

# In Situ Processing of Silicon Carbide Layer Structures

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**A novel route to low-cost processing of silicon carbide (SiC) layer structures is described. The processing involves pressureless liquid-phase cosintering of compacted powder layers of SiC, containing alumina ( $\text{Al}_2\text{O}_3$ ) and yttria ( $\text{Y}_2\text{O}_3$ ) sintering additives to yield an yttrium aluminum garnet (YAG) second phase. By adjusting the  $\beta$ : $\alpha$  SiC phase ratios in the individual starting powders, alternate layers with distinctively different microstructures are produced: (i) "homogeneous" microstructures, with fine equiaxed SiC grains, designed for high strength; and (ii) "heterogeneous" microstructures with coarse and elongate SiC grains, designed for high toughness. By virtue of the common SiC and YAG phases, the interlayer interfaces are chemically compatible and strongly bonded. Exploratory Hertzian indentation tests across a bilayer interface confirm the capacity of the tough heterogeneous layer to inhibit potentially dangerous cracks propagating through the homogeneous layer. The potential for application of this novel processing approach to other layer architectures and other ceramic systems is considered.**

## I. Introduction

RECENTLY, considerable interest has developed in the processing of *in situ*-toughened (or self-reinforced) silicon carbide (SiC) ceramics by pressureless liquid-phase sintering.<sup>1-3</sup> By judiciously adjusting the relative amounts of  $\alpha$ - and  $\beta$ -phase in the starting SiC powders, and with appropriate yttria ( $\text{Y}_2\text{O}_3$ ) and alumina ( $\text{Al}_2\text{O}_3$ ) additives, the process produces a microstructure of platelet SiC grains with yttrium aluminum garnet ( $\text{Y}_3\text{Al}_5\text{O}_{12}$ —YAG) second phase.<sup>1</sup> A combination of weak interphase boundaries and high local thermal expansion mismatch stresses toughens the microstructure, by enhanced deflection and bridging of cracks.<sup>4</sup> The same combination, while diminishing intrinsic strength (from enhanced flaw severity) and wear resistance (from enhanced grain removal), increases the resistance to strength degradation from extraneous contact damage.<sup>5</sup> Accordingly, the new approach offers a potentially low-cost route to the processing of damage-tolerant ceramics, avoiding more expensive conventional hot-pressing alternatives.

This concept of microstructural design for enhanced damage tolerance raises an attractive prospect in the light of another recent development, in ceramic *layer* structures.<sup>6-9</sup> This other

development alternates layers of homogeneous microstructures, to preserve surface strength and wear resistance, with heterogeneous microstructures, to enhance subsurface toughness. Layer composites prepared in this way show uncommonly high damage resistance in Hertzian indentation tests.<sup>9,10</sup> Unlike more traditional layer structures, which promote either *toughness* by interlayer crack deflection<sup>11,12</sup> or *strength* by incorporating macroscopic compressive residual stresses,<sup>13-21</sup> the new approach deliberately seeks to produce strong interlayer bonding and to eliminate residual macroscopic stresses. Accordingly, any attendant counterproductive aspects of weak interlayers and residual stresses, such as enhancement of delamination failure modes, can be avoided. The proposed pressureless liquid-phase sintering route offers a novel opportunity for economical *in situ* processing of SiC interlayer structures of this kind, with strong interfaces, minimal residual stress, and, moreover, with high chemical compatibility.

In the present note we describe initial results in this direction, with the fabrication of SiC bilayers. Emphasis is placed on the simplicity of the powder preparation and sintering. The interlayer interface is demonstrated to be well-defined, with continuous SiC matrix phase across the boundary. Hertzian indentation microprobe tests across the bilayer interface indicate the capacity of the heterogeneous layer to arrest cracks formed in the homogeneous layer without attendant delamination.

## II. Experimental Procedure

Submicrometer size powders of  $\beta$ -SiC,  $\alpha$ -SiC,  $\text{Y}_2\text{O}_3$  (respective grades B10, A10, Fine; H. C. Starck, Berlin, Germany), and  $\text{Al}_2\text{O}_3$  (grade AKP-30, Sumitomo Chemicals, Tokyo, Japan) were used as starting materials. Two batches of powder were prepared as previously described,<sup>1</sup> with appropriate amounts of  $\text{Al}_2\text{O}_3$  and  $\text{Y}_2\text{O}_3$  to yield 20 vol% YAG in the final microstructures. The first batch contained only  $\alpha$ -SiC starting powder, as prescribed for the evolution of a "homogeneous" microstructure with fine, equiaxed grains. The second batch contained  $\beta$ -SiC powder doped with 0.5 vol%  $\alpha$ -SiC "seeds," for the growth of a "heterogeneous" microstructure with relatively coarse, elongate  $\alpha$ -SiC grains.

