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Thermal diffusivity of ceramics at the neutron irradiation temperature estimated from post-irradiation measurements at 123–413 K

Masafumi Akiyoshi

Kyoto University, Yoshida-Honmachi, Sakyo-ku, Kyoto 606-8501, Japan

A B S T R A C T

In ceramic materials, heat is mainly carried by phonon. During and after irradiation, phonon is scattered by neutron-induced defects, and the neutron-irradiated specimen showed severe degradation in thermal diffusivity. On the other hand, phonons are also scattered among each other independent of any defects, and the scattering increases with the increase in the measurement temperature. In this study, the thermal diffusivity of heavily neutron-irradiated ceramics was measured at the temperatures of 123–413 K. The dependence of thermal diffusivity on measurement temperature was approximated with a function, and with several fitting parameters, the thermal diffusivity at irradiation temperature was estimated without measurement at elevated temperature that causes annealing effect. With several assumptions, the thermal diffusivity at the irradiation temperature has been considered to represent the thermal diffusivity during irradiation. Furthermore, the estimated thermal diffusivity during irradiation was almost independent of the irradiation temperature.

1. Introduction

Some structural ceramic materials like SiC possess many superior properties for the future nuclear applications, where they would be exposed to high fluence of 14 MeV neutrons at temperatures up to 1400 K [1–3]. High fluence of neutron irradiation induces many defects and produces various changes in the physical property. Thermal diffusivity is one of the most important factors for blanket material of the future fusion reactor and other nuclear applications, such as high-temperature gas cooling fission reactor. It has also been reported that the thermal diffusivity of neutron-irradiated ceramics showed significant degradation [4–10].

In ceramic materials, unlike metals, heat is mainly carried by phonon, and phonon transportation is scattered by lattice defects (phonon–lattice scattering). Hence, neutron-irradiated specimens show severe degradation in the thermal diffusivity. On the other hand, phonon is scattered among each other (phonon–phonon scattering), independent of any defects. Thermal diffusivity of ceramics is mainly affected by these two scatterings. The former, phonon–lattice scattering, is not affected by measurement temperature, but the density of the defects is dependent on the irradiation temperature. At higher irradiation temperature, the recombination is more likely and the density of the defects is lower. On the other hand, the frequency of phonon–phonon scattering increases with the measurement temperature even in the non-irradiated specimens.

In most of the earlier studies, post-irradiation measurements were operated at room temperature. After irradiation, the defects in the specimens were quenched and maintained at the same density, but the frequency of the phonon–phonon scattering at room temperature was different from that at the irradiation temperature. In our earlier work, we measured the neutron-irradiated specimen at elevated temperatures, and using an approximation function, the thermal diffusivity at the irradiation temperature was estimated [11]; however, the number of measurement temperature was only five and the irradiation condition used was only three for each material. After the annealing at the irradiation temperature, the measurements at room temperature were performed, which showed a little recovery in the thermal diffusivity. Heavily neutron-irradiated specimens in this work are very precious and the number is quite limited, and hence, we avoided the measurement at elevated temperature that annealed out the defects. In our study, the measurements of thermal diffusivity were carried out at 123–413 K.

2. Experimental

Typical structural ceramics, α-Al_2O_3, AlN, β-Si_3N_4, and β-SiC specimens, were irradiated in the experimental fast reactor JOYO enclosed in the same capsule. Several capsules were irradiated to different neutron fluence ($0.4–7.3 \times 10^{26}$ n/m^2) at different temperatures (646–1039 K). These irradiation conditions are listed in Table 1. Properties of each specimen before the irradiation have been reported in our earlier studies [11,12]. The dimensions of

E-mail address: akiyoshi@nucleng.kyoto-u.ac.jp

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Table 1
Neutron-irradiation conditions for each specimen and the obtained fitting parameters *n*, *k*, and the thermal diffusivity at the irradiation temperature, α_{irr}.

