POSTIRRADIATION EXAMINATION OF BERYLLIUM PEBBLES — D. S. Gelles (Pacific Northwest National Laboratory)*

OBJECTIVE

The purpose of this work was to determine the effects of irradiation on prototypic beryllium pebbles. Such pebbles are under consideration as the neutron multiplier medium in the European Fusion Technology Program Helium Cooled Pebble Bed (HCPB) Blanket.

SUMMARY

Postirradiation examinations of COBRA-1A beryllium pebbles irradiated in the EBR-II fast reactor at neutron fluences which generated 2700-3700 appm helium have been performed. Measurements included density change, optical microscopy, scanning electron microscopy, and transmission electron microscopy. The major change in microstructure is development of unusually shaped helium bubbles forming as highly non-equiaxed thin platelet-like cavities on the basal plane. Measurement of the swelling due to cavity formation was in good agreement with density change measurements.

PROGRESS AND STATUS

Introduction

A large number of beryllium specimens have been irradiated in U.S. DOE Office of Fusion Energy Sciences (OFES) experiments in the Experimental Breeder Reactor II (EBR-II) in Idaho Falls, ID, and the Fast Flux Test Facility (FFTF) in Hanford, WA. Due to lack of funding, most of those specimens have never been tested, but DOE still retains an interest in the results. With the increased difficulty for performing irradiation experiments, due in part to the shutdown of FFTF and EBR-II and other reactors worldwide, these specimens are becoming very valuable. The German fusion materials development program centered at Forschungszentrum Karlsruhe INR has been concentrating efforts on design of a fusion blanket, using beryllium pellets, an intermediate product in the production of high grade beryllium, as a neutron multiplier. An EBR-II test, called COBRA-1A, included four beryllium pebble product forms, and thereby provides fast neutron irradiated materials for postirradiation testing. Therefore, Forschungszentrum Karlsruhe INR has concluded that it is efficient use of their available funds to invest in the testing of some of these specimens. This report provides results for that testing program.

The program is divided into three phases:

I. Characterization of beryllium pebbles irradiated in the COBRA 1-A experiment, including density change measurement, microstructural examination, helium and tritium release response on heating and neutronic analysis.

II. Removal of beryllium cylindrical specimens with 97 and 100% of theoretical density from capsules irradiated in FFTF experiments and containing lithium as a heat transfer medium.

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III. Characterization of the FFTF irradiated beryllium specimens. Due to funding limitations, this is to be restricted to density change measurements.

Experimental Procedure

EBR-II

The COBRA-1A vehicle, irradiated in row 2 of EBR-II starting on November 26, 1992 with run 168, contained a number of beryllium specimens in canisters C03 and D03 located in below core positions in capsules B-390 and B-391, respectively. Both capsules were of weeper design, so that reactor coolant was in direct contact with each canister, and canisters were helium filled to provide a heat transfer medium. Experimental details are provided in reference [1]. Capsules B-390 and B-391 were removed from reactor upon removal of the COBRA-1A2 vehicle on September 26, 1994. At that time, EBR-II ceased to operate. Neutron dosimetery and damage calculation for these irradiations are included in reference [2].

Canisters C03 and DO3 contained beryllium in several product forms: 1 mm pebbles, 3 mm pebbles, 5 mm pebbles, 7.6 mm diameter x 20 mm right cylinders and 7.7 mm diameter x 6 mm right cylinders. (The cylinders were of two densities, 100% and 97% and were used for other purposes.) Pebbles were from two sources, Brush Wellman in all sizes and Nippon Gaishi Co. (NGK) at 1 mm. Pebble details, as supplied by the manufacturer are provided in Table 1.

