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Saturation in degradation of thermal diffusivity of neutron-irradiated ceramics at 3×10^{26} n/m^2

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Abstract

Unlike metals, heat in ceramic materials is mainly carried by phonons, and neutron-irradiated ceramics show serious degradation in thermal diffusivity. In this work, typical structural ceramics were neutron-irradiated in the fast reactor JOYO, to fluences of 0.4–8.0 × 10^{26} n/m^2 at 646–1039 K, and thermal diffusivity was measured by the laser flash method at room temperature. It showed that the thermal diffusivity of neutron-irradiated ceramics saturated at neutron dose around 3 × 10^{26} n/m^2, and was simply determined by the irradiation temperature. All tested materials showed this behavior, with the saturation value different for each material. In ceramic materials thermal diffusivity is proportional to the mean free path of phonons. From the saturation value of thermal diffusivity at the lowest irradiation temperature of 775 K, the calculated mean free path of phonons related to neutron induced defects was roughly equal to the lattice constant of each material.
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1. Introduction

Ceramic materials have superior properties for many applications, and some of them are expected to be used as blanket materials for future fusion reactors where they would be exposed to a high fluence of 14 MeV neutrons at temperatures up to 1300 K [1,2]. High fluence neutron irradiation of ceramics induces physical property changes, such as swelling, degradation of mechanical strength, thermal diffusivity and thermal conductivity. Thermal conductivity and diffusivity is one of the most important factors for the blanket or divertor design in fusion reactors and other nuclear applications. In ceramic materials, heat is mainly carried by phonons unlike metals, and neutron irradiation induces many crystalline defects that influence the phonon transfer. It has been reported that thermal diffusivity and conductivity showed significant decrease after neutron irradiation [3–8].

In our previous works [6,8], α-Al_2O_3, AlN, β-Si_3N_4 and β-SiC specimens that were irradiated to neutron fluences of 0.4–1.4 × 10^{26} n/m^2 showed very

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low thermal diffusivity. In this work, specimens were neutron irradiated to 2.8–8.0 × 10^{26} n/m^2 at nearly the same irradiation temperature. The dependence of thermal diffusivity on neutron fluence and irradiation temperature were measured and analyzed for each material.

2. Experimental

Specimens were irradiated in the experimental fast reactor JOYO using CMIR-4 and -5 rigs, and four kinds of materials, α-Al_2O_3, AlN, β-Si_3N_4 and β-SiC were enclosed in the same capsule. The capsules were irradiated to different neutron fluences at different temperatures. Irradiation conditions for each specimen are listed in Table 1 and additional details were reported in our previous papers [9,10]. In our previous works [6–8], it was reported that specimens irradiated to neutron fluences of 0.4–1.4 × 10^{26} n/m^2 (correspond to specimen ID T7x) showed very low thermal diffusivity. In this work, specimens irradiated to 2.8–8.0 × 10^{26} n/m^2 at higher temperatures (correspond to specimen ID T5x) were measured. The dimensions of the specimens were 3 mm diameter and 0.5 mm height for T5x specimens and 10 mm diameter and 2 mm height for T7x specimens.

Thermal diffusivity was measured by the laser flash method with the $t_{1/2}$ analysis using a specially ordered ULVAC TC-7000 that can measure 3 mm diameter × 0.5 mm thick (T5x) specimens. The 10 mm diameter × 2 mm thick (T7x) specimens, as reported previously, were measured using a Rigaku FA8510B. The measurements were performed at room temperature. AlN and α-Al_2O_3 specimens were coated using a carbon spray and baked on a hot plate at about 373 K before the measurement to reduce their translucence.

3. Result

Fig. 1(a)–(d) show the dose dependence of normalized thermal diffusivity for each material. In this work, unirradiated specimens of both 10 mm and 3 mm diameters were measured with the same measurement system (Ulvac TC-7000). The result required a significant dimensional correction; that is, 3 mm diameter specimens showed different thermal diffusivity than 10 mm diameter specimens. The 10 mm specimens are usually used as standard; and the T7x specimens are 10 mm diameter × 2 mm thick, but T5x specimens are 3 mm diameter × 0.5 mm thick. In Fig. 1 the neutron irradiation effects are given as normalized thermal diffusivity by the unirradiated specimen, defined as $\alpha_{irr}/\alpha_0 \times 100\%$, where α_{irr} represent the thermal diffusivity after neutron irradiation and α_0 for an unirradiated specimen of the same size. The α-Al_2O_3 specimens showed relatively small change and maintains normalized values of 20–35%, while AlN: 2–4%, β-Si_3N_4: 12–29%, β-SiC: 9–14%, relatively. But it mainly depends on the thermal diffusivity before the irradiation. The absolute thermal diffusivities of neutron-irradiated and unirradiated specimens measured at room temperature are given in

