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Effects of Grain-Boundary Structure on the Strength, Toughness, and Cyclic Fatigue Properties of a Monolithic Silicon Carbide

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An *in situ*-toughened silicon carbide (ABC-SiC) has been examined in the as-processed condition, where the grain-boundary films are predominantly amorphous, and following thermal exposure at a temperature of 1300°C, where the films become fully crystalline. Previous work has shown that, at elevated temperatures (up to 1300°C), once the grain-boundary films crystallize *in situ*, only a marginal reduction in strength, fracture toughness, and cyclic-fatigue crack-growth properties is observed, in compared with those of the as-processed microstructure at 25°C. In the present study, the effect of such crystallization on the subsequent strength, toughness, and fatigue properties at 25°C is examined. Little or no degradation is observed in the room-temperature properties with the crystallized grain-boundary films/phase; in fact, although the strength shows a small reduction (~3%), the fracture toughness and fatigue-crack-growth threshold both increase by ~20%, compared with that of the as-processed structure with amorphous grain-boundary films.

I. Introduction

THE use of commercial SiC for high-temperature structural applications has been limited, to date, by its very low fracture toughness ($K_c \approx$ 2–3 MPa·m$^{1/2}$); however, SiC ceramics with toughnesses as high as 9 MPa·m$^{1/2}$ recently have been processed using the *in situ* toughening route.[1–4] However, maintenance of such good fracture resistance at elevated temperatures has been a problem with this approach; indeed, the development of good low-temperature toughness with correspondingly good high-temperature creep/oxidation resistance often is a compromise in monolithic ceramics. An additional concern with such toughened ceramics is that they can be susceptible to premature failure via cyclic fatigue, even at ambient temperatures. The dependency of the resulting fatigue-crack-growth rates on the applied stress intensities can be quite high; therefore, predicted lifetimes of components manufactured from such ceramics are a very strong function of the applied stresses.[5]

Recent studies[3,4,6,7] on one such *in situ*-toughened SiC that was developed by De Jonghe and co-workers[4] and involves sintering additions of aluminum, boron, and carbon (the so called "ABC-SiC") have indicated that good low-temperature toughness ($K_c \approx$ 6–9 MPa·m$^{1/2}$) and fatigue-crack-growth resistance can be combined with excellent elevated-temperature properties, at least with respect to toughness,[6,7] creep,[7] and fatigue behavior[6] up to 1300°C. Unlike typical behavior in Al_2O_3 and Si_3N_4 ceramics, these properties can be ascribed to the fact that, at temperatures greater than ~1100°C, the amorphous film that is 1–2 nm thick along the grain boundaries fully crystallizes *in situ* or during prior thermal exposure (see Fig.1). Correspondingly, fracture toughness and fatigue-crack-growth rates at 1200°-1300°C are only marginally worst than at room temperature, with no evidence of softening or viscous flow of the grain-boundary phase nor creep cavitation within this phase;[6] moreover, the creep properties up to 1400°C exceed those of commercial SiC (Hexoloy™).[7]

Indeed, the grain-boundary structure of ABC-SiC can be altered (crystallized) by heat treatment. A recent study[8] on the structural evolution in grain boundaries on heating has revealed that, in as-processed ABC-SiC, amorphous grain-boundary films were observed for most (>90%) grain boundaries that have been imaged, whereas crystalline grain-boundary films were observed only occasionally. However, after prolonged heat treatment at 1100º–1300ºC in argon, *all* the imaged grain boundaries exhibited crystallized interlayers; in fact, no amorphous films were observed at any grain boundaries in our transmission electron microscopy (TEM) study. Quantitative X-ray energy-dispersive spectroscopy (EDS) revealed an aluminum- and oxygen-rich composition, in comparison to the SiC matrix; this composition was similar to that of the amorphous films before annealing. The crystallized grain-boundary phase was identified as a 2H-wurtzite structure with an Al(Si)-O-C composition. This 2H-wurtzite structure is very closely related to the 2H-SiC structure, in terms of crystallography and lattice parameters. Therefore, epitaxial 2H-wurtzite grain-boundary films often were formed on the basal-planes of the adjacent α-SiC grains, as illustrated in the "pre-exposed" micrograph in Fig. 1. Because of such structural similarity, it would be difficult to distinguish the grain-boundary films from that of the interior SiC matrix when only a one-dimensional lattice was observed via high-resolution transmission electron microscopy (HRTEM) (see the pre-exposed micrograph in Fig. 1). Nevertheless, high aluminum and oxygen concentrations in these grain boundaries confirm the existence of the films. Actually, when the imaging zone axis is suitable to reveal two-dimensional lattice, the crystallized boundary films can be recognized by different stacking sequence of the atomic layers, compared with those of the 4H- or 6H-SiC matrix. Details of the characterization of grain-boundary structural evolution on heating is described elsewhere.[8]

