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Study of indentation induced cracks in $MoSi_2$-reaction bonded SiC ceramics

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Abstract. **$MoSi_2$–RBSC composite samples were prepared by infiltration of Si–2 at.% Mo melt into a preform of commercial SiC and petroleum coke powder. The infiltrated sample had a density > 92% of the theoretical density (TD) and microstructurally contained SiC, $MoSi_2$, residual Si and unreacted C. The material was tested for indentation fracture toughness at room temperature with a Vicker's indenter and K_{IC} was found to be 4·42 MPa√m which is around 39% higher than the conventional RBSC material. Enhancement in indentation fracture toughness is explained in terms of bowing of propagating cracks through $MoSi_2$/SiC interface which is under high thermal stress arising from the thermal expansion mismatch between $MoSi_2$ and SiC.**

Keywords. **Reaction bonded SiC; indentation hardness; fracture toughness.**

1. Introduction

A reaction bonded silicon carbide (RBSC) material contains genetically a substantial amount of silicon (Popper 1960; Forrest *et al* 1972) which limits its practical usable temperature to around 1200°C. Its strength drops abruptly at ~ 1300°C (Chakrabarti *et al* 1994) because of the residual silicon (m.p. 1410°C) becoming soft. Presence of silicon also affects its room temperature strength, toughness and hardness (Chakrabarti *et al* 1997) properties. The oxidation and corrosion resistances are adversely affected (Becher 1983, 1984; Ferber and Tannery 1983, 1984) because of the free silicon in the RBSC. Efforts are presently afoot to convert this detrimental Si-phase in RBSC to a highly refractory $MoSi_2$ phase (Messner and Chiang 1990; Lim *et al* 1989; Singh and Behrendt 1995; Chakrabarti and Das 1996) in making $MoSi_2$–RBSC materials. The high refractoriness of the $MoSi_2$ (m.p. 2050°C), good thermodynamic compatibility with SiC (Gac and Petrovic 1985), ability to undergo brittle to ductile transition at around 1000°C (Schlichling 1978) and the exceptional oxidation resistance (Huffadine 1960), make the incorporation of $MoSi_2$ in RBSC very attractive.

The Si/SiC interface has been observed to be very weak in the conventional RBSC material (Nees and Pages 1986). Conversion of Si to $MoSi_2$ is associated with the replacement of the Si/SiC interface with an $MoSi_2$/SiC interface. Therefore, the interaction of a propagating crack-front with $MoSi_2$/SiC interface is a matter of great concern particularly at room temperature, since both $MoSi_2$ and SiC are known to have a poor fracture toughness. The aim of the present investigation is to examine the interaction of indentation cracks with the $MoSi_2$/SiC interface with an assessment of fracture toughness (K_{IC}) of $MoSi_2$/RBSC material at room temperature.

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2. Experimental

2.1 *Material*

The carbonaceous SiC compacts used in this study for preparing $MoSi_2$–RBSC material, were made by pressing intimate mixture of commercial raw SiC (1200 grit; SiC 98·20%, free C 0·25%, SiO_2 0·50%, Fe_2O_3 0·04% w/w, Grindwell Norton Ltd., India) and petroleum coke (sp. surface area of 4·03 m^2/g; ash-content 0·68 w/w, Assam Carbon Products Ltd., India) powders. The compacts were infiltrated with Si–Mo melt (~ Si – 2 at.% Mo) made from elemental powders of Mo (2–4 μm; purity 99·9 + % w/w, Johnson Mathey GmbH, Karlsruhe, Germany) and Si (– 200 B.S.S.; Si 99·41%, Fe 0·22%, Al 0·08%, Ca 0·004%, Mg 0·2% and Na + K 0·09% w/w, Indian Metals and Ferro Alloys, India). The fabrication details are described elsewhere (Chakrabarti and Das 1996). The fired densities of the infiltrated samples were measured and the samples were found to be quite dense (> 92% T.D.). Microstructurally the material contains SiC, $MoSi_2$, residual Si with minor amount of unreacted C.

2.2 *Sample preparation*

5 × 5 × 3 mm specimens were cut from the infiltrated samples, the end surfaces of each being ground flat. The specimens were embedded in resin and polished up to 1 μm finish. These were taken for indentation and other investigations.

