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The effect of helium generation and irradiation temperature on tritium release from neutron irradiated beryllium

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Abstract

The effect of neutron irradiation condition on tritium release from beryllium is described in this paper. Beryllium samples were irradiated in the SM reactor with neutron fluence ($E > 0.1$ MeV) of $(0.37–2.0)\times10^{22}$ cm^{-2} at 70–100 and 650–700 °C. Mass-spectrometry technique was used in out of pile tritium release experiments during stepped-temperature anneal within a temperature range from 250 to 1300 °C. The total amount of helium accumulated in irradiated beryllium samples varied from 521 to 3061 appm. The first signs of tritium release were detected at temperature of 406–553 °C. It was shown that irradiation temperature and helium generation level significantly affect the tritium release. A fraction of 44–74% of tritium content in samples irradiated at low temperature (70–100 °C) is released from beryllium at an annealing temperature below 800 °C, whereas for samples after high temperature irradiation (650–700 °C) tritium release did not exceed 14%. Majority of tritium (~68%) is released within a temperature range from 800 to 920 °C. The increase of helium generation from 521 to 3061 appm results in lowering the temperature of maximal tritium release rate and the upper temperature of tritium release from beryllium by 100–130 and 200–240 °C, correspondingly. On the basis of data obtained, the diffusion coefficients of tritium in beryllium were calculated.

1. Introduction

Knowledge of tritium release behavior in irradiated beryllium is an important aspect of beryllium application in future fusion devices. The tritium behavior in irradiated beryllium is a complex function dependent both on conditions of the irradiation (helium inventory, irradiation temperature, duration, etc.), and properties of beryllium (density, grain size, beryllium oxide contents, etc.). In spite of the fact that during recent years considerable progress was achieved in understanding of the above-stated problem [1–8], many aspects of tritium and helium behavior are poorly known. This paper presents the recent results of experiments on investigation of the effects of helium generation and irradiation temperature on tritium and helium release from beryllium.

2. Materials and experimental procedure

The study was performed for TE-56 and TshG-56 beryllium grades irradiated in high flux channels of the SM reactor. The irradiation parameters and some initial characteristics of beryllium are presented in Tables 1 and 2.

Tritium release kinetics was analyzed in a stepped isothermal annealing mode. Gases liberated under

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Table 1
Characteristics of beryllium

Grade of Be	Density, g/cm^3	Grain size, μm	Be, wt%	BeO, wt%
TE-56	1.856	22–25	98.63	1.48
TshG-56	1.85	22–25	99.10	0.95

sample heating were collected in a closed volume equipped with omegatron mass-spectrometer (OMS). The OMS was calibrated against H_2 and ^{4}He standard leaks. Released amounts of ^{4}He were calculated from reading the OMS signal at mass 4. For evaluation of tritium released, only the mass 6 alone was used, since the fraction of tritium released in form of HT molecules was found to be negligible in our experimental conditions. A temperature was elevated by 20–170 °C with every step of multi-stage annealing. A heating duration was between 0.6 and 4 h, and the temperature ranged within 200–1300 °C. At the final step of multi-stage annealing the samples were melted and the total amount of helium and tritium was measured.

3. Experimental results

3.1. Tritium release

The results of the measurements on a fraction of the total quantity of tritium released as a function of temperature and duration of annealing are presented in Fig. 1; the tritium release rate is shown in Fig. 2.

The highest initial temperature of tritium release ($T_s = 553$ °C) was found for sample #1. When the temperature increased to 600 °C the tritium release rate grew more than by a factor 30 and was constant up to 700 °C. At $T = 760$ °C the tritium release rate increased again by a factor 2.5 and then at 850 °C increased additionally by a factor 2.2, approaching the maximum value ($T_{max.rate}$) (Table 3). At the following steps of annealing up to 1000 °C, where tritium release was completed, the release rate decreased continuously, but remained rather high. Detritization of sample #1 completed at $T = 1000$ °C (T_{upper}).

