JOURNAL OF MATERIALS SCIENCE 6 (1971) 324–331

Temperature Dependence of Fracture Effects in Self-bonded SiC

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The effect of temperature on the fracture strength and work of fracture of a self-bonded SiC has been measured from room temperature to 1100° C. Over the same temperature range the work of fracture was observed to increase twofold. These phenomena were accounted for in terms of the behaviour of the free-silicon phase. In addition, electron microscopy of the fractured surface was undertaken. Fracture chips showed that dislocations were generated during the failure process in both the secondary SiC and silicon phases. Stacking faults were observed in the SiC phases, and some of these were shown to have formed during the fracture process.

1. Introduction

The use of higher temperatures in nuclear reactors necessitates the continual search for materials capable of withstanding the demanding conditions and environments. A number of low neutron absorption cross section ceramics have been considered [1] and silicon carbide has been reviewed favourably. Since radiation swelling in SiC is known to saturate [2] out at low fluences and because of its high strength, it has been suggested for use in nuclear fuel applications [3].

The absence of ductility in a ceramic causes some difficulty in determining mechanical properties. In the case of silicon carbide, certain aspects of fracture behaviour have been investigated [4]. Fracture has been shown to be a time dependent phenomenon [5], analogous to static fatigue in oxides, and to be related to damage previously introduced by impact [6]. Impact damage has also been shown to have an effect, but to a lesser extent, on fatigue behaviour [7]. Thus, although the mechanical properties have been reasonably documented, little or no information is present in the literature on fracture mechanisms. At the same time the main part of the investigations referred to have been carried out at room temperature. Thus in this paper the effect of temperature on mechanical properties and fracture mechanisms has been studied.

2. Experimental Techniques

2.1. Mechanical Testing

Specimens 5 × 0.6 × 0.3 cm were machined using diamond impregnated wheels from a large bar of the material.* Care was taken to machine all opposite faces on the specimen parallel. The specimens were fractured under four-point loading using 0.6 cm diameter Lucalox rods as anvils, the loading device being designed so that axial loading was automatically generated (fig. 1).

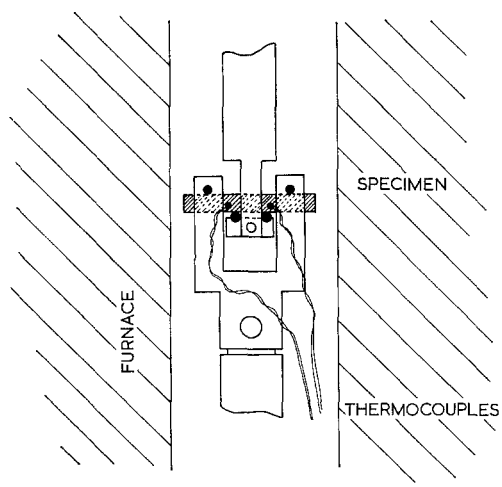

Figure 1 Self aligning four-point bending jig used under tensile conditions.

*Self-bonded silicon carbide, trade name KT silicon carbide

The temperature was monitored by two chromel-alumel thermocouples attached to the specimen. The whole assembly was situated inside a split furnace capable of reaching a temperature of 1200° C, and the temperature controlled by a saturable reactor type controller via a Pt/Pt-Rh thermocouple. The temperature variation aross the bend specimen was less than 2° C and the temperature control was better than $\pm$ 2° C. Before testing, each specimen was allowed to equilibrate at the testing temperature for 30 min. The load was applied at a constant strain rate of 2.5×10^{-3} cm/sec by means of an Instron tensile testing machine.

Rectangular work of fracture specimens 2.5 $\times$ 0.5 $\times$ 0.5 cm were also cut from the same material. A "Vee"-shaped notch was cut into the bar so that in the process of fracturing, the crack was stable and fracture did not occur catastrophically. Three-point loading was used, the centre loading point being at the base of the notch as shown in fig. 2. The apparatus consisted of high density alumina push rods supporting Lucalox anvils mounted in the same furnace as was used for the four-point bending experiments. The alumina push rods were mounted on a base plate such that any alignment necessary could be made with four screws situated on the walls of the base plate as shown in the lower part of fig. 2. The whole assembly was then mounted in the Instron tensile testing machine.

