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Characterization of β-Silicon Carbide for Potential Use as Irradiation Temperature Monitor

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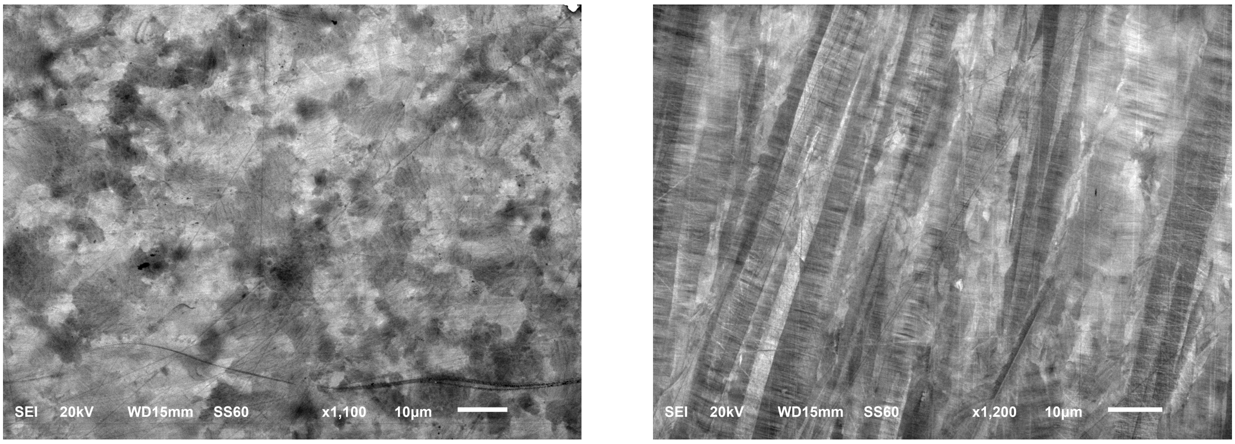
Abstract. Post-irradiation data on the neutron-induced swelling behaviour and resistivity changes in silicon carbide often does not show a clear trend. This makes a quantitative comparison between different studies difficult. To address the diverging results after irradiation in different studies, a thorough reference study is performed on high quality β-silicon carbide. The results show the response to neutron irradiation may be significantly influenced by structural defects present before irradiation. These findings open a way to improve the accuracy of silicon carbide irradiation temperature monitors.

Introduction

SiC temperature monitors are cost effective and easy to use in nuclear reactors, because these monitors require no wires and can be relatively small. During neutron irradiation defects are generated in SiC, which change material properties such as: dimensions and resistivity. These defects can be annealed out of the material at a specific elimination temperature. The annealing out of defects in SiC is accompanied by a (partial) restoration of the material properties to the pre-irradiation conditions. Only defects with an elimination temperature above the irradiation temperature will survive the irradiation process. The irradiation temperature can be retrieved by studying the recovery of material properties. When the annealing temperature exceeds the irradiation temperature, defects start to annihilate and a change in material properties can be observed. The post-irradiation data on irradiation swelling and resistivity, often does not show a clear trend [1, 2, 3, 4, 5]. Until now these studies irradiated the as-received material, and little attention was paid to the initial characterization. This is already recognized by Snead [6], who recommends the use of theoretically dense β-SiC. This study reports on the microstructure and thermal stability of as-received β-SiC material and looks at the effects of a varying irradiation temperature and neutron fluence on the post-irradiation material properties.

Table 1. Irradiation conditions with crystal damage in displacement per atom in steel (dpa).

Campaign	Crystal damage [dpa]	Irradiation temperature [°C]
Nomad	0.025	100
	0.075	100
Motore	0.5	255
		310
		410
	1	255
		310
		410



(a) Topview of a sample with bar geometry. (A)

(b) Sideview of a sample with bar geometry. (B)

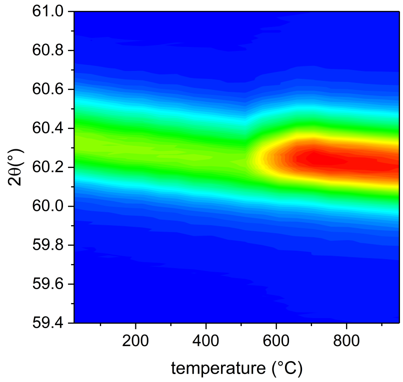
Fig. 1: SEM micrographs of two sides of a SiC bar. Each side with it's own microstructure A or B.

