Journal of Nuclear Materials 386–388 (2009) 874–877

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

Tritium release from beryllium articles for use in fusion devices

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ARTICLE INFO

PACS:
81.05.Bx
81.40.Wx
81.70.Jb

ABSTRACT

Results obtained on radiation and magnetic field (MF) effects on tritium release at annealing of the beryllium pebbles from the EXOTIC-8-3/13 irradiation are presented in this study and compared with those for other irradiated beryllium materials. Abundance ratios of chemical forms of tritium in the EXOTIC-8-3/13 beryllium pebbles were determined: T_2 – 65%, T^0 – 23%, T^+ – 12%. A complete detritiation of these pebbles was achieved at 1123 K for 240 min; MF of 2.35 T had no appreciable effect on the tritium release. At 991 K for 240 min, the degree of detritiation was 96.6% without MF; MF of 2.35 T decreased it to 86.7%. At 940 K for 47 min, the degree of detritiation was 60%, 5 MeV fast-electron radiation of 14 MGy/h increased it to 76%, but the simultaneous action of the fast-electron radiation and MF of 1.7 T increased it to 88%.

1. Introduction

Beryllium is foreseen to be used as the first wall material according to the current ITER design [1], and as neutron multiplier according to the reference concept of the Helium-Cooled Pebble-Bed (HCPB) blanket in the European Breeding Blanket Programme for the DEMO design [2,3]. Tritium accumulation in beryllium articles of fusion devices under operation conditions is an important technological and environmental issue. Under the real operating conditions of the HCPB, the multiplier Be pebbles will be subjected to action of a high temperature up to 920 K, intense fast neutron radiation of 2.4 MW m^{-2} (i.e. 10^{18} n m^{-2} s^{-1}), and a high magnetic field (MF) of 7–10 T [2,3]. Results obtained on radiation and MF effects on tritium release at annealing of the beryllium pebbles from the EXOTIC-8-3/13 irradiation are presented in this study and compared with those for other irradiated beryllium materials.

2. Experimental

2.1. Samples investigated

The beryllium pebbles irradiated in the EXOTIC-8-3/13 experiment were investigated in this study. These pebbles of a diameter 0.1–0.2 mm have been manufactured by spraying molten beryllium in an inert atmosphere (inert gas atomization process, IGA) at Brush Wellman Inc. [4,5]. Their grain size is 40 to 200 μm [5]. The main impurities are 3400 ppm BeO, 100 ppm Mg [5]. The irradiation programme EXOTIC (EXtraction Of Tritium In Ceramics) was carried out in the High Flux Reactor (HFR) in Petten, the Netherlands [5–7]. The pebbles were irradiated for 449.8 days in the HFR at temperatures 800–900 K with a neutron fluence of 2.7×10^{25} m^{-2} (E > 0.1 MeV) of a fast fission spectrum [5]. The ^{4}He content of 285 appm and the ^{3}H content of 1.16 appm (i.e. 138 MBq/g) at the end of the irradiation in the year 2000 were calculated on the basis of the irradiation history [5].

2.2. Dissolution method of tritium analysis

In order to determine the total tritium activity and abundance ratios of chemical forms of tritium (T_2, T^0, T^+) in irradiated beryllium pebbles, two weighed amounts of the pebbles were dissolved in pure 2 mol/L H_2SO_4 and in the solution 2 mol/L H_2SO_4 + 0.5–1 mol/L $Na_2Cr_2O_7$ in a special setup [8]. Beryllium dissolving in pure acid forms hydrogen – 1 molecule of H_2 corresponds to 1 Be atom. The rate of hydrogen evolution was measured with a catarometer. The tritium forms T_2 and T^0 of the activities A_{T_2} and A_{T^0}, respectively, localized in a Be sample transfer as T_2 + HT into a flow of carrier gas, where the tritium activity released, $A_{T\_gas\_acid} = A_{T_2} + A_{T^0}$, was determined with a gas flow-through proportional meter TEM 2100A with a detector DDH 32. T^+ localized in a Be layer remains in the solution. After the Be pebbles had completely dissolved, the tritium activity in the solution, $A_{T\_sol\_acid}$, was measured with liquid scintillation method: $A_{T\_sol\_acid} = A_{T^+}$, where A_{T^+} – the activity of T^+ in the sample dissolved. In the second dissolution experiment the scavenger of H^0 (T^0) – 0.5–1 mol/L $Na_2Cr_2O_7$ decreases H^0 (T^0) by 90%: $H^0 + Cr_2O_7^{2-} \rightarrow H^+$ (solution) + Cr^{3+}. Then the activity of the tritium released into a gas phase and retained in the solution are the respective sums: $A_{T\_gas\_Cr(VI)} = A_{T_2} + x \cdot A_{T^0}$ and $A_{T\_sol\_Cr(VI)} = A_{T^+} + (1-x)\ A_{T^0}$, where $x = n_{H_2}/n_{Be}$ (x was found to be 0.1); n_{H_2} – the molar amount of the

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doi:10.1016/j.jnucmat.2008.12.193

Table 1
The total tritium activity, A_T, and abundance percentages of chemical forms of tritium localized in irradiated beryllium samples.

