A modified simple interface fracture test for ceramic environmental barrier coating on ceramic matrix composite

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A new interface fracture test was developed for ceramic environmental barrier coatings (EBCs) on ceramic matrix composites (CMCs). The proposed test modifies an existing test in order to simplify specimen preparation. An asymmetric double cantilever beam (ADCB) test specimen with a stiffener adhering to the coating was used, and a wedge was inserted into the notch to open it. A formula to calculate interface toughness using the notch opening load and displacement was derived; it was expanded so that the toughness could be obtained from the wedge insertion load and displacement. The interface toughness of a model EBC/substrate system consisting of a mullite layer and Si bond coat on a monolithic SiC plate was measured to examine the validity of the test. The obtained interface toughness was in good agreement with that obtained by the original test in the previous study. The interface to be fractured was controllable by adjusting the position of the notch.

Key-words: Interface toughness, Interface fracture test, Coating adhesion, Environmental barrier coating (EBC), Ceramic matrix composite (CMC)

1. Introduction

Environmental barrier coatings (EBCs) are essential for applying SiC fiber-reinforced SiC matrix composites (SiC/SiCs) to hot sections of jet engines since SiC/SiCs are rapidly oxidized and volatilized in water-vapor-rich atmospheres of combustion gas.1–5 Usually EBCs are multilayer coating systems designed to meet the demands of protective coatings: a top coat providing water shielding composed of Ba$_2$SrAl$_2$Si$_2$O$_8$ (BSAS) or rare earth silicates (RE$_2$Si$_2$O$_7$ and RE$_2$SiO$_5$) (RE: rare earth elements like Y, Sc, and Yb); an intermediate layer of mullite or mullite/BSAS serving as an oxygen barrier that also relaxes thermal expansion mismatch; and a Si bond coat layer for good adhesion to the substrate. These constitutive materials have different Young’s moduli and coefficients of thermal expansion (CTE); thermal stresses are induced on the order of several hundreds of MPa after processing and during heat cycles in use.6,7 Thus, thermomechanical durability is required for EBCs as well as chemical stability in the environment, even in quite severe heat cycle conditions ranging from ambient temperature to service temperatures of 1300–1400 °C.

EBCs usually have larger CTE than SiC/SiC substrates because the main components of EBCs are oxides; in-plane tensile stress is induced in the coating when it is cooled. Such a stress condition with a brittle coating and substrate can induce several kinds of coating damage,5,9 as shown in Fig. 1: edge delamination, internal fracture in the coating or substrate parallel to the interface, and transverse cracking through the coating thickness followed by interface deflection or penetration to the substrate. This situation is completely different from that in thermal barrier coating (TBC) systems on super alloy substrate; in-plane compressive stress is generated in the coating layer on the ductile substrate, in which the main delamination mode is buckling.8

The damage mechanism of a realistic EBC system was reported to be the ingress of the gaseous environment to the interface between the mullite and Si bond coat layer through a transverse through-thickness crack (mud cracks in the top coat and intermediate coat) and the edge of the specimens.9–12 This results in the formation of thermally grown oxide (TGO) of β-cristobalite (SiO$_2$) at the interface, and the volume change of the TGO during cooling causes microcracking and consequently interface degradation. By this degradation of the interface, edge delamination and deflection of transverse cracking at the interface, which are illustrated as (a) and (c) in Fig. 1, are enhanced in those studies. Thus, the evaluation of delamination resistance as a result of such degradation is a key issue, as well as the initial resistance as processed. A quantitative evaluation with physically meaningful parameters, e.g., interface toughness, is required for the design and processing selection of EBCs. More importantly, the interface
toughness may be time-dependent because the degradation is accompanied by the progress of gaseous diffusion and the formation of chemical product. The change in interface toughness with service time is also important for the life prediction of EBCs and the safe usage of SiC/SiCs.

Interface toughness depends strongly on the phase angle, $\psi$, which is the measure of the stress state near the interface crack tip. In dealing with interface fractures like edge delamination and crack deflection of a transverse crack at the interface in EBCs, the importance of interface fracture tests reproducing relatively low phase angles increases compared with the case we deal with the fractures in TBC/superalloy systems. While there have been well-designed tests for TBCs that reproduce stress states with relatively high phase angles ($45°-60°$), tests for low phase angles have been lacking.

