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Effects of carbon on the microstructure and fracture toughness of composite ceramic materials

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In brittle composites, spontaneous microcracking might occur due to the presence of residual stresses. As microcracking has a significant influence on the properties of composite ceramic materials, it is important to understand the mechanism of microcrack formation in order to introduce a controlled amount of microcracking. However, there are fairly few materials where toughening by microcracking has been rigorously verified. Fully validated examples of microcracking toughening are restricted to the composite materials such as zirconia toughened alumina (ZTA) [1], SiC/TiB_2 [2] and some of the liquid-phase sintered SiC ceramics [3].

The appropriate addition of additives plays an important role in improving physical and mechanical properties of ceramic composites [4, 5]. Various additives such as the rare earth, the oxide of alkaline metals, metallic elements and carbon, and so on, have got wide applications in both structural and functional ceramics. Specifically, the addition of carbon is beneficial for sintering and improvement of the properties of SiC/TiB_2/TiC ceramic composite [6]. Moreover, it was reported by Sigl *et al.* [7] that the fracture toughness of B_4C/TiB_2 ceramic was notably increased because of the addition of a certain amount of free carbon.

On the other hand, Al_2O_3/TiC composite is one of the popularly used structural ceramic materials. Since the 1970s, a great deal of research work has been done focusing on the microstructure, mechanical property and engineering performance of Al_2O_3/TiC composite [5, 8–10]. Sintering aids like MgO, TiO_2, Y_2O_3, TiH_2, Ni and Mo, etc. has been adopted in the fabrication of Al_2O_3/TiC composite for high density and properties [10–14].

In the present study, a certain amount of free carbon is purposely incorporated into Al_2O_3/TiC ceramic composite. Attempts have been made to investigate its influence on the microstructure and fracture toughness of the material.

High purity Al_2O_3 and TiC powders were used as the starting materials with average sizes of 0.5 and 0.8 μm, respectively. Al_2O_3 was blended with TiC (30 vol.%) and doped with different amounts of phenolic resins to yield samples with varying amounts of free carbon additives in the as-sintered bodies. The mixtures were subsequently homogenized with alcohol media in a ball mill for 80 hr. After milling, the slurry was screened and dried in vacuum. The phenolic resins were completely pyrolized to amorphous carbon at 1000 °C under the protection of N_2 atmosphere. Bodies were then formed by hot pressing technique in an N_2 atmosphere under 35 MPa for 35 min at 1750 °C in a graphite mould. All specimens were sintered to be at least 98% of their theoretical density (determined by Archimedes' technique). Sintered bodies were then cut with a diamond wheel into samples. AT0 indicates the Al_2O_3/TiC ceramic material without carbon addition, AT1C indicates the Al_2O_3/TiC ceramic material with 1 vol.% free carbon.

Three point bending method was used to measure the flexural strength with a span of 20 mm and a cross head speed of 0.5 mm/min. The test bars were carefully ground and polished into a size of 3 mm thick, 4 mm wide and 30 mm long with an average surface roughness, R_a of 0.1 μm. The edges of the tensile surfaces were chamfered. The fracture toughness was estimated by measuring crack lengths generated by a Vickers' indenter with a load of 196 N [15]. Data for Young's modulus, flexural strength and fracture toughness were gathered on five specimens.

Microstructures of the material were observed with scanning electronic microscope (SEM, model HITACHI S-570). Samples used for analysis with transmission electron microscope (TEM, model HITACHI H-800) were first ground and polished mechanically to be less than 50 μm, and then ion thinned to perforation.

It is shown in Table I that the fracture toughness of the composite increases quickly at the beginning period of the increase in the volume fraction of free carbon. But at a higher carbon content (higher than 2.0 vol.%), the fracture toughness tends to be saturated. When the carbon content is equal to 2.0%, the fracture toughness of the composite amounts to 5.9 $MPam^{1/2}$, which is approximately 20% higher than that (4.9 $MPam^{1/2}$) of the corresponding Al_2O_3/TiC ceramic material without carbon. However, the flexural strength decrease slowly with the increase in the volume fraction of free carbon resulted possibly directly from the low mechanical properties of free carbon. The flexural strength corresponding to the 2.0 vol.% addition of free carbon is 761 MPa.

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TABLE I Mechanical properties of Al_2O_3/TiC ceramic material with different contents of free carbon

Content of free carbon (vol.%)	0	0.5	1.0	1.5	2.0	3.0	4.0
Flexural strength (MPa)	802 ± 34	795 ± 52	786 ± 49	773 ± 37	761 ± 28	722 ± 44	674 ± 47
Fracture toughness (MPam$^{1/2}$)	4.9 ± 0.2	5.5 ± 0.1	5.7 ± 0.2	5.8 ± 0.3	5.9 ± 0.2	5.9 ± 0.2	6.0 ± 0.1

