used with JUPITER-II project: (a) system for deuterium/tritium permeation experiments in Flibe; (b) setup for Flibe Purification and REDOX experiments.

Before work on tritium interactions with Flibe began, reference measurements were made on the experimental system without Flibe present using D₂. Single probe permeation experiments were conducted at 550 to 700°C. Double probe permeation experiments were performed at 600 and 650°C. The source-side pressure of deuterium (Probe 1) was 690 torr. Analysis of both transient and steady-state permeation behavior indicated good agreement with the best available Ni permeability data. Subsequent experiments were conducted with 400 mL of Flibe in the crucible Comparison of permeation data from Flibe experiments with double-probe reference experiments (no Flibe) provided qualitative picture of impact of Flibe on the permeation process.

Results from initial permeation experiments provide qualitative agreement with predictions based on early solubility and diffusivity data. Some of the permeation data through Flibe indicated that observed deuterium transport rates in Flibe were much faster than expected. These observations may be due to convection currents in the Flibe resulting from a small temperature gradient.

More recent information on the progress of this work can be found in the papers of Anderl et al. at and Fukada et al. at this workshop [12,13].

6. PEBBLE BED THERMAL CONDUCTANCE

Beryllium pebble beds have been proposed for use in fusion blankets to serve various purposes. The experimental data on bed effective thermal conductivity and the bed-clad interface thermal conductance coefficient is important for the design and analysis of fusion blankets. Work has been underway at the University of California Los Angeles (UCLA) to investigate and quantify experimental data on the effective thermal conductivity of a beryllium-helium pebble bed, on interface thermal conductance between beryllium pebbles and SiC, and on the effects of external applied pressure on these thermal properties. Beryllium as a metal has a high thermal conductivity (183 W/m.K at 25°C) in its typical solid form, but when it is used in a pebble form with a gas, the bed effective conductivity becomes much smaller.

The beryllium pebbles are contained in a cylindrical container made of stainless steel-316 with a 60-mm height, 101.6-mm inner diameter and

6.1 Experimental Apparatus

An experimental apparatus was designed and constructed to conduct the required experiments. Figure 11 shows a schematic drawing of the apparatus, while its picture is also given in Figure 12.



Figure 11. Schematic drawing of the experimental apparatus.



Figure 12. The experimental apparatus.

108mm outer diameter. Two columns of thermocouples are placed inside the bed at two different azimuthal angles. Each column has six thermocouples uniformly distributed along the bed axial direction. The thermocouples are placed normal to the direction of heat flow in order to minimize their effect on the bed's isotherms. The thermocouples' probes are placed 10 mm from the bed centerline. A copper heating block, placed above the container, is used to uniformly transfer the heat from the heater to the beryllium pebble bed. An 800-W heater is attached to the heating block in order to supply the bed with the required heat. At the bottom, the container is closed by a SiC disc, which is used as a clad material for the bed.

6.2 Experimental Procedure

Two groups of experiments were conducted. Experiments in the first group investigated bed effective thermal conductivity and the interface heat conductance as a function of the bed mean temperature. For this group, helium (at atmospheric pressure) was used as a cover gas. The bed mean temperature ranged from 130° C to 420° C.

Experiments in the second group were conducted to study the effect of the externally applied pressure on bed effective thermal conductivity and interface heat conductance. The external pressure ranged from 0.4 to 2.0 MPa and helium was used as a cover gas. The pressure was applied using a manually driven hydraulic press and measured with a load cell.

The packing fraction of the pebble bed was 60.6% for all runs. The bell jar was evacuated by a mechanical pump to produce a vacuum and then filled with the cover gas. The measured temperature signals were captured by a data acquisition system and stored in a personal computer. Beryllium pebbles of 2-mm diameter were used for all runs.

6.4 Results

The results of this work show that heating increases the effective thermal conductivity for the bed. For a beryllium pebble bed with 2-mm diameter pebbles and a packing fraction of 60.6% and helium at atmospheric pressure, the effective conductivity increases from 2.29 to 3 W/m.K with the increase of bed mean temperature from 130 to 420°C.

The experimental results are in good agreement with the predictions of available models as well as previous experimental results obtained by Dalle Donne [14], as shown in Figure 13. Those models include the Hall and Martin [15], Schlunder, Zehner, and Bauer (SZB) [16], and Slavin et al. [17] models.