Experimental Methods

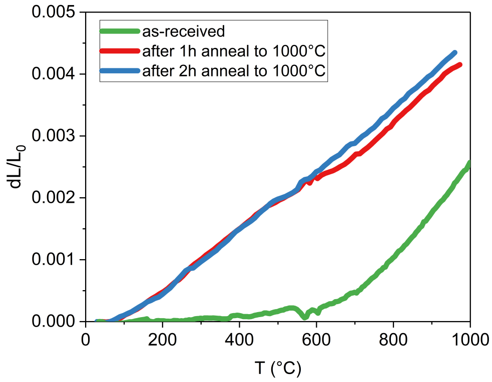
High-density chemical vapour deposited (CVD) β-phase (cubic structure) SiC is used with geometries: bars (1x1x12 mm^3) and discs (Ø5x1 mm^3). The reference study included a microscopic characterization using scanning and transmission electron microscopy (SEM, TEM) before and after annealing treatments up to 1000°C, for 1h at a ramp rate of 1°C/s, under vacuum. Also, the resistivity, length and lattice parameter are measured. To study the influence of irradiation temperature and a varying neutron dose two irradiation campaigns were set-up, using dedicated irradiation capsules, in the Belgian Reactor 2 (BR2) at SCK•CEN in Mol, Belgium. In this study relative low levels of crystal damage are chosen. This introduces exclusively simple point defects during neutron irradiation [7]. The irradiation conditions are listed in Table 1. Irradiation swelling is measured as the dimensional expansion and the lattice expansion of the samples. Dimensional changes are measured with a Mitutoyo PJ300 profile projector, lattice expansion is measured with an X-ray diffractometer using a CuKα source (40kV, 45mA) with a stepsize of 0.02°. In addition to these techniques a NETSCH-DIL402C dilatometer is used to study the thermal expansion in function of annealing temperature. Resistivity is measured using a Jandel Microposition four-point probe with 0.5 mm probe spacing and a load of 10g.

Results

SEM micrographs of SiC bars, in figure 1, show a laminar structure. The left image is taken on the top side of the bar (1a), the right by rotating the bar 90° to obtain a side view (1b). The microstructure of the discs is identical. The grains in the as-received samples have sizes varying between one and ten microns diameter. TEM analysis shows the as received material contains stacking faults. In addition to the microstructure the thermal stability is of great interest. As the material will be irradiated, and annealed afterwards. Any instabilities present before the irradiation, will cloud the measurements taken during, or after post-irradiation annealing. Instabilities are observed as distortions from the expected linear trend in XRD and dilatometry measurements, shown in figure 2. The XRD profile displayed in figure 2a shows the evolution of the (220)-peak with increasing temperature. The figure shows peak broadening, a shift towards higher 2θ values, and a general increase in intensity that is not expected. Identical measurements on other samples showed similar behaviour but at different starting temperatures for the instability. In figure 2b shows the instabilities observed in dilatometry. The observed instabilities are no longer present after annealing for 1h at 1000°C as shown in figure 2b. No changes in XRD or dilatometry are observed in additional measurements at high temperatures or after additional annealing. After annealing TEM micrographs show no changes in the microstructure compared to the as-received microstructure. After irradiation, irradiation swelling is measured using dimensional and

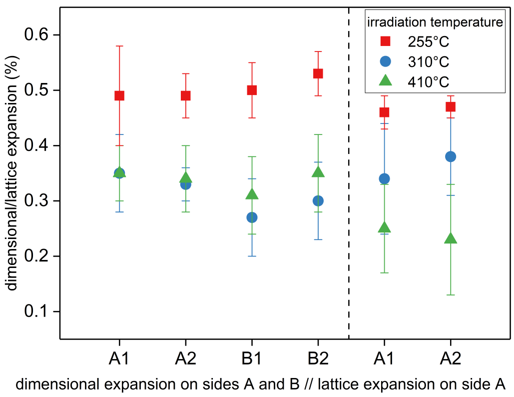


(a) XRD contour of (220) on side A.

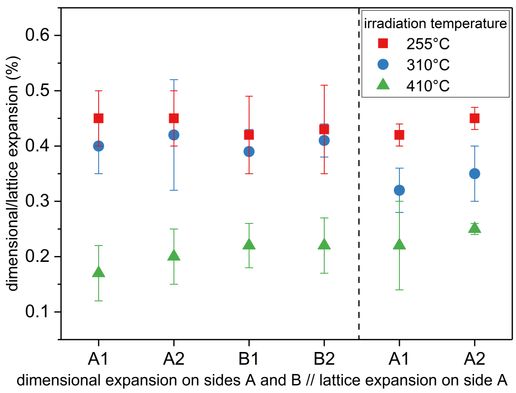


(b) Length changes in function of temperature.

Fig. 2: XRD and dilatometry measurements of the observed instabilities in as-received material. On the right side, dilatometry also shows results after different annealing treatments.



(a) Crystal damage level: 0.5 dpa.



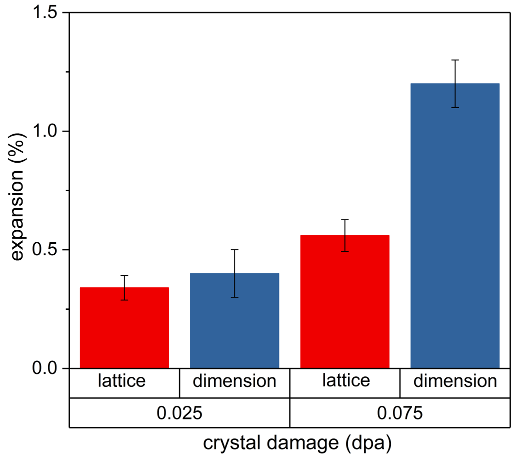
(b) Crystal damage level: 1 dpa.