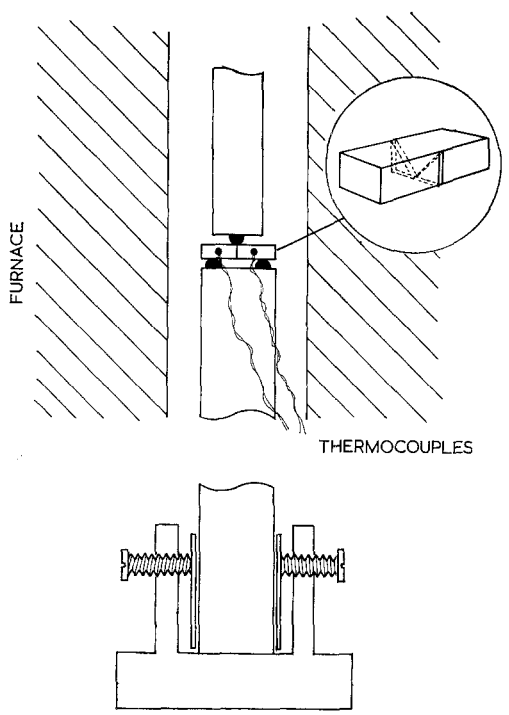

Figure 2 High temperature work of fracture jig. Any alignments necessary can be made with the screws in the base of the apparatus.

2.2. Electron Microscopy

Unshadowed two stage plastic-carbon replicas were made of the fractured SiC surfaces, and the fracture chips found on the replica were carefully examined. The first replica taken was used to obtain the maximum number of fracture chips, whereas usually it is discarded. The replicas were then examined in an Hitachi HU200 electron microscope at 200 kV. The replicas were held in a specimen stage capable of $\pm$ 30° tilts in two directions at right angles, and the fracture chips examined using transmission techniques.

Sections of the fracture surface were cut from the test specimen using diamond impregnated wheels. The fracture surface of these specimens were then vapour coated with a 100 Å layer of gold and examined directly in a Stereoscan scanning electron microscope.

3. Mechanical Properties

3.1. Fracture Strength

The results of the four-point bend tests are best shown graphically, the effect of temperature on the maximum outer fibre stress at fracture being illustrated in fig. 3. Each point is the mean of ten tests, apart from the results at room temperature and 700° C when thirty tests were carried out. The bars about the points represented one

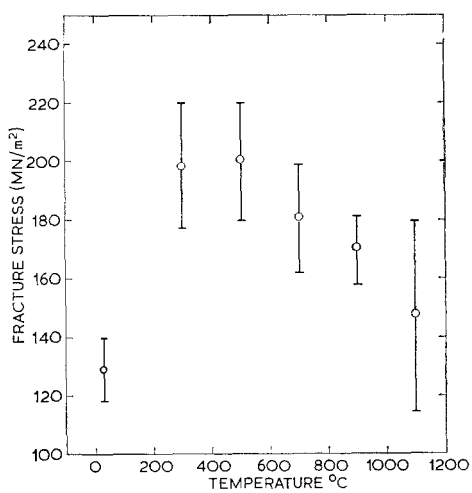

Figure 3 Temperature dependence of the fracture stress under four-point loading.

standard deviation about the mean, so that it is apparent that there is significant scatter in results over the whole temperature range. An important feature of the data is the increase in strength above room temperature and the gradual decrease in strength above 500° C.

3.2. Work of Fracture

The temperature dependence of the work of fracture is shown in fig. 4, each point representing the mean value of six tests. The value obtained at room temperature is less than reported previously [8] and is a consequence of a modification to the notch shape. It has been shown that the work of fracture decreases to a limiting value on lowering the notch height [9], and for the results reported here all notches were cut to two-thirds of the beam height.

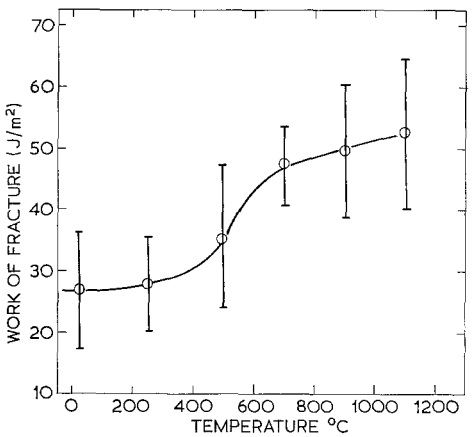

Figure 4 Temperature dependence of the work of fracture.