Complete crystallization of the grain-boundary films at elevated temperatures (the pockets at triple junctions are already crystalline after processing) is clearly highly beneficial to the subcritical crack-growth properties at high temperatures, as was shown for ABC-SiC by Chen *et al.*,[6] with respect to toughness and cyclic-fatigue resistance. However, the presence of amorphous grain-boundary films is considered to promote intergranular fracture, which, in turn, leads to grain bridging, which is the basis of the low-temperature toughening in ABC-SiC (and other monolithic ceramics, such as coarse-grained Al_2O_3 and grain-elongated

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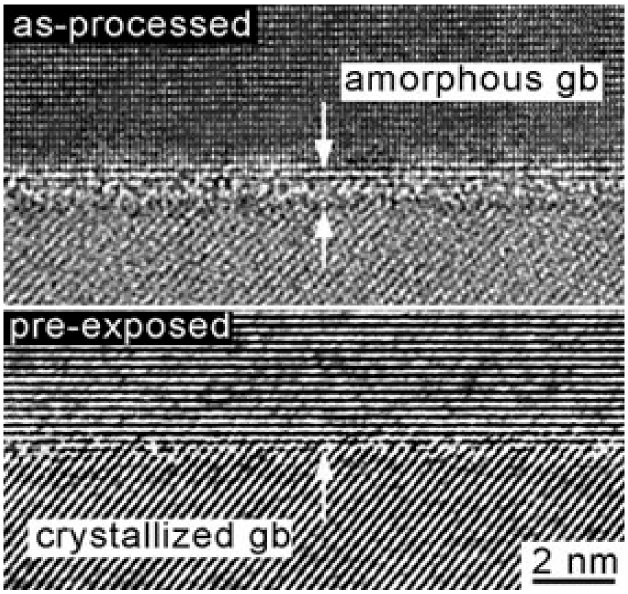

Fig. 1. HRTEM micrographs of the grain boundaries in ABC-SiC (as-processed and pre-exposed at 1300°C, showing amorphous and crystallized grain-boundary structures, respectively).

Si_3N_4); therefore, concern about whether the crystallized grain-boundary films can compromise the subsequent low-temperature mechanical properties become manifest. Accordingly, the objective of the present study is to clarify the effect of crystallization of the grain-boundary films on the *room-temperature* properties of ABC-SiC, specifically with respect to strength, fracture toughness, and cyclic fatigue-crack growth resistance.

II. Experimental Procedures

Monolithic ABC-SiC—with a composition of SiC-3Al-0.6B-2C (by weight percent)—was processed via hot pressing at 1900°C for 1 h under a pressure of 50 MPa. The as-processed microstructure consists of 5 vol% β–phase (3C polytype) and 95 vol% α–phase (4H, 6H) in the form of elongated platelike grains (~5.2 µm long, ~1.5 µm wide); aluminum- and oxygen-containing amorphous films 1–2 nm thick were formed along grain boundaries, with strips of crystalline phase ($Al_8B_4C_7$, Al_4O_4C, Al_2O_3) at the triple junctions.[3,4,6] The mechanical properties of this as-processed microstructure (hereafter referenced as "amorphous gb") were compared to the same material following thermal exposure for 85 h in flowing gaseous argon at 1300°C. The microstructure of the pre-exposed material (hereafter referenced as "crystallized gb") was identical to that of the as-processed condition, with the notable exception that the glassy grain-boundary films/phase was fully crystallized (see Fig. 1).

Fatigue testing was performed using disk-shaped compact-tension DC(T) specimens (28 mm wide, 3 mm thick), which were cycled at a loading frequency of ν= 25 Hz (sinusoidal wave) at a load ratio (ratio of minimum load to maximum loads) of R = 0.1. Following completion of the fatigue tests, specimens were monotonically loaded to failure, to measure the resistance curve (R-curve) and the fracture toughness (K_c). The strength was evaluated in four-point bending beam specimens with dimensions of 3 mm × 4 mm × 30 mm, the major and minor spans were 25.4 and 10.2 mm, respectively. Additional details of these test procedures are described elsewhere.[3,4,6]

III. Results and Discussion

The effects of high-temperature exposure on the flexure strength and fracture toughness of ABC-SiC are listed in Table I.

