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Effects of neutron irradiation at 70-200 °C in beryllium

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Abstract

At present beryllium is considered one of the metals to be used as a plasma facing and blanket material. This paper presents the investigations of several Russian beryllium grades fabricated by HE and HIP technologies. Beryllium specimens were irradiated in the SM reactor at 70–200 °C up to a neutron fluence $(0.6-3.9) \times 10^{22}$ cm⁻² (E > 0.1 MeV). It is shown that the relative mass decrease of beryllium specimens that were in contact with the water coolant during irradiation achieved the value >1.5% at the maximum dose. Swelling was in the range of 0.2–1.5% and monotonically increasing with the neutron dose. During mechanical tensile and compression tests one could observe the absolute brittle destruction of the irradiated specimens at the reduced strength level in comparison to the initial state. A comparatively higher level of brittle strength was observed on beryllium specimens irradiated at 200 °C. The basic type of destruction of the irradiated beryllium specimens is brittle and intergranular with some fraction of transgranular chip. © 2002 Published by Elsevier Science B.V.

1. Introduction

The assumed range of working temperatures of beryllium as a plasma facing material in the up-to-date concept of some ITER components makes up 200–500 °C, in particular, that of the first wall 230–280 °C [1]. According to the general ideas on mechanisms of radiation damage of metals and according to a number of papers on beryllium radiation resistance [2–4], degradation of the physical-mechanical properties of this metal occurs to the largest degree in the lower boundary of the working temperature range, i.e. at irradiation temperatures up to 200–250 °C. It was of special interest, therefore, to obtain new data on effect of low-temperature neutron irradiation on the significant properties of beryllium grades to be used in fusion. The present paper describes the investigation results of four Russian beryllium grades fabricated by different technologies and irradiated in the SM reactor at 70-200 °C.

2. Material and experiment technique

The paper used four beryllium grades, two of which, TE-56 and TE-30, were fabricated by hot extrusion (HE), and two, TIP and DIP – by hot isostatic pressing (HIP). The chemical composition and grain size of materials under study are presented in Table 1.

The cylindrical billets were used to produce the specimens of two types: for mechanical tensile tests in the form of cylindrical dumb-bells with the working part of \emptyset 3 × 10 mm and for compression tests in the form of cylinders, \emptyset 6 × 8 mm. The specimens of anisotropic TE-56 and TE-30 grades were cut in two orientations – along and across the extrusion axis.

The specimens were irradiated into irradiation facility in investigation channels of the SM reactor core, at

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Grade	Technology	Average grain size (µm)	Chemical composition (mass %)							
			Be	BeO	0	Fe	Al	Si	С	Mg
TE-56	He	25	98.6	1.48	0.98	0.17	0.026	0.016	0.08	No data
TE-30	HE	15	98.1	2.5	1.66	0.11	0.015	0.013	0.088	0.002
TIP DIP	HIP HIP	12 13	98.8 98.6	1.3 2.0	0.89 1.3	0.13 0.028	0.013 0.005	0.013 0.013	0.07 0.067	0.0066 0.0016

 Table 1

 Chemical composition and grain size of beryllium grades

Table 2Irradiation parameters of beryllium

Grade	Medium	Temperature (°C)	Neutron fluence ($E > 0.1$ MeV, $\times 10^{22}$ cm ⁻²)	Damage dose (dpa)	Helium content (appm)
TE-56	Water	70	0.59–3.88	3.1–20.6	860–9990
	Helium	200	1.4	7.6	3210
TE-30	Water	70	2.3	12.1	5060
	Helium	200	1.4	7.6	3220
TIP	Water	70	1.8	9.8	4160
	Helium	200	1.4	7.6	3220
DIP	Water	70	1.9	10.2	4220
	Helium	200	1.4	7.6	3210

70 °C in the fluence range of $(0.6-3.9) \times 10^{22}$ cm⁻² (E > 0.1 MeV) in the medium of the primary circuit water coolant, at 200 °C up to fluence of 1.4×10^{22} cm⁻² (E > 0.1 MeV) in helium of technical purity. The irradiation parameters of the materials are summarized in Table 2.

