Delamination Resistance of Two Hybrid Ceramic-Composite Laminates

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Hybrid laminar composites that are comprised of alternating layers of ceramic sheets and fiber-reinforced ceramic-matrix composite (CMC) layers exhibit attractive mechanical properties, including a high first cracking stress and a high strain to failure. To achieve these properties, a strong bond must exist between the ceramic and CMC layers; otherwise, delamination will occur readily between the layers. The present study focuses on the delamination resistance of such laminates at ambient and elevated temperatures. The delamination resistance of interfaces that have been subjected to mixed-mode loading has been measured for two different hybrid composites by using edge-notched flexure specimens. At low temperatures, delamination occurs by a process that involves multiple matrix cracking within the CMC layers normal to the fibers, followed by cracking of the matrix parallel to the fibers at or near the ceramic/CMC interface. The corresponding fracture energies are typically in the range of about 100-300 J/m², comparable to the delamination resistance of the CMC itself. At elevated temperatures, delamination occurs via cavitation and rupture of the matrix within the CMC layers at or near the ceramic/CMC interface, with an attendant loss in toughness (to about 10-30 J/m²). The loss in toughness occurs most rapidly at temperatures that are close to the strain point of the matrix phase; this represents the life-limiting temperature for this class of composites.

I. Introduction

HYBRID laminar composites are fabricated by bonding together alternating layers of a monolithic ceramic and a fiber-reinforced ceramic-matrix composite (CMC) at elevated temperatures, using the matrix phase of the CMC as the bonding agent. A variety of such composites have previously been fabricated and their properties have been characterized. A distinct advantage of this compositing scheme over conventional fiber CMCs is that the constituent layers can be first fabricated independently of each other, following a route that optimizes their respective mechanical properties. Subsequently, the layers can be selected and combined in such a way that the ceramic layers impart a high cracking stress (provided that they are stiff and strong) and the CMC layers provide a high strain to failure and good damage tolerance. This process

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II. Experimental Procedure

Two different hybrid laminates were studied. Both were fabricated with dense SiC sheets that were about 0.5 mm thick (Hexpoloy SA, Carborundum Co., Niagara Falls, NY). The CMC layers were comprised of either Nicalon™ SiC fibers (Nippon Carbon, Tokyo, Japan) within a glass matrix (aluminoisoclastic glass, Corning 1723, Corning, NY) or of Nicalon fibers in a glass-ceramic matrix (calcium aluminoisolate glass-ceramic, Corning-CAS, Corning). Table I lists the strain, annealing, and

3Usually, the key changes in the flow characteristics of glasses are characterized by the strain point, the annealing point, and the softening point; these correspond to viscosities of 10^14 s, 10^13 s, and 10^10 s, respectively. Physically, the strain point represents the temperature below which the glass behaves essentially elastically, the annealing point is the temperature at which internal stresses are relaxed within a period of minutes, and the softening point is the temperature at which the glass readily flows at low stress.

4CMC layers were supplied as unfired fiber-matrix prepreg (Corning).
Table I. Critical Temperatures for CMC Matricesa

<table>
<thead>
<tr>
<th>Matrix</th>
<th>Strain point (°C)</th>
<th>Annealing point (°C)</th>
<th>Softening point (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glass (Corning 1723)</td>
<td>−665</td>
<td>−710</td>
<td>−910</td>
</tr>
<tr>
<td>Glass-ceramic (Corning</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CAS)</td>
<td>−1140</td>
<td>−1245</td>
<td>Not available</td>
</tr>
</tbody>
</table>

aData supplied by the manufacturer (Corning, Corning, NY). bApparent value based on beam-bending viscosity of bulk beams.

indicate that, for the systems that are under consideration, first-ply delamination is more likely to occur in the specimen configuration in which one fiber layer is sandwiched between two ceramic layers (Fig. 1(a)), relative to the configurations that contain a larger number of alternating layers, such as that which is shown in Fig. 1(b). The analysis that is presented in the Appendix neglects residual stresses, because their effects have been shown to be negligible for the systems that are studied herein.

