Nuclear Materials and Energy 16 (2018) 24–28

Contents lists available at ScienceDirect

Nuclear Materials and Energy

journal homepage: www.elsevier.com/locate/nme

Physical property changes of neutron-irradiated aluminum nitride and their recovery behavior by annealing using a step-heating dilatometer

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ARTICLE INFO

Keywords:
Aluminum nitride
Neutron irradiation
Physical property
Recovery behavior
Activation energy

ABSTRACT

Aluminum nitride (AlN) is a candidate tritium permeation, electric insulation and corrosion barriers for several kinds of blankets such as molten salt–cooled (FLiBe) or liquid metal (Li-Pb or Li)-cooled blankets because of its desirably low dielectric constant and tritium diffusion. Commercially available AlN ceramic specimens were neutron-irradiated at two different fluences but the same irradiation temperature in the Japan Materials Testing Reactor. Specimen swelling was found to be slightly different for both conditions, with higher dose causing greater swelling. All irradiated specimens consisted from hexagonal AlN phase with α-Al_2O_3 phase occurring on the surface after long-time post-irradiation annealing in He atmosphere. The a- and c-axis experienced isotropic increase and degree of unit-cell volume change was almost the same with the macroscopic volume increase obtained from the length change. This result indicates uniform distribution of Frankel pairs. After step-wise thermal annealing by using a dilatometer up to 1673 K for 6 h at each step, the maximum recovery was found at 1573 K. Based on the recovery rates at each step by first-order analysis, macroscopic length recovery during annealing can be divided into three regions with different activation energies, low temperature (373–523 K) with ~4.5 eV, intermediate temperature (523–873 K) with 0.5–1.0 eV, and high temperature (873–1273 K) with 2.0–2.5 eV. Over 1273 K, a slight increase of length was observed. It is thought that the expansion is due to oxidation.

1. Introduction

Due to the increased importance of minimizing the radioactive waste burden of fusion reactors, the development of nonmetallic structural materials is required. For this specific application ceramic materials with high thermal conductivity and good mechanical stability in nuclear environments are under consideration for a wide range of application in fusion power systems. Aluminum nitride (AlN) is one of the promising material in magneto-hydrodynamic (MHD) and/or corrosion barrier for liquid metal (Li and Li-Pb)-cooled blanket because of its excellent electrical and chemical properties. There are advantages to reduce MHD barrier pressure drop and high chemical stability in liquid lithium that exhibits high reactivity at high temperatures [1]. In the case of FLibe-cooled branket, tritium permeation barrior is required, and for this perpose AlN is also expected to be applied. Moreover, AlN properties of high thermal conductivity for about 170–220 W/m·K at 298 K and low thermal expansion coefficient of 4.3–4.6 × 10^{-6}/K from 298 to 673 K are approximately comparable to silicon carbide (SiC) which is widely used in nuclear applications [2].

Presently, the most easily accessible fusion reaction is that of the deuterium and tritium isotopes which yield about 80% of the energy in the form of a 14.1 MeV neutron [3]. This high energy neutron, through a series of elastic collisions in structural materials, produces a host is displacement cascades, displacing any and all atoms for which sufficient energy in excess of the displacement energy is imparted. In general, modification in microstructure of irradiated materials can be occurred by many factors; namely temperature, primary knock-on atom energy, displacement dose, damage rate, crystal structure, solute additions, and transmutant elements such as H and He [4–8]. By the study of Yano et al. [9] of neutron irradiation effects on AlN properties, at lower neutron fluence than 2.4 × 10^{24} n/m^2, the swelling occurred isotopically because Frenkel pairs distributed uniformly. On the other hand, at higher fluence, the anisotropic change occurred with the larger expansion in the c-axis than the a-axis because of the interstitial loop formation vertical to the c-axis. Similar results were found in subsequent studies in 1993 and 2000 of the irradiated specimens up to 10^{25}–10^{26} n/m^2 [10,11]. The only observable defect by transmission electron microscopy in the mentioned study is an interstitial dislocation

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https://doi.org/10.1016/j.nme.2018.05.027
Received 29 November 2017; Received in revised form 22 April 2018; Accepted 31 May 2018

loops lying on the (0001) basal plane, which causes expansion in the c-axis. This phenomenon is similar to BeO (beryllium oxide), which have the same hexagonal wurtzite crystal structure, and to α-Al_2O_3 (aluminum oxide, alumina) with hexagonal corundum structure.

For designing radiation-resistant materials, very efficient recovery of the defects should be focused. According to a study of Yano et al. [12], the specimen, which was irradiated at low neutron fluence of 8.3×10^{22} n/m^2 at 373 K, starts to recover gradually from close to the irradiation temperature to 1100 K. The recovery was expected to be occurred mainly by recombination of vacancies and interstitial atoms. Moreover, it has been explained that the recombination occurs by correlated pairs which is the recombination of the same pair of vacancy and interstitial in both short and long range and can be describes by first-order reaction kinetics [13–15] as shown in Eq. (1);

$$\ln[L] - \ln[L_0] = -k^*t, \tag{1}$$

where L is a length of the irradiated specimen, L_0 is a length of the unirradiated specimen, k^* is a rate constant, and t is an annealing time at a constant temperature. Meanwhile, a smaller number of uncorrelated pairs, which is the recombination of the different initial pairs in long distance, can be occurred and described by second or higher order reaction kinetics. The annealing can drive the potential to VI-recombination which brought the shrinkage of the materials occurred during the annealing.