For sample #2 tritium release started at $T_s = 406$ °C. At the subsequent steps of annealing the tritium release rate grew up more than by a factor 1.4–4.3 at each temperature step and reached the maximal value at 920 °C. With further increase of temperature the tritium

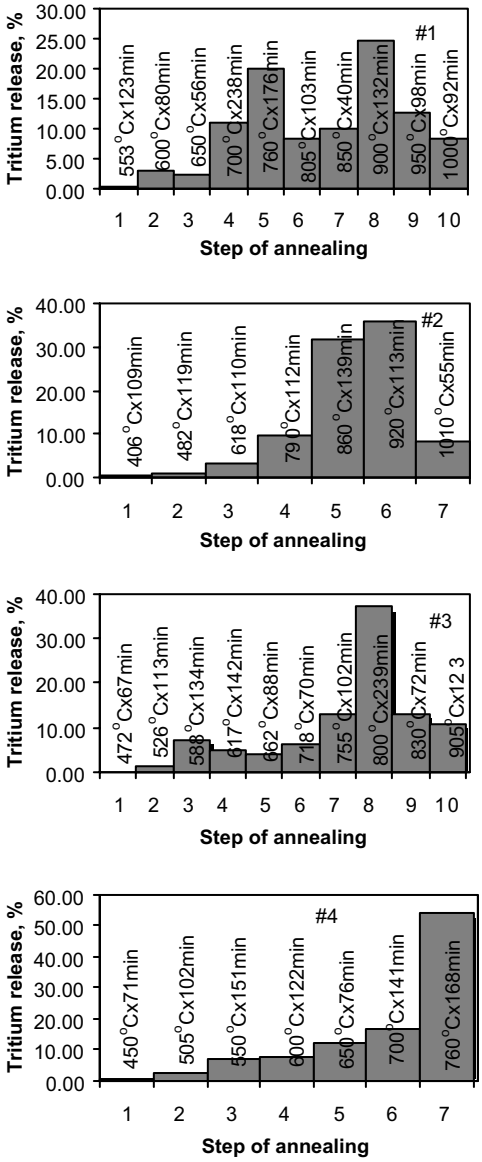

Fig. 1. Tritium release (samples # 1–4).

Table 2
Irradiation parameters of beryllium

Sample	Shape of sample	Neutron fluence, $E > 0.1$ MeV, cm^{-2}	T_{irr}, °C
#1 (TE-56)	Irregular	0.5×10^{22}	70
#2 (TshG-56)	Disk $d = 6$ mm, $h = 0.5$ mm	$(0.37–0.59)\times10^{22}$	650–700
#3 (TE-56)	Disk $d = 6$ mm, $h = 0.5$ mm	0.9×10^{22}	70
#4 (TE-56)	Irregular	2×10^{22}	70–100