The geometrical dimensions were measured with a special instrument, the measurement error did not exceed 0.1–0.15%. The mass and density of specimens were measured at hydrostatic balance, the measurement error was 0.2%. Mechanical tensile tests were conducted at the tearing machine in 10^{-7} bar vacuum, mechanical compression tests – in air, in both cases the motion speed of the active grip made up 1 mm/min. The error made up 5% and 10% for strength and plastic properties, respectively.

3. Experimental results

3.1. Corrosion in water under irradiation

During irradiation a part of specimens under study were washed by distilled water of the SM reactor primary circuit. Some characteristics of the primary circuit water that were observed in the process of the experiment are summarized in Table 3.

The measurement results of the relative mass decrease for the TE-56 beryllium grade depending on the Table 3

Some characteristics of the SM reactor primary circuit water during experiment

umerical values
1
6–6.0
5–2.0
20
10
5
1

neutron irradiation dose and test duration are presented in Fig. 1(a). On the whole, it can be stated that the mass decrease grows with increasing dose and test duration. However, there is no strict data monotony. In particular, there is no strict correlation between the measurements on specimens with different geometry, which is especially noticeable at the dose of 3.9×10^{22} cm⁻² (6080 h). It can be related to different dynamics of the water flow along the specimens in the form of dumb-bells and cylinders, in particular, turbulence in the contraction area in the working part of the specimen for mechanical tensile tests. Probably, corrosion tests of cylindrical specimens are more correct, as they have an easier geometry. The comparative mass decrease of the beryllium grades for fluences $(1.9-2.4) \times 10^{22}$ cm² (2720 h) is



Fig. 1. Corrosion of beryllium in water under irradiation at 70 °C: (a) dose dependence of the relative mass decrease of the TE-56 grade; (b) comparative mass decrease of some beryllium grades (TE-56, TE-30, TIP, DIP) in water under irradiation at 70 °C up to fluence $(1.9-2.4) \times 10^{22}$ cm⁻² (E > 0.1 MeV) (2720 h).

presented in Fig. 1(b). No considerable differences in corrosion rates between the grades is observed. The TIP beryllium grade has the minimum value of the relative mass decrease.

3.2. Swelling

The swelling dose dependence of the TE-56 grade after irradiation at 70 °C determined by the results of geometrical dimensions and density is presented in Fig. 2(a). On the whole, growth of swelling with increasing neutron fluence is observed. Some differences in the swelling values can be noted by the measurements of geometrical dimensions and density. However, these differences are not so large, therefore they do not affect the tendency of monotonic swelling increase with increasing dose. The comparative swelling value of the beryllium grades at the irradiation temperatures of 70 and 200 °C is presented in Fig. 2(b). All beryllium grades have less than 1% swelling, in this case considerable spread in data is observed. Evidently, the TE-30 grade has the least swelling, it makes up 0.2-0.6%, the remaining beryllium grades have a bit higher swelling. There is no strict dependence of swelling on the irradiation temperature, though for isotropic TIP and DIP

grades the irradiation at 200 °C leads to comparatively higher volume changes, but the total insignificant effect does not allow definite confirmation of this at present. Though with due account of much less irradiation dose at 200 °C the indicated tendency is most likely.

3.3. Mechanical properties

The specimens of all beryllium grades in irradiated state at room test temperature exhibit a totally brittle fracture. Fig. 3 presents the dose dependences of brittle strength of the TE-56 grade. The brittle strength decreases with increasing dose according to the results of tensile tests (Fig. 3(a)) and compression tests (Fig. 3(b)). In this case in the dose range up to 1×10^{22} cm⁻² (E > 0.1 MeV) there is the maximum (2–3 times higher) decrease of strength as compared to the initial state. With further increase of the dose there is stabilization of brittle strength of irradiated beryllium at 230 and 130 MPa according to the tensile test results for specimens cut along and across the extrusion axis, and at 500 and 700 MPa – according to compression tests, where the dependence of brittle strength on cut-off direction about the axis is reverse.



Fig. 2. Swelling of beryllium grades: (a) swelling dose dependence of the TE-56 grade under irradiation at 70 °C; (b) comparative swelling of some beryllium grades (TE-56, TE-30, TIP, DIP) under irradiation at 70 °C up to fluence 2.4×10^{22} cm⁻² (E > 0.1 MeV) and at 200 °C up to fluence 1.4×10^{22} cm⁻² (E > 0.1 MeV).



