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Fabrication and characterization of laminated SiC ceramics with self-sealed ring structure

Z. B. YU, V. D. KRSTIC
Centre for Manufacturing of Advanced Ceramics and Nanomaterials, Department of Mechanical Engineering, Queen's University, Nicol Hall, 60 Union Street, Kingston, Ontario, Canada, K7L 3N6

One of the most creative achievements of researchers in the structural ceramics field is ceramic-matrix composite materials. Like so many materials science accomplishments, uni-directionally aligned continuous fibre reinforced ceramic composites [1–3] or laminated plate-form ceramic composites [4–12] actually partially imitate the structure of a natural material, in this case wood, either in one or two dimensions. The advantage of layered structures is that they perform the function of fibre-reinforced ceramic composites, but are much easier to fabricate. Plate-form laminated ceramic composites with a weak interface such as the SiC/graphite system, usually show a high work of fracture and fracture toughness as high as 14 $MPam^{1/2}$ [4, 5]. However, the major problem associated with the plate-form laminate is that it possesses two delamination directions, which prevents the laminates from enjoying widespread applications as structural components. Therefore other structures are needed, either at the micro- or meso-/macro-level, to address this problem. From the meso-/macro-structure point of view, one possible solution to the intrinsic delamination problem in plate-form ceramic laminates is to once again look to a natural material and imitate the ring structure of wood in three-dimensions. This strategy reduces the potential delamination directions in a laminated ceramic material from two to zero when the structure of a laminate changes from plate form to a self-sealed ring, i.e., a highly anisotropic structure.

The objective of the present work is to create a ceramic laminate by imitating the concentric cylinder tree trunk structure and to examine the delamination resistance and fracture behaviour of the structure. A simple shaping technique, a modified slip casting method, has been used to achieve a self-sealed ring structure for a variety of ceramic laminates. In our experiment, the silicon carbide/carbon system was chosen as an example to demonstrate this structure. This system has two characteristics: (1) the graphite not only happens to be a successful sintering aid for SiC but also provides a weak interface [4, 5] and (2) our previous research [4] showed that the carbon layers can be converted into porous silicon carbide layers. Hence a single-phase SiC ceramic with a better oxidation resistance can be obtained while at the same time retaining a suitably weak interface.

SiC slurry was prepared by mixing 88 wt% of SiC, 8 wt% of Al_2O_3 and 4 wt% of Y_2O_3 with water with solid to liquid ratio of 30/70 by volume. The concentration of carbon in the water based graphite slurry was between 2.5 and 5% by volume. SiC/graphite laminates were slip-cast with alternate SiC and graphite layers in a plaster of Paris mould with a casting chamber 10 mm in diameter and 50 mm in length. All the laminates were fabricated in such a way that the outermost layer and core were SiC. The thickness of the SiC layer was varied from 50 to 600 μm and that of the graphite layer from 5–20 μm by adjusting the viscosity of the slurry and casting time. The number of SiC layers in the structure was varied from 20 to 25. After slip casting, the green bodies were slowly dried in air for 48 h and sintered at temperatures ranging from 1,800 to 1,850°C for 1 h in an Ar atmosphere at 1 atm pressure. Bulk densities were measured by the Archimedes method. The theoretical density of the laminates was calculated on the basis of the rule of mixtures.

Three-point bending tests on specimens with a span of 25.4 mm were conducted using an Instron machine (8502) at room temperature. The crosshead speed was 0.06 mm/min. Since ceramic rods with circular crosssections were used for the mechanical property tests, the work of fracture/failure work was used to characterize the fracture resistance of the silicon carbide/carbon laminates. The total work per unit volume during a bending test can be written as:

$$W = \frac{\int_0^{y\ \max} F(y)\mathrm{d}y_{\max}}{\pi r^2 L} \tag{1}$$

where $y_{\max}$ is the roller displacement, $F(y)$ is the load, and r and L are the radius of the specimen and loading span of three-point bending test, respectively. As the load $F(y)$ cannot be expressed as a single and simple function of y in the entire roller displacement range, the calculation of the work of fracture for the laminated composites was done by measuring the area under the load-displacement curve. To achieve higher accuracy, the area under the load-displacement curve was divided into tiny rectangular strips each with a width of about 100 μm and multiplied by the corresponding

