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Critical assessment of beryllium pebbles response under neutron irradiation: Mechanical performance and tritium release

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Abstract

The two still open issues related to beryllium for fusion applications are its mechanical response under neutron irradiation and the kinetics of the tritium release as a function of neutron fluence and temperature. The EXOTIC-7 as well as the ''Beryllium'' experiments carried out in the HFR reactor in Petten are considered as the most detailed and significant tests for investigating the beryllium pebble response under neutron irradiation. This paper reviews the present status of beryllium post-irradiation examinations performed at the Forschungszentrum Karlsruhe with samples from these irradiation experiments, emphasizing the effects of irradiation on essential material properties and trying to elucidate the processes controlling the property changes.

1. Introduction

For the Helium Cooled Pebble Bed (HCPB) blanket, which is one of the two reference concepts studied within the European Fusion Technology Programme, the neutron multiplier consists of a mixed bed of about 2 and 0.1–0.2 mm diameter beryllium pebbles. The main structure of the pebble bed is given by the larger pebbles with a packing factor of 63.3%. In the space between them are placed the smaller beryllium pebbles with a packing factor of 17.5%. Both kind of pebbles are fabricated by melting, however for the larger ones a relatively inexpensive intermediate product of the beryllium fabrication route (Brush–Wellmann Company) has been chosen [1]. Beryllium has no structural function in the blanket, however microstructural and mechanical properties are important, as they might influence the material behavior under neutron irradiation.

The EXOTIC-7 [2] as well as the ''Beryllium'' [3] experiments carried out in the HFR reactor in Petten are considered as the most detailed and significant tests for investigating the beryllium pebble response under neutron irradiation. The beryllium irradiated in both the EXOTIC-7 and the ''Beryllium'' experiments consists of a mix of about 2 mm and 0.08–0.18 mm diameter beryllium pebbles. However, while in EXOTIC-7 the beryllium pebbles were – during irradiation – in contact with Li_4SiO_4-pebbles, in the ''Beryllium'' experiment only pure beryllium pebble beds were irradiated.

For some post-irradiation examinations (PIE) the components of the EXOTIC-7 mixed beds had to be separated. The 2 mm diameter Be pebbles were separated from the small (0.1–0.2 mm) Li_4SiO_4 and Be pebbles by sieving. The small Li_4SiO_4 and Be pebbles were separated by a wet procedure using an organic liquid with a density in-between that of the Li_4SiO_4 and Be pebbles. The visual inspection of the Li_4SiO_4 pebbles showed that only a small number of pebbles were fractured, whilst the beryllium pebbles look almost all intact.

2. Microstructural analysis

The bigger unirradiated beryllium pebbles usually show a relatively large number of indentations on their external surface. This is probably due to the fact that

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during the fabrication process very hot (or still partially molten) beryllium pebbles come in contact with cold and already solidified ones. With optical microscopy, a quite strong variation of coarse pores was observed. Some pebbles show big voids which seemed to be generated during the cooling phase of the fabrication process. Relatively often a coarse porosity with a pore size of 0.1–0.2 mm has been observed. On the other hand, most of the pebbles reveal a very small micro-porosity usually oriented along the crystal axis showing a very fine dendritic or cellular structure. In general, it has been observed that the porosity of small pebbles is always smaller than that of the bigger ones, which clearly confirms that coarse porosity is generated during the cooling phase of the fabrication process. Furthermore, a large number of pebbles present, near the external surface, a dense region the depth of which usually reaches 0.2–0.3 mm. The metallographic structure of both smaller and bigger pebbles shows the presence of large grains, in some of the smaller pebbles as large as the pebble diameter [4].

Insoluble impurities have been usually observed on the grain boundaries, while iron and chrome are almost exclusively present in solid solution in the beryllum-matrix. The external surface of the large unirradiated Be pebbles is usually covered by a 2 μm thick SiO_2 layer and/or a fluorine layer (probably BeF_2) the thickness of which is generally lower than 2 μm. Two extraneous phases are present in almost all the analyzed pebbles. Mostly a round bright phase looking like an eutectic stored in the beryllium matrix and occasionally a dark square phase which appears as a primary precipitated phase has been observed.The dominant precipitated phases are $Be_{13}Mg$, Be_{13}(Mg, Zr, U), Mg_2Si and Al_2O_3 [5].

