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Desorption of tritium and helium from high dose neutron irradiated beryllium

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Abstract

The effect of high dose neutron irradiation on tritium and helium desorption in beryllium is described. Beryllium samples were irradiated in the SM and BOR-60 reactors to a neutron fluences ($E > 0.1$ MeV) of $(5–16) \times 10^{22}$ cm^{-2} at 70–100 °C and 380–420 °C. A mass-spectrometry technique was used in out of pile tritium release experiments during stepped annealing in the 250–1300 °C temperature range. The total amount of helium accumulated in irradiated beryllium samples varied from 6000 to 7200 appm. The first signs of tritium and helium release were detected at temperature of 312–445 °C and 500–740 °C, respectively. It is shown that most tritium (~82%) from sample irradiated at 70–100 °C releases in temperature range of 312–700 °C before the beginning of helium release (740 °C). In the case of beryllium sample irradiated at 380–420 °C, tritium release starts at a higher temperature ($T_s > T_{ann} = 445$ °C) and most of the tritium (~99.8%) is released concurrently with helium which could be considered as evidence of co-existence of partial amounts of tritium and helium in common bubbles. Both the Be samples differ little in the upper temperatures of gas release: 745 and 775 °C for tritium; 1140 and 1160 °C for helium. Swelling of beryllium starts to play a key role in accelerating tritium release at $T_{ann} > 600$ °C and in helium release – at $T_{ann} > 750$ °C.

1. Introduction

Beryllium is considered as a main candidate material for armor of the first wall and as a neutron multiplier and moderator of the breeding blanket for DEMO. The efficiency of beryllium for fusion applications will strongly depend on its behavior under neutron irradiation. The most important consequences of neutron irradiation of beryllium are helium induced swelling and tritium retention and release. Tritium and helium behavior in irradiated beryllium is complex, depending on both the conditions of irradiation (helium inventory, irradiation temperature, duration, etc.) and properties of beryllium (density, grain size, beryllium oxide contents, etc.). In spite of the fact that considerable progress has been achieved in understanding of the above-stated problem in recent years [1–11], nevertheless direct experimental data on tritium and helium

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behavior in irradiation conditions which are close to the temperature-dose operating regime expected for the blanket of DEMO are still unavailable. This paper presents some new experimental results on investigation of tritium and helium behavior in beryllium.

2. Materials and experimental procedure

The study was performed for TE-400 grade samples (#1) irradiated in the fast reactor BOR-60 and TE-56 grade samples (#2) irradiated in the SM reactor. The irradiation parameters and some initial characteristics of beryllium are presented in Tables 1 and 2.

Tritium release kinetics were analyzed during stepped isothermal annealing. Gases liberated under sample heating were collected in a closed volume equipped with an omegatron mass-spectrometer (OMS). The OMS was calibrated against H_2 and ^4He standard leaks [8]. Released amounts of ^4He were calculated from reading the OMS signal at mass 4. Only mass 6 was used for the evaluation of tritium release because the fraction of tritium released in form of HT molecules was found to be negligible in our experimental conditions. Temperature was elevated by 20–170 °C in every step of multi-stage annealing. The heating duration at each step ranged between 0.9 and 40.5 h for annealing temperatures from 250 °C up to the melting point of beryllium. At the final step of multi-stage annealing, the samples were melted and the total amount of helium and tritium was measured. A microstructure study of samples after irradiation was carried out using the JEM 2000 FS-II transmission electron microscope. To estimate swelling evolution during stepped annealing, several samples of OE-56 grade were subjected to short-term vacuum annealing in the temperature range of 300–1200 °C with the duration of 1 h at each step.

Table 1
Characteristics of beryllium samples

Grade of Be	Density (g/cm^3)	Grain size (μm)	Be (wt%)	BeO (wt%)
TE-400	1.845	~80–100	~97.4	~2–2.3
TE-56	1.856	22–25	98.63	1.48

Table 2
Irradiation parameters of beryllium samples

Sample (grade)	Neutron fluence, $E > 0.1$ MeV (cm^{-2})	T_{irr} (°C)	t_{irr} (years)
#1 (TE-400)	1.6×10^{23}	380–420	12
#2 (TE-56)	5×10^{22}	70–100	2.5

3. Experimental results

3.1. Helium release

Measured helium releases depending on annealing temperature and duration are presented in Figs. 1 and 2 and Table 3.

Helium desorption from sample #1 (TE 400) started as early as at 500 °C (T_s). However, helium release rate still remained rather small up to the temperature of 775 °C and the total amount of release did not exceed 0.2%. Then helium release rate sharply increased by a factor ~100 at 775 °C and remained at this level during all further annealing steps up to 1140 °C (T_{upper}) when helium release came to an end. Maximum rates (V_{max}) were observed in the temperature range from 880 to 925 °C. About 20% of helium was released from beryllium during annealing at 775 °C for 10.5 h and ~88% during annealing at 775–925 °C with a total duration of 40.5 h.