Fig. 3: Dimensional expansion and lattice expansion measured after neutron irradiation (Motore) on SiC bars. A1 and A2 are the two sides with microstructure A, whereas B1 and B2 stand for the two sides with microstructure B. Dotted lines separate dimensional and lattice expansion measurements.

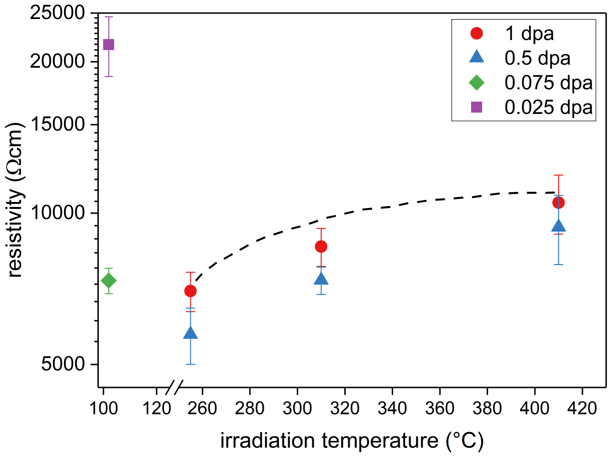
lattice parameters measurements. The lattice parameters are measured on the side with microstructure A. The results of the XRD measurements are displayed in figures 3 and 4a. After 0.5 and 1 dpa of damage (figure 3a and 3b), a decrease in swelling with increasing irradiation temperature was measured. The dimensional and lattice expansion measurements give similar results for 0.5 and 1 dpa. In the Nomad campaign the dimensional and lattice expansion measurements do not give exactly the same results, but they both show an increase in expansion with increasing crystal damage. Post-irradiation resistivity measurements are displayed in function of irradiation temperature for different levels of crystal damage in figure 4b. The Nomad results show a decrease in resistivity with increasing neutron dose. The dotted line shows the trend between the different data points.

Discussion

The thermal stability analysis of as-received β-SiC material shows that due to initial defects, annealing measurements can become scattered and unexpected swelling behaviour appears. The material can be stabilized by annealing to 1000°C for 1h. As annealing eliminates defects in the material, the pre-irradiation structural defects present in the material cause the instabilities that are observed. These defects are not the stacking faults that are introduced during CVD growth. As these defects are still observed after annealing in TEM. In general, it can be concluded from the measurements that with an



(a) Post-irradiation swelling.



(b) Post-irradiation resistivity measurements.

Fig. 4: Post-irradiation swelling data from Nomad and post-irradiation resistivity measurements.

increase in irradiation temperature, a decrease in swelling is observed. Also, at low levels of crystal damage an increase in damage leads to an increase in swelling. At higher levels of crystal damage this is not clearly observed as the level of crystal damage has saturated in this region. The resistivity decreases with an increase in crystal damage, but increases when the irradiation temperature is increased. Resistivity measurements confirm defect saturation above 0.5 dpa, as there are no significant differences at identical irradiation temperatures. The results can be attributed to the effect of annealing out simple point defects which are introduced during neutron irradiation. The higher the amount of neutrons a sample receives, the higher the level of crystal damage, and the more defects. These defects cause the material to swell and act as dopants in the pure SiC matrix, hence decreasing it's resistivity. When the irradiation temperature is increased, part of the simple point defects which are introduced are eliminated. This decrease in defect concentration, decreases the amount of swelling we observe, but it also lowers the concentration of dopants, and thus increases the resistivity.

Conclusion

Until now little attention was paid to the initial characterization of SiC temperature monitors. The response to neutron irradiation may be significantly influenced by structural defects that are present in the material before irradiation. This is confirmed by the pre-irradiation thermal instabilities observed in β-SiC, which is not related to the observed high density of stacking faults. Our findings can explain existing inconsistencies in the published results and open a way for improving the measurement accuracy of these irradiation temperature monitors.

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References

[1] H.Suzuki, T.Iseki, and M.Ito: Journal of Nuclear Materials 48.3 (1973): 247-252

[2] H.Miyazaki, et al.: Journal of Nuclear Science and Technology 29.7 (1992): 656-663.

[3] T.Suzuki, et al.: Fusion technology 27.3 (1995): 314-325.

[4] H.Huang, and N.Ghoniem: Journal of nuclear materials 250.2-3 (1997): 192-199.

[5] Y.Katoh, H.Kishimoto, and A.Kohyama: Journal of nuclear materials 307 (2002): 1221-1226.

[6] L.L.Snead, A.M.Williams, and A.L.Qualls: Effects of Radiation on Materials: 21st International Symposium. ASTM international, 2004.

[7] Y.Katoh, et al.: Journal of nuclear materials 351.1-3 (2006): 228-240.