EFFECT OF THE HEAT TREATMENT ON THE FRACTURE TOUGHNESS AND R-CURVE BEHAVIOUR OF SILICON CARBIDE SINTERED WITH Al_2O_3 AND Y_2O_3

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Abstract

R-curve behaviour and fracture toughness of silicon carbide ceramics sintered with Al_2O_3 and Y_2O_3 additives have been studied as a function of the heat treatment. The study showed that the heat treatment at 1650°C/5 h had no influence on the fracture toughness values but the treatment at 1850°C/5 h significantly increases fracture toughness because the $\beta \rightarrow \gamma$ phase transformation takes place and is accompanied by an increase of the aspect ratio of SiC grains. The increasing R-curve (stress intensity factor versus crack length) was found in the coarsest microstructures. The mechanisms of toughening in this case were mainly crack deflection and crack branching.

Keywords: silicon carbide, fracture toughness, toughening mechanisms, R-curve behaviour

INTRODUCTION

Silicon carbide is an important structural material because of its unique combination of properties, such as high temperature strength, resistance to wear, corrosion, and thermal shock. Interest in liquid-phase sintered SiC has grown continually over recent years because such materials are easier to process and seem to have mechanical properties superior to the solid-state sintered SiC [1]. However, the low fracture toughness and extremely flaw-sensitive strength of SiC at room temperature have made it unsuitable for use in many structural applications. Since the innovative approach initiated by Omori and Takei [2], who found that SiC could be fabricated at relatively lower temperatures (1850°C to 2000°C) with a liquid oxide phase, a number of attempts have been made to improve the fracture toughness of SiC. These efforts of microstructural toughening of SiC include adding liquid-forming additives, such as Al_2O_3, Al_2O_3-Y_2O_3 , Al_2O_3-Y_2O_3-CaO and Al-B-C, controlling the initial α-SiC content of the starting powder, seeding techniques for preferential grain growth and heat treatments for controlled grain growth [3-9].

One promising method to increase the toughness of SiC is to develop in situ or self-reinforced materials though $\beta \rightarrow \alpha$ phase transformation. The microstructure of LPS-SiC materials can be changed radically by varying the ratio of α-SiC to β-SiC in the starting powder and by applying a suitable post-sintering heat treatment. The fracture toughness of LPS-SiC strongly depends on the aspect ratio of the grains. Some recent publications focus on the relationship between toughness, toughening mechanisms, and microstructure in toughened SiC ceramics. Crack bridging, crack deflection, and pullout are suggested as operating toughening mechanisms in SiC ceramics [10-12]. On the other hand, coarsening resulting from thermal treatment at high temperature also leads to an increase of the critical flaw which diminishes flexural strength.

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In this work, the effect of heat treatment on the fracture toughness and R-curve behaviour of LPS SiC produced with addition of Al_2O_3 and Y_2O_3 was studied and compared, both on as-hot pressed and on annealed materials.

EXPERIMENTAL PROCEDURE

Powder batches were produced using β-SiC powder (HSC-059, Superior Graphite). The sintering additives were Al_2O_3 (A 16 SG, Alcoa) and Y_2O_3 (grade C, H.C. Starck). A combination of 91 wt.% β-SiC powder with 3 wt.% Al_2O_3 and 6 wt.% Y_2O_3 was ball milled in isopropanol with SiC balls for 24 hours. The suspension was dried and subsequently sieved through 25 μm sieve screen in order to avoid hard agglomerates. The samples were hot pressed at 1850°C/1 h under mechanical pressure of 30 MPa in a N_2 atmosphere. Hot pressed samples were subsequently annealed under various time/temperature conditions given in Table 1. After sintering and annealing, the specimens were cut, polished to a 1 μm finish and plasma etched.

The microstructure of polished and plasma-etched specimens was studied using scanning electron microscopy (JEOL JSM-7000F).

Hardness was determined by Vickers indentation (hardness testers LECO 700AT) under a load of 50 N for 10 s. Fracture toughness was determined by indentation technique [13] and by the Single Edge V- Notched Beam (SEVNB) method [14].

The variation of fracture toughness with indentation load (R-curve-like behaviour) was estimated by changing the indentation load over a range of 5–100 N. An observation of in situ crack propagation under the load was performed using PHYSIC INSTRUMENTE and optical microscope LEICA, LEITZ DM RME.