The specimen loading and capsule position within EBR-II were somewhat different for the two capsules. Radiographs of the capsules are provided in Figure 1. In both cases, the bottom of the capsule is at the right and careful observation reveals that in each case the bottom contains a layer of 1 mm pebbles, with the remaining pebbles distributed amongst the cylindrical specimens. The 1 mm pebbles in capsule C03 were from Brush Wellman whereas the 1 mm pebbles in capsule D03 were from NGK. Because reactor neutron flux varied as a function of vertical position, minor differences in dose developed for the 3 mm pebbles in C03 and the 5 mm pebbles in D03. In each capsule, only seven 5 mm pebbles were included whereas about 75 3 mm pebbles and over 150 1 mm pebbles from each manufacturer were irradiated. Accumulated fluences were estimated based on capsule position and information in reference 2, giving 3.77×10^{22} n/cm^2 (E > 0.1 MeV) for Brush Wellman 1 mm pebbles, 3.62×10^{22} n/cm^2 for NGK 1 mm pebbles, 4.31×10^{22} n/cm^2 (E > 0.1 MeV) for Brush Wellman 3 mm pebbles, and 4.88×10^{22} n/cm^2 (E > 0.1 MeV) for Brush Wellman 5 mm pebbles. The operating temperatures are estimated as follows: 379°C for C03 and 378°C for D03. The irradiation temperatures are therefore described as 380°C in the remainder of this report.

In a companion report, [3] the production of ^{4}He and tritium from beryllium in the COBRA-1A2 irradiation based on calculations is provided.

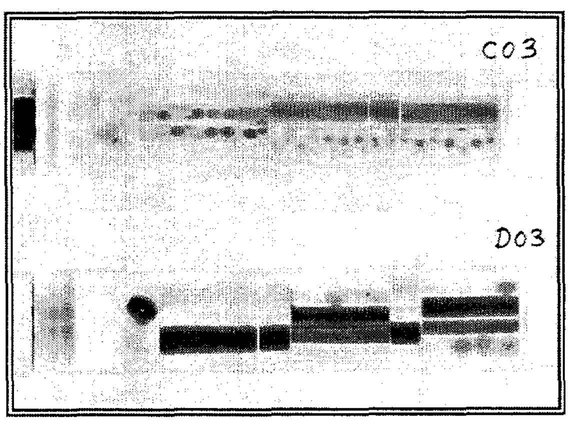

Figure 1. Specimen loading in the COBRA-1A vehicle as shown by neutron radiography with the bottom of the subcapsule filled with 1 mm pebbles located at the right.

Table 1. Pebble details

Type	Supplier & Batch #	Size	Major Impurities	Minor impurites (in ppm)
1 mm	Brush Wellman 826-64-5	1 ± 0.2 mm -16 +20 mesh	0.094 wt% BeO 0.047 % C 0.06 % O_2	750 Mg, 565 Fe, 380 Si, 365 Al, 110 Cr, 105 Zr, 95 Mn, 90 Ti, 80 Ni, 70 Zn, etc.
3 mm	Brush Wellman 826-64-3	3 mm -6 +7 mesh	0.020 wt% BeO 0.038 % C 0.013 % O_2	2400 Mg, 375 Al, 370 Fe, 250 Si, 105 Zr, 90 Mn, 85 Cr, 85 Zn, 75 Ti, 65 Ni, etc.
5 mm	Brush Wellman 826-64-4	5 mm -3½ +4 mesh	0.094 wt% BeO 0.047 % C 0.06 % O_2	420 Fe, 400 Mg, 390 Al, 230 Si, 95 Mn, 90 Zr, 80 Zn, 75 Cr, 70 Ti, 60 Ni, etc.
1 mm	Nippon Gaishi Co. 4310	1 ± 0.3 mm 0.72 to 1.10 mm	1.51 wt% BeO 0.023 % C	1100 Fe, 780 Al, 300 Si, 170 Mg

FFTF

A description of specimen loadings for all molybdenum alloy (TZM) subcapsules containing beryllium specimens in depleted lithium and progress on disencapsulation and density change measurements on selected specimens are provided in a companion report. [4]

Experimental Results

Optical Microscopy (OM) and Scanning Electron Microscopy (SEM)

In order to gain an understanding of pebble to pebble conformity and determine surface degradation due to irradiation, several specimens were selected for optical examination and one 3 mm pebble of each of the irradiated and unirradiated conditions was then examined by SEM. Figure 2 provides examples of OM; in each case different pebbles are shown with 1 mm pebbles above and 3 mm pebbles below. The figure demonstrates that beryllium pebbles vary significantly in size and many contain surface irregularities, including depressions and seams.