Of particular interest is the rise in work of fracture over the temperature range 400 to 600° C, the average value increasing by approximately 100%. No distinct differences in the work of fracture curves were observed until testing took place at 1100° C, when the load was found to increase after the initial crack had propagated, thus lowering the applied load. This behaviour, which is shown diagrammatically in fig. 5, was repeated several times before fracture was complete, indicating crack stopping and starting was taking place.

4. Results and Discussion

4.1. Metallography

The microstructure of a typical area of self-bonded silicon carbide is shown in fig. 6. The white etching phase amounting to approximately 12 vol %, consists of free silicon. The light grey angular shaped areas are the primary grains of silicon carbide, with the darker etching secondary silicon carbide forming a complete layer between them and the free silicon. In addition to these phases there are nearly always small amounts of free graphite and porosity present.

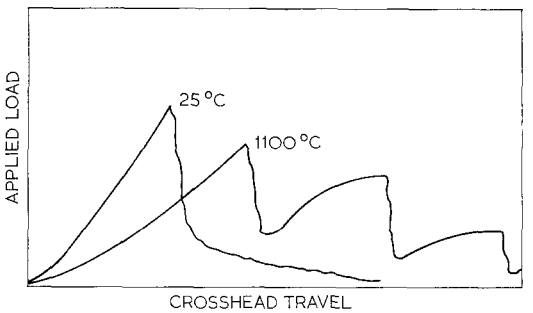

Figure 5 Diagrammatic representation of work of fracture curves at room temperature and 1100° C.

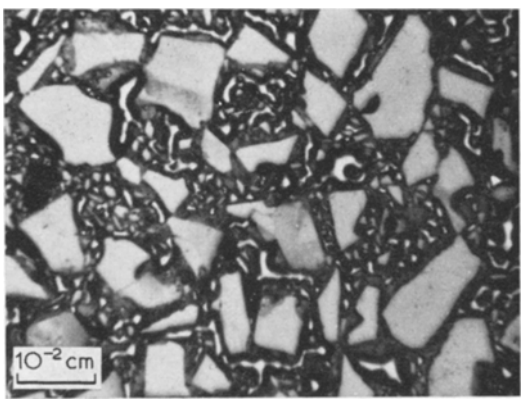

Figure 6 Optical micrograph of self-bonded silicon carbide. Electrolytically etched in saturated KF + 2% HF (× 150).

4.2. Electron Microscopy

Examination of chips of material on replicas taken from the fracture surfaces of the SiC showed a limited amount of dislocation movement had occurred over the whole temperature range. Such dislocation movement was observed in both the free silicon and β SiC phases. Deformation in silicon has been investigated by Sylwestrowicz [10] and the electron microscopy summarised by Alexander and Haasen [11]. The electron microscopy of fracture chips of β SiC has been described earlier [12] where it was

shown that the Burgers vector of the dislocation $\mathbf{b} = a/2\ (1\,1\,0)$ and the slip plane (1 1 1).

Generally any α SiC chips examined were too thick for transmission to occur, and were only transparent near the chip edge, which precluded detailed examination. However, these α SiC chips were seen to contain large numbers of contrast fringes typical of stacking faults or microtwins (fig. 7), which in the micrograph can be seen intersecting a grain boundary. It can be seen that the fringes extend through the fracture chip and that the bounding partial dislocations at the edge of the fringe are not visible. Together with the absence of any slip traces, this would appear to indicate the faults are grown in rather than introduced during the fracture process.

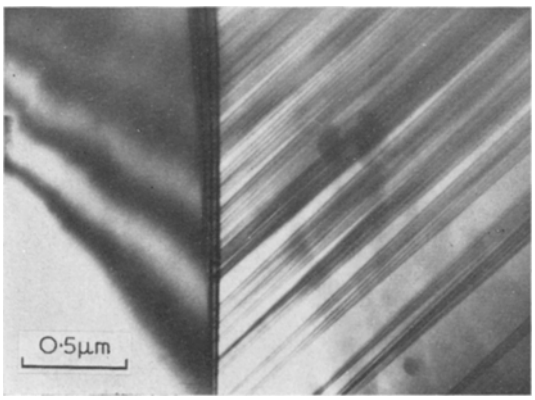

Figure 7 Transmission electron micrograph showing stacking fault fringes intersecting a grain boundary in a fracture chip of α SiC. Thickness contour fringes can be seen in the other grain.