Approximately 4 g of the powder from the first batch was placed in a cylindrical graphite die assembly (25 mm diameter) and was pressed lightly (<5 MPa). After removing the die plunger, 4 g of powder from the second batch was laid down over the first compact in the die cavity, and the compact was uniaxially pressed at 50 MPa. The bilayer compact was then extracted from the die and cold-isostatically pressed at 350 MPa in a wet bag.

The ensuing green bilayer pellet was packed in loose SiC powder (600 grit Crystolon, Norton, Worcester, MA) in a graphite crucible with a screwable lid. Layers of 15  $\mu\text{m}$   $\text{Al}_2\text{O}_3$  powder (Buehler, Lake Bluff, IL) were packed near the bottom

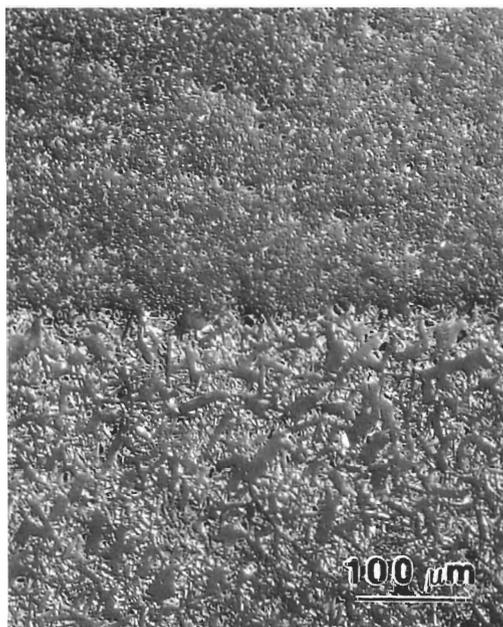
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**Fig. 1.** Nomarski micrograph of bilayer SiC composite, showing "homogeneous" layer at top, "heterogeneous" layer at bottom, with well-defined interface. The microstructures consist of SiC grains (gray) with YAG second phase (light), with some porosity (dark). The relatively elongate nature of the SiC grain structure in the heterogeneous layer is evident.

and the top of the crucible cavity. The crucible was then closed tightly and inserted into a graphite furnace (Labmaster, Thermal Technology, Santa Rosa, CA). Sintering was carried out at 1900°C for 0.5 h in a flowing argon gas atmosphere, with heating and cooling rates 600°C/h and 1200°C/h, respectively. The as-sintered pellet was then repacked in SiC powder in the graphite crucible, but this time without any Al<sub>2</sub>O<sub>3</sub>, and subjected to a further heat treatment at 2000°C for 3 h. On cooling, density was measured using Archimedes' principle.

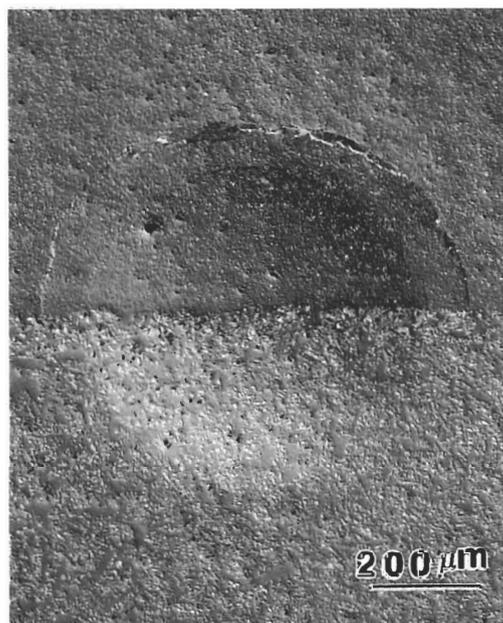
A cross section normal to the cylindrical faces of the pellet was cut using a diamond saw. The section on one of the sawn faces was polished to 1 μm finish and gold-coated for characterization, using Nomarski illumination in an optical microscope.

To investigate the bilayer cracking characteristics, Hertzian indentations were made symmetrically across the trace of the interlayer interface on the polished section, in the same manner as described in a previous study on glass/glass-ceramic bilayers.<sup>10</sup> The tests were performed using a tungsten carbide sphere of radius 3.18 mm at contact load 1500 N on a universal testing machine (Model 1122, Instron, Canton, MA). The indentation sites were examined in the optical microscope.

Vickers indentations (Model 3212, Zwick, Ulm, Germany) with radial cracks were made on the cross-section surface to check for macroscopic residual stresses,<sup>10,22</sup> using a contact load 20 N. These were located in the homogeneous layer close to (but not intersecting) the interface, and aligned with the impression diagonals parallel and perpendicular to the interface. Again, the indentations were examined in the optical microscope.