2. Materials and methods

AlN specimens (Shapal, Tokuyama Soda Co. Japan) used in this study are commercial products with the same name "Shapal". However, they might be produced in slightly different fabrication procedures. "AlN Shapal, unirradiated specimen" and "irradiated U704" produced in 1986 have grain size for 4–8 µm. "Irradiated 89F5U" specimens produced in 1989 have grain size for 15–20 µm. Grains are equiaxed in both products. The latter shows higher thermal conductivity, but metallic impurities are the same level with the former. Then, both specimens were prepared in a 4 mm × 2 mm × 25 mm bar. All faces were polished with engraved identification numbers. The specimens were neutron-irradiated in the Japan Materials Testing Reactor (JMT) with two different fluences at almost same temperature at around 373 K. The 89F5U specimens were irradiated up to a neutron fluence of 4.4×10^{23} n/m^2 (E > 0.1 MeV) (capsule irradiation in helium atmosphere), and the U704 specimens were irradiated up to a fluence of 1.8×10^{23} n/m^2 (E > 0.1 MeV) (Hydro-rabbit irradiation). Both irradiations were conducted under constant reactor power but without temperature control and temperature monitor. The irradiation temperature of the 89F5U specimens was obtained from the length recovery of concurrently irradiated SiC by ourselves.

Specimen length was measured before and after the irradiation by using a point-type micrometer at room temperature on an accuracy of 1 µm. The average length change of 10 bars is listed. The micrometer is calibrated by using a 25.000 mm ZrO_2 standard. An X-ray diffractometer (XRD, Phillip PW-1700, Cu-Kα 0.15418 nm) was operated for both phase analysis and lattice parameter measurement with 40 kV and 40 mA from 10 to 80°/2θ using a curved graphite monochrometer to eliminate Kβ radiation. XRD patterns were obtained from the specimen surfaces. After that, the reported XRD patterns were calculated for d-spacing value using Bragg's law.

To study recovery, specimens were cut into half along their length and annealed in the dilatometer (NETSCH, DIL 402C, Germany) which was calibrated using standard sapphire rod. The measurement program and procedures were referred to the previous study of Idris et al. in recovery measurements of neutron-irradiated SiC [16]. The annealing conducted from 323 K up to 1673 K with heating rate of 5 K/min and kept at each 50 K for 6 h step-wisely under 50 ml/min helium gas flow. The length change during the annealing was continuously recorded. Accordingly, there are three specimens in total, unirradiated AlN, irradiated 89F5U, and irradiated U704. They were annealed under the same temperature profile but different maximum temperatures as mentioned in Table 1. Finally, the results of recovery behavior at each isothermal annealing step were analyzed by a first-order reaction kinetics as shown in Eq. (1), and activation energies for length recovery were obtained by Arrhenius' plot as shown in Eq. (2).

Table 1
Irradiation and annealing conditions of AlN specimens.

Code	Type	Neutron fluence (n/m^2) (E > 0.1 MeV)	Irradiation temperature (K)	Maximum annealed temperature (K)
AlN Shapal (1986)	Unirradiated	–	–	1673
89F5U (1989)	Irradiated	4.4×10^{23}	373	1673
U704 (1986)	Irradiated	1.8×10^{23}	373	1573

$$\ln[k^*] = \ln[L] = -\frac{E_a}{kT} \tag{2}$$

where k^* is a rate constant, L is a length of irradiated specimen, E_a is an activation energy (eV), k is a Boltzmann constant, 8.617×10^{-5} eV, and T is an absolute temperature (K).

3. Results and discussion

The length change of AlN induced by the neutron irradiation is shown in Table 2. Data were an average value of 10 bars. The results show a general effect of irradiation on materials is swelling. The 89F5U specimens received higher neutron fluence and showed a slightly larger percentage of swelling as 0.140(3)% while the U704 specimens of the lower neutron fluence showed the smaller percentage of 0.108(9)%. In the present case, the estimated irradiation temperature is the same, it could be said that the higher fluence 89F5U specimens showed slightly larger swelling. If the concentration of defects is saturated, irradiation-temperature affects the resulted swelling and the specimens irradiated at higher temperature generally show smaller swelling as in the case of SiC [17]. Present results on AlN swelling indicated concentration of defects is not reached saturate-level at least in the case of the lower fluence specimens.

XRD patterns of the specimens before and after post-irradiation annealing of the 89F5U and U704 specimens with those of unirradiated AlN were observed (not shown here). Those of annealed specimens were obtained after the dilatometer measurements. Before annealing, the patterns can be clearly indexed to hexagonal AlN phase (JCPDS: 01–076–0703) with the strong and sharp peaks that clarify high crystallinity of the specimens. However, after the annealing, the patterns show the mixture of the hexagonal AlN phase and alpha alumina phase (α-Al_2O_2, JCPDS: 01–075–1865), which indicated the occurrence of oxidized layer on the surface of the specimens.

Lattice parameters and percentages of lattice parameter change due to the irradiation and the annealing are listed in Table 3. The lattice expansion of the irradiated specimens 89F5U and U704 were 0.20% and 0.13% in the a-axis and 0.24% and 0.14% in the c-axis, respectively. Moreover, after the annealing until the highest temperature of

Table 2
Macroscopic length change by neutron irradiation.

Specimen	Macroscopic length change (%)	Standard error	Volume change (%)	Standard error
89F5U	0.140	0.003	0.420	0.009
U704	0.108	0.009	0.324	0.027