ID	Dose (10^{26} n/m^2)	T_{irr} (K)	α-Al_2O_3			AlN			β-Si_3N_4			SiC		
			n	α_{300}	α_{irr}	*n*	α_{300}	α_{irr}	*n*	α_{300}	α_{irr}	*n*	α_{300}	α_{irr}
				(10^{-6} m^2/s)			(10^{-6} m^2/s)			(10^{-6} m^2/s)			(10^{-6} m^2/s)	
Non-irradiated			2.30	11.8	–	1.08	99.1		1.06	25.0		1.05	41.0	
T51	2.8	775	0.50	2.28	1.18	–	1.73	–	0.50	4.70	2.37	–	3.81	–
T53	3.9	864	0.66	2.42	1.14	0.33	1.66	0.95	0.57	4.82	2.13	0.81	3.85	1.32
T55	4.2	1004	0.76	3.52	1.30	0.41	2.06	1.04	0.56	5.88	2.41	0.68	4.36	1.55
T57	3.7	1011	0.51	3.55	1.73	0.31	2.14	1.21	0.58	5.98	2.40	0.64	4.48	1.65
T71	0.5	646	–	0.00	–	0.70	3.31	1.93	0.59	4.00	2.54	0.82	4.76	2.53
T72	1.4	668	0.78	3.09	1.75	0.55	2.23	1.44	0.68	4.78	2.77	0.82	4.87	2.52
T73	0.4	853	0.95	4.14	1.70	0.56	4.17	2.33	0.63	7.17	3.71	0.84	5.54	2.31
T52[a]	5.3	775	0.40	1.97	1.07	0.29	1.59	0.88	0.31	3.92	2.45	0.37	3.53	1.95
T54[a]	7.3	835	0.52	2.30	1.11	0.37	1.65	0.91	0.40	4.28	2.46	0.42	3.72	1.92
T58[a]	6.9	1039	0.51	2.91	1.29	0.27	2.04	1.10	0.34	4.44	2.39	0.38	3.68	1.81

[a] Result from the previous work [11].

the specimens were ϕ 3 × 0.5 mm and ϕ 10 × 2 mm for the T5x and T7x specimen IDs, respectively.

In this study, the thermal diffusivity was measured by the laser-flash method using specially ordered measurement system that can measure φ 3 mm disk (ULVAC-RIKO Inc. TC-7000L/Special), and was analyzed with the $t_{1/2}$ method. All the specimens were coated using graphite spray and baked on a hot plate at about 400 K to avoid transmission of laser flash. Temperature of a specimen after a laser flash was measured by φ 0.05 mm *K*-type thermocouple that was attached using electroconductive epoxy (Circuit works CW2400) below 400 K to avoid annealing. Measurements at low temperature were carried out in He atmosphere (<10^4 Pa), and a cryostat was cooled by liquid nitrogen. Measurements were taken at every 20 K from 123 K to 413 K. In addition, measure-