We previously proposed an asymmetric double cantilever beam (ADCB) interface fracture test applicable to ceramic EBCs on SiC/SiC substrates. The test is conducted with a small, simple-shape block specimen having a notch and a pre-crack. It can delaminate the interface without loading the SiC/SiC substrate, which may have a weaker interlaminar strength than the interface. We can obtain interfacial toughness with a relatively low phase angle ($<35°$), unlike other interface fracture tests designed for TBCs on metal substrates. The test has a fairly wide application range for coatings with various Young’s moduli, strengths, and thicknesses, but several problems still remain: difficulty in applying to an interface with very high interface toughness (over several tens of J/m²), a slightly tangled pre-crack introduction procedure, and a need for precise notch machining. In this study, we propose a simpler interface fracture test, a modification of the original test.

2. Proposed test

Two modifications of the previously proposed test are done in this study. The specimen preparation procedure in the original test includes the introduction of indentation cracks at the interface to be fractured, creation of pre-crack from the indentation crack and pre-crack dying. The introduction of controlled pre-crack is effective for accurate and reproducible measurement of interface toughness but the procedure is somewhat complicated. In the modified test, a hard plate stiffener is adhered to the coating. A schematic illustration of the proposed test is shown in Fig. 2.
expected to fracture. The notch depth is denoted as \( L \) and the coating thickness remaining after notch machining is \( h_B \). A wedge with a tip angle of \( 2\theta \) is inserted into the notch to open it, which provides a cantilever-like bending of the coating in the notched part, producing a mode-I-rich mixed mode stress concentration at the crack tip. Interface toughness can be calculated from the strain energy stored in the cantilever (the coating and stiffener in the notched part).

Consider the situation in which a wedge is inserted into the notch with a load \( P \) and displacement \( u \) as shown in Fig. 2, and it generates both a load to open the notch, \( F \), and a notch opening displacement, \( \delta \). \( F \) and \( \delta \) are related to the following formula by a basic elastic theory:

\[
F = \frac{3E'I}{L^2} \delta \tag{1}
\]

\[
E'I = \frac{b}{3} [E_A'((\eta_0 + h_0 + h_A)^3 - (\eta_0 + h_B)^3) + E_B'((\eta_0 + h_0)^3 - (\eta_0)^3)] \tag{2}
\]

\[
\eta_0 = \frac{E_A'h_A^3 + E_B'h_B^3 + 2E_A'h_Ah_B}{2(E_A'h_A + E_B'h_B)} \tag{3}
\]

\[
E' = \frac{E}{1 - \nu^2} \tag{4}
\]

where \( E'I \) is the equivalent flexural rigidity of the combined bar of stiffener and coating and \( \eta_0 \) is the distance from the edge of the coating to the neutral axis. \( E \) is Young’s modulus and \( \nu \) is Poisson’s ratio; subscripts A and B designate the stiffener and the coating, respectively.

The strain energy stored in the coating, \( U \), is given by

\[
U = \frac{1}{2} \int_0^L M^2 \, dx = \frac{F^2L^3}{6E'I}. \tag{5}
\]

Here \( M \) is the moment applied to the coating.

Ignoring the strain energy in the substrate because the thickness and Young’s modulus of the substrate are much larger than those of the coating, the energy release rate when the crack propagates from the notch tip, \( G \), is given by

\[
G = - \frac{1}{b} \left[ \frac{\partial (-U)}{\partial c} \right] = \frac{L^2}{2E'Ib} F^2. \tag{6}
\]

Combining Eq. (6) with Eq. (1), \( G \) can be expressed in the forms of

\[
G = \frac{3}{2bL} F \delta \tag{7}
\]

or

\[
G = \frac{9E'I}{2bL^2} \delta^2. \tag{8}
\]

Interface toughness is defined as the critical energy release rate where the crack starts to propagate from the notch tip. Substituting the load and displacement at the critical point, \( F_c \) and \( \delta_c \), into any of Eqs. (6)-(8), we obtain the interface toughness.