Figure 13. Comparison of the experimental results of the bed effective conductivity with model predictions and Dalle Donne experimental values.

The ratio of solid to gas conductivity (k_s/k_g) plays an important role in the heat flow mechanism across the pebble bed. When this ratio is high (such as with beryllium and helium) the contact area characteristics have a significant impact on the effective conductivity of pebble beds. For the same beryllium-helium bed, effective conductivity was measured as a function of externally applied pressure (from 0.4 to 2.0 MPa) with four different bed temperatures (100, 180, 270, and 350°C). When the external pressure was increased to 2 MPa with these four temperatures, the effective conductivity increased by factors of 2.53, 2.30, 2.18, and 2.11 respectively, relative to that with no pressure. This is shown in Figure 14.

Similar to the bed effective conductivity results, the interface thermal conductance increased when the bed mean temperature increased. It increased by a factor of 1.89 with an increase in the bed temperature from 130 to 420°C. These results are shown in Figure 15. Figure 16 shows the effect of externally applied pressure on the interface thermal conductance coefficient, h. The increase in the interface conductance values with pressure shows variations similar to those of the effective thermal



Figure 14. Effect of external pressure on effective thermal conductivity



Figure 15. Interface conductance coefficient as a function of the bed mean temperature

conductivity. As the external applied pressure was increased to 2 MPa, the interface conductance increased by a factor of 2.5 relative to that with zero pressure (for bed mean temperature, $T_m = 270^{\circ}$ C).

Similar to the effects of externally applied pressures, one may expect that effective thermal conductivity would increase during blanket operation due to irradiation swelling and/or differential expansion. Interpretation of the applied pressure to operational parameters, such as the amount of swelling and differential thermal stresses is complex and should be addressed in future studies.



Figure 16. Effect of externally applied pressure on Be/SiC interface conductance.

7. BIOLOGICAL EFFECTS

An area of research that has received considerable attention in recent years is the physiological consequence of beryllium exposure. Accounts of very minimally exposed persons contracting chronic beryllium disease (CBD) and becoming seriously ill have appeared from many sources. The disease is prevalent among workers in beryllium metal, beryllium oxide, and copper alloys. The continuing incidence of CBD among workers in facilities that meet the beryllium air concentration level of $2 \mu g/m^3$ has led regulators to recommend substantial reduction in allowable air concentrations.

The vast majority of beryllium research being conducted in the U.S. at the present time is in support of better understanding of the biological aspects of beryllium exposure. One such will be mentioned here.

A group of researchers that include participants from Los Alamos National Laboratory, Johns Hopkins University, the National Institutes of Occupational Safety and Health (NIOSH), the University of Oklahoma, and Brush Wellman, Inc. have been investigating specific surface area of beryllium and some of its compounds [18], The work is based on the hypothesis that the risk of disease is increased by long particle dissolution times in the body. That, in turn, depends on specific surface area of the particles ingested. Earlier animal toxicity studies [19] demonstrated that higher surface area beryllium oxide particles were more soluble and more toxic than lower surface area beryllium oxide particles. In-vitro studies [20, 21] found a strong correlation between particle specific surface area and cytotoxicity.

This group of researchers used estimates from geometric relationships and actual measurements using gas adsorption techniques to compare specific surface areas of beryllium metal, beryllium oxide, and beryllium-copper alloy. They concluded that specific surface area of the beryllium forms they investigated is very much a function of the way in which they were manufactured. Beryllium oxide specific surface area was relatively insensitive to particle size, consistent with the particles of beryllium oxide being clusters of smaller, relatively constant sized grains. For beryllium metal, specific surface area was greater than that predicted by the smooth spherical model, but the trends were clearly the same. Beryllium metal particles are hardly spherical, but they do tend to have similar shapes from small to large particles, giving the trend. On the other hand, the specific surface area of copperberyllium alloy was almost insensitive to particle diameter. This is consistent with the ultra-fine cluster morphology resulting from vaporization and condensation in the arc furnace in which they were made.

A recommendation from this group was that instead of specifying allowable air concentrations in terms of beryllium mass per unit volume, the specification should probably be given in terms of aggregate surface area per unit volume. Considerable work remains to validate models that could be used in such a specification, however.