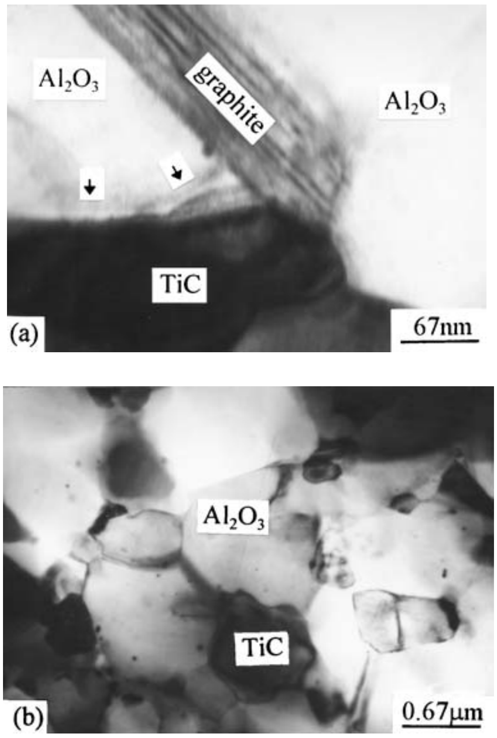

Figure 1 Microstructures of AT0 and AT1C ceramic materials under TEM (a) AT1C; (b) AT0.

Fig. 1 gives the TEM morphologies of the spontaneous microcracks in AT1C composite. It is noted that the observed microcracks have been formed spontaneously upon cooling from sintering temperature. An approximately strip-like graphite particle with a width of about 100 nm is distributed at the interface of Al_2O_3 and TiC grains (Fig. 1a). A microcrack at the phase boundary of Al_2O_3 and TiC can obviously be observed which seems to be originated from the end of the graphite particle. But the kind of microcracking can rarely be discovered in AT0 ceramic material without free carbon (Fig. 1b).

It is suggested from complete experiments that spontaneous microcracking usually appears in the vicinity of graphite particles. Once the free carbon is incorporated, spontaneous microcracking will occur in the material, and the density of spontaneous microcracks will increase with increase in the volume fraction of free carbon. This result gives credence to the assumption that the mechanical preparation technique, utilized for TEM-foil preparation, does not cause microcracking during grinding and polishing. The occurrence of microcracking is therefore unequivocally attributed to the existence of the graphite and then to the weak interfaces associated closely with the graphite.

Fig. 2a and b are the typical morphologies of indentation cracks observed with SEM in AT0 and AT1C ceramic composites. It is shown that the crack in AT0 material is relatively wide and straight and no obvious crack deflection, crack bridging and crack branching happen. But for AT1C ceramic, the crack is nearly the same just within a length from the very beginning to point A. From point A to B it narrows gradually. Nearly after point B, a series of complex crack propagation patterns take place. About eight secondary cracks, almost parallel to each other, can be seen near the end of the main crack in the area D. Crack deflection, crack branching and crack bridging exist together as a result of the interactions at the beginning and/or the end of secondary cracks.

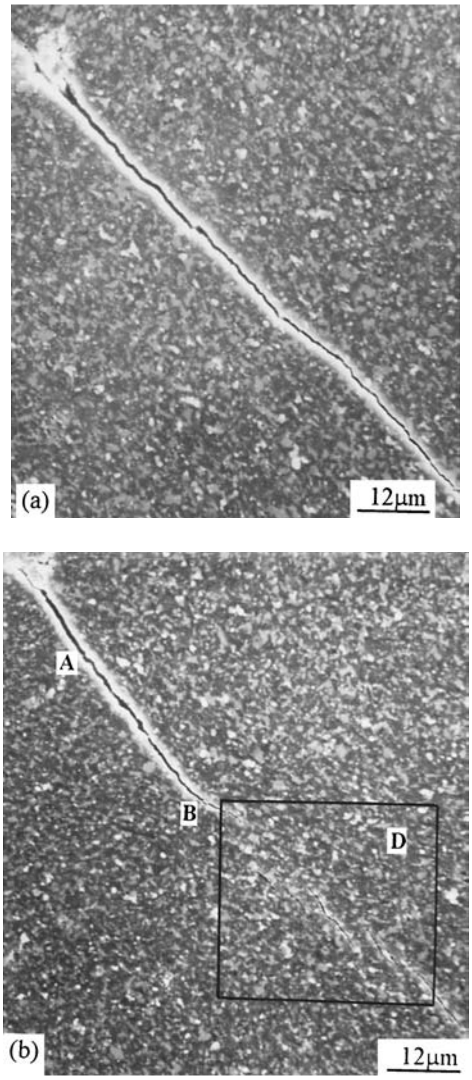

Figure 2 Indentation cracks in AT0 and AT1C composites (a) AT0; (b) AT1C.

It can be summarized that the addition of free carbon can noticeably increase the fracture toughness of ceramic composites. The fracture toughness of the developed carbon containing Al_2O_3/TiC ceramic material is 5.9 MPam$^{1/2}$, which is approximately 20% higher than that of the corresponding material without any carbon. The added free carbon existing in the form of graphite can result in not only the formation of spontaneous microcracks but also the formation of weak interfaces. The existence of weak interfaces is an effective approach to achieve higher fracture toughness of ceramic materials.

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