Table 1
Measured thermal diffusivity (α) and calculated mean free path of phonon related to irradiation induced defect (λ_d) and the irradiation condition

ID	Neutron fluence (10^{26} n/m^2)	Irradiation temperature (K)	Thermal diffusivity α (10^{-6} m^2/s) λ_d (nm)			
			α-Al_2O_3	AlN	β-Si_3N_4	β-SiC
3 mm diameter × 0.5 mm thick disk specimen						
unirradiated			7.68	61.6	25.7	33.4
			2.29	19.1	8.16	8.79
T51	2.8	775	1.85	1.42	3.80	3.07
			0.73	0.67	1.42	0.89
T52	5.3	775	1.60	1.30	3.18	2.84
			0.60	0.41	1.15	0.82
T53	3.9	864	1.96	1.36	3.90	3.10
			0.79	0.43	1.46	0.90
T54	7.3	835	1.87	1.35	3.46	3.00
			0.74	0.43	1.27	0.87
T55	4.2	1004	2.86	1.69	4.76	3.51
			1.36	0.54	1.86	1.03
T56	8	950	2.83	1.81	4.10	3.14
			1.34	0.58	1.55	0.91
T57	3.7	1011	2.88	1.74	4.83	3.60
			1.37	0.55	1.89	1.06
T58	6.9	1039	2.37	1.67	3.59	2.96
			1.02	0.53	1.33	0.86
10 mm diameter × 2 mm thick disk specimen						
unirradiated			11.8	99.1	25.0	41.0
			3.52	30.1	7.95	10.8
T71	0.5	646	—	3.31	4.00	4.76
			—	1.06	1.51	1.42
T72	1.4	668	3.09	2.23	4.78	4.87
			1.25	0.71	1.88	1.46
T73	0.4	853	4.14	4.17	7.17	5.54
			1.90	1.35	3.20	1.69

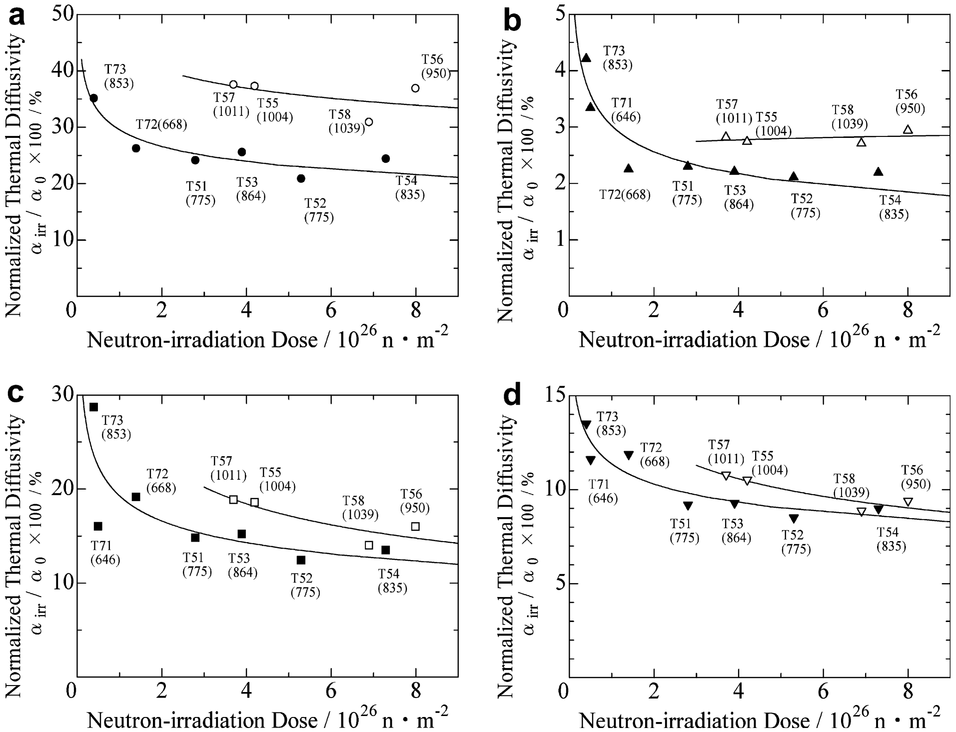

Fig. 1. The dose dependence of normalized thermal diffusivity for each material. The normalized thermal diffusivity by the unirradiated specimens was defined as $\alpha_{irr}/\alpha_0 \times 100\%$ where α_{irr} represent the thermal diffusivity after neutron irradiation and α_0 for unirradiated specimen. A number in parentheses under a specimen ID gives the irradiation temperature (K). Specimens they were irradiated at relatively high temperatures are shown by open symbols, and irradiated at lower temperatures shown by solid symbols. (a) α-Al_2O_3, (b) AlN, (c) β-Si_3N_4 and (d) β-SiC.