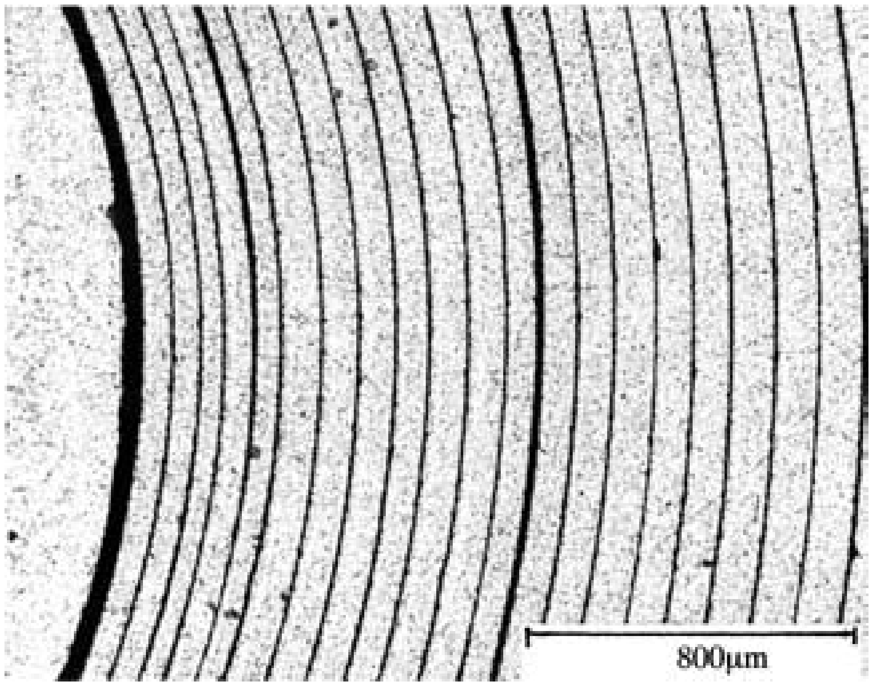


Figure 1 Reflected light micrograph of a SiC/C laminate sintered at 1,850°C for 1 h. Grey layers are dense SiC layers and black layers are porous SiC layers.

load according to the Equation:

$$W = \frac{\left(\sum_{y=0}^{y\ \max} F(y)\Delta y\right)}{\pi r^2 L} \tag{2}$$

The bending strength was calculated from the Equation:

$$\sigma = 3FL/\pi r^3 \tag{3}$$

The cross-sectional structure of a sintered laminate with the SiC/C thickness ratio of about 20:1 is shown in Fig. 1.

Except for the first few outer layers, the inner layers are homogeneous, and the interface uniformity between SiC and graphite layers is in the micrometer range.

With this modified slip casting method, the layered ceramics can be tailored to have structures with different thicknesses of SiC and graphite layers. As expected, the longer the slip casting time, the thicker the layer produced, as shown in Fig. 2a. The largest thickness of SiC layer produced was approximately 600 μm and the smallest was approximately 50 μm. In addition to the slip casting time, the viscosity and the solid-to-liquid ratio of the slurry also play an important role in determining the layer thickness. During the casting of thicker layers, the thickness of previous layers becomes the dominating factor controlling the thickness of new layers. As the layer thickness increases, the removal of water through the wall of the green body becomes more difficult, and for a given slip casting time, the layer thickness becomes smaller. This phenomenon explains the non-linear relationship between the layer thickness and the casting time. It was noted that structural features such as layer thickness and layer thickness ratio of SiC/C etc. have a strong effect on the final properties of the laminates as well as on the sintering behavior.