8. DISPOSAL ISSUES

In the U.S., the active high-power reactors using beryllium components include the Advanced Test Reactor (ATR) at the Idaho National Engineering and Environmental Laboratory (INEEL), the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory (ORNL), and the Missouri University Research Reactor (MURR) at Columbia, Missouri. Because of swelling caused by production of helium and tritium from the beryllium itself, the beryllium must be replaced periodically. That irradiated beryllium must be subsequently disposed of.

In addition to the currently operating reactors, several decommissioned reactors have beryllium reflectors that will eventually require disposal. Some of these are the Engineering Test Reactor (ETR) and the Materials Test Reactor (MTR) at the INEEL, and the Plum Brook Reactor at Sandusky, Ohio. Approximately 31 other reactors worldwide use beryllium components in fission reactors and may face the same disposal issues. The International Thermonuclear Experimental Reactor (ITER) is also planning to use beryllium as a first wall coating, and beryllium is anticipated as a neutron multiplier in tritium breeding blankets.

Though swelling will probably not be a reason for replacement of beryllium components in fusion reactor systems, its sputter removal from plasmafacing surfaces and re-deposition to other parts of the reactor may require periodic clean-up. At the end of reactor life, the beryllium must be disposed of. Thus, as with fission reactors, progress in disposing of irradiated beryllium will be of benefit to fusion.

Work to characterize irradiated beryllium has been performed over the last few years. Several concerns are associated with disposal of the beryllium following irradiation. First historically was concern over production of ¹⁴C, an activation product generated from a fast neutron (n, p) reaction with¹⁴N (a common impurity in beryllium metal) and the production of ⁹⁴Nb from a ⁹³Nb impurity. More recently discovered is the presence of transuranic isotopes brought about by neutron absorption in ²³⁸U and subsequent reactions. Natural uranium occurs as an impurity in the beryllium metal procured for the ATR.

9. MANUFACTURING

With the tightening of occupational exposure guidelines, Brush Wellman, the only source of beryllium metal in the U.S., has elected to no longer reduce metal from ore [22]. An interesting technology is being investigated with the hope that it will enable direct reduction of beryllium metal without some of the hazardous steps formerly employed in its production. This process was invented in the late 1930s, but it has seen a recent resurgence. The current version, called FFC for Fray, Farthing and Chen, who developed the method in 1993, is effectively electrolytic reduction from the oxide in a molten salt bath.

This process is presently primarily directed at titanium [23]. When titanium oxide is immersed in CaCl and some of the oxygen is removed, and the oxide becomes conductive. Electric current then reduces the oxide directly to metal. The current research is exploring whether the same effect may take place in beryllium [24].

Another interesting development is the interest by designers of future reactors in the alloy AlBeMet [25]. This material is an alloy consisting of 62% beryllium and the remainder 1100 series aluminum. The cost is 50 - 60% less than pure beryllium. It retains many of the neutronic advantages of beryllium, but it has properties that in some ways are preferable to those of beryllium.

10. HOPED FOR ACTIVITY

We hope to see a resurgence of research on beryllium technology directly applicable to fusion in the years ahead. As has been mentioned, this will depend strongly on the re-entry of the U.S. to the ITER Project. Work required to solve disposal issues for beryllium components in fission reactors around the world will also be beneficial for fusion.

10.1 Fabrication

Techniques for plasma spraying beryllium insitu in tokamaks would be highly beneficial. Presently, designs are limited to tiles that can be prepared in a controlled environment such as a glovebox. This may greatly facilitate surface replacement and repair if it can be done economically.

10.2 Decontamination and Recycle

There is a need for a process that can return irradiated and other contaminated beryllium to the supply stream. Several possibilities have been proposed and partially investigated. These may include distillation, melting and slagging, zone refinement, and FFC processes. Failing that, methods of storage or entombment that will prevent dispersion of radioactive components in the future may also be needed.

10.3 Concentration Monitoring

Laser fluorescence techniques have been developed that will give beryllium concentrations in air by passing a known quantity of air through a filter, then analyzing the material caught on the filter [26]. That technology is only moderately successful because of particle size effects and the relatively long times required for sampling. To get enough material to analyze at the 0.2 μ g/m³ now recommended as the 8-hour average concentration workers are exposed to [27], sample collection for

several hours is needed. It would be useful to have an instrument that will give accurate measurements of airborne beryllium concentrations below the 0.2 μ g/m³ action level.

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