Generation of stacking faults appears to have taken place in the secondary SiC phase during the fracture process at 300° C, as can be seen in fig. 8. The partial dislocations bounding these stacking faults are shown in fig. 9, individual stacking faults and their partial dislocations being readily identifiable by the many markers in the fracture chip. Also of interest in the micrograph are the two types of slip traces, faint ones which are difficult to see, and which could be due to the oxide film [13] and a strong trace possibly due to a dislocation lying beneath the oxide foil.

At higher temperatures (900° C) tangles of dislocations are readily observed in the fracture chips of SiC, which often appear to be associated with particles of foreign matter. A fracture chip of SiC examined from a 1100° C test specimen

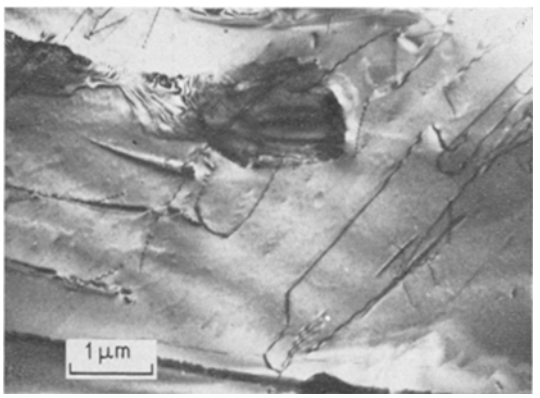

Figure 8. Transmission electron micrograph showing stacking faults believed to have been generated during the fracture process. The stacking faults lie along the $\langle\bar{1}01\rangle$.

also showed dislocation tangles, and in this particular case (fig. 10) individual dipoles and jogs can be readily distinguished. This would suggest that at the higher temperatures the Peierls force in SiC is lowered sufficiently for slip to be taking place on a number of systems, with the resulting dislocation interaction, under the action of the stress concentrations occurring near the crack tip [12].

Fracture chips of silicon were not as prevalent as the SiC chips; however, dislocations were seen in the silicon phase at all test temperatures, usually associated in tangles (fig. 11). Occasionally hexagonal networks were found in the chips (fig. 12), usually above 500° C. These could either be due to recovery of dislocation tangles formed during the fracture process or be intrinsic sub-

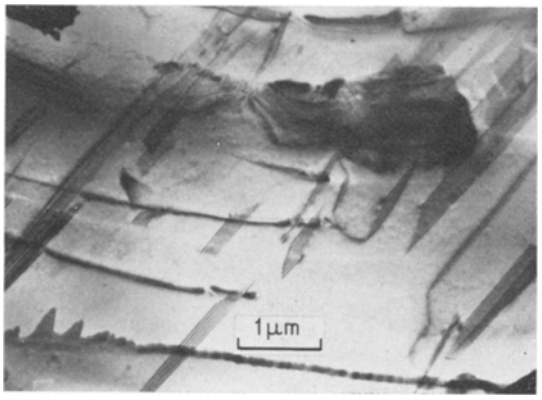

Figure 9 Partial dislocations bounding the stacking faults in fig. 8, showing a typical wavy appearance. The long unit dislocations lie along the $\langle\bar{1}01\rangle$.

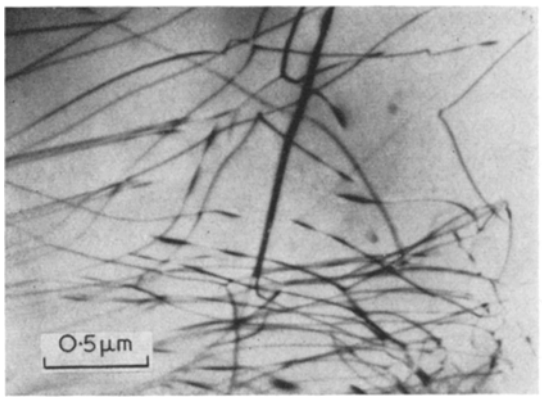

Figure 10 Tangles of dislocations containing dipoles and jogs formed during fracture at 1100° C (β SiC).