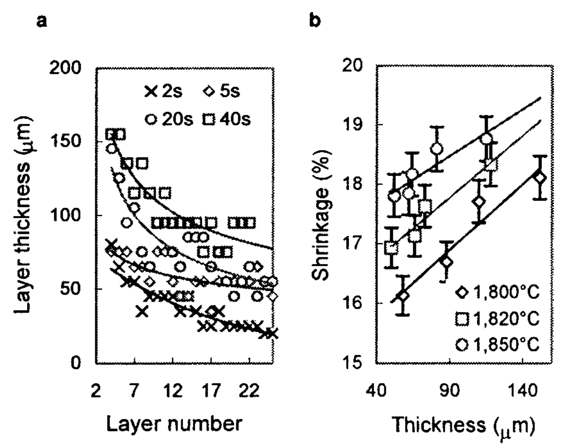
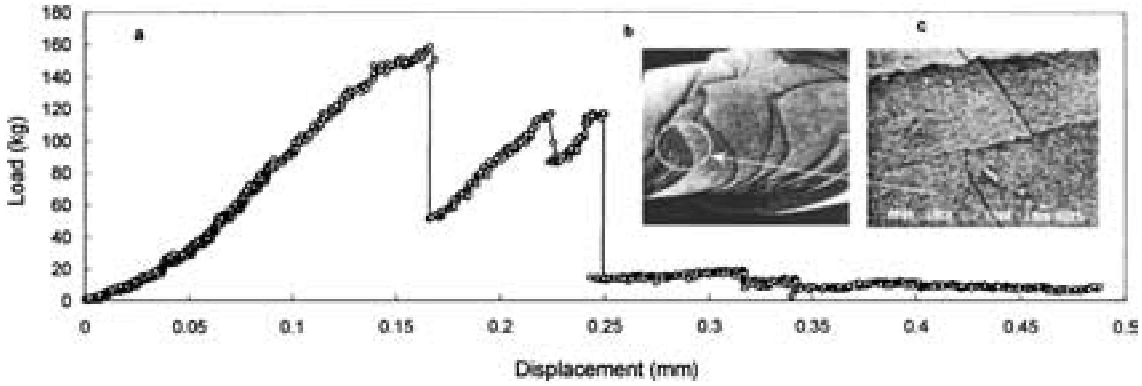


Figure 2 Relationships between: (a) Layer thickness and casting time and (b) Shrinkage and layer thickness.

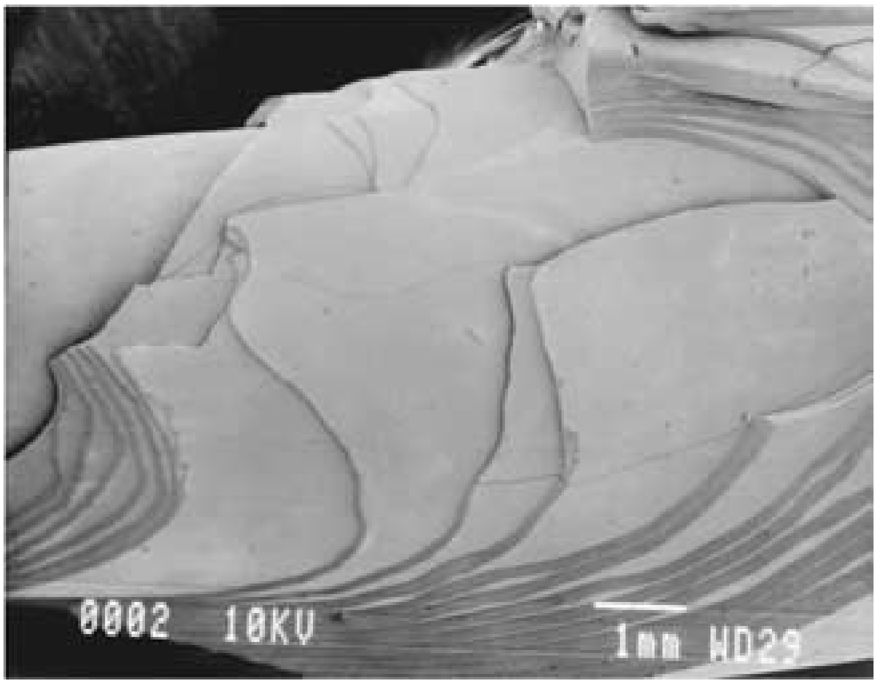
The apparent densities of the laminates sintered at different temperatures are approximately 95% of theoretical density (TD), which is lower than the ∼97%TD obtained for monolithic SiC ceramic. Examination of porosities in the laminates revealed that the open porosity decreased with increasing sintering temperature. The monolithic SiC ceramic specimen showed a lower (<0.5%) open porosity than the laminated ones (0.5–1.8%); and laminates with thicker SiC layers had a lower open porosity than those with thinner SiC layers because of a relatively high fraction of pores in the latter. This effect can also explain the shrinkage of SiC/C laminates as presented in Fig. 2b.

