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Effect of microstructure and grain boundary chemistry on slow crack growth in silicon carbide at ambient conditions

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Abstract

Silicon carbide (SiC) is being used increasingly as a room temperature structural material in environments where moisture cannot always be excluded. Unfortunately, there have been almost no reports on slow crack growth (SCG) in SiC at room temperature. To address this gap, SCG in SiC was studied using constant stress rate and double torsion tests in water. SiC based materials were produced with a wide range of grain boundary chemistries and microstructures, which may affect their slow crack growth behaviour. To clarify the role of chemistry and microstructure respectively, solid state (SS) sintering with carbon and boron along with liquid phase (LP) sintering using oxides additives were used to produce materials with fine and coarse grains. The LP-SiC was three times more sensitive to SCG than SS-SiC materials. Moreover, the larger grained material with a higher toughness was less sensitive to SCG than the materials with fine grains.

Keywords: Slow crack growth; SiC; Toughness; Grain morphology

1. Introduction

Silicon carbide has attracted much attention as a candidate material for high temperature applications. Therefore there is a wide body of research on the mechanical behaviour of SiC at elevated temperature and its interaction with several combustion environments. It is well known that in these environments, the oxidation of SiC causes slow crack growth [1–7].

However, SiC's properties such as high specific stiffness, low thermal expansion and high thermal conductivity, make it an interesting structural material for many technologies that do not involve high temperatures. For example, silicon carbide is playing an increasingly more prominent role in the scientific missions of the European Space Agency. A prime example is the 3.5 m diameter silicon carbide main mirror on ESA's Herschel telescope [8]. Such instruments tend to be manufactured over relatively long periods, and components sometimes need to be stressed for prolonged times on earth in environments where moisture cannot always be excluded. Moreover, SiC is becoming a potential candidate for biomedical applications such as orthopaedic implants [9,10] and for mechanical seals in hydraulic systems [11], where the components are stressed for extended periods of time in wet environments.

Unfortunately, there have been almost no studies of the SCG in SiC at room temperature. In itself this is not surprising as the covalent bonds of silicon carbide should make it immune to corrosive attack by water [12,13]. However, all practical silicon carbides are sintered using a diverse range of additives and it is well documented that these additives modify the grain boundary chemistry [14,15]. As cracks tend to propagate through grain boundaries, their chemistry is likely to have large influence on environmentally assisted failure [16]. In addition to additives, different thermal treatments may lead to a wide range of microstructures that can also affect slow crack growth [13].

The aim of this paper is to quantify the effect of grain boundary chemistry and microstructure on the sensitivity of SiC to SCG. Two typical silicon carbide materials were considered: a solid state sintered material (SS-SiC) with additions of boron and carbon and a liquid phase sintered material (LP-SiC) with additions of alumina and yttria. To determine the effect of

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microstructure, we studied a fine grain and a larger grain material for each chemistry. The results provide guidance for the selection of materials and the fabrication of silicon carbide parts designed to work under wet environments.

2. Experimental methods

2.1. Material processing

For the SS-SiC materials, 3 wt% of carbon and 0.2–0.5 wt% boron (Grade II, H.C. Starck, Germany) was used [17]. Fine grains were achieved by hot pressing α-SiC (UF-25, H.C. Starck, Germany) at 2050 °C for 30 min with 0.2 wt% boron. After several trials, microstructures with elongated grains were obtained by increasing the boron content to 0.5 wt% and hot pressing β-SiC (BF-17, H.C. Starck, Germany) at 2150 °C for 3 h. The carbon was added in the form of a phenolic resin (CR-96, Novolak, Crios Resinas, Brazil) with a 50% carbon yield after pyrolysis at 400 °C for 1 h in an argon atmosphere.

For the LP-SiC materials, a mixture of 6 wt% of alumina (AKP-30, Sumitomo, Japan) and 4 wt% yttria (Grade C, H.C. Starck, Germany) was used as sintering additives. The fine equiaxed-grain material was produced by hot pressing α-SiC at 1950 °C for 30 min, whereas the larger grains were obtained by hot pressing β-SiC at 2050 °C for 3 h [18–21].

The SiC powder and additives were wet mixed using methyl ethyl ketone (VWR, London, UK) for 24 h using 10 mm diameter milling media (Union process, Akron, USA) of either alumina for batches containing oxide additives or silicon nitride for batches with non-oxide additives.

