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Influence of high dose neutron irradiation on thermal conductivity of beryllium

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Abstract

The thermal conductivity of beryllium is a very important physical characteristic, which considerably determines the serviceability of fusion reactor components where this material is supposed to be used. The neutron irradiation leads to a decrease of the thermal conductivity, the quantitative characteristic of the effect considerably depending upon the irradiation temperature and dose. So, after the irradiation in the SM reactor at a temperature of 70 °C up to neutron fluence of $(1–6) \times 10^{22}$ cm^{-2} ($E > 0.1$ MeV) the effect reaches hundreds of percents, after irradiation in the BOR-60 reactor at a temperature of 400 °C up to fluence of 1.6×10^{23} cm^{-2} ($E > 0.1$ MeV) the effect reaches only tens of percents. One hour annealing of beryllium at 500 °C after the low temperature irradiation leads to a partial thermal conductivity recovery.
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1. Introduction

At present beryllium is considered as one of candidate materials for some fusion reactor components (first wall, divertor, blanket) [1–3]. Under the influence of a high-energy particle fluence a considerable degradation of physical–mechanical properties of this metal occurs and the knowledge of the degradation degree is necessary for serviceability validation of the fusion reactor under design. Thermal conductivity is one of the most important physical characteristics, especially for materials used for plasma facing components. This paper presents the results of the investigation of the high dose neutron irradiation effect upon the thermal conductivity of beryllium.

2. Materials, technique of the experiment

Beryllium of TE-56 and TE-400 grades fabricated using the hot extrusion technology was used in this work. The content of basic impurities in the TE-56 grade is: O – 0.98; BeO – 1.48; C – 0.08; Fe – 0.17; Al – 0.026; Si – 0.016; Cr – 0.041; $B < 0.0009$ mass%. The TE-400 grade belongs to obsolete materials with O and C contents of about of 1.5–1.8 mass% and with high contents of metallic impurities. Specimens in the form of disks were used: for the TE-56 with 7 mm in diameter and 0.6 mm in thickness which were produced in the initial condition before the irradiation; for the TE-400 – with 11 and 2 mm, correspondingly, produced after irradiation from the irradiated cylinders with 11 mm in diameter and 100 mm in length.

The specimens of the TE-56 grade were irradiated in the experimental channels of the fuel assemblies of the research SM reactor. During irradiation the specimens were cooled by the primary circuit water. The TE-400 specimens were irradiated in the BOR-60 reactor in helium medium as part of the sealed neutron source. The irradiation parameters of the examined specimens are presented in Table 1. Then some of the irradiated specimens of the TE-56 beryllium grade were annealed in 1×10^{-7} bar vacuum at 500 °C during 1 h.

The examinations before and after irradiation were performed on the experimental facility using the pulsed method by measuring of thermal diffusivity of beryllium specimen in the direction of the extrusion axis. In detail

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Table 1
Irradiation parameters of beryllium

Beryllium grades	Reactor, position	Medium	T_{irr} (°C)	F (cm^{-2}) ($E > 0.1$ MeV)	D (dpa)	Helium content (appm)
TE-56	SM, core	Water	70	$(1.0–6.0) \times 10^{22}$	5.3–32.0	2100–12 000
TE-400	BOR-60 reflector, 7th row	Helium	400	1.6×10^{23}	94	9800

the theory of the method is presented in [4]. Fig. 1 presents the principle scheme of the facility, which consists of the cell (2) for the disposition of specimen (1) and heating up to the predetermined temperature, of chromel–alumel thermocouple (4,5) that directly contacts the specimen surface, of measuring circuit and of intensifier. A short-term light pulse (3), generated by the flash-lamp heats the frontal side of the specimen, and heat propagates through the specimen. The temperature of the back side of the specimen is measured by the thermocouple and the time dependence curve of the specimen response temperature is built. The curve is used to determine the time, which is necessary for the increase of the backside temperature by a value, equal to the half of its maximum raise followed by calculation of the thermal diffusivity. Then the following formula was used for the specimen thermal conductivity calculation:

$$\lambda = \alpha dc, \tag{1}$$

where α is the thermal diffusivity, m^2/s; d is density, kg/m^3; c is specific heat capacity, J/kg K.

The specific heat of beryllium in the initial state was taken from [5] (1760–2380 J/kg K at temperature of 20–200 °C). It was used for the calculations of irradiated specimens assuming that under irradiation the specific heat changes are rather low. The density of the initial and irradiated specimens was measured using the hydrostatic method. The results are presented in Table 2. Measurements of the thermal diffusivity were performed in the temperature range from room temperature up to 230 ° C under heating in air.

