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Neutron irradiation damage in aluminum oxide and nitride ceramics up to a fluence of 4.2×10^{26} n/m^2

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Abstract

High-purity Al_2O_3 and AlN ceramics were concurrently irradiated in the JOYO experimental fast reactor up to 4.2×10^{26} n/m^2($E > 0.1$ MeV) at 650–1008 K. Changes in macroscopic length and lattice parameter were measured. Macroscopic swelling of Al_2O_3 was observed to be 1.2–2.3% and was larger at higher irradiation temperature. The change in lattice parameter of Al_2O_3 was very little compared to the macroscopic length change. The *a*-axis remained at the unirradiated value, whereas the *c*-axis expanded slightly. Microstructural observations showed the formation of dense and tangled dislocations and a dense array of microvoids. Macroscopic swelling of AlN was observed to be 1.8–2.1%. The change in lattice parameter of AlN was also very little compared to the macroscopic length change. The *a*-axis expanded very slightly, whereas the *c*-axis contracted. The formation of a high density interstitial dislocation loop on the basal plane was observed. The thermal diffusivity reduced by 64–74% and 96–97% in alumina and in AlN, respectively. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

Ceramics will be used for fusion reactors, as electric insulators for toroidal current breaks, neutral beam injectors and superconducting magnets; or RF windows; and many diagnostic windows. In solid breeder concepts, lithium oxides or silicates, zirconates and beryllium oxide will be used as breeding and neutron multiplier materials. Furthermore, in liquid breeding/cooling blankets, ceramic coating should be necessary to suppress MHD loses. For all of these applications, ceramics should maintain adequate electrical, thermal and mechanical properties during their design lifetime under severe neutron irradiation conditions. The evaluation of heavy neutron irradiation on the physical properties of ceramics is therefore very important.

High-purity alumina is the most typical insulator material and also a candidate insulator for fusion reactors [1–6]. A large number of neutron irradiation experiments on several kinds of alumina ceramics and single crystals have examined swelling, mechanical properties and microstructure after relatively low doses [7,8] and high doses [9,10]. Furthermore, in situ electrical property measurements have also been conducted [11,12]. Many basic properties have been clarified, such as the anisotropy of swelling, mechanical degradation as a result of anisotropic swelling, type of induced defects and radiation induced conductivity (RIC) through these studies. Despite this relatively large amount of neutron-induced property change data, it is still important to obtain further irradiation data, particularly at high neutron fluences.

Aluminum nitride ceramics with very high thermal conductivity and high electrical resistivity have been developed only recently; therefore, neutron irradiation data on AlN are very limited compared with those of alumina [13–20], particularly at high fluences. From these limited data, it appears that the anisotropic

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features of the irradiation response are similar to alumina.

In this paper, aluminum oxide and aluminum nitride specimens, both of which are the technically important ceramics containing Al as a commom component with different interatomic bonding nature, were irradiated concurrently in the neutron fluence range of 0.4–4.2 × 10^{26} n/m^2 and the macroscopic length, lattice parameter and thermal diffusivity changes were measured. The responses are analyzed based on crystal chemistry.

2. Experimental procedures

High-purity Al_2O_3 (Type NSAR, >99.9%, Nippon Steel) and AlN (Type Shapal SH-15, >99.5%, Tokuyama) ceramics were concurrently irradiated with fast neutrons in the JOYO fast experimental reactor in Japan up to fluences of 0.4–4.2 × 10^{26} n/m^2 ($E > 0.1$ MeV) at 650–1008 K. The irradiation conditions are listed in Tables 1 and 2. The temperature of specimens during irradiation was estimated based on TED temperature monitors. Relative densities of these ceramics were higher than 99.5%. The specimens irradiated were 1.2 × 1.2 × 15 mm^3 for length measurement; 10 mm in diameter and 2 mm in thickness for thermal diffusivity measurement; and 3 mm in diameter and 0.5 mm in thickness for transmission electron microscopy (TEM). Macroscopic length, lattice parameter and thermal diffusivity were measured by micrometer, X-ray diffractometry and laser-flash method, respectively. Microstructure was observed by TEM. To determine the lattice parameter of alumina and AlN, X-ray diffraction with a Cu K_α source was used, and diffraction from (220), (223), (134), (21·10), (404), (318) and (410) planes of alumina, and (210), (211), (114), (212), (105), (300), (213), and (302) planes of AlN were precisely measured using an internal standard of Si at 26°C. The experimental error of the measurement was less than 0.01% for the specimen with broadened peaks.