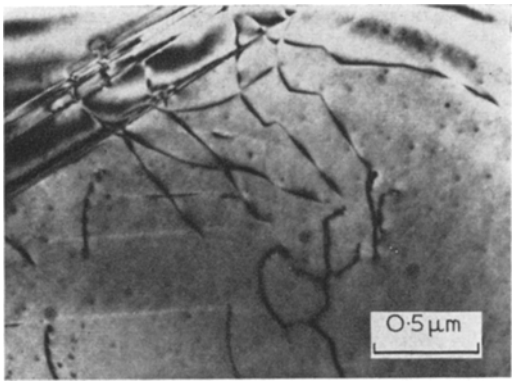

Figure 11 Dislocations in the silicon phase. Fracture chip viewed in transmission.

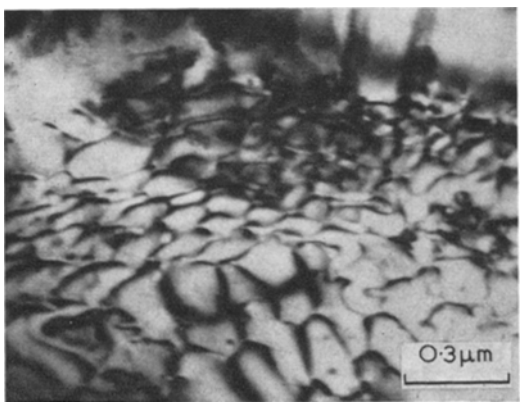

Figure 12 Hexagonal network in a silicon chip from a specimen fractured at 900° C.

grain boundaries. Because of the lack of representative area and the difficulty of working with fracture chips neither possibility could be confirmed.

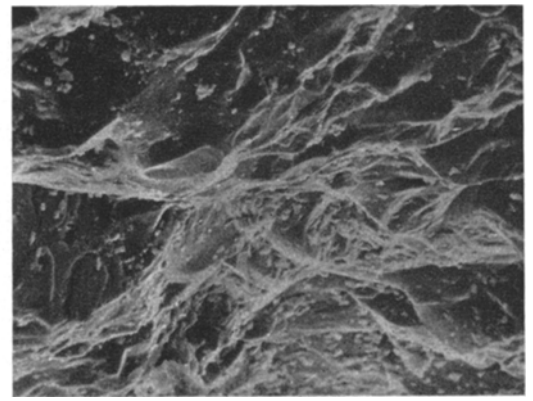

Figure 13 Scanning electron micrograph of the surface of self-bonded SiC fractured at low temperature (× 250).

Direct observation of the surface using scanning electron microscopy showed little change in the fracture surface with temperature except at 1100° C. Fracture was mainly intergranular with few grains showing evidence of transgranular fracture (fig. 13). Small particles of material were seen on the surface at all temperatures up to 1100° C. At this temperature the surface appeared relatively clean (fig. 14).

Figure 14 High temperature (1100° C) fracture surface of self-bonded SiC. Scanning electron micrograph (× 500).

4.3. Brittle Fracture

It is apparent from the results of the bend tests that there is a maximum fracture strength for self-bonded SiC in the region of 500° C, the mean value being significantly higher than that measured at both room temperature and 1100° C. At all temperatures the work of fracture is low (< 60 J/m^2) and for the composite ceramic, failure is thus brittle. Modified versions of the

Griffith [14,15] equation have been developed and utilised to account for the fracture of MgO and SiN by Evans *et al* [16, 17], and certain of the work on Al_2O_3 has been described in a similar fashion by Gutshall [18] and also by McKinney [19]. Any explanation of the fracture mechanisms must therefore account for the brittle nature of failure and the increase in work of fracture at $\sim 500°$ C.

The fracture stress can be related to the crack length and crack initiation energy by means of the Griffith equation. A modification due to Orowan [15] includes a term allowing for limited plastic strain in or near the crack tip.

$$\sigma_f = \sqrt{\frac{E\gamma_I\rho}{\pi a_o c}} \quad (1)$$

where σ_f = fracture stress, E = elastic modulus, γ_I = fracture initiation energy, a_o = atomic radius, and ρ = crack tip radius.