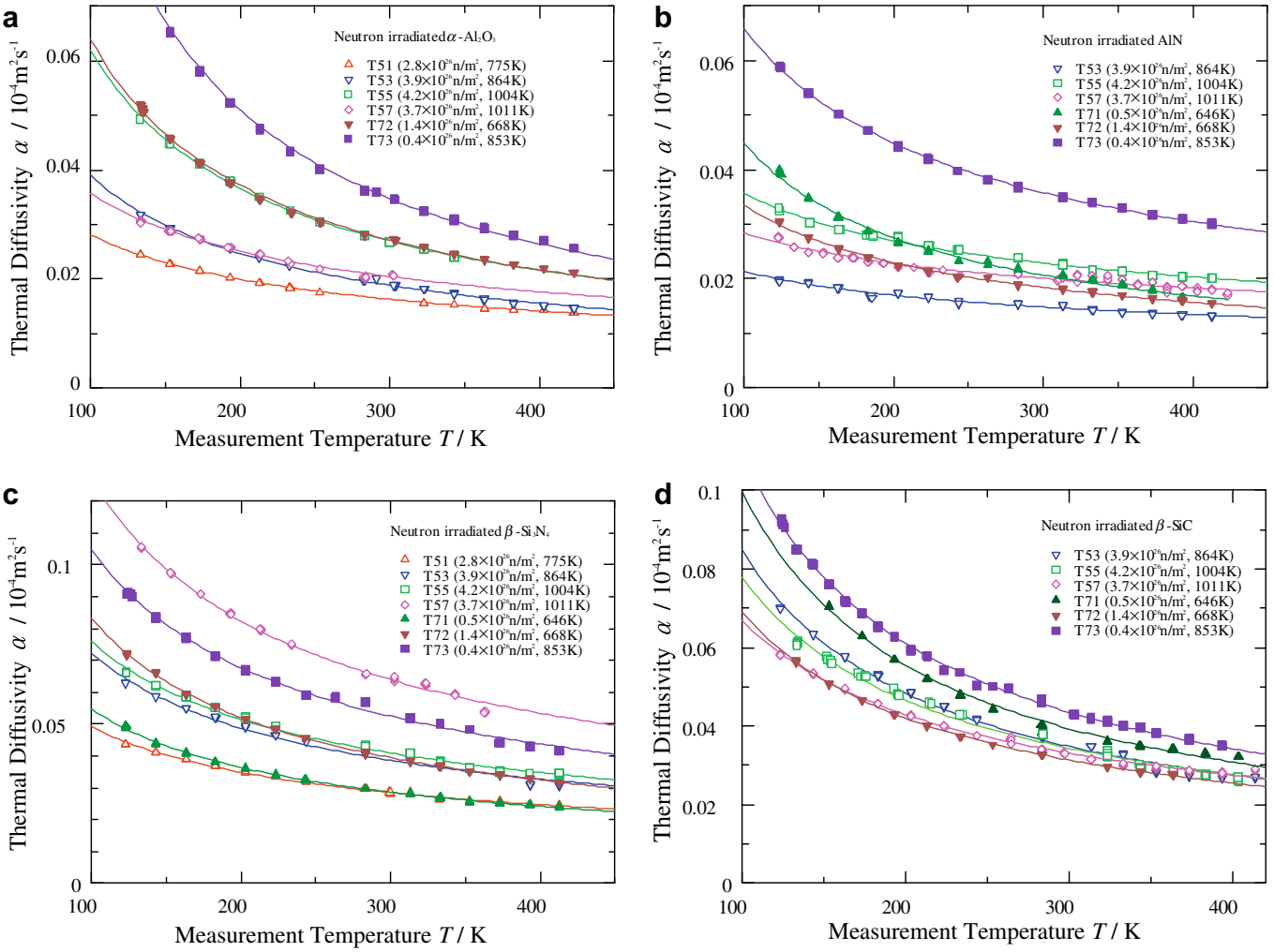

Fig. 1. The dependence of thermal diffusivity of neutron-irradiated ceramics on measurement temperature from 123 K to 413 K. (a) α-Al_2O_3, (b) AlN, (c) β-Si_3N_4, and (d) β-SiC.

ments using infrared sensor that produces more reliable values were taken at room temperature for all the specimens [4,8,9,11,13]. Further, measurements at elevated temperatures for T52, T54, and T58 specimens were obtained from our earlier works [4,11].

3. Result

Fig. 1(a)–(d) shows thermal diffusivity of neutron-irradiated ceramics (α-Al_2O_3, AlN, β-Si_3N_4, and β-SiC) measured at 123–413 K. Thermal diffusivity, α (m^2/s), of neutron-irradiated ceramics depend on the measured temperature T (K) as $\alpha = k/T^n$, where k is the constant that is related to the absolute value, and n is the constant that represents the state of induced defects. In addition, the thermal diffusivity at 300 K (α_{300} (m^2/s)) was measured by infrared sensor, and the thermal diffusivity at irradiation temperature, α_{irr} (m^2/s), was estimated as $\alpha_{irr} = \alpha_{300}\,(300/T_{irr})^n$, where T_{irr} is the irradiation temperature. Fig. 2(a)–(d) illustrate the dependence of α_{irr} on T_{irr}, plotted with the results from the earlier work, which were obtained by measurements at elevated temperature [11]. In Table 1, parameter n, α_{300}, and α_{irr} are presented with irradiation conditions for each specimen. For each material, the trend of estimated thermal diffusivity at irradiation temperature in this study agreed with the result of the earlier work, except T7x specimens. The T7x specimens were irradiated to 0.4–1.4 × 10^{26} n/m^2 and the degradation of thermal diffusivity was not saturated with neutron dose; however, the T5x specimens were irradiated to 2.8–8.0 × 10^{26} N/m^2 and saturated [4].

