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Effects of helium production and radiation damage on tritium release behavior of neutron-irradiated beryllium pebbles

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Abstract

The tritium release from neutron-irradiated beryllium pebbles, irradiated under different helium production (0.5–1.0×10^3 appm He) and dpa (4.2–8.6) conditions, was studied. From these results, it was clear that the apparent diffusion coefficient at 600°C was significantly affected by irradiation conditions, but returned to normal values at 900°C, apparently due to thermal annealing. Multiple peaks in the tritium release curve at 900°C were observed.

1. Introduction

Tritium release behavior from neutron-irradiated beryllium pebbles is important to evaluate tritium inventory in a fusion reactor blanket. From recent studies, it has become clear that tritium release from beryllium was significantly affected by a surface oxide layer [1,2], the grain size of the specimen [3], and the connection of helium bubbles, which lead to a burst release [3,4]. Mechanical properties are affected by both helium production and damage by neutron irradiation [5]. However, the effect of both parameters on the tritium release behavior of beryllium pebbles is not clear. Hence, tritium release from neutron-irradiated beryllium pebbles, irradiated to different helium levels and dpa were studied.

2. Experimental

The specimens were beryllium pebbles produced by the Rotating Electrode Method (REM pebbles, NGK Insulators). The diameter of pebbles was about $\varnothing$ 0.8 mm, and the grain size was about 0.5 mm. The chemical compositions of beryllium pebbles are listed in Table 1. There were three neutron-irradiation conditions, summarized in Table 2 [5–7]. Detailed information on the beryllium pebbles has been reported in [8–10]. Specimens P1 and P2 were irradiated in Japan Materials Testing Reactor (JMTR) in an alumina container in a sealed capsule with He (6 N purity). Specimen P3 was irradiated in EBR-II (Experimental Breeder Reactor) in a TZM container in a sealed capsule with He (6 N purity). The apparatus for tritium release measurements is shown in Fig. 1 [3]. He + 1%H_2 carrier gas was used for the tritium release experiments, and the flow controlled carrier gas was introduced at 50 cm^3/min into a small electric furnace. The carrier gas was divided into two measuring lines: IC1 for HT + HTO and IC2 for HT. The delay time for tritium measurement was about 2 min. Beryllium pebble specimens were heated on a BeO plate in an alumina crucible in a small electric furnace that was installed in a cylindrical quartz glass tube (inner diameter 30 mm, outer diameter 34 mm, length 100 mm). Ten pebbles were heated to 300°C, 600°C, 900°C and 1000°C, and were held for 20 min at each temperature.

3. Results and discussion

Results of the tritium release measurement are shown in Fig. 2. The tritium release curve for P1 is not shown

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Table 1
Chemical composition of Be pebble

Element (wt%)					
Be	BeO	Fe	C	Al	Mg
98.2	1.51	0.11	0.023	0.078	0.017

Table 2
Neutron-irradiation condition

Specimen	T_{irr} (°C)	Fluence $\times 10^{21}$ (n/cm^2)[a]	Helium $\times 10^3$ (appm)	dpa	He/dpa $\times 10^2$	D_{ave} (mm)[b]
P1	445	3.0	0.7	4.2	1.7	0.793
P2	616	4.3	1.0	6.0	1.7	0.814
P3	383	4.3	0.5	8.6	0.6	0.805
Ref. [2]	200	0.085	0.002	0.1	2.0	0.871

[a] Fast neutron energy: $E > 1$ MeV.
[b] Average diameter of 10 pebbles.

