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Beryllium R&D for blanket application

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Abstract

The paper describes the main problems and the R&D for the beryllium to be used as neutron multiplier in blankets. As the four ITER partners propose to use beryllium in the form of pebbles for their DEMO relevant blankets (only the Russians consider the porous beryllium option as an alternative) and the ITER breeding blanket will use beryllium pebbles as well, the paper is mainly based on beryllium pebbles. Also the work on the chemical reactivity of fully dense and porous beryllium in contact with water steam is described, due to the safety importance of this point. © 1998 Elsevier Science B.V. All rights reserved.

1. Introduction

The four ITER partners plan to use beds of beryllium pebbles as the neutron multiplier of their DEMO relevant blankets and thus for their blanket modules to be tested in ITER. Recently it has been decided to use this solution for the ITER breeding blanket as well.

Because it is important to achieve a high beryllium density in the blanket a binary bed of larger and smaller beryllium pebbles is used, which allows the bed to achieve a packing factor of about 80%. The larger pebbles (diameter $\approx$ 2 mm) are fabricated by melting and are a relatively inexpensive intermediate product of the beryllium fabrication route, called Magnesium Reduction Method (MRM) [1]. On the other hand the smaller pebbles can be produced either by Melting and Spraying (MS) or by the Rotating Electrode Method (REM).

Beryllium has no structural function in the blanket, however microstructural and mechanical properties are important, as they influence the material behavior under neutron irradiation.

The helium produced mainly by the reaction ^{9}Be(n,2n)2 ^{4}He is the dominant cause of beryllium swelling and embrittlement under irradiation, which are the major lifetime limiting factors for the material. The tritium inventory in beryllium, produced by transmutation reactions, could constitute a safety hazard in case of accidental temperature excursions in the reactor. Furthermore, the chemical interaction between beryllium and structural material as well as the chemical reaction occurring when steam comes in contact with fully dense or porous beryllium are dealt with in the paper due to their safety relevance.

2. Pebble characterization and mechanical behavior

The larger 2 mm MRM (FZK) pebbles have a relatively large number of surface indentations. Optical microscopy revealed large pores of various size and a microporosity usually oriented along the crystal axis perpendicular to the basal plane. The average open porosity of the pebbles is 0.57% and the closed porosity is 0.84% [2]. The smaller MS pebbles (0.1–0.2 mm) have an average open porosity of 0.86% and an average closed porosity of about 2.4%. Insoluble impurities have been usually observed on the grain boundaries, while iron and chrome are almost exclusively present in solid solution in the beryllium-matrix. The external surface of the large beryllium pebbles is usually covered by a 2 μm thick SiO_2 layer and/or a fluorine layer (probably BeF_2) with a thickness generally lower than 2 μm. Two extraneous phases are present in almost all the analyzed pebbles.

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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 [3].

Table 1 shows the chemical composition of the REM and MRM (JAERI) pebbles [4,5] and of the MRM (FZK) and the MS pebbles [2]. The difference of BeO content in the two qualities of MRM pebbles is considerable, pointing out that this type of material could not be very well characterized in some cases. Brush Wellman is prepared, in case of large orders of this kind of pebbles, to produce them by melting under vacuum and leaving the liquid beryllium percolate through a perforated metal sheet and fall in a 10 m high tower (shot method). With this method the pebbles would have better defined properties and could be produced in larger quantities [6].

The basic characterization results of the REM1 (produced with a hot-pressed beryllium electrode) and REM2 (vacuum cast beryllium electrode) are shown in Table 2. Almost all REM pebbles have a big pore at their center, produced by the volume reduction due to solidification (pebble density = 0.97% TD). The sphericity and the grain size were measured from the cross section of beryllium pebble by optical microscopy. The specific surface is measured by the BET method with nitrogen gas.

The mechanical behavior of the unirradiated MRM (FZK) pebbles has been investigated by submitting them to compressive loads 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 show 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 has been observed [2]. Pebbles loaded with 800 N (deformation up to 25%) or more, reveal cracks on their "meridian" planes [2]. Microhardness measurements have been also 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.