Table I. Fracture Toughness and Strength in As-Processed ("amorphous gb") and 1300°C Pre-exposed ("crystallized gb") ABC-SiC

Sample	Fracture toughness (MPa·m$^{1/2}$)		Strength at 25°C (MPa)
	25°C	1300°C	
As-processed	6.2	4.3	620 ± 15
Pre-exposed	7.2	5.2	604 ± 17

Crystallization of the grain-boundary glassy phase apparently causes little or no degradation in the mechanical properties. Although the room-temperature strength is reduced marginally (by ~3%), the toughness actually increases, at both 25° and 1300°C, by ~ 20%. Similarly, no degradation in the room-temperature fatigue-crack-growth-rate properties is observed (see Fig.2); in contrast, the fatigue threshold (ΔK_{th}) (and, hence, the entire growth-rate curve) are increased by ~20%, with no visible change in slope (which is given as the exponent m in the Paris law: $da/dN = C'\Delta K^m$).

Scanning electron microscopy (SEM) micrographs of the trajectories of the fatigue cracks at 25°C in both the as-processes (amorphous gb) and pre-exposed (crystallized gb) microstructures, which are shown in Fig. 3, indicate that the crack paths in both structures are fully intergranular, with evidence of shielding from grain bridging in the crack wake. Corresponding fatigue fracture surfaces (shown in Fig. 4) exhibit extensive debris from wear and abrasion of the crack faces during load cycling; this phenomenon is characteristic evidence of ceramic fatigue in the form of a cyclic-induced degradation of the grain-bridging zone. Clearly, the ambient-temperature toughening mechanism of grain bridging, and the associated cyclic-fatigue mechanism (which results from the progressive degradation in such bridging), apparently are unchanged by the crystallization of the grain-boundary films.

The reason why crystallization of the grain-boundary films should lead to a slight improvement in the toughness and fatigue resistance at room temperature is unclear. However, as stated previously, the mechanisms of both fracture and fatigue are associated with grain bridging, as a result of the frictional tractions that are generated via contact between opposing crack faces.[9] The pullout resistance, which is represented by the bridging stress, acts to reduce the near-tip crack-driving force for crack extension; its magnitude is linearly proportional to the frictional coefficient between the sliding grain faces.[10] Crystallization of the grain-boundary phase is expected to increase the frictional coefficient, and the resulting increase in the bridging stress may well provide the origin of the enhanced grain bridging.

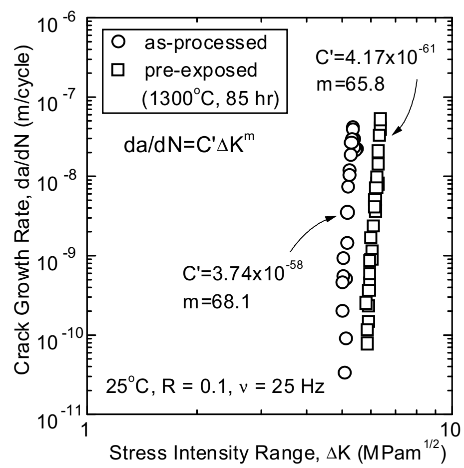

Fig. 2. Comparison of cyclic-fatigue crack-growth rates (da/dN) at 25°C for the as-processed (amorphous grain boundary) and 1300°C pre-exposed (crystallized grain boundary) microstructures in ABC-SiC, as a function of the applied stress-intensity range (ΔK), with R = 0.1 and ν = 25 Hz.

