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Effect of porous interlayers on crack deflection in ceramic laminates

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Abstract

Ceramic layered systems with interlayers of various porosities were fabricated using tape casting technique. Submicron size alumina powders were used to make the tapes for both the strong dense laminae and the weak porous interlayers. Porosity was introduced into the interlayers by the addition of PMMA powders. The pores generated were found to be spherical and uniformly distributed. The crack deflection capability of the layered systems with interlayers of different porosity were then investigated. To facilitate the study, the fracture energies of the different porosity monolithic porous layers, and also the dense alumina layer, were quantified using four point bending tests. It was observed that there exists an optimum porosity that the crack deflection, hence the fracture energy of the system, can be maximized. Theories proposed in the literature on crack deflection in layered systems were also discussed and compared with the present experimental findings.

Keywords: Al_2O_3; Crack deflection; Fracture; Laminates; Porosity

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1. Introduction

Ceramic layered systems have attracted wide attention in recent years as such configurations have shown to be effective in improving the toughness of the ceramic components.[1,2] It is noted that such enhancement in fracture property is mainly attributed to the crack deflection capability in the interlayers of such systems. Both experimental and theoretical works reported in the literature have indicated that the ability to deflect crack depends on the fracture energy ratio of the interlayer and the laminae in the layered systems.[3–7] Cook and Gordon[8] have analyzed the problem based on the stresses at a crack tip and suggested that a crack will be deflected at an interface if the strength of the interface is about 1/5 of that of the matrix. Based on an energy approach, Kendall,[3] on the other hand, has proposed that crack deflection will occur if the fracture energy of the interface is less than 10 to 20% of the matrix, where the exact value depends on the thickness ratio of the interface and the matrix layer. In a theoretical study by He and Hutchinson[4] for layered systems without the presence of residual stresses, they proposed that the critical interface to bulk fracture energy ratio for crack deflection is 0.25. In their subsequent work,[9] they incorporated the effect of in-plane and residual stresses in their model to illustrate the influences of these stresses in crack deflection of layered systems. The results of their studies were also found to be consistent with that computed numerically by other researchers.[10]

The above studies have shown that to build a toughness enhanced layered system, not only a weak interface is required to promote crack deflection, a chemically compatible interface is also required to avoid the building up of internal stresses. An easy way to construct such a system has been proposed by Clegg et al.,[1,11] where a porous interlayer of the same material as that of the bulk is to be employed as the weak interface. In their work, natural starches, such as rice and potato starch, were used to generate the pores in the porous interlayer via burnt-off during sintering. In the present work, however, PMMA powders are used to generate the desired porosity in the porous interlayers. It is found that the PMMA powder particles produce uniformly distributed spherical pores after they burnt out during sintering. Both the dense and porous layers were fabricated using tape casting technique.

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2. Experimental procedure

The ceramic powders used in the present work were fine-grained high purity alumina powders (AKP 30, Sumitomo Chemicals, Japan) with a average particle size of 0.4 μm. The PMMA powders (Acrylic Powder, Buehler) used to generate pores in the porous interlayers, on the other hand, have an average particle size of 40 μm. Tapes for forming the respective layers in the layered systems were produced by tape casting of a aqueous slurry containing 58 wt.% of alumina powders, 1 wt.% of fish oil, 6.5 wt.% of polyethylene glycol 400, 4.5 wt.% of benzyl butyl phthalate, 7 wt.% of polyvinyl butyral and 23 wt.% of ethanol (99.86%). For the porous tapes, various volume percent of PMMA powders were added to the slurry with respect to the amount of the alumina powder in the slurry. The different volume percent added were 40, 50, 60, 70 and 80 vol.%. The slurries were then de-gased in a pressure de-gas system and finally tape casted using a continuous feed tape casting machine (Unique, USA) onto a polypropylene carrier tape running at a speed of 20 cm/min. The thickness of the dense and porous tapes fabricated was both 0.3 mm. After drying, the tapes were cut into a rectangle of size 60×4 mm. The dense and porous layers were then stacked and pressed together at room temperature to a final thickness of 3.3 mm. For layered systems, dense and porous layers were stacked alternately. The stacked green samples were then trimmed to 50×4×3.3 mm block before sintering at 1550 °C for 3 h. The sintered layered systems and the monolithic samples were finally subjected to microstructural examinations and four point bending tests with a loading span of 30 mm[11] to evaluate the fracture energies of the samples. Microstructural studies were performed on the fine-polished cross-section face of the samples using Scanning Electron Microscope (Jeol, JSM5310).

