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Microstructure and Helium Release Behavior of Neutron-Irradiated SiC Containing B_4C

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Abstract

SiC containing B_4C as sintering additive of three kinds of ^{10}B concentration were neutron-irradiated in the Japan Materials Testing Reactor up to a fluence of 2.8×10^{24} n/m^2 (>0.1 MeV) at 300°C. Microstructure observation on the as-irradiated and annealed samples was conducted. He bubbles were observed in the samples annealed at 1300°C, but for 700°C-annealed samples. The latter samples contained black-dot like contrasts in grains. The relation between the mobility of He atoms and vacancies, and bubbles at grain boundary was discussed.

Keywords:
microstructure, helium release, SiC, B_4C, swelling

1. Introduction

Study on effects of neutron irradiation onto silicon carbide (SiC) ceramics would be very important because SiC is one of the candidate materials for use in the structure and blanket materials for fusion reactors [1-3]. He effects on thermal and mechanical properties, and microstructure of neutron-irradiated SiC have been studied by several authors, using $^{10}B(n,\alpha)^7Li$ nuclear reactions. Carey *et al.* [4] observed that SiC containing ^{10}B exhibited volume swelling up to 7%, which was considerably larger than that of SiC without ^{10}B. Correli *et al.* [5] mentioned that burnup of B caused a drastic reduction of fracture strength. Harrison *et al.* [6] observed that He bubbles were formed at grain boundary of SiC ceramics annealed at 1500°C.

Holt *et al.* [7] measured two release peaks of He at 1350 and 2400°C from CVD-SiC irradiated by 14 MeV neutrons. Sasaki *et al.* [8,9] investigated He release rates and microstructure of neutron-irradiated SiC. He release mechanism and its temperature dependence were also reported. The present authors [10] examined He release behavior and change in physical properties of neutron-irradiated SiC ceramics containing B_4C. In the present study, microstructural observation by a Transmission Electron Microscope (TEM) was carried out on neutron-irradiated SiC ceramics containing B_4C after annealing at several temperatures.

2. Experimental Procedure

The specimens were sintered α-SiC ceramics containing B_4C using various concentration of ^{10}B. Each set of these sintered samples was made from the mixture of α-SiC powder and B_4C powder, by hot-pressing at 2050°C for 0.5 h. Three kinds of B_4C were used, i.e., with 0.4 wt.% of B_4C with 19.6% of ^{10}B, 0.4 wt.% of B_4C with 91.6% of ^{10}B, and mixture of the two B_4C (56.8% of ^{10}B), namely natural B_4C, enriched B_4C and mixed B_4C, respectively. Calculated helium production amounts from them were 100, 280 and 460 appm, and estimated He concentration was 1.43×10^{15}, 3.73×10^{15} and 5.88×10^{15} atom/mg, for natural, mixed and

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enriched B_4C, respectively. These samples ($2 \times 5 \times 20$ mm^3) were neutron-irradiated in the Japan Materials Testing Reactor up to a fluence of 2.8×10^{24} n/m^2 (E > 0.1 MeV) at 300°C in He-filled capsule. After the neutron irradiation, the specimen was cut into small bulk ($2 \times 1.5 \times 1$ μm^3) and crushed into powder, with average particle size of 1 mm. He release rates up to 1800°C with heating rate of about 8°C/min were continuously measured.

Specimens for TEM observation were made from sintered specimens with the size of $1 \times 2 \times 0.15$ mm^3. The irradiated specimens were heated up to 700, 1300 and 1500°C, at a rate of 30°C/min and kept for 1 h. After the heat treatment, those specimens were mechanically polished and thinned by an Ar ion-beam under an accelerating voltage of ~5 kV. The microstructural observation was performed with a TEM (Hitachi Ltd., H-9000) operating at 300 kV.

3. Results and Discussion

Figure 1 shows the He release profiles from bulk specimens and powders containing various concentration of ^{10}B in B_4C.

The He release peaks from SiC powder were observed at around 250, 1160 and ~1800°C. The continuous peak was also observed at around 650~1350°C. The quantity of He release from SiC containing enriched B_4C was the highest. Comparing these profiles from previously reported release profiles from the neutron-irradiated SiC containing B [8,9], the difference of peak position is observed, particularly the peak at ~1160°C.

Bubbles were not observed for the as-irradiated SiC, regardless of the ^{10}B concentration.