Beryllium sample	A_T	T_2 (%)	T^0 (%)	T^+ (%)	Reference
EXOTIC-8-3/13 pebbles	4–18 MBq/g	65	23	12	This paper
BERYLLIUM pebbles	0.6–1.5 GBq/g	85	10	5	[8]
JET tile "*A*"	31.7 kBq/cm^2	44	42	14	[10]
JET tile "*B*"	4.8 kBq/cm^2	73	16	11	[10]

hydrogen evolved at the dissolution; n_{Be} – the molar amount of the beryllium dissolved. The contents of T^0, T_2, T^+ (Bq g^{-1}) in a sample were determined separately from the corresponding differences in the activities: $A_{T^0} = (A_{T\_gas\_acid} - A_{T\_gas\_Cr(VI)})/(1\text{-}x)$; $A_{T_2} = A_{T\_gas\_acid} - A_{T^0}$; $A_{T^+} = A_{T\_sol\_acid}$.

2.3. Measurement of tritium release rate at annealing

Annealing of Be samples was performed in a continuous flow of the purge gas He + 0.1% H_2 of the rate 14.3 ± 0.5 L/h without and in MF of 1.7 T and/or 5 MeV fast-electron radiation of the dose rate P = 14 MGy h^{-1} in a special rig [9] or in a continuous flow of the purge gas argon of the rate 15 ± 1 L/h in another setup where MF up to 2.35 T can be applied without the fast-electron radiation. The tritium released was measured continuously with a meter TEM 2100A with a detector DDH 32. In the experiments, the sample temperature was increased linearly with time at the rate 5–6.7 K/min from a room temperature to the end temperature selected from the range 773–1123 K, which was kept constant within ±5 K for 47 to 240 min. Then the heater was switched off, and the sample was left to cool in a flow of the purge gas. The radioactivity of tritium released was calculated as Bq g^{-1} to 1 g of the sample of Be pebbles.

3. Results and discussion

3.1. Chemical forms of tritium

Using the dissolution method, abundance ratios of chemical forms of tritium localized in the EXOTIC-8-3/13 irradiated Be pebbles were determined. These values are compared in Table 1 with other values determined for the BERYLLIUM irradiated Be pebbles and subsurface layers of the plasma facing surface of the JET D–T plasma exposed upper belt limiter Be tiles. It should be noted that the EXOTIC-8-3/13 Be pebbles were found to be inhomogeneous with respect to the total tritium activity, A_T, which was found in the range 4–18 MBq/g. This inhomogeneity of the sample limited the accuracy of the determination of the abundance ratios of chemical forms of tritium as the identity of the two samples is essential for this determination. We can see that the most part of tritium in the EXOTIC-8-3/13 Be pebbles, similarly to the BERYLLIUM pebbles and the JET tile "*B*", is localized as molecular tritium T_2, but the abundance ratios of T^0 and T^+ in the EXOTIC-8-3/13 Be pebbles are about by a factor of 2 greater than those in the BERYLLIUM pebbles.