Table 1. The β-Si_3N_4 specimens maintained the highest thermal diffusivity after neutron irradiation, compared to α-Al_2O_3, AlN and even β-SiC.

In Fig. 1, the number in parentheses under a specimen ID shows the irradiation temperature (K). Specimens were separated into two groups by irradiated temperature, and specimens irradiated at relatively high temperature (950–1039 K) are shown with open symbols in Fig. 1, while solid symbols represent lower temperatures (646–864 K). With this grouping, the data shows that the dose dependence of thermal diffusivity saturated near 3×10^{26} n/m^2 for all materials tested.

The thermal diffusivity saturation with fluence was simply determined by the irradiation temperature, and increased slightly with an irradiation temperature increase. Fig. 2(a) and (b) show the relationship of the normalized thermal diffusivity and irradiation temperature for each material. In this figure, a number in parentheses under a specimen ID gives the neutron fluence (10^{26} n/m^2). It is easy to see that T7x specimens with solid symbols showed higher thermal diffusivity than the other specimens irradiated to more than 3×10^{26} n/m^2 presented with open symbols. This verifies that T7x specimens were not saturated at the neutron fluence achieved, while specimens irradiated to more than 3×10^{26} n/m^2 reached saturation and the thermal diffusivity was determined only by the irradiation temperature.

4. Discussion

In Fig. 1(c) and (d), the T71 specimens showed relatively low thermal diffusivity at that neutron dose. T71 specimens were irradiated at 646 K and the microstructure of a β-Si_3N_4 specimen was observed by HREM in our previous work [11]. It showed that the T71 β-Si_3N_4 specimen contained no obvious interstitial dislocation loops but poorly defined clusters or small rudiment of loops, while the T72 specimen irradiated at 668 K contained clear dislocation loops. In addition, the annealing behavior of swelling and thermal diffusivity in T71

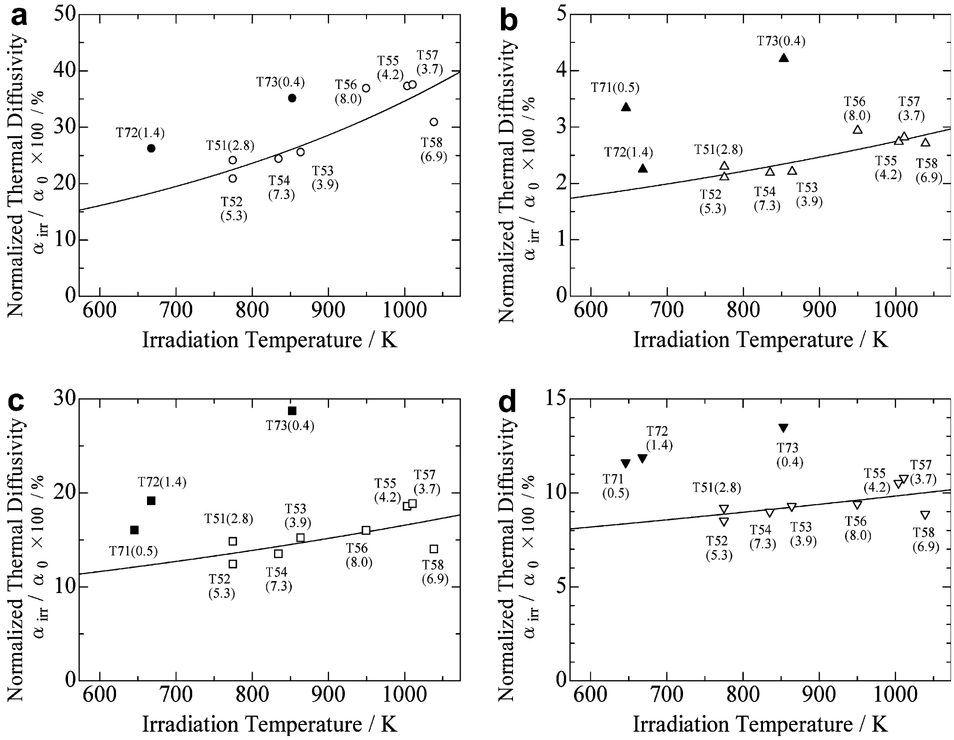

Fig. 2. Dependence of normalized thermal diffusivity on irradiation temperature for each material. A number in parentheses under a specimen ID gives the neutron fluence (10^{26} n/m^2). Specimens they were irradiated to relatively high neutron fluences ($>3\times10^{26}$ n/m^2) are shown with open symbols, and to lower fluences shown with solid symbols. (a) α-Al_2O_3, (b) AlN, (c) β-Si_3N_4 and (d) β-SiC.