One 3 mm specimen of each of the unirradiated and irradiated pebbles were chosen for SEM examination, and the results are provided in Figure 3 showing an unirradiated pebble above and an irradiated pebble below at several magnification levels. Comparisons between these pebbles reveal that both contain two types of shallow surface depressions, on the order of 100 μm in diameter and on the order of 5 μm in diameter, and specimen #6 has many particles adhered to the surface. The adhered particles are assumed to be dust and dirt accummulated during handling, and therefore both samples contain similar features. Evidence for fine surface porosity

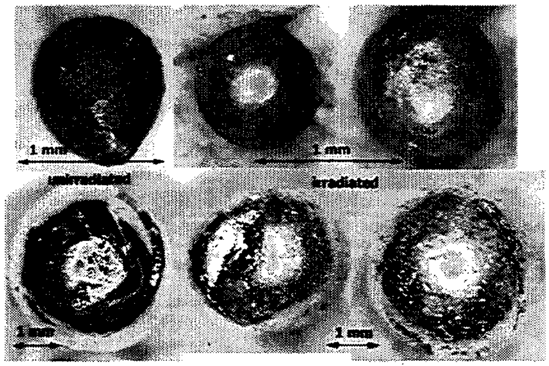

Figure 2. Optical photomicrographs of unirradiated and irradiated beryllium pebbles

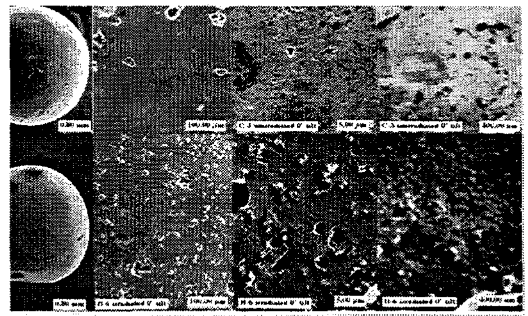

Figure 3. SEM photomicrographs of unirradiated and irradiated 3 mm beryllium pebbles showing the surface features at increasing magnification

due to irradiation was not found.

Swelling in 3 mm pebbles

Four irradiated pebbles from the C03 capsule were selected for density measurement and compared with four unirradiated specimens. Specimens were weighed in air and in water at least three times, until reasonable consistency was obtained because imperfect wetting and air bubble adherence were found to be problems. The results of the measurements are listed in Table 2 so that the density following irradiation (V) was found to be 1.79474 ± 0.01130 and the unirradiated density (V_0) was 1.80969 ± 0.03370. The resultant swelling $(V-V_0)/V_0$ was 1.45%, of the same magnitude as the uncertainty for the unirradiated density (1.86%) but larger than the uncertainty for the irradiated density (0.6%). The large uncertainties may either be due to the surface conditions (bubble adherence) or variations in impurity concentrations within the pebbles. However, given the larger scatter in the unirradiated samples, the latter cause is more likely.

Table 2. Density measurements for 3 mm pebbles

Sample	Condition	density g/cm^2	standard deviation
1	380°C, 4.31x10^{22} n/cm^2	1.78818	± 0.02987
2	"	1.8114	± 0.0096
3	"	1.79206	± 0.02958
4	"	1.7873	± 0.02691
5	unirradiated	1.83124	± 0.02032
6	"	1.77046	± 0.01331
7	"	1.8435	± 0.0152
8	"	1.79354	± 0.00661

Transmission Electron Microscopy (TEM)

Several 3 mm pebbles were ground to disks approximately 0.12 mm thick and thinned by electropolishing in a solution of 75 parts nitric acid, 15 parts sulfuric acid, 15 part hydrochloric acid in 750 parts ethylene glycol at 9 to 13 V with the solution cooled to -20°C. Pitting presumed to be due to internal flaws was found to be a problem, but suitably thin areas were obtained in several specimens prepared for examination following brief repolishing of pitted specimens. Two unirradiated specimens and two irradiated specimens were thinned and polished providing one good specimen and one poor specimen with still another specimen providing sufficient thin area to confirm observations on the other two specimens.

Examinations revealed several noteworthy features. In the irradiated specimens, grain size was found to be large, with a low dislocation density but containing many small features tentatively identified as bubbles lying as plate-like features on the basal plane. Several examples were also found of large regions as big as 5 μm in diameter with amorphous diffraction patterns. Such regions were found to be rich in Si, Al, Mg and Mn, indicating that impurities were non-uniformly distributed and that the impure regions became amorphous due to irradiation. In comparison, the dislocation density was greater in the unirradiated condition, with evidence of subgrain development. Also, a region containing impurity was found. Therefore, irradiation is found to reduce the dislocation density significantly, to form a fine distribution of unusually shaped bubbles and to amorphize large regions of impurity.