Because of the relatively low irradiation temperatures in both EXOTIC-7 and "Beryllium" experiments ($T_{irr} = 410$–550°C), no remarkable thermally induced changes in the beryllium structure (macroscopic and microscopic) have been observed after the neutron irradiation. However, although the fast neutron fluence was almost the same in both experiments ($\Phi \approx 1.3 \times 10^{21}$ cm^{-2}; $E_n > 1$ MeV), the beryllium pebbles from EXOTIC-7 showed strong irradiation induced structural changes. In particular a new type of porosity was observed in a boundary layer of the pebbles not deeper than 40–50 μm. The new pores are of an oblong shape, are oriented along the crystals and have diameters of up to 2 μm as shown in Fig. 1. This new porosity is essentially due to the contact between beryllium and Li_4SiO_4 during irradiation. According to previous studies [6] it is in fact expected that, if beryllium is in direct contact with ceramics during irradiation, a fraction of the tritium as well as helium ions produced in the Li_4SiO_4 during irradiation is implanted in a surface layer of beryllium the depth of which is about 40 μm.

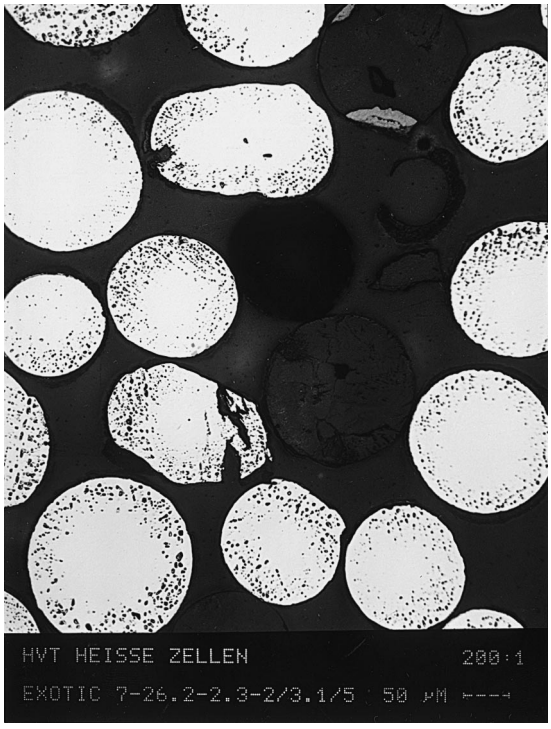

Fig. 1. Irradiation induced porosity in beryllium pebbles from the EXOTIC-7 experiment. This porosity is essentially due to the contact between beryllium and Li_4SiO_4 during irradiation.

Etching the microsections reveals a polygonization pattern consisting of bundles of oblong lines, mostly aligned in the same direction. Along with the line structures of polygonization, etching causes a dense network of etching pits. A striking phenomenon are narrow strips of approx. 2 μm width on both sides of the grain boundaries, which remain free from etching attack.

3. Mechanical behavior

The mechanical behavior of a significant number of unirradiated pebbles with the larger diameter has been investigated by submitting them to compressive loads of up to 1600 N at room temperature. The plastic deformations of the pebbles have been measured and correlated with the applied loads. Relatively large variations have been observed in the mechanical response of the pebbles. However, probably due to the very small amount of BeO impurities, all the pebbles showed a high ductility at room temperature. Pebbles loaded up to 400 N show diameter reductions up to 13% but, in spite of evident large plastic deformations, no fracturing or crack formation was registered [4]. On the other hand, pebbles loaded with 800 N (diameter deformation up to 25%) or more, reveal cracks on their "meridian" planes [4].

Microhardness measurements were performed in the microsections prepared by metallographic techniques. On the basis of hardness levels, techniques were developed to record the tendencies of irradiation-induced changes in the mechanical properties, i.e. plasticity and elasticity [7]. The hardness measurements were initially conducted with a holding time of 5 s. Afterwards, the same position was subjected to pressure once again, but with a holding time of 60 s. The increase in area, expressed in percent, produced by the second impression was used as a benchmark for the subsequent evaluation of the plastic deformation of the material.

