The structure of slow crack interfaces in silicon nitride

N. J. TIGHE
National Bureau of Standards, Center for Materials Science, Washington, DC 20234, USA

Fracture interfaces formed in silicon nitride at high temperatures were studied using light and electron microscopy. The structure of the fracture interface depended on the type of silicon nitride fractured. High-purity, reaction-bonded silicon nitride always formed flat, relatively featureless, fracture surfaces. Fracture occurred by a brittle mode even at the highest temperature (1500 °C) studied. The critical stress intensity factor for reaction-bonded silicon nitride (~ 2.2 MN m⁻³/²) is relatively low and is insensitive to temperature. By contrast, hot-pressed silicon nitride gave evidence of plastic flow during fracture at elevated temperatures. Crack growth in magnesia-doped, hot-pressed silicon nitride occurs by creep, caused by grain boundary sliding and grain separation in the vicinity of the crack tip. As a consequence of this behaviour, extensive crack branching was observed along the fracture path. The primary and secondary cracks followed intergranular paths; sometimes dislocation networks, generated by momentary crack arrest, were found in grains bordering the crack interface. As a result of the high temperature, cracks were usually filled with both amorphous and crystalline oxides that formed during the fracture studies. Electron microscopy studies of the compressive surfaces of four-point bend specimens gave evidence of grain deformation at high temperatures by diffusion and dislocation motion.

1. Introduction
Hot-pressed and reaction-bonded silicon nitride materials are being developed for use in gas turbine engines which have service temperatures as high as 1400 °C. The strength and fracture toughness data obtained for these materials show that most currently available compacts may not be useful because their strength degrades during exposure to such temperatures [1–7]. In order to improve the high temperature behaviour, it is essential to understand why the strength and fracture toughness degrade during high temperature exposures and why the two types of silicon nitride behave differently.

Although the strength of magnesia-doped hot-pressed silicon nitride (HPSN) is two to three times greater than reaction-bonded silicon nitride (RBSN) at 25 °C, the strength of reaction-bonded silicon nitride changes little with exposure to high temperature, while that of hot-pressed silicon nitride degrades at temperatures above ~1200 °C. At 1400 °C the strength of both materials is ~250 MPa. From measurements of crack velocity and fracture toughness Lange [5], and Evans and Wiederhorn [4] correlated the strength degradation of magnesia-doped hot-pressed silicon nitride with the onset of slow crack growth. They suggested that slow crack growth occurred as a result of plastic deformation or stress corrosion in the neighbourhood of the crack tip. Reaction-bonded silicon nitride did not exhibit slow crack growth under similar test conditions.

In this paper we characterize fracture and slow crack growth and the strength degradation in terms of the microstructural changes which occurred in 4-point bend and double torsion specimens during experiments at 1400 °C. The fracture interfaces and the regions near fracture interfaces in these
specimens were examined by light and electron microscopy. Earlier microscopy studies [8, 9] showed that hot-pressed silicon nitride was composed primarily of blocky $\alpha$-$\text{Si}_3\text{N}_4$ grains ($\sim 1 \mu\text{m}$ diameter) and column-like $\beta$-$\text{Si}_3\text{N}_4$ grains ($\sim 1 \mu\text{m} \times 5 \mu\text{m}$). In addition, it contained micron-sized grains of $\alpha$- and $\beta$-$\text{SiC}$, and $\text{Si}_2\text{N}_2\text{O}$. Furthermore, glass was occasionally found at triple junctions and as micron-sized regions. The microstructure is therefore heterogeneous with respect to composition. The reaction-bonded silicon nitride [10] was composed of ultra-fine $\alpha$-$\text{Si}_3\text{N}_4$ grains ($\sim 0.02 \text{ nm}$) and larger $\beta$-$\text{Si}_3\text{N}_4$ and $\text{SiC}$ grains ($\sim 1$ to $5 \mu\text{m}$). The $\text{SiC}$ phases occurred in both materials because they were hot-pressed or reaction-bonded in a carbonaceous atmosphere. Both materials oxidized at the test temperature; a complex crystalline and amorphous oxide layer was formed on the surface [11].

2. Experimental procedure

2.1. Materials

The hot-pressed and the reaction-bonded silicon nitride materials examined in this study are currently under consideration for turbine engine applications. Two hot-pressed compositions* (HS130 and NC132) of magnesia-doped $\text{Si}_3\text{N}_4$ were obtained in the form of billets $75.5 \text{ cm} \times 75.5 \text{ cm} \times 2.5 \text{ cm}$. The manufacturer listed the following impurities (wt%): 0.20 $\text{Al}$, 0.05 $\text{Ca}$, 0.40 to 0.64 $\text{Fe}$, 0.70 to 0.74 $\text{Mg}$, 2.6 to 2.7 $\text{W}$. Two sets of reaction-bonded silicon nitride specimens (NC350) were obtained as plates $7.6 \text{ cm} \times 2.5 \text{ cm} \times 0.2 \text{ cm}$. Test specimens were cut from the billets and ground into plates $7.6 \text{ cm} \times 2.5 \text{ cm} \times 0.2 \text{ cm}$ for double-torsion tests and into bars $5.5 \text{ cm} \times 1.0 \text{ cm} \times 5.5 \text{ cm}$ for 4-point bend tests. The double-torsion specimens were grooved along their length by sawing through approximately 1/2 the plate thickness. The specimens were notched at one end of the groove.

2.2. Mechanical testing

Stress intensity factors and crack velocity data were obtained on the double-torsion specimens using the procedure for propagating a controlled crack at high temperature as used by Evans [7]. In this method, the grooved and pre-cracked double-torsion specimens (Fig. 1) were held in a silicon carbide jig with the grooved side up. The load was applied at a constant displacement rate to a ram, which straddled the central notch at $P$. The load was increased until the critical stress for crack initiation occurred and the crack propagated along the groove at constant load. Displacement rates from $5 \times 10^{-5}$ to $5 \times 10^{-2} \text{ cm/min}$ were used. The stress intensity factors were calculated from the load and specimen dimensions. The crack velocity was obtained from measurements of the crack length as a function of time at constant load. Very low crack velocities were obtained by applying a constant load for fixed time intervals up to 48 h. Crack lengths that were measured using a fluorescent dye penetrant were in good agreement with measurements of the crack tip displacement after the plate was broken into two parts.

2.3. Microscopy

Transverse and parallel sections of the test specimens were examined using light and electron microscopy. Specimens were prepared for transmission electron microscopy by standard ion thinning procedures [12]. In this method, disks 3 mm in diameter were cut from polished thin plates 50 to 100 $\mu\text{m}$ thick, using a diamond core drill. These disks were then polished to a thickness of approximately 50 $\mu\text{m}$ and ion-thinned using argon ions. Copper single-hole grids were cemented onto the specimens with epoxy cement to strengthen and to support the thin foil. The thin foils were examined in an electron microscope operating at 200 kV. The double-torsion specimens and the bend specimens were examined with a light microscope in

*The $\text{Si}_3\text{N}_4$ materials were obtained from the Norton Co., Boston, Mass, USA.