Examples of the structure found following irradiation are shown in Figures 4 and 5. The dislocation structure is shown in Figure 4 showing a region in bright field in a) and another region in dislocation dark field contrast in b). The bright field image demonstrates a loose tangle of dislocations, whereas the dark field image also shows a high density of smaller features in weaker contrast. Previously, these weaker features had been identified as c-type dislocations loops. [5] However, when the foil was tilted to bring these weaker features edge-on, they were found to image as cavities. A sequence is provided in Figure 5 using void contrast showing these features

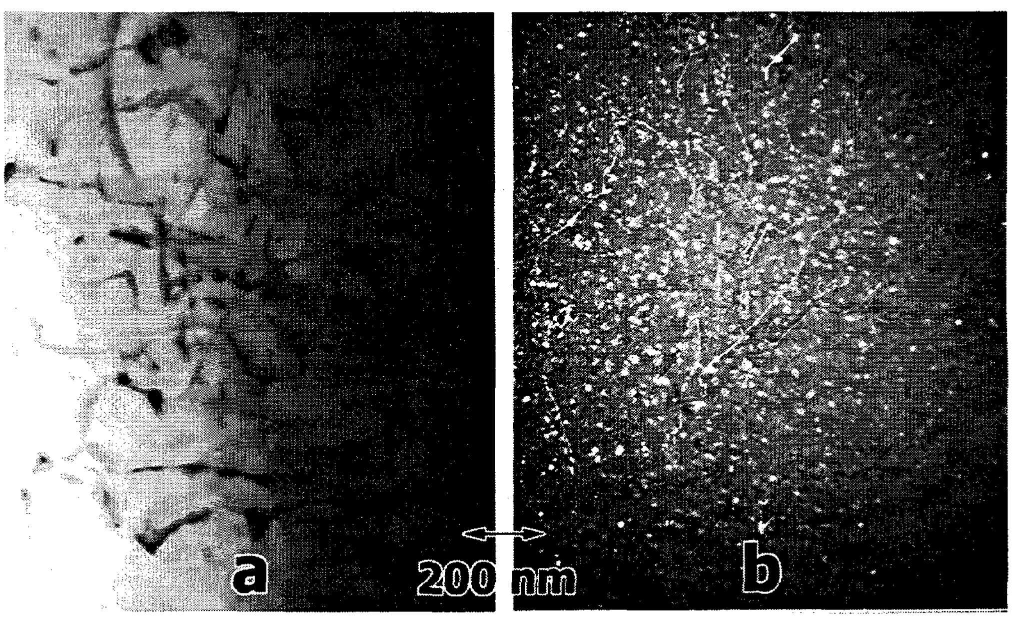

Figure 4. Microstructures in an irradiated 3 mm beryllium pebble showing a) the dislocation structure in $(1\bar{2}10)$ bright field contrast and b) (0001) dislocation dark field contrast

edge-on in (a), tilted off 7° in (b) and tilted off 12° in (c). Figure 5d provides the unusual situation where the features are shown in strong contrast with the features tilted edge-on and demonstrates that the features are probably internally stressed. It can therefore be concluded that the weaker features are probably unusually shaped helium bubbles, the planar surfaces of which can be imaged weakly in dislocation contrast.

<u>Discussion</u>

Effects of Irradiation on Microstructure

It was previously assumed that irradiation would cause microporosity, and c-type loop formation. The added complexity of neutron damage near pebble surfaces was expected to enhance transmutation possibly leading to enhanced surface microporosity. The present SEM examination appears to indicate that surface roughening due to irradiation is negligible to resolutions on the order of 100 nm. TEM did not provide microstructural information regarding cavitation at grain boundaries, but it now appears that swelling in the form of helium bubbles takes on a unusual highly non-equiaxed geometry of thin platelets on the basal plane. Bubbles are approximately 25 nm in diameter and 4 nm in thickness. These features had not been identified previously as bubbles, perhaps because the helium levels were lower, estimated at 80 appm helium following irradiation at 400°C to 8 dpa ($1.8x10^{22}$ n/cm^2) below core in FFTF. In the present experiment, the fluence was approximately $4.3x10^{22}$ n/cm^2 below core in EBR-II, a similar reactor, and therefore the helium level is expected to be about 3 times larger, and is estimated at 3000 appm. It can be noted that the earlier experiment produced significantly coarser damage structure, possibly because the irradiation temperature was higher. Careful review of the earlier micrographs shows examples of contrast similar to that found in the present experiment. The differences in interpretation may be compatible if it is possible that c-type loops accumulate gases such as helium a tritium, eventually becoming highly anisotropic bubbles.