3. Results and discussions

3.1. Sintering behavior

Fig. 1 shows the cross-section SEM micrograph of the layered system formed with 60% PMMA addition. It can be seen that the layers were very well integrated without any delamination between the dense and porous layers. The dense layers have been fully densified during the sintering process and microstructural examination using SEM and image analyzer showed that a relative density of more than 97% has been achieved. It is also observed that the addition of PMMA produced uniformly distributed large spherical pores throughout the porous layer. The average size of these large pores (70 μm) is measured to be more than two orders of magnitude larger than the average grain size of alumina (0.4 μm). These large pores introduced from the burnt-out PMMA powders are termed as "macropores" in the subsequent discussions for clarity. It is also noted from the microstructural examinations that the actual volume fraction of porosity in the porous layer, which is mainly contributed by the large macropores since the matrix is almost fully densified, is lower than that of the initially added PMMA volume percent. For example, in the sample shown (Fig. 1), although the added amount of PMMA was 60 vol.%, the porosity result obtained from image analysis showed a porosity of 48.7%. Similarly, the final porosity after sintering for samples with other volume fraction of PMMA addition was found to be lower than the actual PMMA volume fraction (Table 1).

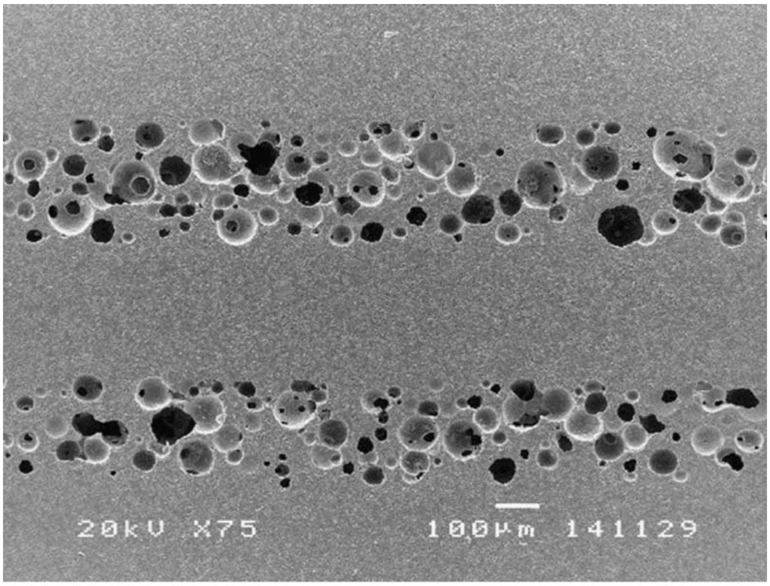


Fig. 1. SEM cross-section micrograph of a layered system with 60 vol.% PMMA.

Table 1
Porosity volume fraction of the porous interlayers after sintering for various amount of PMMA additive

Volume fraction of PMMA (%)	Porosity V_p (%)
40	30.2
50	39.8
60	48.7
70	57.6
80	65.2

Table 2
Fracture energies for the monolithic samples with different amount of porosity

Porosity V_p (%)	Fracture energy (J/m^2)
Dense	62.3
30.2	37.9
39.8	18.2
48.7	11.2
57.6	8.4
65.2	5.8

This phenomenon, nevertheless, is expected as in the process of powder consolidation, natural small pores between the particles will form. During the densification of these natural small micropores, the particles in general have gone through some rearrangement and certain degree of local densification. After the micropores have fully sintered and resulted in the formation of a dense matrix around the large macropores, the macroporous system becomes stable.[12] Researchers[13,14] have shown that pores with coordination numbers smaller than a critical number will sinter, otherwise, they are thermodynamically stable and will remain. In our present situation, the size of the induced macropores are two orders in magnitude larger than the grain size surrounding them, and resulted in a coordination number much larger than the critical number. Therefore, all the macropores should remain stable after the matrix has fully densified. This, in turn, is consistent with our experimental observation in the present work.

3.2. Crack deflection

It is noted from theoretical models in the literature[3–7] that crack deflection in layer systems is mainly determined by the relative fracture energy of the adjacent layers, which is, in turn, dependent on the volume fraction of porosity present in the layers. As a result, in our work, we first determined the fracture energy of the different porosity materials using homogeneous monolithic samples by four point bending test. These results are summarised in Table 2, and will be discussed later on their effects to crack deflection in layered systems. Next, layered systems with interlayers of different volume fraction porosity, from 30.2 to 65.2% volume percent porosity after sintering, were fabricated and their fracture energies evaluated using four point bending tests. The crack propagation results of the layered systems with different porosity interlayers are shown in Fig. 2. It can be seen from Fig. 2(a) that interlayers containing porosity volume fraction of 30.2% did not show any effective crack deflection to provide substantial toughening. However, when the porosity volume fraction of the interlayers increases to 39.8%, crack deflection were observed as shown in Fig. 2(b). It is also further noted that as the porosity of the interlayer continues to increase, the amount of deflected crack propagation also increases in the porous interlayer.