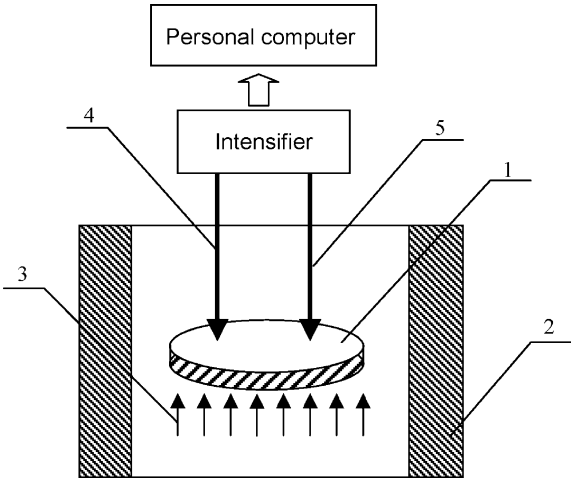

Fig. 1. Principal scheme of the pulsed facility for thermal conductivity measurement of materials: (1) flat specimen; (2) heater for temperature setting of specimen; (3) 1.2 ms duration pulse from the flash lamp; (4) chromel contact; (5) alumel contact.

3. Results and discussion

The results of the investigation of the TE-56 beryllium grade thermal conductivity are presented in Fig. 2. In the initial state at room temperature beryllium has a thermal conductivity of 230 W/m K, and with increasing measurement temperature, its monotonous decrease occurs almost linearly. The irradiation leads to an abrupt fall of the thermal conductivity of beryllium up to values of about hundreds of percents. So, at neutron fluence of 1.0×10^{22} cm^{-2} the value of the thermal conductivity does not exceed 70–80 W/m K. At the same time the dependence of the thermal conductivity on temperature that was observed in the initial condition

Table 2
Density of irradiated beryllium

Beryllium grades	Density (kg/m^3)					
	Initial state	After irradiation at F (cm^{-2}) ($E > 0.1$ MeV)				
		1.0×10^{22}	2.4×10^{22}	2.4×10^{22} + ann. 500 °C, 1 h	6.0×10^{22}	1.6×10^{23}
TE-56	1858.4	1854.5	1852.0	1854.1	1835.1	–
TE-400	1851.2	–	–	–	–	1787.1

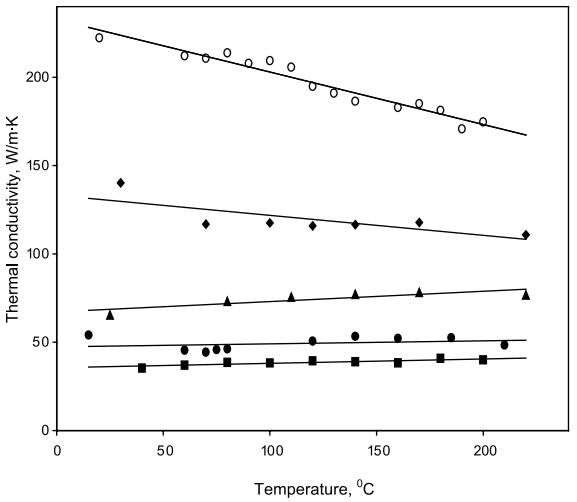

Fig. 2. Dependence of the thermal conductivity of TE-56 beryllium grade upon testing temperature: (◯) initial state; (▲) $T_{irr} = 70$ °C, $F = 1.0 \times 10^{22}$ cm^{-2} ($E > 0.1$ MeV); (●) $T_{irr} = 70$ °C, $F = 2.4 \times 10^{22}$ cm^{-2} ($E > 0.1$ MeV); (■) $T_{irr} = 70$ °C, $F = 6.0 \times 10^{22}$ cm^{-2} ($E > 0.1$ MeV); (◆) $T_{irr} = 70$ °C, $F = 2.4 \times 10^{22}$ cm^{-2} ($E > 0.1$ MeV) + annealing at 500 °C, 1 h.

disappears. Actually, the thermal conductivity of beryllium after irradiation has a constant value in the temperature range up to 250 °C and does not depend on the measurement temperature. An increase of the dose leads to a further decrease of the thermal conductivity of beryllium but in the comparatively lower degree. Fig. 3 presents the dose dependence of thermal conductivity changes for the measurement temperature that is approximately equal to the irradiation temperature, i.e. 70–100 °C. At the maximum accumulated dose of 6.0×10^{22} cm^{-2} ($E > 0.1$ MeV) the thermal conductivity decreased to 30 W/m K. The post-irradiation one-hour annealing at 500 °C of beryllium, irradiated up to a fluence of 2.4×10^{22} cm^{-2} ($E > 0.1$ MeV), leads to a

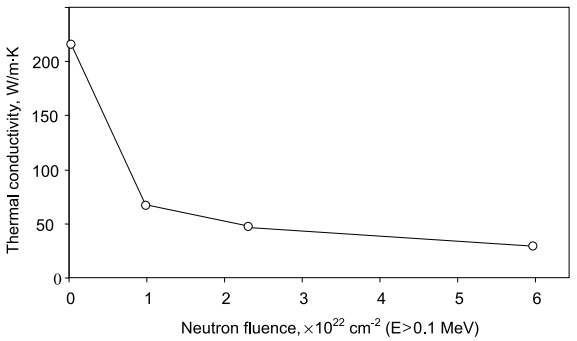

Fig. 3. Dependence of the thermal conductivity of irradiated TE-56 beryllium grade upon neutron fluence for $T_{irr} = 70$ °C and testing temperature range of 70–100 °C.