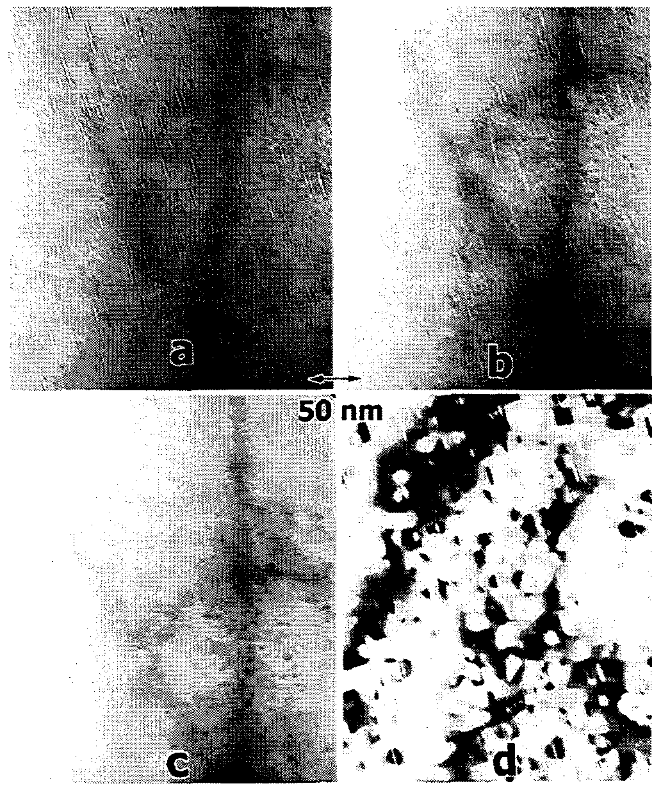

Figure 5. Void swelling in an irradiated 3 mm beryllium pebble tilted parallel to the basal plane so that the voids are edge on in a), tilted off 7° in b), tilted off a further 5° in c) and shown edge on in strong multibeam contrast in d)

In the current work, the bubbles were measured to provide diameter, number density and an estimate of swelling. It was found that the bubbles ranged in diameter from 5 to 25 nm, with a mean diameter of 11.9 nm at a number density of $2.0x10^{16}$ cm^{-3}. If one were to assume spherical voids with these diameters, this corresponded to 2.5% swelling, but as the bubble shape was flattened, with an aspect ratio for thickness to diameter of about 4/25 or .16, the actual swelling can be estimated at about 0.4 of the spherical value or about 1%. Therefore, the measured density change of 1.45% appears to be largely due to the development of internal bubbles estimated at 1% swelling.

Swelling may also originate from the transformation of impurity regions from a crystalline to an amorphous phase. It is difficult to estimate the volume fraction of impurity regions or to estimate the resultant swelling due to phase transformation, so the magnitude of the swelling change cannot be estimated at this time.

If much of the helium and tritium is bound in bubbles, then release of these gases from heated specimens can be expected to occur only after bubble coarsening and therefore will be observed mainly at higher temperatures. Examination of heated specimens may allow verification of such a prediction.

CONCLUSIONS

Post irradiation examinations of COBRA-1A beryllium pebbles irradiated in the EBR-II fast reactor at neutron fluences which generated 2700-3700 appm helium have been performed. Measurements included density change, optical microscopy, scanning electron microscopy, and transmission electron microscopy. The major change in microstructure is development of unusually shaped helium bubbles forming as highly non-equiaxed thin platelet-like cavities on the basal plane. Measurement of the swelling due to cavity formation was in good agreement with density change measurements.

FUTURE WORK

This work is completed.

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