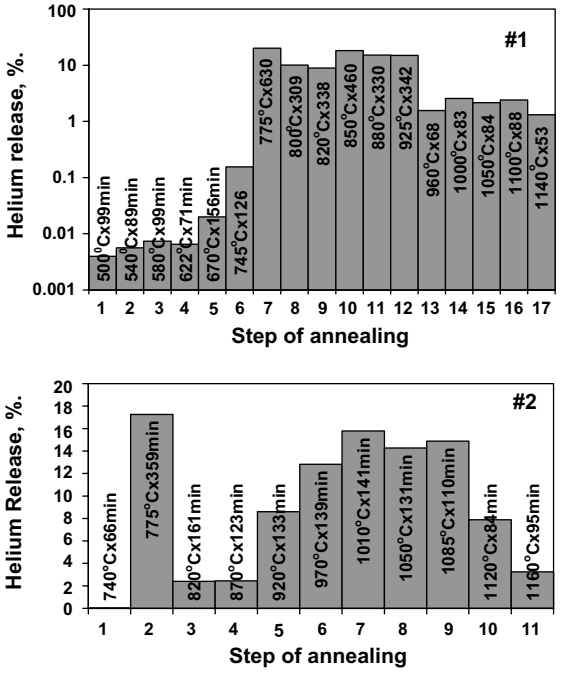

Fig. 1. Helium release (samples # 1–2).

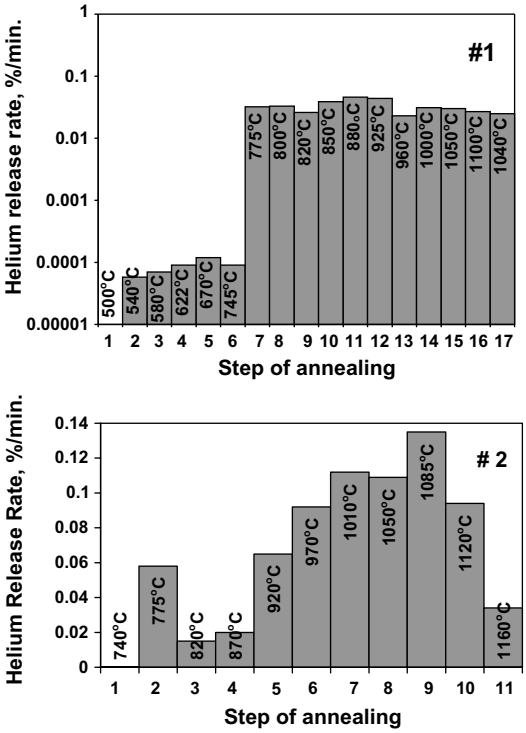

Fig. 2. Helium release rate (samples # 1–2).

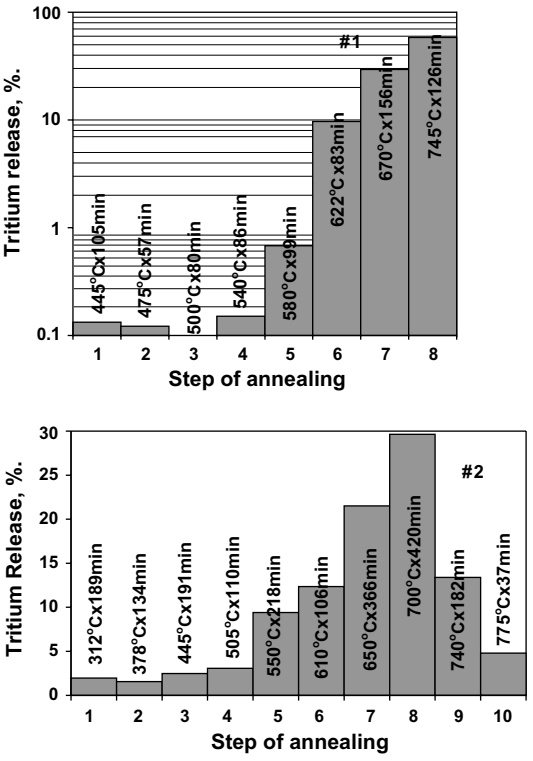

Fig. 3. Tritium release (samples # 1–2).