The mechanical properties of the pebbles from the "Beryllium" irradiation experiment [7] do not significantly change after irradiation. On the contrary, pebbles from the EXOTIC-7 experiment [8] show an increase up to about 30% of their elastic behavior range and a decrease up to about 50% of their plastic behavior range,

Table 1
Chemical compositions of beryllium pebbles (%)

	BeO	Al	Fe	Mg	Si	Mn	Cr	F	Zn	Co
REM 1	1.36	0.048	0.084	0.031	0.022	0.002	0.003	0.05	0.001	0.0006
REM 2	0.57	0.044	0.033	0.004	0.088	0.010	0.005	0.03	0.001	0.0005
MRM (JAERI)	1.31	0.041	0.062	0.242	0.038	0.009	0.005	0.18	0.08	0.0020
MRM (FZK)	0.08	0.025	0.073	0.16	0.020	0.011	0.015	0.099	0.012	0.0005
MS	0.34	0.04	0.090	0.01	0.03	–	–	0.008	–	–

Table 2
Basic characterization

Item	Value	
	REM1	REM2
Pebble diameter [a] (mm)	0.809 ± 0.040	0.808 ± 0.039
Pore diameter [a] (mm)	0.179 ± 0.024	0.184 ± 0.025
Density (g cm^{-3})	1.796 (97.2%TD)	1.791 (96.9%TD)
Sphericity [b] (%)	1.6	1.1
Specific surface area (cm^2 g^{-1})	4.54×10^{-3}	5.47×10^{-3}
Surface roughness (μm)	$R_a = 0.68$ $R_{max} = 3.81$	$R_a = 0.45$ $R_{max} = 2.36$
Grain size (mm)	0.53	0.53

[a] Average ± s.
[b] $(D_{max}-D_{min})/D_{av}$.

due to the tritium implanted in the Be from the adjacent lithiated ceramic pebbles [2].

The compression tests for the unirradiated pebbles and for pebbles irradiated in JMTR at 330°C and with a neutron fluence of 1.3×10^{21} n/cm^2 ($E > 1$ MeV) were carried out at room temperature. The results of these tests indicated that the fracture loads (about 150 N) of REM1 pebbles of 0.9 mm in diameter were unaffected by irradiation, while the fracture displacement decreased from 0.17 to 0.11 mm showing a certain degree of embrittlement.

3. Pebble bed heat transfer parameters

The heat transfer parameters of the binary pebble bed, namely the thermal conductivity and the heat transfer coefficient to the containing wall have been obtained by measurements performed at FZK. These two parameters have been correlated with the percental "interference" (thermal expansion of the bed minus thermal expansion of the containment) of the pebble bed with the containing wall and with the average bed and the wall temperature, respectively [9].

The experiments were performed with a rigid containment and thus they are applicable only to the cases where very small deformations of the bed containment are expected. Experiments with the measurement of a further parameter, namely the pressure exerted by the expanding bed on the containing wall, are required for the applications in case of deformable walls. These experiments are planned by FZK.

4. Swelling and tritium release

The helium produced mainly by the reaction ^{9}Be(n,2n)2 ^{4}He is the dominant cause of beryllium swelling and embrittlement, which are the major lifetime limiting factors for the material. Furthermore, the tritium inventory in beryllium, produced by transmutation reactions, could constitute a safety hazard in case of accidental temperature excursions in the reactor. The helium bubbles and the oxygen present in the material appear to be the main causes of tritium retention in irradiated beryllium.

The computer code ANFIBE, developed at FZK, is able to model the precipitation of helium atoms into intragranular bubbles, the migration of these bubbles to the grain boundaries to form intergranular bubbles, the growth and coalescence of these bubbles and their interlinkage with pores. The description of the helium behavior in beryllium accounting for the beryllium properties (mainly surface tension, temperature and irradiation induced creep) allows to calculate the volume swelling. Furthermore, the code models the tritium diffusion in the beryllium and the trapping of the tritium by the helium bubbles (physical trapping) and by the beryllium oxide (chemical trapping) [10]. The comparison of the ANFIBE calculations with the experiments available from the literature, including the recently performed Post Irradiation Experiments for irradiated beryllium pebbles performed by Battelle PNNL in collaboration with FZK and JAERI [11] shows a quite good agreement.