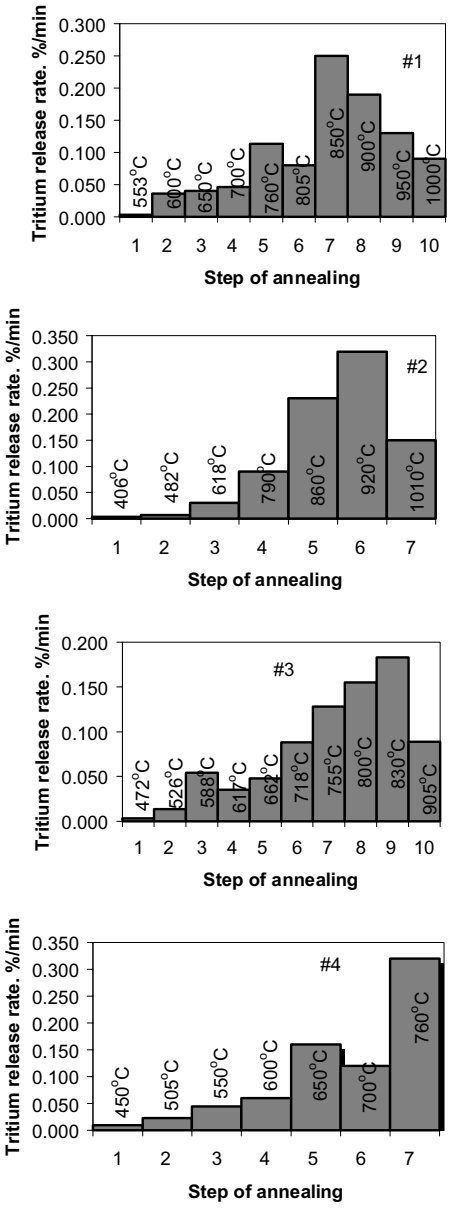

Fig. 2. Tritium release rate (samples # 1–4).

release rate fell down and detritization completed at $T = 1080$ °C. Majority of the tritium contained in the sample (~68%) was allocated within a temperature range from 800 to 920 °C. 90% of the tritium was released when the temperature was increased to $T = 1010$ °C.

For sample #3 the tritium release started at $T = 472$ °C. At an annealing temperature of 588 °C more than 10% of the tritium inventory was released from the sample, and at an annealing temperature of 800 °C about 37% tritium was released from the sample after 4 h of exposure. The integrated tritium release was thus 74%. About ~90% of the tritium release occurred at a temperature of 830 °C and maximal tritium release rate corresponded to this temperature (Table 3). Detritization of sample #3 was completed at $T = 905$ °C.

For sample #4 an intensive tritium release was already observed at 450 °C. At the subsequent steps of annealing the tritium release rate grew up by a factor 1.5–2.5 at each temperature step and reached a maximal value at 760 °C, that coincided with the maximal tritium release rate from the sample (760 °C). After 3 h of exposure about 54% of tritium contained in a sample was allocated from beryllium at $T = 760$ °C.

3.2. Diffusion of tritium

Using kinetic curves of gas release, the diffusion coefficients of tritium were calculated for samples #2 and #3 (Fig. 3) on the base of a solution of a differential equation for a thin plate [9]. The results show that at temperatures below 900 °C the diffusion mobility of tritium in sample #2 is lower than that in sample #3.

They also show, that at post irradiation annealing the retention of tritium in beryllium is higher for the samples irradiated at high-temperature (650–700 °C) than for the samples, which were irradiated at low (50–100 °C) temperatures.

4. Discussion

Among the factors, which can significantly affect the retention and release of tritium from beryllium, are the factors caused by operation conditions of beryllium in future fusion device. The most important from these

Table 3
Tritium and helium release parameters

Sample	Helium content, appm	Tritium content, appm	T_s, °C (tritium/helium)	T_{upper}, °C (tritium/helium)	$T_{max.\,rate}$, °C (tritium/helium)	Swelling, %
#1	521	6.5	553/553	1000/1180	850/1075	0
#2	601	12	406/694	1080/1215	920/1165	2
#3	1161	20	472/754	905/1245	830/1172	0.1
#4	3061	203	450/760	760/1150	760/1150	0.5–0.7

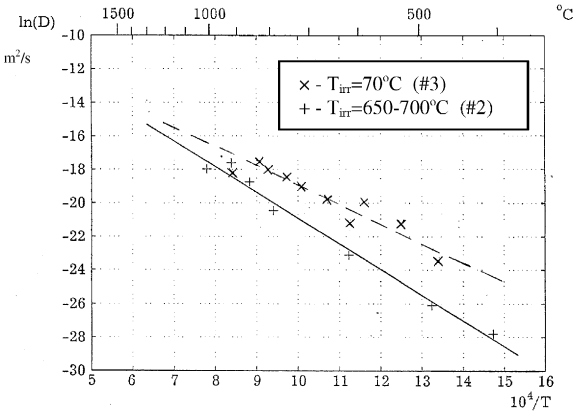

Fig. 3. Apparent tritium diffusion coefficients.

factors are temperature and irradiation dose, and also their influence on the integrity of the beryllium structure (micro-cracks, swelling etc.) under the irradiation and at subsequent annealing.