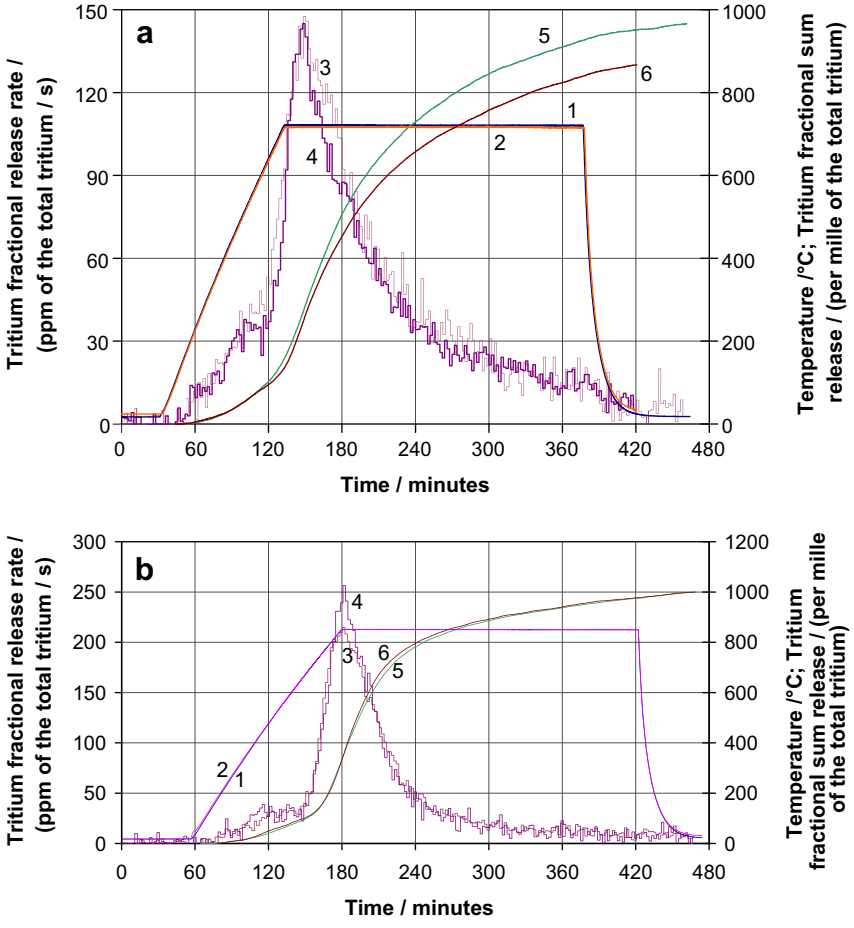

Fig. 1. Tritium fractional release rate (3, 4) and tritium fractional sum release (5, 6) at annealing of the EXOTIC-8-3/13 Be pebbles at a given temperature (1, 2) – a constant rate of temperature increase 6.7 K/min and at a constant temperature of 991 K ± 4 K (a) and 1123 K ± 2 K (b) for 240 min without (1, 3, 5) and in MF of 2.35 T (2, 4, 6).

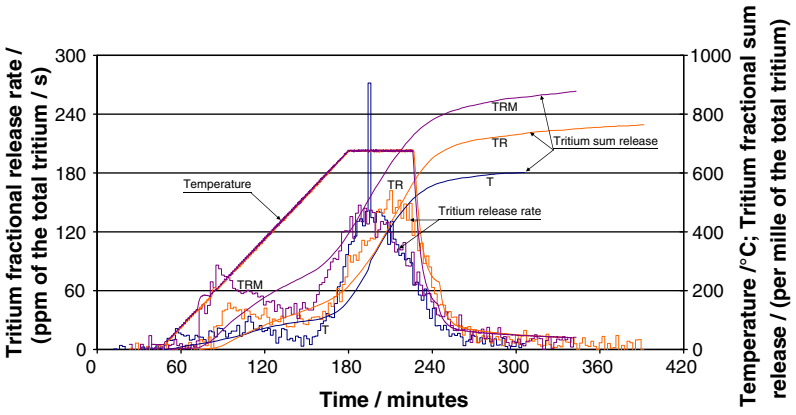

Fig. 2. Tritium fractional release at annealing of samples of the EXOTIC-8-3/13 Be pebbles in a continuous flow of 14.3 ± 0.5 L/h of He + 0.1% H_2.