The blanket-end-of-life beryllium swelling has been calculated for the European HCPB (Helium Cooled Pebble Bed) DEMO blanket with the use of older pebble bed heat transfer correlations obtained with the extrapolation of experimental data obtained for mixed beds of beryllium and Li_4SiO_4 pebbles. The maximum calculated swelling was about 8%, i.e. considerably smaller than the pebble bed void fraction (20%). These calculations should be repeated with the new correlations obtained recently [9] or, even better, with the correlations of the experimental data which should be obtained in 1998.

The Beryllium [7] as well as the EXOTIC-7 [8] experiments carried out in the HFR reactor in Petten are detailed and significant tests for investigating the tritium release kinetics in irradiated beryllium pebbles. The beryllium irradiated in both the Beryllium and the EXOTIC-7 experiments consists of a mix of about 2 mm and 0.1–0.2 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 pebbe beds were irradiated. Release kinetics and total amount of released tritium were determined at FZK by out-of-pile 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 He+0.1 vol% H_2. A series of tests showed that the release characteristics of each type of pebbles are reproducible.

For the pebbles from the Beryllium experiment it has been found that the release from 0.7 mm diameter pieces from broken 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. Release from the small pebbles is faster than that from 0.7 mm diameter pieces from the large pebbles.

The release of all investigated beryllium samples (Be 2 mm, Be 2 mm broken, Be 0.1–0.2 mm) of the EXOTIC-7 irradiation from the mixed beds [8] is very similar. In agreement with previous studies [12,13] the release starts at about 500°C and achieves a maximum at about 700°C. The observed release at about 500°C is probably due to tritium escaping from chemical traps (BeO), while the maximum release at about 700°C is due to tritium escaping from physical traps (He bubbles).

5. Compatibility with structural materials

Under the conditions of high temperature and long heating time typical of a fusion reactor, the chemical interaction between beryllium and structural material could represent a concern. Flament et al. [14,15], Hofmann and Dienst [16] and Terlain et al. [17] showed that out-of-pile the compatibility limit of beryllium for 316 stainless steel and for ferritic martensitic steel are 600°C and 650°C, respectively. The experiments of Kawamura show that this limit is 600°C for stainless steel and between 600°C and 800°C for the martensitic-ferritic steel F82H [18–20].

6. Safety

For any application of Be in a fusion reactor, the possibility of a coolant or gas leak that exposes the beryllium to oxygen or water must always be considered. Be has a very strong affinity for O. Normally a protective oxide film forms on the surface of Be almost immediately after being machined or otherwise fabricated. At elevated temperatures, especially after neutron irradiation, that film loses its protective character, and extended oxidation takes place. In addition to being destructive of the beryllium, if the reaction is with water, the H_2 liberated poses the added threat of a chemical explosion.

Work to explore the rate at which Be oxidation takes place has been ongoing for several years. Pioneering work was done at TRW [21]. Most of the recent work of Be has been done on steam oxidation at the Idaho National Engineering and Environmental Laboratory (INEEL) [21–25] though work has been done in the Russian Federation [26], and some aspects are being considered in other institutions [27,28]. The research has shown that the oxidation rate depends on many factors, but a major one is the amount of interconnected porosity or available surface area over which the reaction can take place. Temperature is obviously also important. Factors that can increase specific surface area include the processes used in manufacturing the beryllium and the generation of internal gas pockets and other defects from neutron irradiation.

As an indicator of the progress made, a series of experiments was conducted at the INEEL [23,29] on Be specimens that had been irradiated in the EBR-II fast-flux reactor. Irradiation was in the COBRA-1A2 test facility in the EBR-II reactor at a nominal temperature of 400°C, to a fast neutron fluence ($E > 0.11$ MeV) of 5.2×10^{22}–6.7×10^{22} n/cm^2. Some specimens were subjected to annealing tests to observe swelling and gas release processes. Others were exposed to steam at elevated temperatures. Control samples, not irradiated, were subjected to the same experiments. Samples were all from the same lot, manufactured from Brush Wellman SP-200-F powder by cold isostatic pressing, sintering in vacuum at 1250°C for 4 h, and hot isostatic pressing at 1000°C and 103 MPa for 3 h to achieve densities of 1.854 g/cm^3. They were subsequently dry machined to cylinders 0.75 cm in diameter. Lengths of 0.635 and 2.032 cm were used in these tests.