Specimens for testing were cut from larger panels, parallel to the fiber direction. Typical specimen dimensions were ~1.7 mm thick × 3.6 mm wide × 50 mm long. One transverse face which exposed the CMC layer was polished to a 1 µm finish to facilitate observations of damage evolution, as described below. One of the SiC layers was notched using a diamond blade to a depth of ~80% of its thickness (not shown in Fig. 1). Sharp precracks were introduced at the tips of the notches by loading the specimens in three-point bending at room temperature. These cracks did not deflect into the ceramic/CMC interface but did travel across the interface into the CMC layer a short distance (~10–20 µm) and then arrested (Fig. 2). For the subsequent calculations of the energy release rates, the crack depth was considered to be equal to the thickness of the outer SiC layer, which was measured with a micrometer prior to processing.

The precracked specimens were subsequently loaded in four-point flexure, using inner and outer loading spans of 19 and 39 mm, respectively. The tests were performed at temperatures ranging from ambient to 910°C for the glass-matrix composite and to 1350°C for the glass-ceramic-matrix composite. The effects of oxidation embrittlement that occur in Nicalon-containing CMCs were precluded by testing in a stagnant argon environment. Prior to testing, the furnace was evacuated to ~5 × 10⁻⁶ torr (~6.7 × 10⁻⁴ Pa) and subsequently backfilled and flushed with argon three times. The tests were performed in a hydraulic testing machine (Model 810 with a Centorr vacuum furnace, MTS Systems, Eden Prairie, MN). Prior to loading, the specimens were heated at a rate of 10°C/min to the prescribed temperature and held at that temperature for 10 min. The majority of the tests were conducted at a crosshead displacement rate of 0.05 mm/min. Some of the elevated temperature tests were performed at displacement rates of 0.001, 0.08, and 1.0 mm/min.

Damage evolution was monitored in two ways. At room temperature, the polished surfaces were viewed using in-situ stereomicroscopy. At high temperatures, it was accomplished by interrupting the tests, cooling the specimens rapidly to ambient temperature (>50°C/min), and examining them via either optical microscopy or scanning electron microscopy (SEM). After examination, the specimens were placed back

![Fig. 1. Testing geometries for beams with (a) one fiber layer sandwiched between two dense ceramic layers (SiC for present experiments) and (b) two fiber layers sandwiched between three ceramic layers.](image-url)
III. Results and Observations

(1) Laminates with Nicalon/1723 Glass CMC

Figure 3 shows the curves of nominal bending stress versus crosshead displacement for the edge-notched specimens. The curves generally exhibit (i) an initial linear elastic region, (ii) a transient region in which damage occurs ahead of the precrack and the response gradually softens (manifested in a decreasing tangent modulus), and (iii) a plateau (steady-state) stress at which extensive delamination occurs at or near the ceramic/CMC interface. At room temperature, the initial nonlinearity in the stress–displacement response was associated with the formation of matrix cracks that were oriented roughly normal to the interface (Fig. 4(a)). These cracks were typically ~200 μm in length and spaced ~50–100 μm apart. At higher stress levels (approaching the plateau), delamination cracks were observed emanating from the precrack at or near the CMC/ceramic interface. In some instances, the delamination crack followed a somewhat tortuous path through the CMC layer, which resulted in some fiber bridging by inclined fibers (Fig. 4(b)). Similar bridging processes have been observed previously in CMCs under transverse Mode-I loading conditions. However, most of the delamination crack followed a path either along the interface or through the glass matrix near the interface (within ~10 μm). Subsequent examination of the delaminated SiC surfaces revealed remnants of the glass, which was consistent with

![Image of microscopic view of a precrack](image)

Fig. 2. Micrograph of a precrack emanating from a notch and penetrating into the CMC layer.

![Image of stress-versus-displacement curves](image)

Fig. 3. Nominal-stress-versus-displacement curves for SiC/glass-CMC hybrid composites tested at the temperatures noted.

![Image of delamination in SiC/Nicalon 1723 glass laminate](image)

Fig. 4. Delamination in the SiC/Nicalon 1723 glass laminate at room temperature, showing (a) accompanying Mode I cracking in the glass matrix and (b) matrix cracking combined with fiber bridging.
the observations of the dominant crack propagating partially through the CMC itself rather than along the interface. The delamination resistance was in the range of 140–210 J/m². These values are only slightly lower than those that were measured using double-cantilever-beam specimens (in Mode I) of the CMC itself (≈250 J/m²).