RESULTS AND DISCUSSION

The microstructures of hot pressed SiC-HP (Fig.1a) and SiC-AN1650 material annealed at 1650°C consist of fine submicron-sized equiaxed SiC grains with a low aspect ratio (1.03). No visible effect of the heat-treatment at 1650°C was found on the microstructure of the material.

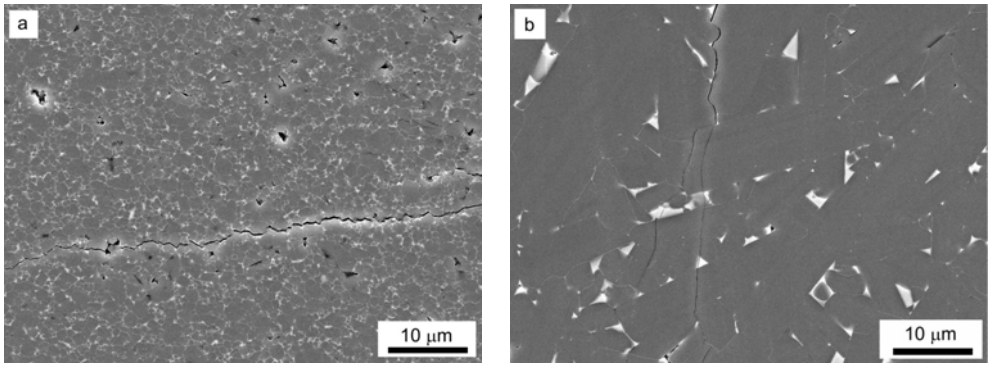

Fig.1. Crack path after annealing, a) SiC-HP; b) SiC-AN1850, crack branching and crack deflection.

The microstructure of the SiC material significantly changed after post-sintering high temperature treatment at 1850°C (Fig.1b). It has bimodal distribution and consisted of elongated SiC grains with higher aspect ratio (4.41) and smaller equiaxed SiC grains [15].

The above results clearly demonstrate that β-α phase transformation is key for the growth of high aspect ratio SiC grains.

The mechanical properties of prepared materials are summarized in the Table 1. Vickers hardness values were in interval from 21.9 ± 0.9 to 23.3 ± 0.1 GPa and did not show any significant difference between sintered and annealed samples.

Tab.1. Mechanical properties of sintered and annealed SiC ceramics

Material	Conditions	HV 5 [GPa]	$K_{IC,ind}$ [MPa·m$^{1/2}$]	K_{IC} [MPa·m$^{1/2}$]
SiC-HP	HP:1850°C/1h	22.2 ± 2.3	2.9 ± 0.2	3.2 ± 0.2
SiC-AN1650	HP:1850°C/1h + AN:1650°C/5h	21.9 ± 0.9	3.3 ± 0.2	3.6 ± 0.5
SiC-AN1850	HP:1850°C/1h + AN:1850°C/5h	23.3 ± 0.1	4.5 ± 0.4	4.7 ± 0.3

The fracture toughness increased with annealing temperature. For fine-grained globular hot pressed SiC, the fracture toughness estimated by SEVNB method was 3.2 ± 0.2 MPa·m$^{1/2}$. After heat treatment at 1650°C/5h it increased to 3.6 ± 0.5 MPa·m$^{1/2}$. The highest fracture toughness value (4.7 ± 0.3 MPa·m$^{1/2}$) was obtained after annealing at 1850°C/5h with plate-like microstructure.

Quinn and Bradt [16] concluded that the indentation technique is not reliable for estimation of fracture toughness of ceramics or other brittle materials, because the crack initiation and propagation is not the same as the crack propagation in the standard fracture toughness tests. However, current comparison identified only small differences between indentation and SEVNB fracture toughness values.

The variation of fracture toughness with indentation load is shown in Fig.2. SiC-AN1850 reveals rising crack growth resistance behaviour with crack length, i.e., R-curve-like behaviour, while toughness of SiC-HP and SiC-AN1650 are not sensitive to indentation load. Figure 3 shows the dependence of stress intensity factor (K_I) on crack length of SiC-AN1850 measured in situ during loading, as demonstrated in Fig.4. Such behaviour was not observed in the fine globular materials.

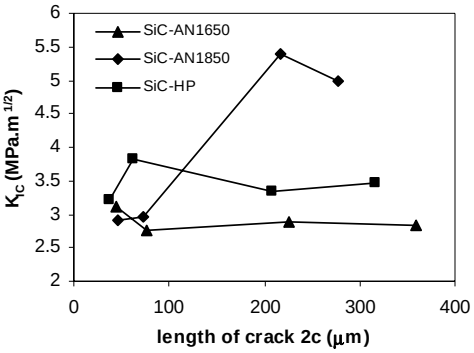

Fig.2. Dependence of indentation fracture toughness on the applied load of as-sintered and annealed SiC ceramics.