Figure 2 (a) and (b) show micrographs around grain boundaries of the irradiated SiC containing natural B_4C and enriched B_4C, respectively, after annealing at 1300°C for 1 h. The bubble size of SiC containing enriched B_4C was larger than that containing natural B_4C, indicating He bubble size is larger with increasing ^{10}B concentration. Furthermore, the bubble size of SiC annealed at 1500°C was generally larger than that of the sample annealed at 1300°C.

The He bubble along grain boundaries was not observed for the three kinds of specimens annealed at 700°C. Figure 3 (a) and (b) are high magnification TEM

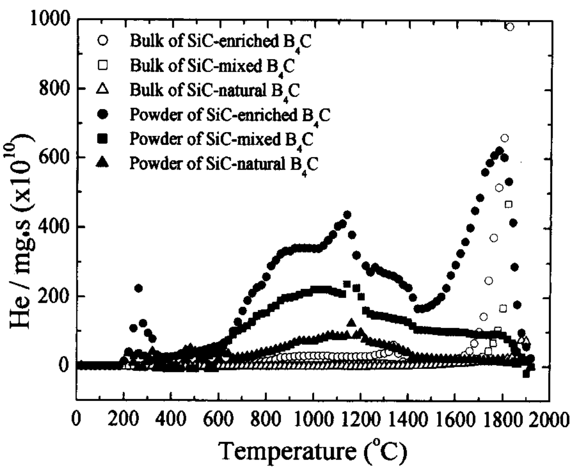


Fig. 1 Helium release rates from 1 mg of irradiated SiC bulk specimens and powder containing natural B_4C (19.6% ^{10}B), mixed B_4C (56.8% ^{10}B), and enriched B_4C (91.6% ^{10}B)

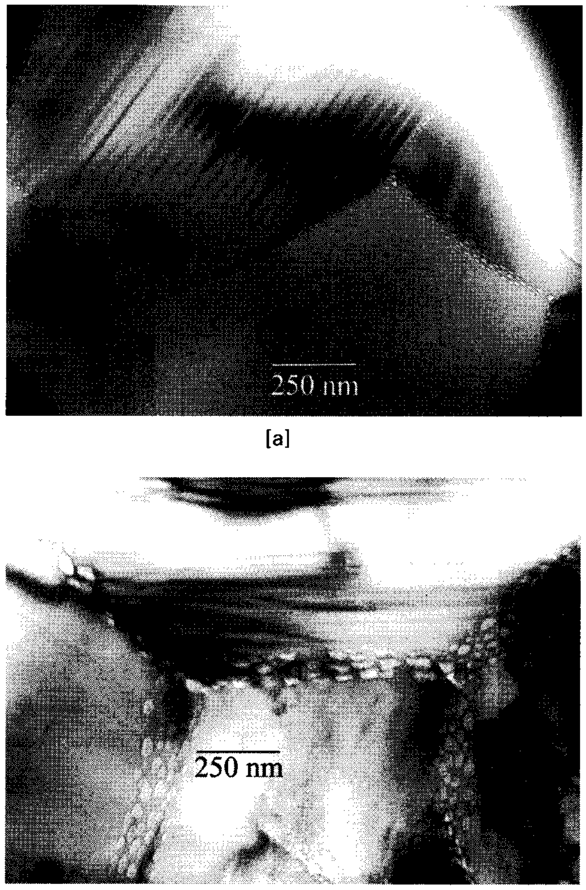
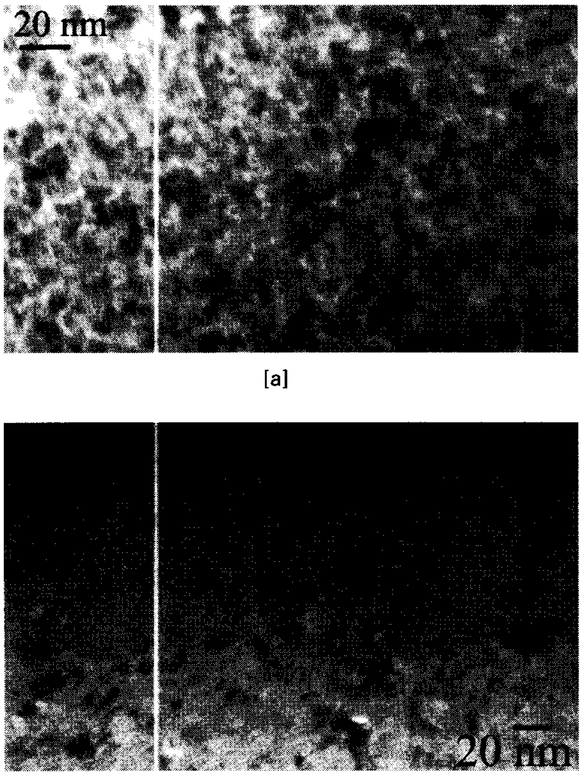