It is worth noting that the higher shrinkage did not result in higher density of the laminates because of the creation of highly porous SiC layers. This is why laminates with thicker SiC layers experienced higher shrinkage.

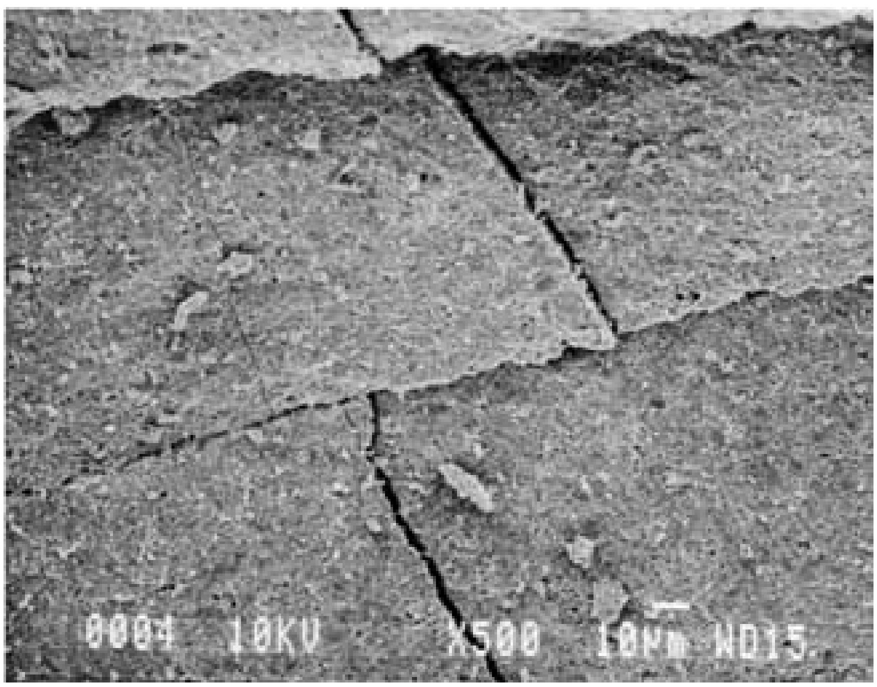
The fracture behavior of the laminates and monolithic SiC ceramics was evaluated in a three-point bend



(a)



(b)



(c)

Figure 3 Fracture behaviour of SiC ceramics with an average thickness ratio of SiC/C of 30:1 sintered at 1,850°C for 1 h. (a) Load-displacement curve, notably similar to those obtained for continuous fibre-reinforced ceramic composites. (b) Fractured cross-sectional surface. (c) Deflected crack between layers. The role of the interface and the unique structure in deflecting cracks and eliminating undesired delamination can be clearly seen.

test and a representative load-displacement curve is presented in Fig. 3.