The increase in fracture stress up to 500° C and its gradual decrease thereafter could be due to healing of the intrinsic surface cracks by an oxidation mechanism, or to a factor such as plasticity of material at the crack tip increasing the crack initiation energy γ_I.

The nature of the four-point bending test assures that the maximum tensile stress occurs on the outer surface of a specimen. Both the SiC and silicon phase are coated at the surface with a thin protective film of SiO_2 [20, 21], hence any Griffith flaw in, or near the surface should be covered with a similar film. Any rise in temperature is likely to increase the rate of healing and hence blunt the crack tip. An investigation into the healing of surface cracks in SiC by an oxidation mechanism has been conducted by Lange [22]. He showed that deep cracks introduced by thermally shocking bars of SiC could be blunted by long term oxidation at 1400° C. The oxidation of surface flaws introduced by diamond machining was also investigated. Such flaws would be more representative of those usually found in test specimens. Heating at 1400° C for times in excess of 100 h was found to increase the flexural strength by 10%, a figure which is within the statistical variation normally encountered. Because of the short duration of the tests in the present investigation and the lower temperatures employed, strengthening due to blunting of crack tips by oxidation is thus considered unlikely.

The increase in the work of fracture at $\sim 500°$ C and the marked change in shape of the load-extension curve at 1100° C (fig. 5) allows the fracture behaviour to be discussed in terms of mechanisms occurring over three temperature ranges.

The crack initiation energy γ_I and the work of fracture γ_f have been measured for a number of ceramics [23], and it was usually the case that $\gamma_f > \gamma_I$. Significantly separated values of γ_f and γ_I were attributed to differences in the mechanisms of crack initiation and propagation. In the present work no difference in the fracture surface was detected over the width of the surface, or with variation in temperature with the exception of tests conducted at 1100° C. It would appear that there is no appreciable difference in initiation and propagation over this temperature range. Hence the γ_I term used in the Griffith equation can be discussed in terms of any increase in γ_f above the room temperature value.

4.4. Fracture below 500°C

It is apparent from the results of the bend tests that there is a maximum in the fracture strength of self-bonded SiC in the region of 500° C. The work of fracture over the temperature range 0 to 500° C has values of 27 to 34 J/m², with a tendency to increase with temperature, although this trend is within the limits of experimental variations. Since the value of γ_f is typical of a brittle material the properties can be correlated by means of the Griffith-Orowan relationship (equation 4).

Factors contributing to the γ_I term of equation 1 have recently been discussed by Evans [24], who showed that the largest contribution was made by the plastic deformation term γ_p for the particular case of large grain size polycrystalline MgO.

He related the individual terms comprising the fracture initiation energy γ_I

$$\gamma_I = \psi\gamma_o + \gamma_p + \gamma_s + \gamma_c + \gamma_b \quad (2)$$

where ψ is a geometrical factor, γ_o is the surface energy, γ_s is a term due to subsidiary cracking, γ_c is due to cleavage step formation and γ_b was introduced to account for blunt cracks.

Transmission electron microscopy has shown dislocation motion to occur in both the SiC [12] and free silicon phases (figs. 10, 11) below 500° C. In addition, large numbers of stacking faults were shown to be generated near the fracture surface. Therefore the γ_p term of equation 2 would be expected to make a significant contribution to γ_I during fracture of self-bonded SiC.

Experimental results indicate that the fracture strength has increased by ~ 1.5 times at 500° C over the room-temperature value. A corresponding increase of 2.25 times should occur in the $E\gamma_I\rho$ term of the Griffith-Orowan relationship for a constant crack length c. Since the elastic modulus is nearly independent of temperature [25] the effective variables are γ_I and the crack tip radius ρ.

Electron microscopy of the fracture surfaces did not reveal any change in the appearance of the fracture surface with temperature in the range 0 to 900° C, hence changes in ψ, γ_s and γ_c would not be expected. The surface energy γ_o should be constant at temperatures well below the melting point. Thus any changes in γ_I used in equation 1 would be expected to occur by way to the plasticity term γ_p. It is suggested that the increase in the fracture strength up to 500° C is due to the increase in γ_p and its effect on the crack tip radius ρ.