Fig. 2 TEM photographs of neutron-irradiated SiC ceramics containing natural B_4C [a] and enriched B_4C [b], irradiated to 2.8×10^{24} n/m^2 (E > 0.1 MeV), annealed for 1h at 1300°C.

micrographs of the samples annealed at 700°C for 1 h, containing natural B_4C and enriched B_4C, respectively. "Black-dot" like contrasts were observed in both the specimens. Neutron irradiation primarily produces pairs of interstitial and vacancy, Frenkel defect, into SiC and it induces volume increase [11,12]. These are mostly annihilated due to migration and recombination during annealing up to 1400°C. Annealing at the temperature range less than 1400°C, residual crystalline defects should be effective to expand unit cell. This expansion is generally isotropic in unit cell if the residual defects are only points or point-like defects. As a matter of fact, the lattice parameter change in the c-axis of the present specimens was larger than that in the a-axis [10], and than the value for SiC without He formation, which showed mostly linear decrease against annealing temperature [5,6,12]. On the other hand, plate-like defects such as small dislocation loops can expand lattice anisitropically, as was observed in AlN [13]. Therefore, slight excess expansion of c-axis may be attributed to the formation of the black-dot defects as shown in Fig. 3.



[a]

[b]

Fig. 3 TEM photographs of neutron-irradiated SiC ceramics containing natural B_4C [a] and enriched B_4C [b], irradiated to 2.8×10^{24} n/m^2 (E > 0.1 MeV), annealed for 1h at 700°C.

Above 1100°C, the formation of this defect should be saturated, i.e., the other migration process will proceed. This could be the process of migration of vacancy, which traps He atom and move to grain boundary to form bubbles. The vacancy migration in SiC crystal was reported to be accelerated at around 1200°C [7,8,14]. Above 1400°C, He release from powder was almost diminished except for the specimen containing enriched B_4C, and release from bulk was happened above 1700°C. It should be explained by the interconnection of grain boundary bubbles to escape He outside sintered body [9].

The amount of macroscopic swelling due to the irradiation was large in the specimen containing large amount of ^{10}B, as shown in Fig. 4 at 25°C. The result clearly shows the effect of He formation in the SiC ceramics. Suzuki *et al.* reported that the volume change of neutron-irradiated SiC containing B was slightly larger than these of SiC without B [14]. It should be induced by the larger swelling of B_4C grains, which were existed at grain boundary [15].

The macroscopic length change started to contract from about 300°C, and continued up to about 1200°C. When the annealing temperature increased more than 1300°C, the contraction changed into expansion. The degree of expansion was larger in the specimen containing enriched B_4C. The expansion of length above 1300°C should be attributed to the He bubble formation along grain boundary (Fig. 2), as indicated in the references [9,13].

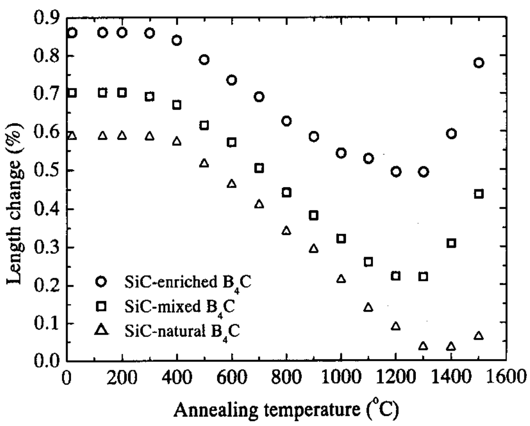


Fig. 4 Length changes of irradiated SiC containing natural B_4C (19.6% ^{10}B), mixed B_4C (56.8% ^{10}B), and enriched B_4C (19.6% ^{10}B), as a function of annealing temperature.

4. Conclusion

The results of this study are summarized as follows;

1) Macroscopic swelling increased with increasing ^{10}B concentration by the irradiation. It expanded by annealing above 1300°C, and has relevance with formation of He bubble along grain boundary.

2) He release started around 600–700°C indicated migration of He atoms. A part of He can form very small black-dot defects into SiC, which was seemed to induce expansion of c-axis length at lower temperature than that of grain boundary bubble formation.

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