3. Results and discussion

3.1. Alumina

The observed length change and lattice parameter change of irradiated alumina is listed in Table 1. Length change varied from 1.20% to 2.28%. Pells summarized the volume expansion of alumina irradiated up to a fluence of 2 × 10^{26} n/m^2 [21] and indicated that the amount of swelling depended on the irradiation temperature; that is higher than 850 K or lower than 650 K. Calculating the volume change as three times the length change and comparing this to the data trend of Pells', it is clear that the present data are consistent with extrapolated lines from Pells data, as shown in Fig. 1. Values for specimens irradiated at 858–1008 K appear to fit the 850–1050 K line, and the 755 K irradiation value appears to fit the 340–680 K line. The higher temperature group is in the void formation regime, and lower temperature group is in the low defect mobility regime [21]. As represented in Fig. 2, an array of small voids was observed in all TEM specimens irradiated at 793, 893 and 1000 K up to 2.8, 3.9 and 3.7 × 10^{26} n/m^2, re-

Table 1
Irradiation condition and macroscopic length change, lattice parameter change of alumina and AlN

Fluence ($E > 0.1$ MeV) (n/m^2)	Irradiation temperature (K)	Macroscopic length change (%)		Lattice parameter change (%)			
		Alumina	AlN	Alumina		AlN	
				a-axis	*c*-axis	*a*-axis	*c*-axis
2.8 × 10^{26}	755	1.20	–	−0.01	0.17	0.12	−0.29
3.7 × 10^{26}	1008	1.96	1.78	–	–	–	–
3.9 × 10^{26}	858	1.77	1.92	–	–	–	–
4.2 × 10^{26}	1004	2.28	2.09	0.01	0.17	0.07	−0.07

Table 2
Irradiation condition and thermal diffusivity change of alumina and AlN. Thermal diffusivities of unirradiated alumina and AlN are 0.118 and 0.986 cm^2/s, respectively

Fluence ($E > 0.1$ MeV) (n/m^2)	Irradiation temperature (K)	Thermal diffusivity (cm^2/s)		Thermal diffusivity change (%)	
		Alumina	AlN	Alumina	AlN
0.4 × 10^{26}	815	0.042	0.040	−64.4	−95.9
0.5 × 10^{26}	650	–	0.033	–	−96.6
1.4 × 10^{26}	667	0.031	0.026	−73.7	−97.4

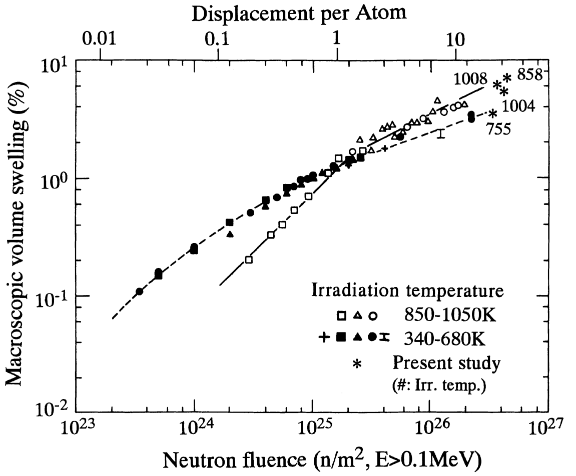

Fig. 1. Macroscopic volume swelling of alumina as a function of neutron fluence. Figure based on Fig. 6 in Pells' review [21].