2.3 *Indentation*

For the fracture toughness evaluation, at least ten indentations were made on the polished surfaces of the samples using Vicker indentation and Wolpert Hardness Tester (Germany) at 9·8 N load. Fracture toughness (K_{IC}) was calculated following 'direct crack measurement' technique according to the equation of Anstis *et al* (1981)

$$K_{IC} = 0{\cdot}016(E/H)^{1/2}(P/c^{3/2}), \quad (1)$$

where, E is the elastic modulus, H the hardness, P the applied indentation load and c the crack length around the indenter. An average value of ten results was taken.

2.4 *Optical microscopy*

The fracture paths were examined using Leitz Ortholux II pol-BK (Germany) optical microscope to have an insight into the toughness determining mechanism. The crack produced from the corners of the Vicker's indenter provided an outline of a propagating crack front and thus furnished profiles of the crack–microstructure interactions.

The volume fractions of the individual phases present in the final material were determined with the help of image analysis system using 'Volume Fraction Application System' software (version 1·13 supplied by Leading Edge Pty. Ltd., Adelaide, Australia).

3. Results and discussion

3.1 *Measurement of fracture toughness*

Table 1 shows the indentation fracture toughness values of the $MoSi_2$–RBSC materials with different compositions. With the gradual increase of $MoSi_2$ content from 3 to 9 vol.%, there was an increase in K_{IC} values from 3·34 to 4·42 MPa√m. Moreover, a comparison of the fracture toughness values between $MoSi_2$–RBSC and conventional RBSC materials showed that the former registered an increase in K_{IC} varying between 34·5 and 38·9% with the corresponding variance in silicon from 13 to 24% v/v. Though the final material still contained free silicon, the present result indicated that fracture toughness could be still improved with a further replacement of Si by $MoSi_2$.

Table 1. Indentation fracture toughness values of $MoSi_2$–RBSC and conventional RBSC materials.

$MoSi_2$–RBSC			Conventional RBSC*	
$MoSi_2$ (vol.%)	Si (vol.%)	K_{IC} (MPa√m)	Si (vol.%)	K_{IC} (MPa√m)
9·07	12·73	4·42	16·5	3·18
4·93	23·72	3·82	24·1	2·84
2·96	16·20	3·34	–	–

*From Chakrabarti *et al* (1994)

3.2 *Study of crack–microstructure interaction*

Figure 1 shows the indentation crack profile in the case of $MoSi_2$–RBSC sample containing 4·93 and 23·72 vol.% $MoSi_2$ and Si respectively. The fracture paths were mostly planar with characteristics of the cracks passing transgranularly through SiC grains.

An occasional propagation of the cracks through Si/SiC interface was also observed, but the deflection was notably low. $MoSi_2$–RBSC sample with 9·07 and 12·73 vol.% $MoSi_2$ and Si respectively demonstrated a propagation of the cracks mostly through $MoSi_2$/SiC interface with significant crack bowing (figure 2). Sample with 2·96 and 16·20 vol.% $MoSi_2$ and Si respectively showed crack propagation which could be compared with the sintered SiC (Faber and Evans 1983).

3.3 *Maximum thermal stress*

The thermal expansion coefficients of $MoSi_2$ and SiC being widely different, substantial amount of thermal stress was developed within and around $MoSi_2$ phase in SiC, as the body cooled down from the fabrication temperature. The stress system around particles in an isotropic medium is theoretically well established and Selsing (1961) has shown that the maximum value of the hydrostatic component of the stress generated in the matrix is given by

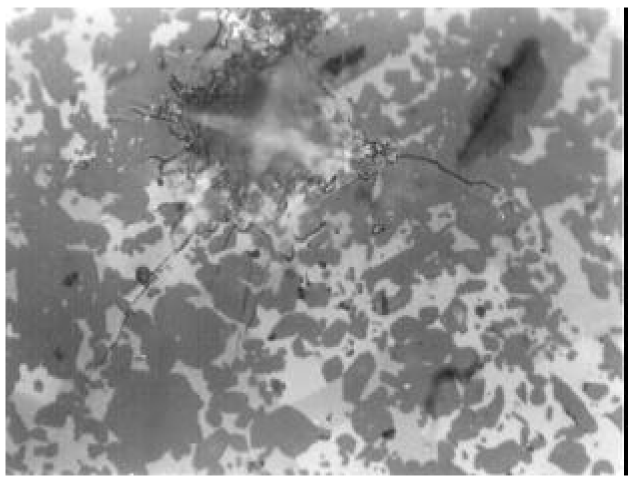

Figure 1. Optical photomicrograph of $MoSi_2$–RBSC composite with 4·93 and 23·72 vol.% $MoSi_2$ and Si showing planar cracks passing mostly through SiC (× 1000).