The mechanical properties of the pebbles from ''Beryllium'' experiment do not significantly change after irradiation. On the contrary, pebbles from EXOTIC-7 show an increase up to about 30% of their elastic behavior range and a decrease up to about 50% of their plastic behavior range. The decrease of the plasticity is much more evident in the smaller pebbles due to the fact that the implantation depth of tritium coming from ceramics (≈40–50 μm) is of the same order of magnitude of the pebbles radius (≈100–200 μm), which results in a higher implantation efficiency than in the case of the larger Be pebbles.

4. Tritium release

Release kinetics and total amount of released tritium were determined by annealing the pebbles by temperature ramps of 5°C/min up to 850°C, keeping constant this temperature for several hours and purging them with 50 SCCM He+0.1 vol% H_2. A series of tests showed that the release characteristics of each type of pebbles is reproducible. The representative release curves for large pebbles, pieces ($\Phi < 0.7$ mm) from crushed large pebbles and for small pebbles of the ''Beryllium'' experiment are shown in Fig. 2. Useful parameters to characterize tritium release kinetics are the fractional release (release R/production P), the temperature T_{max} corresponding to the maximum release rate (during the 5°C/min temperature ramp) and the factor DF by which the release rate decreases (at 850°C within 3 h). For the pebbles investigated in the frame of the present work these parameters are given in Table 1.

From Fig. 2 and the data of Table 1 the following conclusions concerning tritium release kinetics of Be pebbles from the ''Beryllium'' experiment can be drawn:

- Release from 0.7 mm diameter pieces from the large pebbles is faster than that from the whole pebbles (''particle size'' effect). A further decrease of the particle size to ≤0.5 mm diameter indicated no further release improvement.
- Release from the small pebbles is faster than that from 0.7 mm diameter pieces from the large pebbles.
- Release from previously investigated Brush-Wellman samples (B-26, S200-HIP) [6] [8,9] is comparable with that from the large pebbles. Therefore, it can be assumed that the experimentally determined tritium residence times of beryllium B-26 [6] apply also to the large pebbles.

With reference to EXOTIC-7, release of all investigated Be samples (Be 2 mm, Be 2 mm broken, Be 0.1–0.2 mm) from the mixed beds is very similar. As shown in Fig. 3, and in agreement with previous studies [6,10], the release starts at about 500°C and achieves a maximum at about 700°C. The total release of the large Be pebbles (Be 2 mm) is slightly larger ($\approx 6 \times 10^{10}$ Bq/g) than that from Li_4SiO_4 from the same capsules, while that of the small Be pebbles (Be 0.1–0.2 mm) is about a factor 30 larger ($\approx 1 \times 10^{12}$ Bq/g).

According to previous studies [6] it is expected that, if Be is in direct contact with ceramics during irradiation, a fraction of the 2.74 MeV tritons produced in the Li_4SiO_4 is implanted in a surface layer of beryllium (depth ≈40 μm). This leads to an additional inventory which is usually several times larger than that directly produced by the neutrons reacting in the beryllium. Tritium generation data in Be for both EXOTIC-7 capsules 26.2-1 and 28.2 are not yet available. However, with reference to calculations for the ''Beryllium'' experiment [3] one can estimate a ratio He-production/T-production of a about 45. Based on the He-production in Be for the HFR reactor of about 3440 appm per 10^{22} n/cm^2 ($E_n \geqslant 1$ MeV) [11], the total He produced at the end of the EXOTIC-7 experiment (fast fluence 1.2×10^{21} cm^{-2}) should be 450 appm (for both capsules 26.2-1 and 28.2). This leads to a specific tritium production in Be of 1.3×10^9 Bq/g, in reasonable agreement with a value of 2×10^9 Bq/g based on tritium production vs. fast fluence graph [12]. In any case, the neutron-generated tritium in Be is about a factor 50 lower than the released tritium from the larger Be pebbles and about a factor 700 lower than that from the smaller Be pebbles, assuming no release during the irradiation. This high tritium inventory in both the large and small Be pebbles from EXOTIC-7

Table 1
Tritium release characteristics of various beryllium samples

Experiment	Material	Φ [cm^{-2}] ($E > 1$ MeV)	P [Bq/g]	R [Bq/g]	R/P [%]	T_{max} [°C]	DF
	Large pebbles	1.0×10^{21}	2.1×10^9	8.0×10^8	≈38	>850	10
''Beryllium'' Caps. 2	Large pebbles (pieces)	1.0×10^{21}	2.1×10^9	1.4×10^9	≈67	800	50
	Small pebbles	1.0×10^{21}	2.1×10^9	2.0×10^9	≈100	400, 800	5×10^2