At 710°C (the annealing point of the glass), delamination proceeded in a similar fashion (Fig. 5), although the peak stress was considerably higher and no plateau was obtained, because of the formation of an additional crack in the SiC. The delamination resistance that was estimated from the peak load was ≈470 J/m²—approximately three times the corresponding room-temperature value. This increase is thought to be due to the reduction in the flow resistance of the glass and the resulting increase in its toughness. Similar behavior was obtained at 750°C, although delamination and cracking of the SiC both occurred at lower stresses.

At 810°C, a well-defined plateau stress was obtained. In this case, there was evidence of matrix cracks of the type that are shown in Fig. 4(a). Instead, delamination occurred by a process of cavitation and rupture through the glass matrix very near the ceramic/CMC interface (Fig. 6), with a resistance of only ≈20 J/m². Similar observations were made for the specimen that was tested at 910°C (the softening point of the glass), although there was more damage within the bulk of the CMC layer (Fig. 7) and the delamination resistance was reduced even more (to ≈9 J/m²).

Estimates of the steady-state delamination resistance, which were obtained from the peak loads, are plotted in Fig. 8. The data points that are accompanied by arrows indicate that a plateau stress had not been obtained during testing, which suggests that the actual toughness is somewhat higher. Evidently, the interfacial toughness first increases as the temperature increases, up to approximately the annealing point of the glass, but subsequently decreases at higher temperatures, as matrix cavitation becomes the dominant mode of failure.

The effects of the imposed displacement rate (0.001, 0.08, and 1.0 mm/min) on delamination resistance were determined at 865°C, i.e., between the annealing and softening points of the glass matrix. Figure 9 shows the stress-displacement curves and the delamination resistances that have been obtained from these tests. In each of these cases, there was considerable damage in the bulk of the CMC, although failure ultimately occurred at or near the ceramic/CMC interface. The resistance increased approximately an order of magnitude as the displacement rate increased.

(2) Laminates with Nicalon-Glass-Ceramic CMC

Figure 10(a) shows the stress-versus-displacement curves for the laminates that contain the Nicalon–CAS-glass-ceramic CMC. Similar trends were observed in the temperature dependence of the delamination resistance (Fig. 10(b)), although there were some subtle differences in the fracture characteristics. Notably, at low temperatures (<1000°C), delamination proceeded through the CMC itself, at a relatively large distance from the ceramic/CMC interface (≈100–300 µm) (Fig. 11). No steady-state toughness was obtained at these temperatures, probably because of the tortuosity of the crack path and the

![Fig. 5. Low-magnification view of delamination at room temperature.](image)

![Fig. 7. Damage within the bulk of the CMC layer at 865°C.](image)

![Fig. 6. Micrograph of cavitation near the SiC/glass- CMC laminate interface at 810°C.](image)

![Fig. 8. Variation in delamination resistance with temperature for the SiC/glass- CMC system.](image)
occurrence of fiber bridging, which, in turn, result in an increasing fracture resistance with crack growth (i.e., R-curve behavior).

At temperatures in the range of −1000°–1250°C, delamination occurred “cleanly” along the ceramic/CFC interface, with minimal damage or cracking elsewhere in the CMC, and with some evidence of ductile ligaments spanning the crack. This material is likely the intergranular glass phase that is invariably present in glass-ceramic materials. Relatively well-defined plateau stresses were obtained. The corresponding delamination resistance was essentially constant in this temperature range (~20–25 J/m²). Examination of the SiC surfaces after delamination revealed isolated islands of remnant CAS matrix that was bonded to the SiC (Fig. 12). Similar observations were made on the specimen that was tested at 1350°C, although there was evidence of more cavitation damage within the CAS both near the SiC/CFC interface and within the bulk of the CMC layer (Fig. 13). This test was terminated because of the formation of a crack in the SiC on the compressive side of the specimen. The delamination resistance was estimated to be greater than ~6 J/m².

The effects of loading rate on the delamination resistance at
a temperature of 1100°C were also examined. Qualitatively, the trends are similar to those of the glass-matrix system (Fig. 14), although two of the three tests were terminated because of cracking of the SiC.