In practice, however, the measurement of the wedge insertion load \( F \) may be difficult; the measurement of \( P \) would be much easier in most cases. In this study we actually measured \( P \) and \( \delta \) instead of \( F \) and \( \delta_c \), as described in Section 3. \( P \) can be converted to \( F \) in the following way, taking the friction between the wedge and specimen into account: The equilibrium of force between the wedge and specimen during wedge insertion is shown in Fig. 3. The equilibrium equations in the horizontal and vertical direction are given by

\[
F = N(\cos \theta - \mu \sin \theta) \tag{9}
\]

and

\[
P/2 = N(\sin \theta + \mu \cos \theta), \tag{10}
\]

respectively, where \( N \) is the normal component of the reaction and \( \mu \) is the coefficient of friction. From Eqs. (9) and (10), \( P \) and \( F \) are correlated as

\[
F = \frac{1}{2} \cos \theta - \mu \sin \theta \quad P = A \cdot P. \tag{11}
\]

The constant \( A \) is determined by substituting Eq. (1) into Eq. (11), as

\[
A = \frac{3E'I}{L^3} \frac{\delta}{P} = \frac{3E'I}{L^3} C_0 \tag{12}
\]

where \( C_0 \) is the initial compliance in the \( P-\delta \) curve obtained experimentally. When we ignore the effect of friction, the relation between \( P \) and \( F \) can be expressed simply as

\[
F = \frac{1}{2} \frac{\cos \theta}{\sin \theta} P. \tag{13}
\]
Further, when we have difficulties in measuring \( \delta \), the displacement of the wedge, \( u \), can be used as a substitution by the following conversion:

\[
\delta = 2u \tan \theta.
\]

(14)

3. Experimental procedure

3.1 Material

A multilayer coating system consisting of a Si bond coat layer and a mullite layer on a monolithic SiC substrate was used as the test material. We used the same material as in the previous study proposing the evaluation method so that we could compare the present and previous results. The Si bond coat was deposited by atmospheric plasma spray (APS) with a designed thickness of 100 \( \mu \)m. Then the mullite layer was coated by APS on the Si layer with the thickness set at 250 \( \mu \)m. Young’s moduli of the respective layers were measured by nano-indentation, and 126 GPa for the Si bond coat layer and 132 GPa for the mullite layer were obtained. The previous paper contains more details about the processing and characteristics of the material. 19)

The coated material was cut into the specimen with a height \( (L_0) \) of 4 mm and a width \( (b) \) of 3 mm. The width was determined by reference to the bending fracture test standardized in Japanese Industrial Standard 1607 (JIS1607) so that a plane strain could be assured. The surface was finished as cut without polishing. A sintered \( \alpha \)-alumina plate with a thickness of ~250 \( \mu \)m, one plane of which was mirror polished, was prepared and cut into a 4 \( \times \) 3 mm rectangle; it was used as to stiffen the coating. The unpolished plane of the cut plate and the coating surface of the specimen were bonded with epoxy adhesive (Araldite 2011: Huntsman Corporation, The Woodlands, TX, USA). The detail of the bonding procedure is as follows. The epoxy adhesive was spread on both the surface of the specimen and the stiffener, which then bonded together. The side and corner of the specimen and stiffener were exactly lined up using an alignment jig. Then the bonded specimen and stiffener were compressed with a flat punch with a load of \( \sim 100 \) N to remove porosity in the adhesive and extrude the excess adhesive from the sides. The resultant thickness of the adhesive layer was less than 10 \( \mu \)m. The epoxy was hardened at room temperature for a day, and then the excess adhesive on the sides of the specimen was removed with a craft knife.

A notch was introduced using a diamond wire saw (wire diameter: 200 \( \mu \)m) along the interface to a half depth of the specimen, \( L = \sim 2 \) mm. The cutting wire and the interface were aligned using a digital microscope so that the center line of the notch could overlap either the interface between the mullite and the Si layer or between the Si layer and the substrate.

3.2 Interface fracture test

The setup of the test equipment is illustrated in Fig. 4. The wedge was made by cutting a commercial steel craft knife. The thickness of the wedge was 0.5 mm, and its tip was sharpened with a wedge angle \((2\theta)\) of 16°. The test was conducted by inserting the wedge into the notch at a constant speed of 12 \( \mu \)m/min using a universal testing machine (EZ-LX: Shimadzu Corporation, Kyoto, Japan). A load cell with a capacity of 50 N attached to the machine was used to measure the wedge insertion load during the test, \( P \). The notch opening displacement during the test was measured by a spectral interference displacement sensor [SI-F1000V (SI-F10): Keyence, Osaka, Japan]; the distance from the sensor to the upper part of the stiffener surface was measured as the displacement. The mirror-polished surface of the stiffener contributed to stable and reliable displacement measurement.