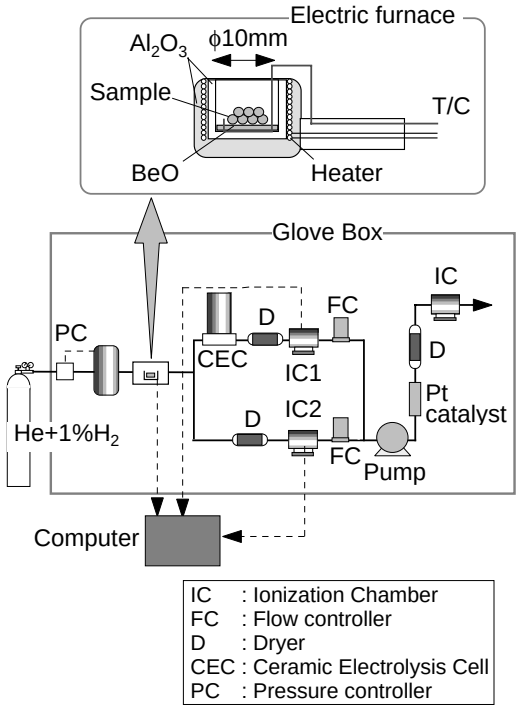

Fig. 1. Apparatus of tritium release measurement.

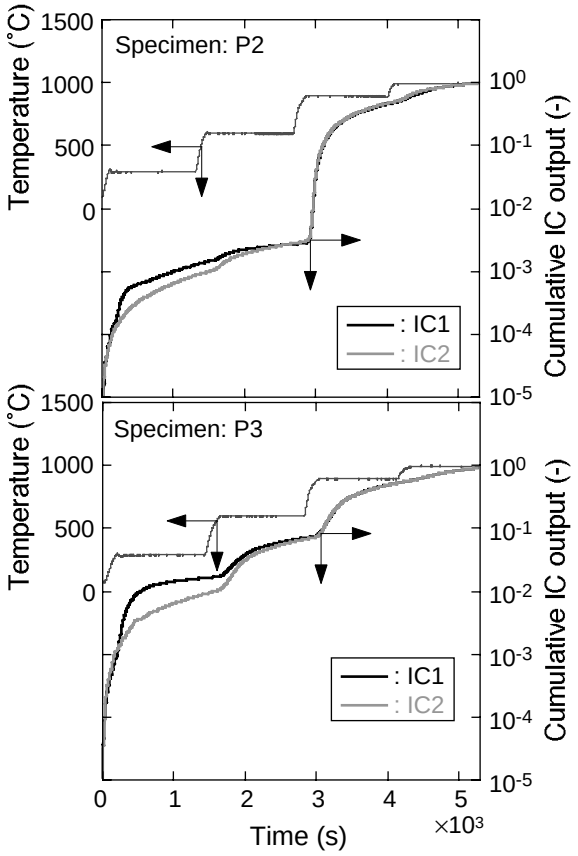

Fig. 2. Result of tritium release measurement.

because it was almost the same as P2. The IC1 signal at low temperature was bigger than that of IC2 for each specimen because tritiated water originated from the surface adsorbed water. It was observed that the tritium release at low temperature for P3 was higher than P1 and P2. Considering that P3 was irradiated to a higher dose at a lower temperature, a greater defect density might be expected; and since the trapping by defects generally restricts tritium release, this result was unexpected.

The tritium release curve for P1 at 900°C is shown in Fig. 3. Multiple peaks in the tritium release curve were observed. Multiple peaks suggest helium release because rapid helium release from neutron-irradiated beryllium

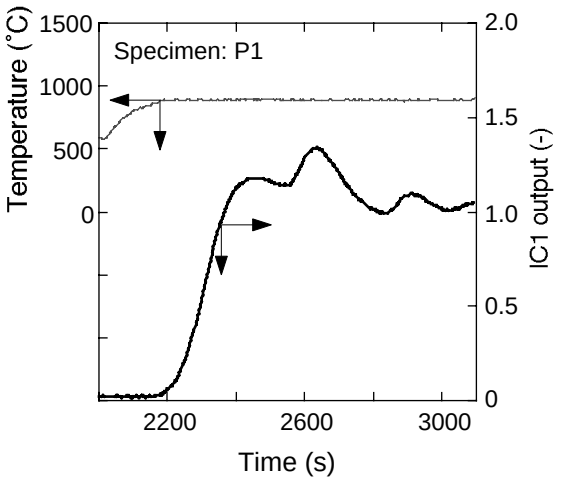

Fig. 3. Multiple peaks of P1 at 900°C.