IV. Discussion

The temperature dependence of the fracture mode and the delamination resistance of the SiC/Nicalon 1723 glass CMC can be correlated with the changes in the viscosity of the glass matrix (Table I). The delamination resistance in this system decreases steeply, by more than an order of magnitude, at a temperature near the annealing point. Near this same temperature, the fracture mode changes from one of brittle fracture of the glass matrix with attendant fiber bridging to one of localized flow, cavitation, and rupture within the glass near the SiC/CMC interface. Similar correlations exist for the laminates that contain the CAS–Nicalon CMC, although the transition seems to occur at a slightly lower temperature in relation to the strain point of the CAS. (The absolute temperature of the transition in the CAS laminate is higher, because of the more refractory nature of the glass-ceramic.) These materials contain some residual glassy phase at the grain boundaries, which seems to control the fracture resistance at higher temperatures; this is believed to be the cause of the lower transition temperature in the CAS system. It is expected that the transition temperature could be elevated further through the use of a matrix material with a higher degree of crystallinity. In addition, the observed increase in crack-growth resistance with increased loading rate is consistent with reported observations of subcritical crack growth that is caused by creep cavitation in glass ceramics, which, again, is consistent with the role of the viscosity of the matrix phase.

Some of the edge-notched flexure tests did not yield definitive values for the delamination resistance, which is a result of cracking of the ceramic sheets prior to the attainment of a steady state. Such tests provide only a lower-bound estimate of the delamination resistance. The incidence of this cracking does not seem to correlate with the test temperature or other characteristics of the test procedure or material properties and is probably associated with the stochastic nature of the strength of the ceramic.

The calculated values of delamination resistance are based on the implicit assumption that delamination occurs in a planar fashion at the SiC/CMC interface and that the zone of damage is confined to a small region that is adjacent to the interface. In some instances, the zone of microcracks extends ~200 μm into the CMC; in others, the delamination crack deviates gradually from the plane of the interface, by as much as ~300 μm. These effects may become important when the corresponding lengths become comparable to the layer thickness (~0.5–1 mm in the present experiments). The magnitude of those effects can be probed experimentally by measuring the delamination resistance in thicker test specimens.

V. Conclusions

The delamination resistance of the hybrid laminates is sensitive to the matrix phase within the CMC, because this is the bonding agent between the ceramic and the CMC layers. For

Fig. A-1. Schematic of a symmetric half of delaminated composite beam subjected to combined axial loading and bending.
Table A-1. Normalized Laminate Stiffnesses

<table>
<thead>
<tr>
<th>Laminate</th>
<th>Region (i)</th>
<th>Region (ii)</th>
<th>Region (i)</th>
<th>Region (ii)</th>
</tr>
</thead>
<tbody>
<tr>
<td>stiffness</td>
<td>$\hat{A}_{11}$</td>
<td>$\hat{B}_{11}$</td>
<td>$\hat{D}_{11}$</td>
<td>$\hat{D}_{11}$</td>
</tr>
<tr>
<td>$\hat{A}_{11}$</td>
<td>$\frac{1}{s+2} + s\hat{\lambda}$</td>
<td>$\frac{s(1-\lambda)}{2(3+2s)^2}$</td>
<td>$\frac{1}{3} \left[ \frac{1}{(3+2s)^2} \right]$</td>
<td>$\frac{1}{12} \left[ \frac{1}{(3+2s)^3} \right]$</td>
</tr>
<tr>
<td>$\hat{B}_{11}$</td>
<td>$\frac{s(1-\lambda)}{2(3+2s)^2}$</td>
<td>$\frac{1}{3} \left[ \frac{1}{(3+2s)^2} \right]$</td>
<td>$\frac{1}{3} \left[ \frac{1}{(3+2s)^3} \right]$</td>
<td>$\frac{1}{12} \left[ \frac{1}{(3+2s)^3} \right]$</td>
</tr>
<tr>
<td>$\hat{D}_{11}$</td>
<td>$\frac{1}{12} \left[ \frac{1}{(s+2)^3} \right]$</td>
<td>$\frac{1}{12} \left[ \frac{1}{(s+2)^3} \right]$</td>
<td>$\frac{1}{3} \left[ \frac{1}{(3+2s)^2} \right]$</td>
<td>$\frac{1}{3} \left[ \frac{1}{(3+2s)^3} \right]$</td>
</tr>
</tbody>
</table>

The equations for $\hat{A}_{11}$, $\hat{B}_{11}$, and $\hat{D}_{11}$ are as follows: $\hat{A}_{11} = A_{11}/(Q_{ch})$, $\hat{B}_{11} = B_{11}/(Q_{ch}^2)$, and $\hat{D}_{11} = D_{11}/(Q_{ch})$, where $Q_{ch} = E_s/(1-\nu_s)$.