4.5. Fracture above 500°C

Above 500° C the fracture stress decreases, and although the work of fracture increases by a factor ~ 2 over the temperature range 400 to 600° C, the measured value ~ 50 J/m^2 indicates that the fracture behaviour is still characteristically brittle. Plastic deformation of single crystals of silicon has been reported by Sylwestrowicz [10, 26] at temperatures as low as 600° C under tensile conditions. In addition Rumsey and Roberts [5] have observed creep deformation to take place in self-bonded silicon carbide containing a free silicon phase at temperatures above 500° C. These temperatures are in good agreement with the temperature at which the work of fracture increases (fig. 4), which would suggest that the free silicon phase is the more important in determining the mechanical properties above 500° C.

The macroscopic fracture process is still brittle above 500° C ($\gamma_f \simeq 50$ J/m^2) and the Griffith-Orowan equation can still be used to relate the fracture stress to c, γ_I and ρ. However, γ_f increase considerably in the region of 500° C. It is suggested this increase is due to plasticity mainly in the silicon phase (figs. 11, 12) [10]. There is evidence to suggest that dislocation movement in the SiC, which has a much higher Peierls stress, can occur near the crack tip [12]. This is also shown in fig. 10 where the jogs and dipoles are indicative of slip on more than one system. Such behaviour was not observed in fracture chips, taken from specimens tested below 900° C, where dislocation movement and stacking fault generation appeared to have taken place on a single slip system in SiC. Because of the unknown nature of the stresses generated at the crack tip, these observations should not be considered typical, but only indicative of possible mechanisms accompanying crack propagation in self-bonded SiC.

In terms of the Griffith-Orowan equation,

$$C \propto \frac{\gamma_I \rho}{\sigma^2}$$

Since π, a_o and E are constant, the value of γ_f has increased considerably, therefore γ_I would be expected to increase similarly with an accompanying increase in ρ. The fracture stress σ decreases above 500° C so that the combination of variables causes an increase in the crack length C. Since the loading is at a constant strain rate, the crack grows with increase in load until, at suitable combinations of critical crack length and stress, catastrophic failure occurs.

Since the critical crack occurs on the surface of the specimen such growth is possible without satisfying Von Mises' criterion [27, 28] for homogeneous deformation, the unconstrained outer surface allowing the crack tip to move without the required five independent slip systems. Thus above 500° C the fracture stress is governed by flow in the free silicon phase, dislocation movements becoming easier with increase in temperature, resulting in a corresponding decrease in fracture stress; the brittle nature of the macroscopic fracture is reflected in the unchanged appearance of fractographs (fig. 13).

4.6. Fracture at 1100° C

At 1100° C a change in fracture behaviour is observed during certain of the work of fracture tests (fig. 5). A crack once started appears to propagate and then stop. The load required to reinitiate crack propagation is almost as high as the initial fracture load. Before the two halves of the work of fracture test specimen were completely separated this process repeated itself several times.

Dislocation movement in silicon occurs readily at 1100° C and deformations up to 30% have been obtained [10] which suggests that the conditions to satisfy Von Mises' criterion are met. The crack path could therefore be reorientated by the silicon phase well below the

initial source of fracture on the surface of the test specimen, and would be expected to take the path of least resistance. Since at 1100° C the silicon/silicon carbide interface appears clean (fig. 14), this would suggest that the crack propagates in the interface region.

5. Conclusions

The fracture stress undergoes a maximum in the region of 500° C. At approximately the same temperature the work of fracture increases two-fold. Thus fracture behaviour can be separated into two regions. Below 500° C fracture is completely brittle whereas above 500° C increasing plasticity in the free silicon phase is believed to affect behaviour. Work of fracture curves at 1100° C indicate a new type of behaviour which, it is suggested, is associated with the separation of the silicon/silicon carbide interface.

Dislocations have been observed in fracture chips of silicon and silicon carbide tested at all temperatures. At the higher temperatures jogs and dipoles were observed in SiC chips indicative of slip on more than one system. In addition, stacking faults were observed in both the primary and secondary SiC; those in secondary SiC were believed to have been introduced by the fracture process.

Acknowledgements

The author acknowledges with gratitude the work of T. D. Clausen, who did much of the mechanical testing, and E. E. Sexton, who prepared specimens for electron microscopy.

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Received 18 August and accepted 20 November 1970.