Helium desorption from sample #2 (TE-56) started (T_s) at a temperature of 740 °C (<0.01%). However, the intensity of helium release increased by a factor of ~300 at an annealing temperature of 775 °C, so more than 17% of the total amount of helium was released during 6 h. With further increase of annealing temperature, the rate of helium release decreased by a factor 3–4, however, the intensity of release was restored to the previous level at annealing temperature of 920 °C. During the next five steps (970, 1010, 1050, 1085 and 1120 °C) the rate of helium release increased additionally by a factor 1.5–2.0 with a peak rate at 1085 °C. Totally about 97% of helium contained in the sample was released in the temperature range 775–1120 °C. Last signs of helium release were revealed at 1160 °C.

3.2. Tritium release

Measured tritium releases depending on annealing temperature and duration are presented in Figs. 3 and 4 and Table 3.

Tritium desorption from sample #1 started at 445 °C (T_s). Within the temperature range 445–540 °C tritium release rate did not change, maintaining a low level. In this temperature range the total tritium release came only to 0.5%. Then release rate strongly increased with each step: by a factor 4 at 580 °C; 15 at 622 °C; 1.8 at 670 °C and 2.6 at 745 °C. The maximum rate corresponded to a temperature of final tritium release – 745 °C. About 59% of the total tritium contained in the sample was released from beryllium at 745 °C during 2 h. Totally about 99.8% of tritium was released in the temperature range 500–745 °C.

Tritium desorption from sample #2 started at 312 °C (T_s). Within the temperature range from 312 to 505 °C the tritium release rate slowly increased. The amount of tritium released at each temperature step did not exceed 1.5–3.0%, and total release was ~9%. When the temperature was increased to 610 °C, the intensity of tritium release grew by a factor 4 approaching the first peak. About 12.5% of tritium contained in beryllium was released after annealing at 610°C for 1.8 h. During the next steps (650, 700, 740 and 775 °C) the rate of tritium release first went down by a factor 2 (650 °C) and then returned to the previous level approaching the second peak at 775 °C. The maximum rate corresponded to the temperature of a final tritium release of – 775 °C.

Table 3
Tritium and helium release parameters of beryllium samples

Sample	Helium content, exp./calcd. (appm)	Tritium content (appm)	T_s (°C) (tritium/helium)	T_{upper} (°C) (tritium/helium)	$T_{max\ rate}$ (°C) (tritium/helium)	Swelling (%)
#1	7184/9770	20	445/500	745/1140	745/880	4–4.5
#2	6007/10228	542	312/740	775/1160	775/1085	0.7–1.2

T_s: starting temperature of gas release, T_{upper}: upper temperature of gas release, $T_{max\ rate}$: temperature of maximum gas release rate.

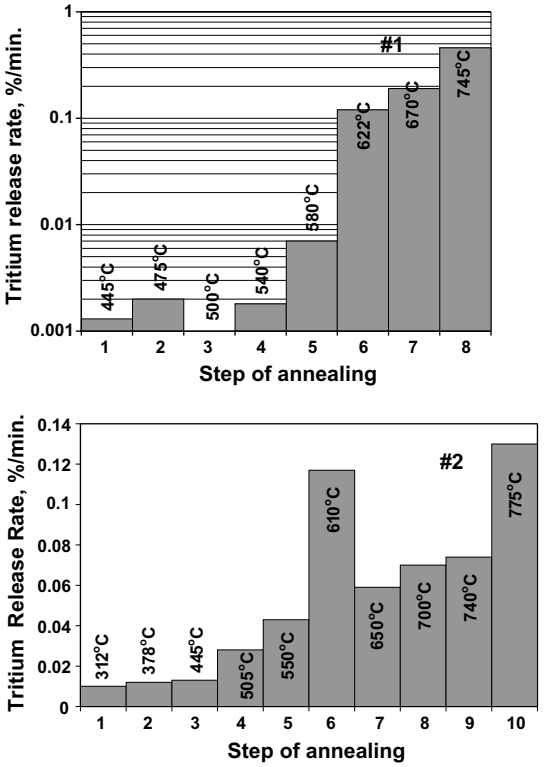

Fig. 4. Tritium release rate (samples # 1–2).

4. Discussion

Operating conditions of future fusion facilities will have an impact on the retention and release of tritium and helium in beryllium. The most important factors are temperature and irradiation dose, which can influence the evolution and integrity of beryllium structure under irradiation (micro-cracks, swelling, etc.).

Helium accumulation in the investigated samples was up to ~30–35% of the level expected for the breeding blanket DEMO end-of-life value. Sample #1 is of particular interest due to the higher irradiation temperature that corresponds to the lower limit of operating temperature (400–450 °C) for neutron multiplier in DEMO blanket.