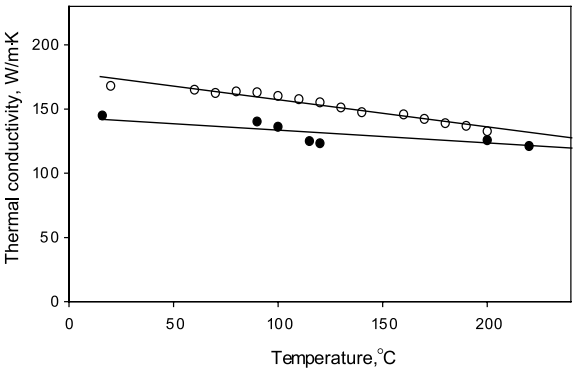

Fig. 4. Dependence of the thermal conductivity of TE-400 beryllium grade upon testing temperature: (◯) initial state; (●) $T_{irr} = 400$ °C, $F = 1.6 \times 10^{22}$ cm^{-2} ($E > 0.1$ MeV).

partial recovery of the thermal conductivity up to a level of about 110–140 W/m K (Fig. 2).

Fig. 4 presents the results of thermal conductivity measurements of the TE-400 beryllium grade in the initial state and after irradiation in the BOR-60 reactor at 400 °C up to a neutron fluence of 1.6×10^{23} cm^{-2} ($E > 0.1$ MeV). The initial thermal conductivity of this beryllium grade is much lower than that of the TE-56 grade. But the irradiation effect here, in its turn, is much weaker, and though the neutron fluence is considerably higher it reaches the value of only some tens of percents.

Coming from the theory that the thermal conductivity of beryllium (like any other material) is determined by electron and phonon conductivity, which directly depends on the free path length of electrons and phonons i.e. on the quantity or volume density of the crystal lattice defects where the electron–phonon scattering occurs. Therefore, a higher impurity content in the matrix of the TE-400 beryllium grade determines a lower value of the thermal conductivity in comparison with the TE-56 grade. The main irradiation effects upon the beryllium microstructure are the generation of large quantity of gas atoms (mainly helium) in the threshold nuclear reactions and also some other radiation damages (point defects, clusters, dislocation loops). Correspondingly, the value of the thermal conductivity of irradiated beryllium is determined by the value of the radiation defects volume density, which, in its turn, depends upon temperature, neutron fluence and other irradiation parameters. Probably, the most significant factor here is the temperature of irradiation or annealing. As the temperature increases, the diffusion mobility of self and foreign atoms increases, the radiation defects configuration changes: their dimensions increase, their volume density decreases. As a result, the free path length of electrons and phonons increases and the thermal conductivity value increases along with it. Consequently, an increased irradiation temperature

leads to a comparatively lower value of the thermal conductivity decrease effect related to the initial value, and high-temperature annealing leads to the initial thermal conductivity recovery (partial, as a result of the radiation defects availability, which were absent in the initial state). It seems that the radiation-induced changes in beryllium during low-temperature irradiation, such as generation of complex systems of helium atoms and dislocation loops of high density, lead to a considerable growth of the thermal resistance value and, as a result, a considerable degradation of the thermal conductivity characteristics occurs. The usage of the post-irradiation annealing at temperatures exceeding the irradiation temperature allows the structure formed under the irradiation to be effected and changed. The one-hour annealing of beryllium at 500 °C leads to a considerable transformation of the dislocation structure and, in particular, the evolution of loops in the dislocation net and also to the formation of the smallest gas bubbles [6]. In such a way a decrease of the efficiency of the considerable amount of barriers, which complicated the heat transfer along the material bulk, occurs in the crystal lattice and, as a result, the thermal conductivity of irradiated beryllium partially recovers after annealing.

4. Conclusions

The investigations of the thermal conductivity of beryllium, irradiated in the SM reactor at 70 °C up to fluences of $(1–6) \times 10^{22}$ cm^{-2} ($E > 0.1$ MeV) and in the BOR-60 reactor at 400 °C up to fluences of 1.6×10^{23} cm^{-2} ($E > 0.1$ MeV) were performed.

The main irradiation effects were:

- Low temperature irradiation leads to the abrupt fall of the thermal conductivity at the beginning stage of irradiation (up to a fluence of 1×10^{22} cm^{-2} ($E > 0.1$ MeV)) with further monotonous slow decrease. The effect of the thermal conductivity fall reaches hundreds of percents.
- Post-irradiation annealing of beryllium at 500 °C during 1 h leads to the partial recovery of the initial thermal conductivity value.
- The increase of the irradiation temperature up to 400 °C, in spite of the considerably higher neutron fluence, leads to a lower degradation of the thermal conductivity, the effect reaches tens of percents only.

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