The slurries were dried by rotary evaporation (R-20 BUCHI Rotavapor, Switzerland). After drying, the powders were crushed and sieved through a 100 μm sieve. Hot pressing was conducted in 80 mm diameter graphite dies at heating rate of 10 °C min^{-1} under flowing argon gas in a graphite furnace (FCT, Rauenstein, Germany) under a pressure of 25 MPa.

2.2. Characterization

The density was determined by Archimedes method with distilled water as the immersion medium according to ASTM standard C8300-00 [22]. The densities were compared to a theoretical density value of 3.28 and 3.21 g/cm^3 for the LP-SiC and SS-SiC materials respectively.

The microstructure was examined using scanning electron microscopy (SEM, S-3400N, Hitachi, Japan). The specimens were polished to 1 μm using diamond suspension and chemically etched with boiling Murakami's solution [23]. The average grain size of 300 grains was calculated using the linear intercept method [24]. For the large grain samples, the average grain length and width was calculated after measuring 300 grains.

The structures and chemistries of the grain boundaries were characterized by Transmission Electron Microscopy (TEM). Cross-sectional TEM foils were prepared by focused ion beam (FIB) milling (Helios NanoLab 600). TEM work was carried out on a FEI Titan 80-300 S/TEM operated at 300 kV. Energy dispersive X-ray spectroscopy (EDS) and electron energy loss spectroscopy (EELS) were performed in STEM mode, and the incident angle α and collection angle β for EELS acquisitions were ~10 and ~14 mrad, respectively.

The Young's modulus was measured by means of the impulse excitation method [25]. The resistance to fracture was measured in two ways. Firstly, toughness measurements in three point bending were performed according to ASTM 1421 using the single notched edge beam (SENB) method [26] with root notch radius of 15 μm. Then *R*-curve measurements were carried out *in situ* in the SEM (vacuum level of 1×10^{-3} Pa) using a constant moment double cantilevered test rig, built in line with the set-up proposed by Sorensen et al. [27]. The silicon carbide specimens were 65 × 10 × 5 mm. One side of the samples was polished to facilitate the observation of the crack tip. The speed of the stage motor was 0.1 mm min^{-1}. Every time crack propagation was observed, the applied load was recorded and a SEM micrograph was taken.

Slow crack growth (SCG) testing was carried out using both the constant stress rate test in four point bending [28] and the double torsion test [29]. For constant stress rate testing, the specimens measured 4 × 3 × 40 mm. The tensile face was polished with 1 μm diamond suspension and all the edges were bevelled to eliminate stress concentrations. A 2 kg Vickers indent was placed in the centre of the tensile face with the indent orientated such that two of the cracks emanating from the corners created a crack perpendicular to the applied tensile stress. The indentations were made in air with a 10 s holding time, immediately prior to starting the bending test the samples were immersed in distilled water. The fracture stress was measured with an inner span of 10 mm and an outer span of 20 mm at cross head speeds ranging from 0.001 to 1 mm min^{-1}. For every test, the fracture force and time were recorded in order to calculate the fracture stress and stress rate. To determine the exponent n of the SCG law, the following relationship was used [12]:

$$v = AK_I^n \tag{1}$$

where, v is the crack velocity, K_I is the applied stress intensity factor, A and n are material and environment dependent SCG parameters. The slope of log fracture stress versus the log stress rate, n' was converted to n using [30]:

$$n = \frac{4n\prime - 2}{3} \tag{2}$$

All fractured samples were checked after the test to confirm that the fracture occurred because of the indent. A total of five specimens were tested at each stress rate. For comparison, one set of indented samples were tested in mineral oil with silica desiccant balls with an estimated humidity of less than 1% at a stress rate of 1 mm min^{-1}.

For double torsion testing, specimens were 40 × 20 × 2 mm polished with 1 μm diamond suspension. A notch 10 mm long (a_0) with a root radius of 0.3 mm was introduced using a diamond blade. At the end of the notch, two Vickers indentations of 10 kg each were placed in order to obtain a well-aligned sharp crack. A sharp pre-crack length (a_i) of 12–13 mm was achieved by loading the specimen slowly at a cross head speed of 0.05 mm min^{-1} followed by fast unloading once the crack