The curve indicates that the fracture behavior of laminated SiC is entirely different from that of monolithic SiC. The former showed non-catastrophic failure with a large crosshead displacement and the latter always shows brittle fracture with a minimal deformation. After the bending tests were complete, the laminated specimens held together in one piece rather than breaking into two pieces as in the case of the monolithic SiC samples. The laminates showed a non-linear load-displacement behavior with some peaks occurring either before or after the maximum load, depending on their structural features, but all exhibited gradual, non-catastrophic failure. These peaks correspond to the failure of either a single layer or multiple layers indicating strong crack deflection caused by the weak/porous interfaces (Fig. 3b and c). It was also noticed that even when the maximum failure load of the solid SiC core had been reached, the laminates with thicker SiC layers could still resist a relatively high load. The thick SiC layered composites also showed a large displacement over a high load range, which can contribute significantly to work of fracture.

Our initial results (Fig. 4) show that the monolithic SiC has a strength of one and a half times that of the laminates.

However, even though the laminates showed a lower strength, their work of fracture was 2–4 times higher than the monolithic SiC. It seems that there exists a threshold value of the thickness ratio between the SiC layer and the carbon layer. Higher ratios (>20) result in materials with higher fracture toughness. The high work of fracture of the laminates is attributed to the self-sealed ring structure, which eliminates the delamination completely. Clearly, the newly formed porous SiC layers serve to deflect the propagating crack leading to a remarkable improvement in fracture resistance.

In summary, this work demonstrated for the first time that, by imitating the concentric circle tree-trunk structure in three-dimensions, the self-sealed layered structure can be created which effectively eliminates undesired delamination problem associated with plate-form laminates. The self-sealed layered SiC ceramics demonstrate graceful failure behaviour almost identical to that of oriented continuous fibre composites. Currently, the experiments are under way to achieve the self-sealed structure in silicon nitride/carbon system which may open the avenue for the design and manufacture of a large number of ceramics with properties of fibre composites without the difficulty and expense of incorporating fibres.

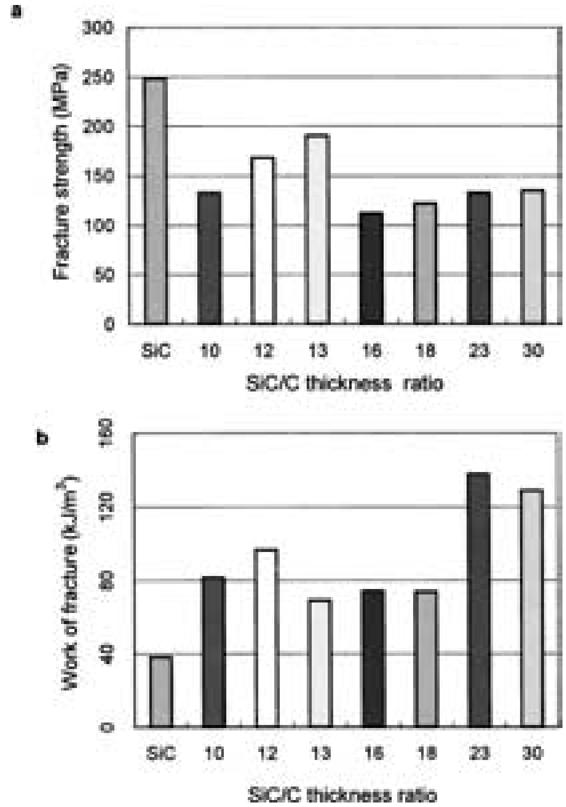


Figure 4 Mechanical properties of laminates: (a) Strength versus composition. (b) Work of fracture versus composition.

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