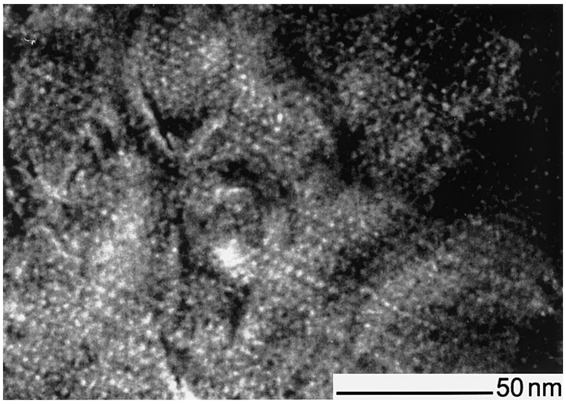

Fig. 2. Under-focused TEM image of void array in alumina irradiated to 3.7×10^{26} n/m^2 at 1000 K.

spectively, as in the cases reported by Clinard et al. [2], Youngman et al. [10] and Kinoshita et al. [22]. The combinations of temperature and fluence in which void formation was observed here were within the void formation regime for alumina summarized by Kinoshita et al. [11,22]. Grain boundary microcracking was also observed in this study. It was not observed whether the specimen irradiated at 755 K up to 2.8×10^{26} n/m^2 contained voids.

The changes in lattice parameter for two specimens of alumina are given in Table 1. In both specimens, the changes were almost equal, with almost no change in the *a*-axis parameter and a 0.17% expansion in the *c*-axis parameter. Corresponding macroscopic length change of one specimen is twice as large as the other. Anisotropy of lattice expansion of alumina was noted earlier by Wilks [23] and later by Clinard et al. [2]. It was observed that expansion of the *c*-axis was larger than that of *a*-axis, and the difference was larger at higher irradiation temperature. While the present results showed anisotropy consistent with these past reports, the amount of change was much smaller than the previous results [2]. It is also unclear that no irradiation temperature dependence was observed on lattice parameter change.

The reduction of thermal diffusivity of alumina by neutron irradiation is listed in Table 2, which indicates reductions of 64–74%. The amount of reduction is similar to previously reported values [21,24].

3.2. Aluminum nitride

Macroscopic swelling of AlN was observed to be 1.8–2.1%, as shown in Table 1. Specimens irradiated at 755 K were broken very easily and could not be used to obtain data. As compared with the previous results [17], the amount of expansion observed here was almost the same.

The change in lattice parameter of AlN was very little compared to the macroscopic length change. The *a*-axis expanded slightly, whereas the *c*-axis contracted. The shrinkage of *c* was similar to or greater than the expansion of *a*. The shrinkage of the lattice parameter by neutron irradiation in AlN was first observed in this study. On the contrary, in the case of irradiation up to 9×10^{25} n/m^2 at 773 K, both the *a*- and *c*-axes length increased [17]. It should be noted that the X-ray diffraction peaks of the present specimens were significantly broadened, making the determination of lattice parameter very difficult. Despite the presence of some uncertainty in lattice parameter, it is clear that a large departure of macroscopic volume change and unit cell volume change probably happens around 3×10^{25} n/m^2 at irradiation temperature of around 770 K, as shown in Fig. 3.

Formation of a high density of interstitial dislocation loops on the basal plane was observed by TEM. The type of dislocation loops was the same as that reported previously [13], but density was higher and the size larger (>10 nm). No voids were observed for specimen irradiated at 793 K up to 2.8×10^{26} n/m^2. Intergranular cracking was also frequently observed.

The thermal diffusivity of AlN decreased more severely than alumina. A reduction of 96–97% was measured, as is listed in Table 2.

3.3. Comparison of irradiation behavior between alumina and AlN

Alumina (α-Al_2O_3) belongs to corundum-type crystal structure based on the closest packing of oxygen anions, and is condensed by ionic interatomic bonding. On the other hand, AlN(2H–AlN) belongs to wurtzite-type crystal structure based on three dimensional connection of AlN_4 tetrahedra, and is condensed by medium of

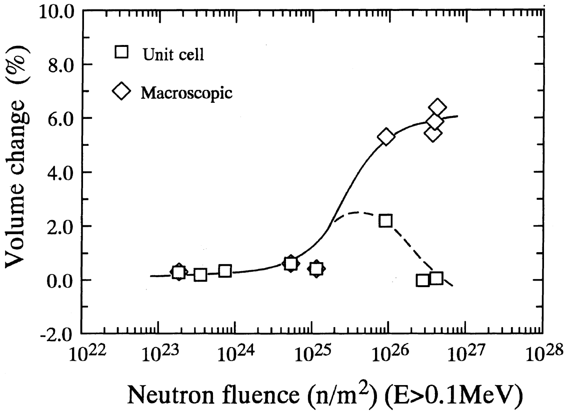

Fig. 3. Effect of neutron fluence on the macroscopic volume and unit cell volume changes in AlN. Figure based on the previous report [17].