Fig. 3. Dose dependence of brittle strength of the TE-56 grade under irradiation at 70 °C ($T_{\text{test}} = 20$ °C): (a) tensile tests; (b) compression tests.

The comparative brittle strength of the beryllium grades irradiated at 70 and 200 °C and subject to tensile and compression tests at the room temperature is given in Fig. 4. Brittle strength of all grades according to tensile tests is in the range of 130–330 MPa, and according to compression tests is 400–900 MPa. The TE-56 grade (compression tests), probably, has comparatively higher brittle strength among anisotropic grades. As to the temperature dependence of brittle strength, excluding the TE-30 grade, the specimens irradiated at 200 °C have comparatively higher strength. It should be noted that firstly, the neutron fluence accumulated at 200 °C is considerably lower, secondly, absolute brittleness of the specimens under study is the cause of instability of the obtained brittle strength values.

3.4. Fractography

The fracture mode of the TE-56 specimen irradiated at 70 °C and tested at room temperature is brittle and intergranular with a minimum quantity of sections



Fig. 4. Brittle strength of some beryllium grades (TE-56, TE-30, TIP, DIP) at 70 °C, $F = 2.4 \times 10^{22}$ cm⁻² (E > 0.1 MeV) and at 200 °C, $F = 1.4 \times 10^{22}$ cm⁻² (E > 0.1 MeV) ($T_{\text{test}} = 20$ °C): (a) tensile tests; (b) compression tests.

having transgranular chip. Increase of irradiation temperature up to 200 °C leads to considerable increase of transgranular chip of the TE-56 specimen. However, the fracture mode of the DIP specimen irradiated at 200 °C also is mainly brittle and intergranular. The average grain size of this grade produced by the HIP method according to the fracture surface is somewhat less than that of the TE-56 grade (HE technology).

4. Discussion

As the review of the literature shows, there are practically no systematic investigations on radiation damage of beryllium at irradiation temperatures up to 200 °C with high neutron doses. Some new papers [5–7] are known, which study the properties of beryllium irradiated with neutrons at higher temperatures and there are some papers [8–10] on low-temperature high-dose irradiation performed long ago and with outdated beryllium grades.

Low irradiation temperatures are always most dangerous for materials from the viewpoint of radiation damage accumulation, which occurs at small diffusion rates and slow annealing. In beryllium, where the most significant damage factor of neutron radiation is the formation of large helium amounts, accumulation of this gas at low-temperature irradiation occurs practically in places of its formation and results in substantial distortions of the crystal lattice and the so-called 'solid' swelling [11,12]. Low-temperature swelling of beryllium is comparatively small (1-2%) at the maximum neutron doses), however, the volume changes lead to the strongest degradation of mechanical properties, which is expressed by embrittlement and decrease in brittle strength of the irradiated material. It is caused by the swelling anisotropy effect of some grains [8], which is observed in beryllium as a metal with HCP lattice. It results in growth of stresses on boundaries, their weakening, which causes prerequisites for crack generation and specimen failure during its loading. The papers [7,13] analyze the 'load-elongation' diagram during mechanical tensile tests of irradiated beryllium and state that irradiation leads to material strengthening and embrittlement, but does not indicate the systematic nature of its brittle strength decrease especially noticeable at reduced irradiation temperatures. There is no doubt that radiation strengthening of beryllium occurs and it is significant in value, as shown by the measurement results of irradiated beryllium microhardness [14]. However, beryllium is a polycrystalline material, therefore, the role of grain boundaries is important, which becomes apparent during mechanical tests. Radiation damages are accumulated in a separate grain under irradiation, which, on the whole, causes the material strengthening (increase of microhardness). But during

mechanical tests the boundary weakening leads to premature failure of the irradiated specimen at the elastic strain section in the absence of plastic strain, i.e. it is absolutely brittle. It is evident that there is such an ultimate dose, at which brittle strength of beryllium specimen can drop to the catastrophically small value, i.e. the specimen failure will take place at the minimum load. The beryllium specimens irradiated at maximum service-life neutron fluences are being investigated currently, the results will be presented.