β-Si_3N_4 specimens was obviously different from that in T72 or T73 specimens [8,11]. The behavior showed that at 646 K interstitial atoms were not mobile to create interstitial dislocation loops or to annihilate with non-neighboring vacancies, therefore many isolated interstitial atoms and vacancies remain. This difference in microstructure of irradiation induced defects caused lower thermal diffusivity of T71 specimen than the other sample series. This also caused significant difference in swelling behavior, and the T71 α-Al_2O_3 specimen was hardly cracked and thermal diffusivity could not be measured.

In addition, T56 and T58 specimens showed a difference in slope of the diffusivity trend of other specimens. It is believed that these specimens were mixed at some process in the specimen treatment. When T56 and T58 are plotted interchanged in Figs. 1 and 2, results were ordered better. The β-SiC specimen for swelling measurement that was irradiated at the same condition as the T58 specimen was annealed and the recovery onset temperature showed some difference from the irradiation temperature of T58 but was close to that of T56. Unfortunately, the swelling specimens that were irradiated at the T56 condition were broken and the swelling could not be measured. The recovery behavior of the β-SiC specimen will be measure using the fractured pieces in future work.

In ceramic materials, the thermal diffusivity is proportional to the mean free path of phonons. This mean free path can be heavily affected by irradiation induced defects, especially vacancies. The mean free path of phonons after irradiation (λ_{I}) is given as $1/\lambda_{\mathrm{I}} = 1/\lambda_0 + 1/\lambda_{\mathrm{d}}$ where λ_0 is for unirradiated material and λ_{d} is related to the irradiation induced defect. Experimentally, λ_0 and λ_{I} were obtained from measured thermal diffusivity before and after irradiation, with the expression $\alpha = 1/3\mu\lambda$. In this expression, μ(m/s) represents the mean phonon speed, and as calculated from $\mu = (E/\rho)^{1/2}$ where E (Pa) is the Young's modulus and ρ (kg/m^3) is the density. In this paper we used Young's modulus and density for unirradiated materials and calculated μ values to be 1.01 (α-Al_2O_3), 0.97 (AlN), 0.94 (β-Si_3N_4) and 1.14 (β-SiC) (10^4 m/s).

In Table 1, λ_{d} or λ_0 are listed with the measured thermal diffusivity. From the saturated thermal diffusivity at the lowest irradiation temperature of 775 K (T52), values of λ_d obtained are α-Al_2O_3: 0.60 nm, AlN: 0.41 nm, β-Si_3N_4: 1.2 nm and β-SiC: 0.82 nm, while λ_0 was 2.3, 19, 8.2 and 8.8 nm for each material (3 mm thick specimens). These λ_d lengths roughly correspond to the lattice constant, $a = b$: 0.476, 0.311, 0.760, 0.436 nm for α-Al_2O_3, AlN, β-Si_3N_4 and β-SiC within a factor of 1.3–1.9. This coincidence is explained because if two vacancies get closer than the lattice constant, they can not act as independent vacancies but act as one divacancy in relatively simple crystal structures. This will cause the saturation in degradation of thermal diffusivity.

Dienst et al. reported 15–30% reduction in Young's Module after 1 dpa neutron irradiation [12,13]. A 20% reduction of Young's Modulus leads to about a 10% increase in λ_d. We measured dynamic micro hardness for the same samples here, but obvious difference was not obtained because of large scatter. Swelling values for the same samples in this paper have already been reported [9], where the linear swelling for α-Al_2O_3 and AlN were about 2% and for β-Si_3N_4 and β-SiC were about 0.3%. But it gives very small change to the λ_d value, and the swelling data were not obtained for the all specimens. Corelli et al. used a velocity of sound in the material as an approximation of the mean phonon speed [14], which is one possible way to estimate the mean speed of phonons in the irradiated specimens. For further detailed treatment, these factors should be considered.

The λ_0 reflects the thermal diffusivity of unirradiated specimens, and is affected by many factors, including grain shape, grain boundary structure, impurities or sintering agents. However, these effects are insignificant in irradiated materials, because dimensions of these defects are large compared to that of irradiation induced defects, which cause the main changes in the mean free path of phonons.

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