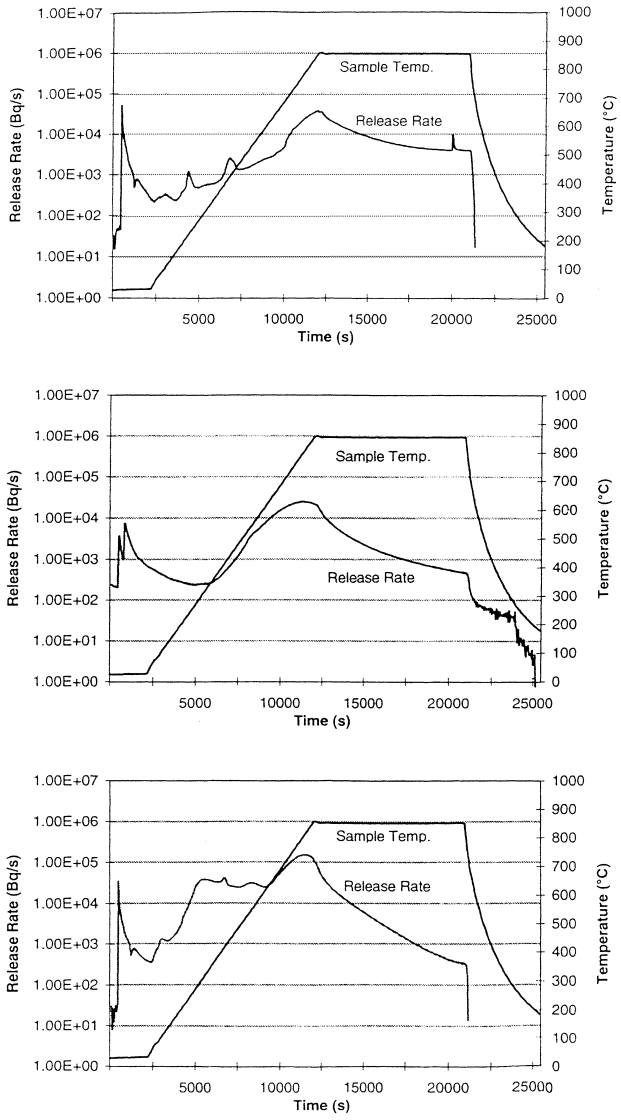

Fig. 2. Tritium release kinetics for large beryllium pebbles (top), pieces from crushed large pebbles (middle) and for small pebbles (bottom) from the "Beryllium" experiment.

capsules 26.2-1 and 28.2 can be only due to implantation from ceramics. Furthermore, the total release from the small Be pebbles is about a factor 15 larger than from the bigger ones. This is probably due to the fact that in the small Be pebbles the implantation depth of tritium coming from ceramics (≈40 μm) is of the same order of magnitude as the pebbles radius (≈50–100 μm), which results in a higher implantation efficiency than in the case of larger Be pebbles.

5. Summary

The behavior of both unirradiated beryllium pebbles and beryllium pebbles irradiated in the EXOTIC-7 as well as in the "Beryllium" experiments has been investigated. The bigger unirradiated beryllium pebbles usually show a relatively large number of indentations on their external surface. Relatively often a coarse porosity with a pore size of 0.1–0.2 mm has been observed. On the other hand, most of the pebbles reveal a very small micro-porosity usually oriented along the crystal axis showing a very fine dendritic or cellular structure. The metallographic structure of both smaller and bigger pebbles shows the presence of large grains, in some of the smaller pebbles as large as the pebble diameter. Two extraneous phases are present in almost all the analyzed pebbles. Because of the relatively low irradiation temperatures in both EXOTIC-7 and "Beryllium"