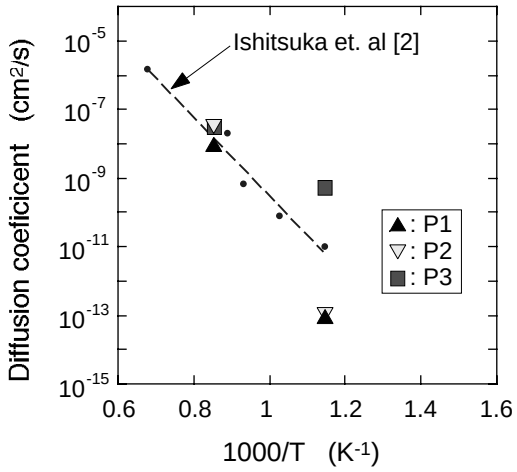

Fig. 4. Apparent diffusion coefficient.

pebbles at 900°C have been reported by Dalle Donne et al. [11].

The apparent diffusion coefficients at 600°C and 900°C were determined by curve fitting using the following simple model for a pebble [12] assuming closed porosity as observed in the pebbles (average pore diameter 0.09 mm [8]):

$$F = 1 - \frac{6}{\pi^2(a^2+ab+b^2)}\sum_{n=1}^{\infty}\left(\frac{b\cos n\pi - a}{n}\right)^2 \times \exp\left\{-\frac{Dn^2\pi^2 t}{(b-a)^2}\right\}. \quad (1)$$

Here, F is the fractional release rate, a the radius of pore, b the average radius of the pebbles, D the apparent diffusion coefficient, and t is the time. In this equation, the only unknown value is D, which can be determined from the experimental data.

The apparent diffusion coefficient is shown in Fig. 4, and the data trend from Ishitsuka et al. [2] for low fluence irradiations at 200°C (see Table 2) is also shown. The apparent diffusion coefficients for P1 and P2 showed similar values, and the value at 600°C was about two orders of magnitude smaller than that of the low fluence data [2]. On the other hand, the apparent diffusion coefficient of P3 at 600°C was about two orders of magnitude larger than that of the low fluence data. However, the apparent diffusion coefficient at 900°C of P3 was near the normal value, presumably as a result of annealing (300°C × 20 min + 600°C × 20 min).

Tritium release behavior of beryllium pebbles at low temperature will be affected by irradiation conditions, i.e., irradiation temperature, surface condition (oxide layer), helium production rate and radiation damage. However, it appears that the difference in irradiation conditions between P1 and P2 – primarily irradiation temperature – was negligible because the tritium release behavior of P1 was similar to P2. While P3 was irradiated at a still lower temperature and to a somewhat greater fluence, the biggest difference between the irradiation conditions of P3 and those of P1 and P2 is He/dpa (see Table 2). These results suggest the possibility that He/dpa may affect the tritium release behavior in beryllium pebbles.

4. Conclusion

The tritium release from neutron-irradiated beryllium pebbles, irradiated under different temperatures, helium production and dpa conditions, was studied, and the following results were obtained.

1. The apparent diffusion coefficient of specimens irradiated at 445°C and 616°C to 4–6 dpa with about 1×10^3 appm He showed similar values, and the value at 600°C was about two orders of magnitude smaller than that obtained from low fluence (0.1 dpa) data.
2. The apparent diffusion coefficient at 600°C of a specimen irradiated at 383°C to 8.6 dpa with 0.5×10^3 appm He was about two orders of magnitude larger than that obtained from low fluence data.
3. The apparent diffusion coefficients at 900°C of all the three specimens were similar to that obtained from low fluence data.

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