4. Discussion

In Table 1 and Fig. 2(a)–(d), α_{irr} represents the thermal diffusivity at the irradiation temperature. Furthermore, α_{irr} even represent the thermal diffusivity during the irradiation based on the following two assumptions: (1) post-irradiated specimens would retain the same amount of defects as during irradiation and (2) defects would remain stable at the measurement temperature.

In this study, the irradiated specimens were cooled in the reactor within 12 h, and the specimens suffered a little irradiation at a lower temperature than the scheduled irradiation temperature. This problem was also discussed in the earlier work about swelling in β-SiC [13]. The JOYO fast reactor has high neutron flux of 5 × 10^{19} n/m^2 s near the core that gives 2 × 10^{24} n/m^2 within 12 h. However, thermal diffusivity was saturated with neutron dose at around 3 × 10^{26} n/m^2 and this effect is considered to be relatively small.

However, more important factor is the amount of transient defects during irradiation. In the specimen during irradiation, the interstitial atoms and vacancies are created by neutron irradiation, and subsequently, most of them recombine in a very short time.

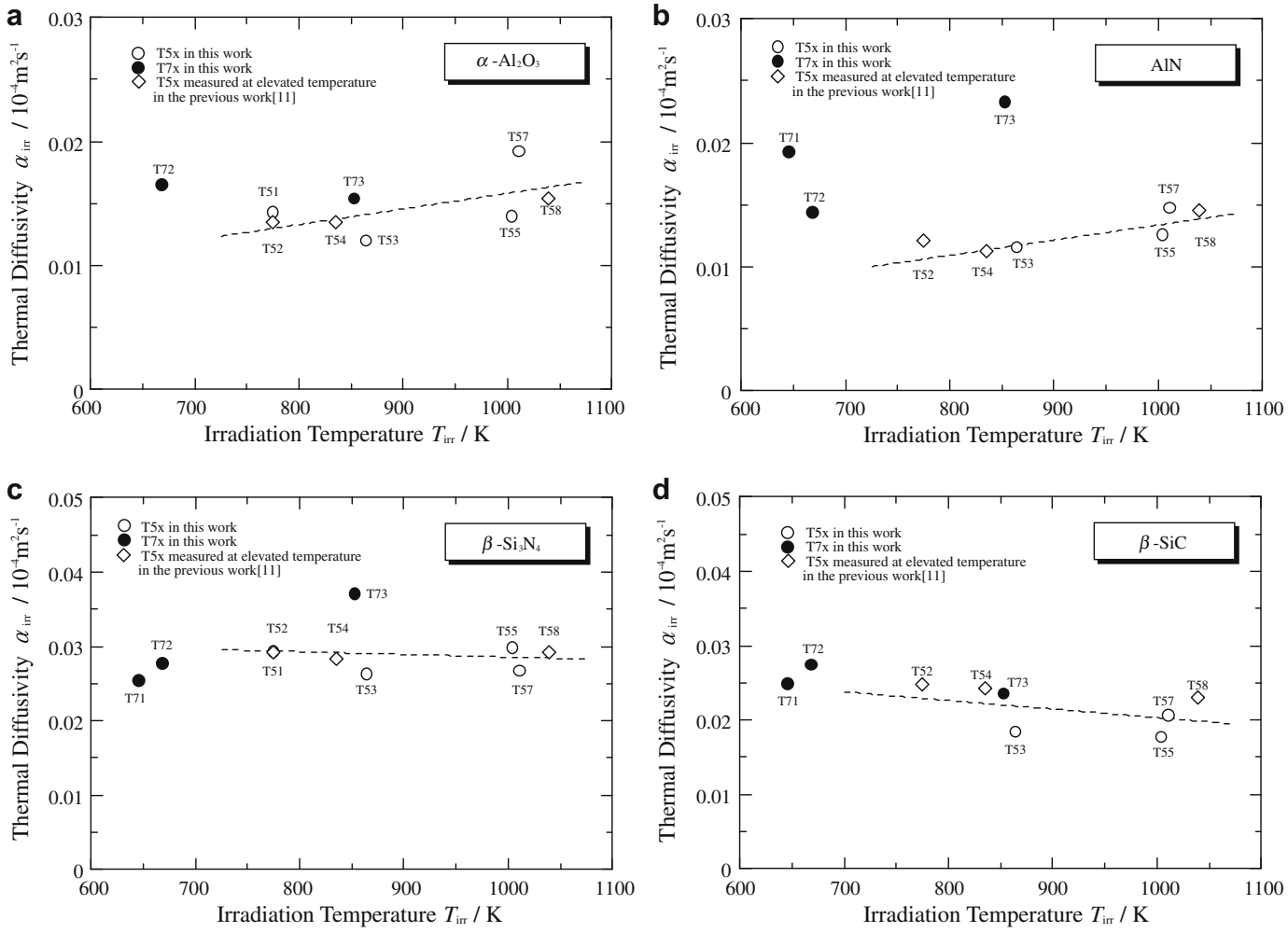

Fig. 2. Estimated thermal diffusivity of neutron-irradiated ceramics at the irradiation temperature, α_{irr}, plotted with the irradiation temperature, T_{irr}. (a) α-Al_2O_3, (b) AlN, (c) β-Si_3N_4, and (d) β-SiC.

Neutron irradiation flux changes the amount of transient defects before recombination, but each atom is displaced 28–42 times during 176 days ($1.8–2.8 \times 10^{-6}$ dpa/s) in the case of the specimens in this study, which indicates that the ratio of the transient defects is very small. Dynamic effect by transient defects during irradiation is an important issue to resolve the defect-production processes [14], but the amount of transient defects is too small to produce significant effect on thermal diffusivity. Nowadays, positron annihilation method is widely used to investigate the irradiation defects, and we are now trying to estimate the effect of transient defects during ion-beam irradiation; however, the result suggests that the transient defects offer almost no contribution to thermal diffusivity.