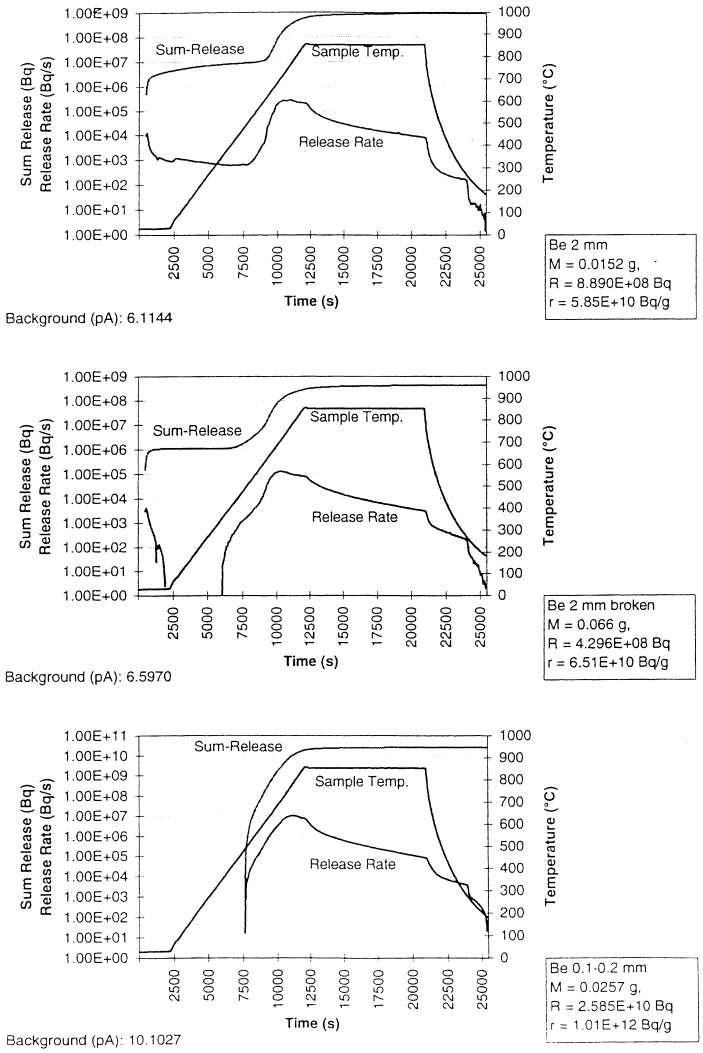

Fig. 3. Tritium release kinetics for large beryllium pebbles (top), pieces from crushed large pebbles (middle) and for small pebbles (bottom) from the EXOTIC-7 experiment (capsule 28.2).

experiments no remarkable thermally induced changes in the beryllium structure (macroscopic and microscopic) have been observed after the neutron irradiation. However, although the fast neutron fluence was almost the same in both experiments, the beryllium pebbles from EXOTIC-7 showed strong irradiation induced structural changes due to the contact with ceramics during the irradiation.

From the mechanical point of view it has been observed that, probably due to the very small amount of BeO impurities, all the pebbles showed a high ductility at room temperature. Pebbles loaded up to 400 N show diameter reductions up to 13% but, in spite of evident large plastic deformations, no fracturing or crack formation was registered. The mechanical properties of the pebbles from "Beryllium" experiment do not significantly change after irradiation. On the contrary, pebbles from EXOTIC-7 show an increase up to about 30% of their elastic behavior range and a decrease up to about 50% of their plastic behavior range. The decrease of the plasticity is much more evident in the smaller pebbles due to the fact that the implantation depth of tritium coming from ceramics is of the same order of magnitude of the pebbles radius, which results in a higher implantation efficiency than in the case of the larger Be pebbles.

Concerning the observed tritium release kinetics of the pebbles from the "Beryllium" experiment it can be drawn that the release from 0.7 mm diameter pieces from the large pebbles is faster than that from the whole pebbles ("particle size" effect). However, a further decrease of the particle size to $\leqslant 0.5$ mm diameter

indicated no further release improvement. With reference to EXOTIC-7, the tritium release of all investigated Be samples from the mixed beds is very similar and, in agreement with previous studies, it starts at about 500°C and achieves a maximum at about 700°C. In any case, the neutron-generated tritium in Be is about a factor 50 lower than the released tritium from the larger Be pebbles and about a factor 700 lower than that from the smaller Be pebbles, assuming no release during the irradiation. This high tritium inventory in both the large and small Be pebbles from EXOTIC-7 is due to implantation from ceramics.

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