Steam reactivity experiments included measurements with an on-line mass spectrometer for H_2 generation rates and an on-line ion chamber for tritium measurements. Ar was used as a sweep gas to carry reaction products through the various measurement stations described in [23,25]. Samples were brought to temperature in the moving Ar atmosphere prior to the introduction of steam. Fig. 1 shows the results of the steam exposure experiments for both irradiated and unirradiated specimens. For experiments at 600°C and below, the H_2 generation for irradiated specimens-exhibited a quasi-parabolic behavior, similar to that seen in unirradiated specimens. A significantly higher oxidation rate was observed for the irradiated specimen exposed to steam at 700°C (experiments INEL96-G3 and INEL 96-WG3 of Fig. 1). It became self-sustaining, causing the temperature to increase to 1000°C or more, and the specimen was almost entirely consumed. This was determined to be due to the high degree of interconnected porosity that develops when irradiated Be is heated to temperatures above 600°C. Irradiated specimens were not exposed to steam at temperatures higher than 700°C. 88% dense beryllium was also tested at INEEL. This porous specimen underwent a self-sustaining oxidation reaction at 600°C and at temperatures above that [22]. In a recent experiment at INEEL, plasma sprayed beryllium at 92–96% density was preheated at 700°C and exposed to steam and underwent a thermal excursion to over 1000°C [24].

7. Conclusions

The problems related to the use of pebbles in the blanket of a DEMO or in the ITER blanket have been defined and are being investigated in several laboratories in Europe, Japan and USA. The characterization and the investigation of the mechanical behavior of irradiated and unirradiated pebbles are being performed in Europe and Japan.

Experiments to determine the heat transfer parameters of binary pebble beds have been performed in Europe. Further experiments are in the planning stage to investigate the effect of the pressure exerted by the expanding bed on the containing walls, besides the already investigated effects of temperature and of the interference of the pebble bed with the containing walls.

Swelling and tritium release can be predicted relatively well by the code ANFIBE also for beryllium pebbles, especially after the PIE of pebbles irradiated in

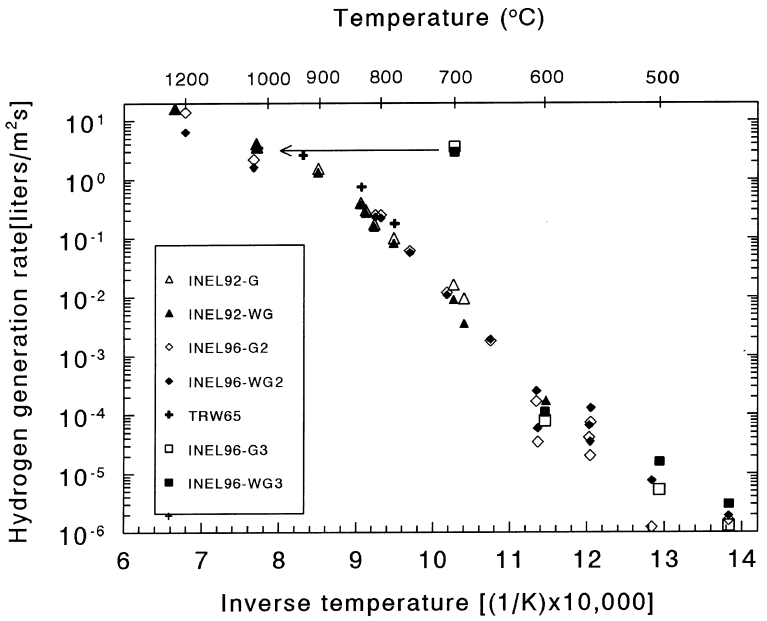

Fig. 1. Comparison of H_2 generation rates for irradiated (INEL96-G3, -WG3) and unirradiated (INEL96-G2, -WG2) [24,29] Be cylinder specimens with unirradiated disk specimens (INEL92-G) [25] with measurements made by Blumenthal and Santy (TRW65) [21]. **G** and **W** in the experiment identifier correspond to analyses based on gas measurements and weight gain measurements, respectively.

two fast reactors in USA have been performed. However, further irradiation experiments of beryllium pebbles are required at higher neutron fluences.

The compatibility of beryllium with structural materials has been investigated out-of-pile. Martensitic steels behave better than austenitic ones.

The chemical reaction of beryllium with steam has been investigated mainly in USA. The experiments indicate that the fully dense beryllium behaves considerably better than the porous one.

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