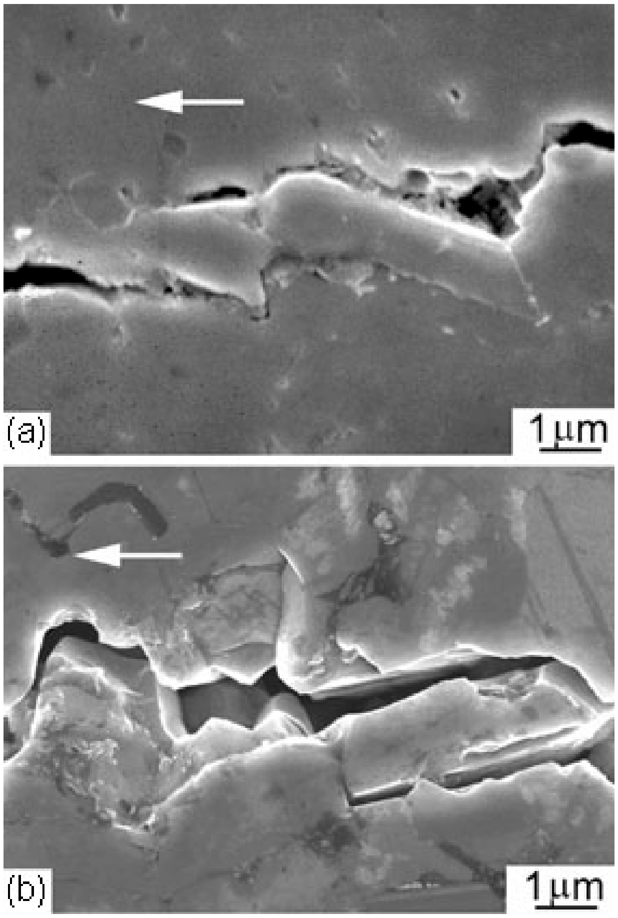

Fig. 3 SEM micrographs of crack trajectories for (a) the as-processed (amorphous grain boundary), and (b) the 1300°C pre-exposed (crystallized grain boundary) microstructures in ABC-SiC, developed during room-temperature fatigue crack-growth, with R = 0.1 and ν = 25 Hz. Arrow indicates the direction of crack growth.

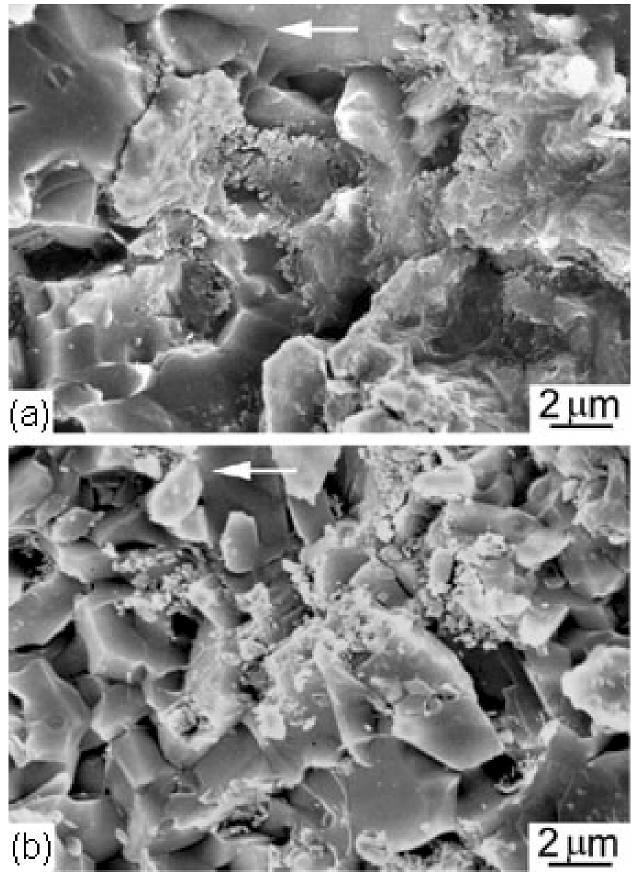

Fig. 4 SEM micrographs of fracture surfaces for (a) the as-processed (amorphous grain boundary), and (b) the 1300°C pre-exposed (crystallized grain boundary) microstructures in ABC-SiC, developed during room-temperature-fatigue crack-growth, with R = 0.1 and ν = 25 Hz. Arrow indicates the direction of crack growth.

IV. Summary and Conclusion

The present work has shown that the crystallization of the grain-boundary films/phase at temperatures of ≥1100°C, which is known to provide good strength, toughness, and resistance to cyclic fatigue and creep in silicon carbide with aluminum, boron, and carbon as sintering aids at elevated temperatures up to 1300°C,[6,7] causes no degradation in the mechanical properties when tested subsequently at room temperature. In fact, in comparison with the as-processed microstructure with amorphous grain-boundary films, the fracture toughness and cyclic-fatigue crack-growth resistance (characterized by the fatigue threshold) actually are ~20% larger, with only a marginal loss (~3%) in strength. Although such crystallization would occur *in situ* during service at elevated temperature, for operation at ambient-temperature, prior heat treatment, similar to that in metallic materials, would seem to provide an effective means of improving the mechanical properties of the ceramic.

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