The second assumption can be clarified very simply, because the measurements were carried out at a very low temperature, compared with the irradiation temperature, and in the earlier works, the point defects were not mobile enough to recombine or make larger structure below 646 K in the case of β-Si_3N_4 [15], and other materials in this study also showed the same tendency. Furthermore, the post-irradiation measurements of thermal diffusivity after isochronal annealing support this assumption [8–10].

Hence, it can be stated that thermal diffusivity at irradiation temperature, α_{irr}, obtained by post-irradiation measurements in this study represents the thermal diffusivity during irradiation. In addition, degradation of thermal diffusivity was saturated with neutron dose above 3×10^{26} n/m^2 [4,5,16], and subsequently, the thermal diffusivity is decided simply by the irradiation temperature. However, at higher irradiation temperature, interstitial atoms and vacancies recombined easily, and phonon–lattice scattering was restrained. On the other hand, phonon–phonon scattering was increased at higher temperature; hence, the thermal diffusivity at the irradiation temperature was balanced at almost the same level within the irradiation temperature of 775–1039 K.

On careful observation of Fig. 2(a)–(d), it can be noted that α_{irr} of α-Al_2O_3 and AlN showed a little increase with the irradiation temperature, while that of β-Si_3N_4 and β-SiC remained almost the same or rather showed a little decrease. Nevertheless, these changes were rather small, and α-Al_2O_3 showed about 1.5×10^{-6} m^2/s, AlN: 1.3×10^{-6} m^2/s, β-Si_3N_4: 2.7×10^{-6} m^2/s, and β-SiC: 2.3×10^{-6} m^2/s.

5. Conclusion

Thermal diffusivity of neutron-irradiated ceramics was measured at 123–413 K. Using the obtained fitting parameters, thermal diffusivity at the irradiated temperature was estimated for specimens irradiated at 646–1039 K. Estimated thermal diffusivities at the irradiation temperature were almost independent of the irradiation temperature within the range of 775–1039 K, i.e., 1.5, 1.3, 2.7, 2.3×10^{-6} m^2/s for α-Al_2O_3, AlN, β-Si_3N_4, and β-SiC, respectively. However, the contribution of transient defects on thermal diffusivity during neutron irradiation is assumed to be very small in this study. Hence, the estimated value is considered to represent the thermal diffusivity during irradiation.

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