### III. Results

Figure 1 is an optical micrograph of the cross section through the SiC bilayer, showing the interface between homogeneous (top) and heterogeneous (bottom) layers. The microstructures of the two layers are the same as those previously obtained in separate processing of the two material components:<sup>1</sup> fine, equiaxed α-SiC grains (dark regions) ≈4 μm diameter, with 20 vol% YAG intergranular phase (light regions), in the homogeneous material; coarse, elongate α-SiC-grains 2–5 μm wide



**Fig. 2.** Nomarski micrograph of Hertzian contact damage in the SiC bilayer composite. Indentation made using tungsten carbide ball, radius = 3.18 mm, at contact load = 1500 N. Note the semicircular ring crack in the homogeneous layer ("brittle" response), arrested as it penetrates the interlayer interface, and the smooth depression in the heterogeneous layer ("quasi-plastic" response).

and 5–40 μm long, again with 20 vol% YAG phase, in the heterogeneous component. At the macroscopic level the interlayer interface is clearly delineated. Microscopically, there is some interpenetration of the SiC grains from one layer into the other. Other than that, the interface appears defect-free. Some fine pores (<10 μm) are observed, consistent with a measured porosity of ≈3%, but these are uniformly distributed through the bilayer.

A Hertzian indentation pattern in the SiC bilayer is shown in Fig. 2. A ring crack has initiated in the homogeneous layer and attempted to run around the contact circle.<sup>23,24</sup> However, the crack has been unable to complete the ring in the adjacent heterogeneous layer. Higher magnification examination confirms that the half-ring crack in Fig. 2 has indeed penetrated the interface into the heterogeneous layer, but that the crack has arrested, supposedly by deflection and bridging by the weakly bonded elongate grains in the heterogeneous microstructure.<sup>4</sup> On the other hand, a residual depression at the contact site is observed in the heterogeneous layer, consistent with a quasi-plastic deformation process in the tougher SiC microstructure;<sup>5,25</sup> this deformation is in turn attributable to microfailure along the weak boundaries between the elongate α-SiC grains and the YAG second phase.<sup>4,25</sup> Most significantly, no indication is found in any of the observations of any delamination at the interlayer interface, confirming the integrity of the interfacial cohesion.

Examination of the Vickers indentations revealed no sign of curvature of parallel-oriented radial cracks toward or away from the adjacent bilayer interface, and no significant difference in the lengths of parallel- and perpendicularly oriented crack arms, confirming the absence of any significant macroscopic residual stresses in the vicinity of the interlayer interface.<sup>10</sup>

### IV. Discussion

We have demonstrated the feasibility of a novel, potentially economical *in situ* processing route to the fabrication of chemically stable SiC layer ceramic composites, without the need for conventionally expensive hot pressing or hot-isostatic pressing. Although in the present work we have explicitly considered only a simple bilayer structure with planar interface, the

approach has generic appeal. Thus, there is no fundamental barrier to extension of the approach to fabrication of hard coatings on tough substrates, or multilayer architectures with several alternating hard and tough layers, or even graded microstructures. Nor is there any obvious barrier to the fabrication of layered nonplanar geometries, e.g., rods and tubes, or balls, for such applications as pipes and bearing elements. To accomplish these more complex geometries, alternative powder consolidation procedures like tape casting, dip-coating, sequential slip casting, powder spraying, electrophoretic deposition,<sup>21</sup> or centrifuging<sup>26</sup> may provide a more convenient precursor step to the final pressureless sintering stage.

We stress control of the  $\beta \rightarrow \alpha$  SiC transformation as a key to our proposed *in situ* layer processing. Thus, while under a given set of sintering conditions normal grain growth occurs in one layer, the transformation is able to proceed in the other. Accordingly, the potential exists for applying our method to other ceramic systems with similar microstructure-evolution characteristics. Silicon nitride, for instance, can be made to undergo analogous  $\alpha \rightarrow \beta$  transformation-assisted anisotropic grain growth.<sup>27,28</sup> Work is under way to explore the *in situ* processing of silicon nitride layer composites.

We would acknowledge the exploratory nature of the Hertzian fracture testing on our SiC layer composite. The advantage of such testing, apart from its obvious simplicity, is that it visually highlights the innate difference in brittleness of the two microstructures on opposite sides of the interlayer interface. It demonstrates the susceptibility of the homogeneous layer to conventional modes of crack propagation and, conversely, the tendency of the heterogeneous layer to energy-absorbing quasi-plasticity. It is implied that the latter layer has the capacity to arrest and contain intrusive cracks from the former layer, from bridging by the weakly bonded elongate SiC grains.<sup>1,4</sup> The Hertzian testing also confirms the capacity of the composite, by virtue of its strongly bonded interface and effectively continuous SiC grain structure, to avoid deleterious interlayer delamination. More detailed information on the fracture and deformation responses of layer structures can be obtained by indenting on *top* surfaces rather than *side* surfaces, i.e., with the contact axis perpendicular rather than parallel to the interlayer interface.<sup>9,10,29</sup> Such tests are yet to be performed on our SiC composites.

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