The hybrid laminates that contain the glass-matrix CMCs, the delamination resistance can be correlated with the viscosity of the glass matrix; it is relatively insensitive to temperature, up to the annealing point of the glass matrix, and subsequently diminishes rapidly with further increases in temperature. For the laminates that contain the more-refractory glass-ceramic, the delamination resistance is maintained to higher temperatures, approaching the strain point of the glass-ceramic. Beyond these temperatures, the failure mode changes to one that involves flow, cavitation, and rupture, which results in substantial reductions in the delamination resistance. In this regime, the resistance is sensitive to the loading rate, which is a consequence of the viscoplastic nature of the matrix phase.

APPENDIX

The steady-state strain energy release rate, $G_s$, for a bimaterial specimen that contains one layer of each material is found elsewhere.\(^3\)\(^7\) Solutions for two other beam geometries that are of greater interest to the present work (see Fig. 1) are presented here. The $G_s$ value is obtained from the difference in the strain-energy densities ahead of and behind the crack tip, using a cutting and pasting operation to simulate virtual crack extension (Fig. A-1). It is expressed as the sum of the strain energy release rate functions that are produced by the applied moment ($G_{sm}$), the axial forces ($G_{sn}$), and the coupled effects of the two ($G_{NM}$). The results are

$$G_s = \frac{S^2h^2 \cos^2 \Phi}{3D} (G_{sn} + G_{NM} + G_{M}) \tag{A-1}$$

where

$$G_{M} = \left( 1 - \frac{\hat{B}_{11}^2}{\hat{A}_{11} \hat{D}_{11}} \right) \left( \frac{1}{8 \hat{D}_{11}} \right) - \left( \frac{1}{8 \hat{D}_{11}} \right) \tag{A-2}$$

and

$$G_{N} = \left( 1 - \frac{\hat{B}_{11}^2}{\hat{A}_{11} \hat{D}_{11}} \right) \left( \frac{1}{8 \hat{D}_{11}} \right) - \left( \frac{1}{8 \hat{A}_{11}} \right) \tag{A-3}$$

Fig. A-2. Variation in the normalized energy release rate, $\hat{G}$, with the layer-thickness ratio $s$ (equal to $t_r/t_c$) and the elastic dissimilarity constant, $\lambda$, in the range of 0.1–0.9. Results were obtained assuming delamination of the top ply in a three-layer composite, as shown in the schematic above the graph.
Fig. A-3. Variation in the normalized energy release rate, $\tilde{G}$, with the layer-thickness ratio $s$ (equal to $t_i/t_c$) for (a) a three-layer composite and (b) a five-layer composite. Results were obtained using elastic dissimilarity constant, $\lambda$, values in the range of 1.0–10.0.

\[
G_{NM} = \frac{1}{\tilde{B}_{11}} \left( \frac{1}{4} \tilde{D}_{11} + \frac{\bar{y}}{\bar{D}_{11}} \right)
\]  
\[
\bar{y} = \frac{\xi}{2(1 + \xi)}
\]
\[
\bar{D} = \left( \frac{E_c}{1 - \nu_c^2} \right) \frac{h^2}{12}
\]
\[
S = \frac{M}{h \cos \Phi} = \frac{Pl}{2bh \cos \Phi}
\]
\[
\xi = \frac{h_1}{h_2}
\]
\[
s = \frac{t_i}{t_c}
\]

\[
\lambda = \frac{E_1^r}{E_c} \frac{1 - \nu_c^2}{1 - \nu_{12}^2 \nu_{21}^2}
\]
\[
\tan \Phi = \frac{Nh}{M}
\]

Here, $E_c$ and $\nu_c$ are the Young's modulus and Poisson's ratio, respectively, of the precracked (ceramic) layer; $P$ is the plateau load, $l$ is the distance between the inner and outer loading points, $b$ is the specimen width, and $h$ is the specimen height. The thickness of the uncracked and cracked layers are, respectively, $h_1$ and $h_2$, and $t_i$ and $t_c$ are the thickness of the ceramic and CMC-reinforced layers, respectively. $M$ is the applied moment, and $N$ is the applied normal force; the normalized stiffnesses $\tilde{A}_{11}$, $\tilde{B}_{11}$, and $\tilde{D}_{11}$ for the two regions that are illustrated in Fig. A-1 are found in Table A-1. Some typical results are plotted in Figs. A-2 and A-3.

References

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