3.2. Tritium release at annealing

In order to investigate possible effect of MF on tritium release from the Be pebbles irradiated in the EXOTIC-8-3/13 experiment, they were annealed at a constant rate of temperature increase 6.7 K/min to 991 and 1123 K and then at a constant temperature for 4 h in a flow of argon of 15 ± 1 L/h, the curves of tritium release are given in Fig. 1. The tritium fractional sum release in Fig. 1 has been calculated for the following values of the initial total tritium activity for 1 g of the sample, MBq/g: 7.98 (a, 5); 9.34 (a, 6); 10.69 (b, 5); 11.52 (b, 6). At heating, at about 900–940 K, the slope of the curves of the tritium release rate increases, which can be explained that additional channels for the tritium release open [4,5]. Subsequent dissolution experiments indicated that no tritium remained in the pebbles after annealing at 1123 K for 240 min. The curves in Fig. 1(b) testify no appreciable MF effect on the tritium release. On the other hand, a larger fraction of the residual tritium of 13.3% had remained after annealing at 991 K for 240 min in MF of 2.35 T in comparison with 3.4% in the case of annealing without MF (Fig. 1(a)), which testifies that under the given conditions MF of 2.35 T decreased the fractional tritium release by about 10%. Experiments in the radiation thermal magnetic rig show that the electron radiation (TR) and the simultaneous electron radiation and magnetic field (TRM) stimulates the tritium release at annealing in comparison with the action of only temperature of 940 K for 47 minutes (Fig. 2). Curve labels in Fig. 2 and category axis labels in Fig. 3 denote the following acting factors: *T* – action of the given temperature; *M* – magnetic field of 1.7 T; *R* – 5 MeV fast-electron radiation of 14 MGy/h. The tritium fractional sum release in Fig. 2 has been calculated for the following values of the initial total tritium activity, MBq/g: 6.20 (T); 5.76 (TR); 6.15 (TRM). Degrees of detritiation obtained in these experiments with the EXOTIC Be pebbles are compared in Fig. 3 with those obtained in previous experiments with samples of different beryllium articles under the following conditions of their annealing: EXOTIC Be pebbles – ramp of 5 K/min to 940 K and at 940 K for 47 min in a continuous flow of He + 0.1% H_2; BERYLLIUM pebbles – a constant temperature 1123 K for 120 min in an ampoule [8]; Be tile "*A*" – ramp of 5 K/min to 773 K and at 773 K for 30 min in a continuous flow of He + 0.1% H_2 [10]; Be tile "*B*" – a constant temperature 773 K for 30 min in an ampoule [9]. We can see from the experimental results obtained that under similar conditions the detritiation of the EXOTIC Be pebbles takes place easier than that of the BERYLLIUM pebbles. That can be explained by the fact that the EXOTIC pebbles have smaller diameter (∅ 0.1–0.2 mm) than the BERYLLIUM pebbles (∅ 2 mm) and therefore a larger ratio of free surface to grain boundary surface. A larger abundance ratio of the atomic form T^0 could also contribute to a higher tritium release of the EXOTIC pebbles. A large degree of detriation of 60% at 940 K for 47 min of the EXOTIC Be pebbles and a lesser abundance of the molecular form T_2 possibly could explain that radiation and MF have relatively lesser effects on the degree of detritiation of the EXOTIC Be pebbles than on the detritiation of the BERYLLIUM pebbles.

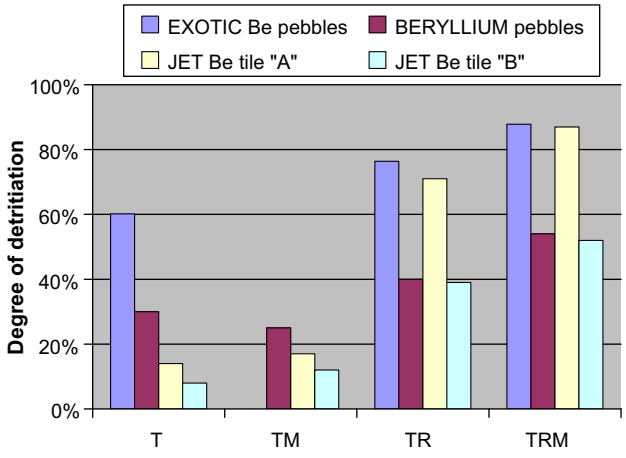

Fig. 3. Comparison of degrees of detritiation of EXOTIC-8-3/13 and BERYLLIUM pebbles and JET Be tiles under the conditions given in text.

4. Conclusion

Abundance ratios of chemical forms of tritium in the beryllium pebbles irradiated in the EXOTIC-8-3/13 experiment were determined: T_2 – 65%, T^0 – 23%, T^+ – 12%. A complete detritiation of these pebbles was achieved at 1123 K for 240 min; MF of 2.35 T had no appreciable effect on the tritium release. At 991 K for 240 min, the degree of detritiation was 96.6% without MF; MF of 2.35 T decreased it by about 10% to 86.7%. At 940 K for 47 min, the degree of detritiation was 60%, 5 MeV fast-electron radiation of 14 MGy/h increased it to 76%, but the simultaneous action of the fast-electron radiation and MF of 1.7 T increased it to 88%. Under similar conditions, the detritiation of the EXOTIC-8-3/13 Be pebbles takes place easier than that of the BERYLLIUM pebbles.

Acknowledgements

This study was carried out with EFDA financial support (Technology Task TW5-TTBB-006-D08).

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