The results of this research show, that at $T < 400$ °C in the helium inventory range of 500–3100 appm, tritium is completely captured in irradiated beryllium. The comparison of the samples irradiated at identical temperature conditions (#1, #3 and #4) shows that the increase of helium generation results in:

- Lowering the initial temperature of tritium release (T_s) from 553 to 450 °C;
- Lowering the upper temperature of tritium release (T_{upper}) from 1000 to 760 °C;
- Lowering the temperature of maximal rate of tritium release ($T_{max.rate}$) from 850 to 760 °C.

For the sample with the maximal helium generation (#4, 3061 appm) the temperature of the maximal rate of tritium release and the upper temperature of tritium release coincided with the initial temperature of helium release from a sample (760 °C). For the other samples $T_{upper} > T_{max.rate}$. This effect, apparently, is caused by distinction in the rate of structural evolution in beryllium (swelling, micro-cracks) during the irradiation and after irradiation annealing.

The results obtained confirm the assumption of study [7] that irradiation temperature significantly affects the tritium release and diffusion mobility of tritium. A fraction of 44–74% of tritium in samples after low temperature irradiation (70–100 °C) is released from beryllium at an annealing temperature below 800 °C, while for samples after high temperature irradiation (650–700 °C) tritium release did not exceed 14%. Majority of tritium (~68%) is released within a temperature range from 800 to 920 °C.

The irradiation temperature influences the upper temperature of tritium release similarly. For samples irradiated at low temperatures detritization completed at lower temperatures of annealing (~905–1018 °C) than for the sample irradiated at high-temperature irradiation (>1050 °C). These distinctions can be explained by the following reasons. In dense beryllium irradiated at low temperatures (70–100 °C) practically all tritium formed during the irradiation remains in beryllium, while during the high-temperature irradiation part of the tritium diffuses out from beryllium. It occurs due to higher diffusion mobility of tritium at high temperatures, and also as a result of an imperfection of structure integrity because of swelling and the formation of helium porosity (Table 3). With higher temperature of irradiation and swelling of beryllium, bigger amount of tritium diffuses out from beryllium during the irradiation [7]. However, tritium remaining in beryllium after the high-temperature irradiation apparently is in a more stable energy condition than after low temperature irradiation [7]. Therefore, a relatively small fraction of tritium (<15%) is released from beryllium irradiated at high temperature when the temperature of post-irradiation annealing $T_{ann} < T_{irr}$, while the remaining fraction will be released at $T_{ann} > T_{irr}$. With higher irradiation temperature and lesser swelling, bigger amount of tritium is retained in beryllium at post irradiation thermal expose.

5. Conclusions

The level of helium generation and irradiation temperature significantly affect the kinetics of tritium release from beryllium at stepped isothermal annealing. The main results are as follows:

1. The increase of helium generation from 521 to 3061 appm results in:
   - Lowering the initial temperature of tritium release by ~100 °C (T_s reduced from 553 to 450 °C);
   - Lowering the upper temperature of tritium release by ~240 °C (T_{upper} reduced from 1000 to 760 °C);
   - Lowering the temperature of maximal tritium release rate by ~90 °C (reduced from 850 to 760 °C).
2. Irradiation temperature significantly affects the tritium release and diffusion mobility of tritium. A fraction of 44–74% of tritium in samples after low temperature irradiation (70–100 °C) is released from beryllium at an annealing temperature below 800 °C, while for samples after high temperature irradiation (650–700 °C) tritium release did not exceed 14%. Majority of tritium (~68%) is released within a temperature range from 800 to 920 °C. Detritization of samples after low temperature irradiation completed in a temperature range from ~905 °C to 1018C, and that for the sample after high-temperature irradiation occurred at a temperature of 1080 °C.

3. The apparent diffusion coefficients of helium and tritium were calculated for a temperature range from 400 to 1000 °C. At temperatures below 900 °C the diffusion mobility of tritium for the sample irradiated at high temperatures (650–700 °C) is lower than for the samples irradiated at low temperatures (50–100 °C).

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