The main distinction in structure of the samples lies in the fact that at an irradiation temperature of 70–100 °C (sample #2) the gas atoms formed are practically immobile (Fig. 5(b)), and material is characterized by the strong high-stress condition with small swelling ~1% (Table 3). At the irradiation temperature ~400 °C (sample #1), slight diffusion processes already take place in beryllium. This becomes apparent by formation of numerous gas bubbles (Fig. 5(a)) and more significant swelling (~4%). In this case a partial stress relaxation takes place already under irradiation and partially open porosity is developed. Moreover, both tritium and helium partly release. That is why under post irradiation annealing the structure of beryllium exposed to low temperature irradiation is strongly modifies due to the intensive recombination of gas-vacancy complexes. That results in the formation of surface micro-cracks during first steps of annealing and

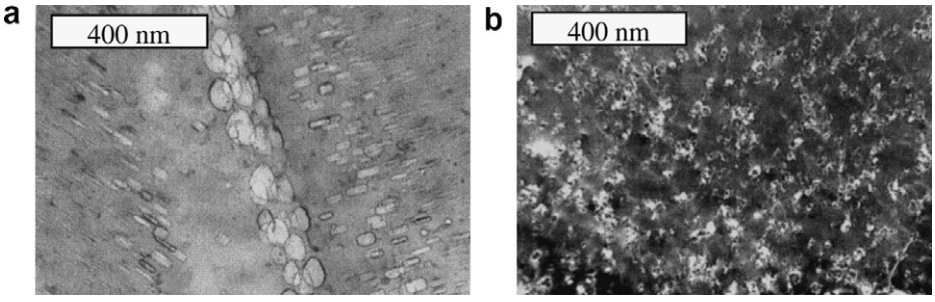

Fig. 5. TEM structure of Be samples after neutron irradiation, ×50 000, a – TE-400 (#1); b – TE-56 (#2).

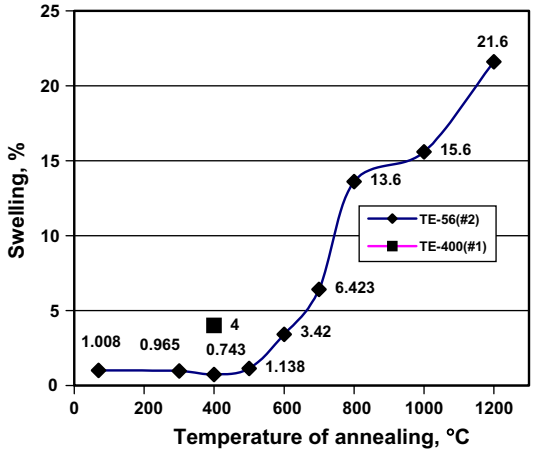

Fig. 6. Swelling of TE-56 sample vs temperature of post irradiation annealing (with duration of 1 h on each step), ■ – swelling of TE-400 sample after irradiation.

more active swelling at further steps (Fig. 6). The difference in structural condition of the samples after irradiation has an impact on the characteristics of helium and tritium release under following stepped annealing.

The basic difference in gas behavior implies that most of the tritium (~82%) releases from the sample irradiated at 70–100 °C (sample #2) in the temperature range of 312–700 °C – before the beginning of helium release (740 °C), i.e. the mechanism of separate tritium release takes place. In the case of beryllium sample irradiated at 380–420 °C (sample #1) tritium release starts at a higher temperature ($T_s = 445$ °C $> T_{irr}$) and a major part of tritium (~99.8%) releases concurrently with helium (i.e., concurrent tritium and helium release mechanism). This fact could be considered as indirect evidence of the co-existence of partial amounts of tritium and helium in common bubbles.

For the both samples, the maximum rate of tritium release corresponds to the upper temperature of tritium release. The samples differ little in upper temperatures of gas release: 745 and 775 °C for tritium, and 1140 and 1160 °C for helium.

Analysis of data on tritium and helium release together with the temperature dependence of swelling for sample #2 (Fig. 6), shows that swelling plays a key role in accelerating gas release from beryllium at annealing temperatures >600 °C for tritium and >750 °C for helium any influence of initial microstructure (grain size etc.) on gas behavior in this experiment is difficult to estimate. However, in our opinion, the distinctions in irradiation temperature of samples are the primary influence on gas behavior.

5. Conclusion

A study was performed on the characterization of tritium and helium desorption from beryllium irradiated at 70–100 °C and 380–420 °C with a helium content up to 6000–7200 appm.

The basic difference in gas behavior is that after the irradiation at 70–100 °C (sample #2), most of the tritium (~82%) releases from the sample in the temperature range of 312–700 °C – before the beginning of helium release (740 °C). After irradiation at 380-420 °C (sample #1), tritium release starts at a higher temperature ($T_s = 445$ °C $> T_{irr}$) and most of the tritium (~99.8%) releases concurrently with helium. This could be considered as evidence for the co-existence of partial amounts of tritium and helium in common bubbles.

Swelling of beryllium starts to play a key role in acceleration of tritium release at $T_{ann} > 600$ °C and helium release – at $T_{ann} > 750$ °C.

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