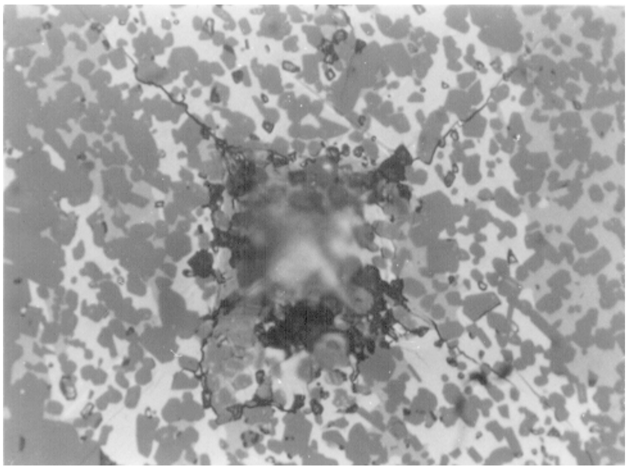

Figure 2. Optical photomicrograph of $MoSi_2$–RBSC composite with 9·07 and 12·73 vol.% $MoSi_2$ and Si, showing crack bowing along $MoSi_2$/SiC interface (× 1000) (dark grains are SiC, light (white) $MoSi_2$, grey Si and occasional black phase, C).

$$P = (\Delta a \times \Delta T)/[(1 + n_1)/2E_1 + (1 - 2n_2)/E_2], \quad (2)$$

where, Δa is the linear thermal expansion coefficient difference of the matrix and the particle, ΔT the temperature difference and $n_{1,2}$ and $E_{1,2}$, the Poisson's ratios and Young's moduli of the matrix and the particle respectively. Substituting $a_1 = 5{\cdot}2 \times 10^{-6}$/°C (Whittemore 1959), $a_2 = 9{\cdot}18 \times 10^{-6}$/°C (Shackelford 1994), $E_1 = 410$ GPa (Carnahan 1968) and $E_2 = 406$ GPa (Huffadine 1960), $n_1 = 0{\cdot}168$ (Carnahan 1968) and $n_2 = 0{\cdot}158$ (Shackleford 1994) and assuming $\Delta T = 1575$°C, (2) yields $P = 1728$ MPa. For Si/SiC system, substitution of $a_2 = 4{\cdot}68 \times 10^{-6}$/°C (Clifford 1971), $E_2 = 160$ GPa (Runyan 1965) and $n_2 = 0{\cdot}42$ (Rochow 1973), the maximum stress (P) developed around Si particles was 708 MPa. Therefore, in $MoSi_2$–RBSC material with greater proportion of $MoSi_2$, the high thermal stress was developed within and around the $MoSi_2$ phase that steers the cracks to propagate around $MoSi_2$ particles and crack bowing along $MoSi_2$/SiC interfaces (figure 2). Consequently, a high K_{IC} of 4·42 MPa√m was achieved. With the decrease in $MoSi_2$ content in the matrix, no such crack bowing was observed because of reduced thermal stress around Si particles and cracks passed preferentially through SiC grains and occasionally through Si/SiC (figure 1) resulting in a decrease in K_{IC} (3·82 MPa√m). With further lowering in $MoSi_2$ content, the fracture path became transgranular compared to the sintered SiC with a low K_{IC} (3·34 MPa√m).

4. Conclusions

(I) The indentation fracture toughness of $MoSi_2$–RBSC composite with 9 vol.% $MoSi_2$ and 13 vol.% Si was measured to be 4·42 MPa√m which was around 39% higher than that of the conventional RBSC material.
(II) The indentation fracture toughness of $MoSi_2$–RBSC material increased from 3·34 to 4·42 MPa√m as $MoSi_2$ increased from 3 to 9 vol.%.
(III) Crack bowing for crack–microstructure interaction, was studied using reflected light optical microscopy (RLM) along $MoSi_2$/SiC interface, which diminished with a decrease in $MoSi_2$-content of the material.
(IV) The crack bowing and increase in K_{IC} could be attributed to the very high thermal stress envisaged to have developed around $MoSi_2$-phase due to wide difference in thermal expansion coefficients of $MoSi_2$ and SiC.

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