The above data show that at the presented damage doses none of the investigated beryllium grades has decisive advantage from the viewpoint of reserving the dimensional stability and degradation degree of mechanical properties (including isotropic grades produced by HIP method). This situation is caused by inherent beryllium anisotropy of the crystal lattice, which cannot be leveled using the HIP technology (provision of equiaxial grains). As the result of anisotropic swelling of separate crystallites, weakening of grain-boundary bond is inevitable in this case too, which leads to generation of grain-boundary cracks. This is a main factor but we know that embrittlement is due to the presence of pinned dislocations full of gas too [12,15].

5. Conclusions

Several beryllium grades (TE-56, TE-30, TIP, DIP) fabricated by HE and HIP technologies were irradiated in the SM reactor at 70-200 °C up to neutron fluences of $(0.6-3.9) \times 10^{22} \text{ cm}^{-2}$ (E > 0.1 MeV) and were later investigated. It is shown that the relative mass decrease of beryllium specimens, that were washed by the primary circuit water during irradiation, monotonically increases with the dose (test duration) and achieves more than 1.5% at the maximum irradiation dose. Swelling of all the investigated beryllium grades is in the range of 0.2-1.5% and also monotonically increases with the neutron dose. The results of mechanical tensile and compression tests evidence that all the investigated irradiated specimens exhibit a totally brittle failure at the reduced strength level as compared to the initial state. On the whole, the beryllium specimens irradiated at 200 °C have a comparatively higher level of brittle strength. The fractographic investigations show that the main failure type of irradiated beryllium specimens is brittle and intergranular with some fraction of transgranular chip.

References

- [1] A. Cardella et al., in: Proceedings of the 4th IEA International Workshop on Beryllium Technology for Fusion, 15–17 September 1999, Karlsruhe, Germany, Proc., p. 192.
- [2] F. Scaffidi-Argentina, G.R. Longhurst, V. Shestakov, H. Kawamura, J. Nucl. Mater. 283–287 (2000) 43.
- [3] V. Barabash, G. Federici, M. Rödig, L.L. Snead, C.H. Wu, J. Nucl. Mater. 283–287 (2000) 138.
- [4] V.P. Chakin, I.B. Kupriyanov, V.A. Tsykanov, V.A. Kazakov, R.R. Melder, in: Proceedings of the 4th IEA International Workshop on Beryllium Technology for Fusion, 15–17 September 1999, Karlsruhe, Germany, Proc., p. 257.
- [5] I.B. Kupriyanov et al., J. Nucl. Mater. 233-237 (1996) 886.
- [6] I.B. Kupriyanov et al., J. Nucl. Mater. 258–263 (1998) 808.
- [7] R. Chaouadi, A. Leenaerts, J.L. Puzzolante, M. Scibetta, in: Proceedings of the 4th IEA International Workshop on Beryllium Technology for Fusion, 15–17 September 1999, Karlsruhe, Germany, Proc., p. 233.
- [8] G.A. Sernyaev, Loss of strength and embrittlement of beryllium under low-temperature irradiation, VANT, in: Nuclear Engineering and Technology, vol. I.2, 1992, p. 48.
- [9] G.A. Sernyaev, Swelling and 'spontaneous' cracking of beryllium under low-temperature irradiation, VANT, in: Nuclear Engineering and Technology, vol. I.2, 1992, p. 35.
- [10] G.A. Sernyaev, On the type of 'dose' dependence of Be ultimate strength under cryogenic irradiation, VANT, in: Nuclear Engineering and Technology, vol. I.3, 1992, p. 38.
- [11] V.P. Goltsev, G.A. Sernyaev, Z.I. Chechetkina, P.G. Averyanov, Swelling of beryllium under low-temperature irradiation, RIAR Preprint P-264, Dimitrovgrad, 1975.
- [12] V.P. Chakin, Z.E. Ostrovsky, these Proceedings.
- [13] D.R. Harries, M. Dalle Donne, F. Scaffidi-Argentina, in: Proceedings of the 4th IEA International Workshop on Beryllium Technology for Fusion, 15–17 September 1999, Karlsruhe, Germany, Proc., p. 211.
- [14] V.P. Chakin et al., Fus. Eng. Des. 58&59 (2001) 535.
- [15] L. Coheur, J.-M. Cayphas, P. Delavignette, M. Hou, in: Proceedings of the 4th IEA International Workshop on Beryllium Technology for Fusion, 15–17 September 1999, Karlsruhe, Germany, Proc., p. 247.