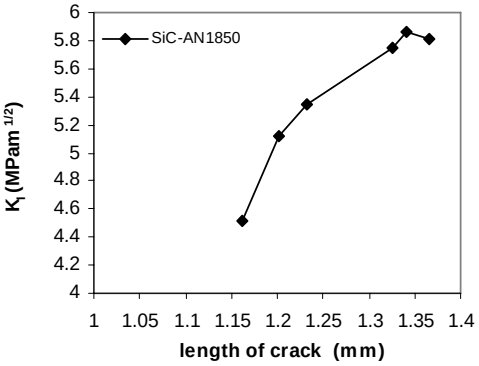

Fig.3. R-curve behaviour of SiC-AN1850.

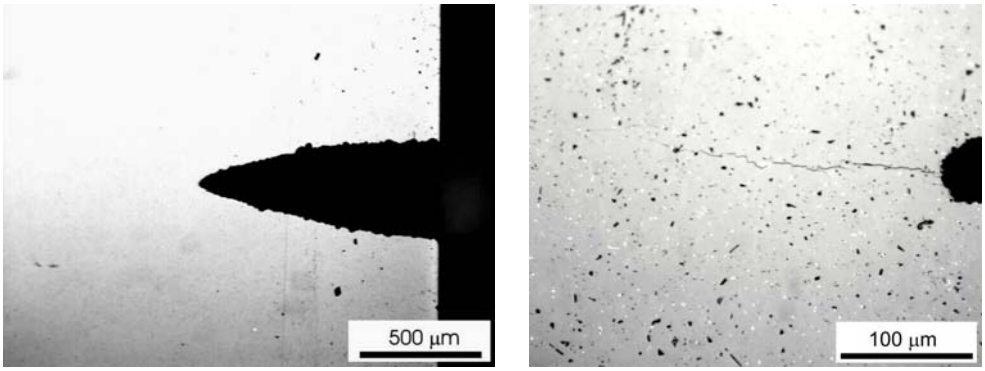

Fig.4. In situ crack path under loading in SiC-AN1850 observed in an optical microscope.

Fractographic observations of the fracture surface and fracture profiles have shown that the crack propagation was controlled by mixed inter- and transgranular fracture in all materials, with slightly higher transgranular portion in the system annealed at 1850°C/h. In materials with fine microstructure and globular grains, the crack propagates mainly intergranularly with relatively small deflection from the main crack direction (Fig.1a). Toughening mechanisms are absent in such microstructures. The crack propagation in the materials with coarse plate-like grains and microstructure is significantly different. The crack deflection is enhanced (often up to approximately 10 μm from the main crack direction), Fig.1b. The toughening mechanisms in the form of frictional and mechanical interlocking of the SiC grains, crack branching and crack deflection have been observed. Such toughening mechanisms are probably responsible for the increase of fracture toughness. This is in agreement with the results of similar investigations [17-19]. This improvement of K_{IC} upon annealing at 1850°C/5h can be directly correlated with the larger grain size and higher aspect ratio, despite some indication of transcrystalline fracture behaviour. In the case of fracture of polycrystalline materials, grain boundaries generally play a role of crack branching or deflection, which results in higher fracture toughness of polycrystalline materials compared to that of single crystals. Process-zones with the mechanisms of grain pull-out and bridging play an important role - to give R-curve behaviour and enhanced fracture toughness, especially in the polycrystalline silicon nitride and silicon carbide with elongated grains [20].

CONCLUSION

The heat treatment at higher temperature results in the β-α phase transformation, which is accompanied by α-SiC grain growth and a change of grain shape of SiC from globular to plate-like.

The heat treatments at 1650°C and 1850°C have no influence on the Vickers hardness values of the studied materials.

The fracture toughness of a hot-pressed SiC estimated by SENVB method was 3.2 $MPa \cdot m^{1/2}$. The fracture toughness increased to 3.6 $MPa \cdot m^{1/2}$ after heat treatment at 1650°C and to 4.7 $MPa \cdot m^{1/2}$ at 1850°C. Higher fracture toughness and R-curve-like behaviour of materials annealed at 1850°C appear to be the result of crack deflection, crack branching and mechanical interlocking of SiC grains.

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