4. Results and discussion

Figure 5 shows an example of the relation between the wedge insertion load, \( P \), and notch opening displacement, \( \delta \), during the test. The load increased linearly with displacement as the wedge was inserted. Then the load deviated from the linear increase at the point when the delamination started, but still continued to increase. After the load reached the maximum, it decreased gradually and finally dropped to almost zero when the coating was completely delaminated. The wedge progressed in the notch smoothly without stick-slip throughout the test.
Figure 6 shows the side surface of the specimen after the test. A crack path can be identified from the side surface observation of the specimen. These paths were classified into three types: the path along the interface between the mullite and Si layers, the path along the interface between the Si layer and the substrate, and the path within the Si layer along the splat boundaries formed during APS processing. These crack paths were dependent on the notch position. Crack initiation tended to occur at a point shifted from the center of the notch to the coating side, where the largest stress concentration ought to occur due to the ADCB geometry of the specimen. This suggested that we can control the interface which should be fractured to some extent by changing the position where the notch is introduced. However, note that the fracture does not necessarily occur at the point where the largest stress concentration is generated but at the weakest point, which satisfies the fracture criteria first.

Interface toughness was determined from the wedge insertion load and notch opening displacement. Assuming that the crack began propagating at the point where the linear load-displacement relation finished, $C(P_c, \delta_c)$ (see Fig. 6), interface toughness $\Gamma_i$ was calculated by substituting $P_c$ and $\delta_c$ into Eqs. (7), (11), and (12), as

$$\Gamma_i = \frac{3A}{2hL} P_c \delta_c. \tag{15}$$

Thus, the obtained toughness is the so-called “initial” interface toughness when the crack starts to propagate. The toughnesses of the three interface fracture paths were determined. At least three successful tests for each type of fracture path were used in the calculations. Exactly, $\Gamma_i$ should be calculated based on the interface fracture actually occurs; e.g., when the fracture occurs at the interface between the Si layer and the substrate, $\Gamma_i$ is the equivalent flexural rigidity of stiffener, mullite layer and Si layer, as done in Ref. 19. However, in this study the effect was ignored because Young’s modulus and the thickness of Si layers is much smaller those of the stiffener.

The measured interface toughness between the Si layer and the substrate was $4.3 \, J/m^2$ with a standard deviation (s.d.) of 0.29; this value was in good agreement with the one obtained in the previous study ($4.1 \, J/m^2$) by the test. Although a pre-crack was not intentionally introduced in this study, the effect of that was likely to be small. In double beam cantilever-type specimens, a crack tends to propagate stably due to the geometric effect. Thus an unintentional very small defect or damage near the notch tip might cause the origin of the stable fracture in an early stage of the loading (before we can detect the end of the linear relation), and the formed crack might act as the pre-crack. The influences of the utilization of the stiffener on test results such as the geometry effect and the phase angle change near the notch tip should be precisely evaluated further; the proposed test seems useful as a simple substitute for the original test. The test is supposed to provide a smaller phase angle than the original test because the existence of a stiffener reduces the asymmetry of geometry and rigidity in the specimen, which would be of help for strict evaluation of interface toughness as a function of phase angle. For the interface fracture between the Si and mullite layers, interface toughness of $9.2 \, J/m^2$ with s.d. of 2.2 was measured; for the fracture within the Si layer, $6.6 \, J/m^2$ with s.d. of 0.31 was measured.

5. Conclusion

A new fracture test measuring the interface toughness of EBCs on ceramic matrix composite substrate was proposed by modifying an existing test. A long notch was introduced instead of pre-crack and a stiffener was applied to reinforce the cantilever part of the coating in the ADCB specimen. The interface toughness obtained by the test agreed well with the one measured in the original test even though the introduction of pre-crack, pre-crack dying, and pre-crack depth measurement were omitted, suggesting the validity of the proposed test. This is advantageous for the simplification of the specimen preparation. Although further analyses for phase angle evaluation and geometry effect are needed for assuring the accuracy of the test, the usefulness of the test was shown.

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