covalent/ionic bonding. Both crystal structures have hexagonal (rhombohedral) unit cells. The macroscopic length changes of both ceramics irradiated under the same conditions were almost the same. On the other hand, changes in lattice parameter of both ceramics were very little and anisotropic. In alumina the *c*-axis expands, while the *a*-axis remains at the unirradiated value, and the *c*-axis contracts while the *a*-axis expands in AlN. Anisotropic changes in lattice parameter of alumina can be related to loop nucleation planes on both $\{1 0 \bar{1} 0\}$ and (0 0 0 1) planes, but mainly (0 0 0 1) plane in heavily irradiated alumina [2,10,11,24]. The same situation, i.e., larger expansion of the *c*-axis than that of *a*-axis and a dense interstitial-loops formation on (0 0 0 1) plane [13], has been observed in AlN irradiated up to 9×10^{25} n/m^2 [17]. The contraction of *c*-axis observed in the present study is unusual. Jack [25] reviewed the polytypes in the Si–Al–O–N system and reported that the thickness of one tetrahedral layer, corresponded to (*c*-axis length)/(number of layer), increased linearly with decreasing M/X, where M and X corresponded to metal ions and anions. This was accompanied by an increase in stacking faults in the basal plane. Based on the microstructural observations, irradiated AlN contains a large number of faulted interstitial loop on the basal plane, so expansion of the *c*-axis is expected, but it is not consistent with the experimental results.

The other difference in the irradiated microstructures is that microvoids are formed only in alumina. Clinard et al. [2] and Kinoshita et al. [11] observed that the formation of unfaulted perfect loops and then dislocations supplied more effective interstitial sinks than faulted loops in alumina, which resulted in a supersaturation of vacancies that migrated to voids or cavities. On the contrary, in AlN interstitial loops remained faulted up to very high fluences in this study. Based on the above explanation, the degree of supersaturation of vacancies is expected to be reduced in AlN. In alumina, unfaulting of 1/3[0 0 0 1](0 0 0 1) faulted loop occurs by a combination of partial shear of $1/3\langle 1 0 \bar{1} 0\rangle$ by changing cation distribution on the cation sublattice [2]. Since alumina ionically bound and only $2/3$ of cation octahedra are occupied, it is possible that the change in cation distribution is relatively easy. On the other hand, in AlN where the bonding is half covalent, redistribution of these ions may be more restricted.

The decrease in thermal diffusivity in AlN by the irradiation was more severe than that of alumina. This may be a result of a high concentration of point defects due to relatively weak sink strength in AlN. High vacancy concentration in AlN may also be the reason for the contraction in *c*-axis parameter.

4. Conclusions

High-purity Al_2O_3 and AlN ceramics were concurrently irradiated in a fast reactor up to 4.2×10^{26} n/m^2 ($E > 0.1$ MeV) at 650–1008 K. Macroscopic swelling of Al_2O_3 was observed to be 1.2–2.3% and was larger at higher irradiation temperature. The change in lattice parameter of Al_2O_3 was very little compared to the macroscopic length change. The *a*-axis remained at the unirradiated value, whereas the *c*-axis expanded slightly. Microstructural observations showed the formation of dense and tangled dislocations and a dense array of microvoids. Macroscopic swelling of AlN was observed to be 1.8–2.1%. The change in lattice parameter of AlN was also very little compared to the macroscopic length change. The *a*-axis expanded very slightly, whereas the *c*-axis contracted. The formation of a high density interstitial dislocation loops on the basal plane was observed. The thermal diffusivity reduced by 64–74% and 96–97% in alumina and in AlN, respectively. Change in irradiation response of two type of crystals should be attributed basically to the nature of interatomic bonding, ionic in alumina and half covalent in AlN, resulting in an ability of unfaulting in interstitial loops only in alumina.

Acknowledgements

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