Theoretical models proposed in the literature have predicted that if there is no elastic mismatch, the interfacial crack will not kink out of the interface when the ratio of the fracture energy, R_i, to that of the matrix (or adjacent layer), R_m, is less than 0.57 [15], i.e.,

$$\frac{R_i}{R_m} < 0.57 \tag{1}$$

In our present studies, the entire layered system can be seen as a composite with Al_2O_3 as the matrix material. Hence, we can assume that there is basically no elastic mismatch between the layers, and the cracks are always moving in the dense Al_2O_3 material, regardless of whether they are in the dense or porous layer. Therefore, for the present configuration, in order for the crack to remain in the porous interlayer, which means that the ligaments of the Al_2O_3 matrix in the porous interlayer must fracture, Clegg [16] has rewritten Eq. (1) to be

$$\frac{R_{\text{lig}}}{R_m} < 0.57 \tag{2}$$

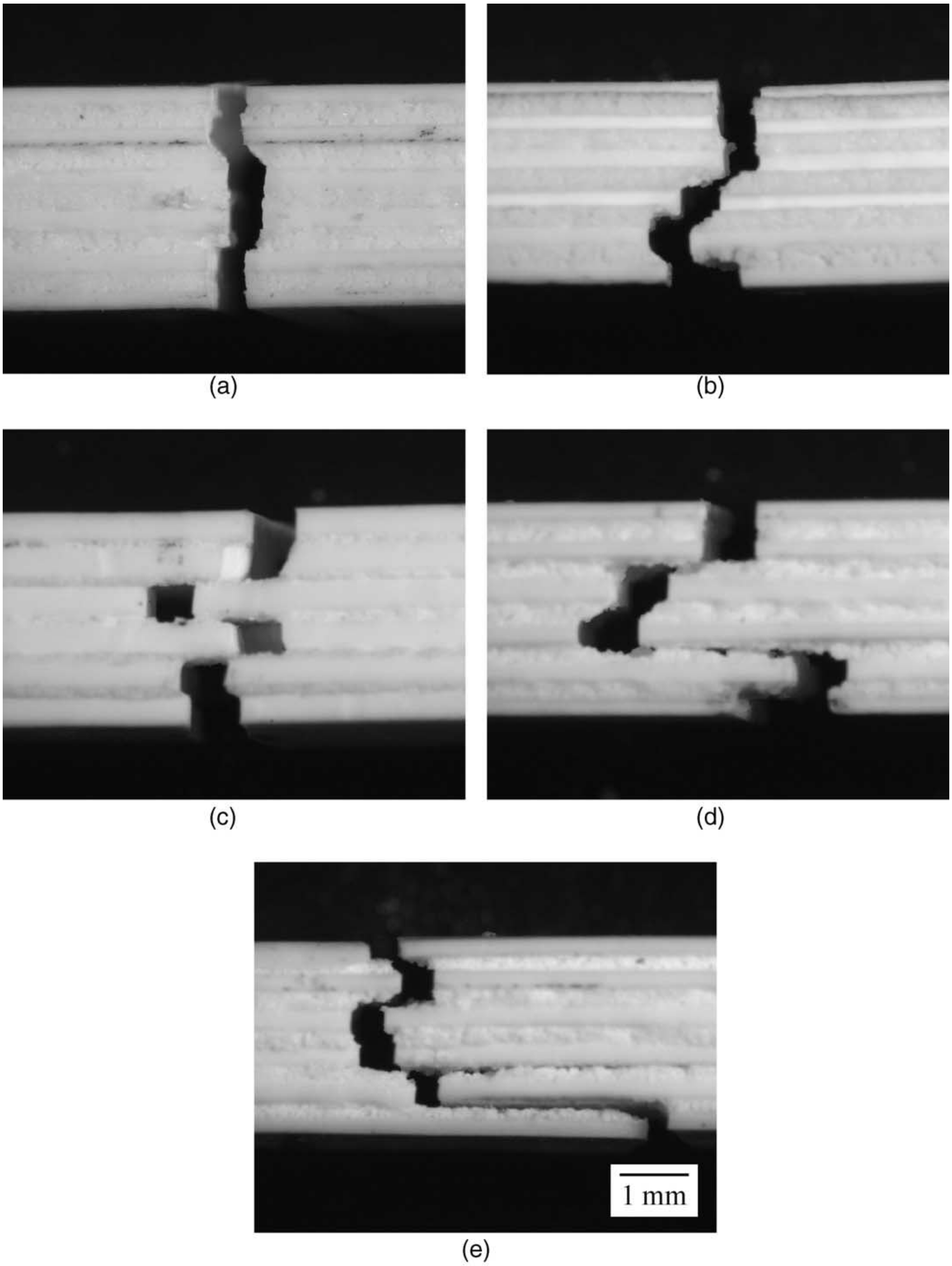
where R_{lig} is the fracture energy of the ligament of the Al_2O_3 matrix in the porous interlayer. Theoretically, the ligament of the Al_2O_3 matrix in the porous interlayer is the same dense material as that of the adjacent dense Al_2O_3 layer in the layered system. Hence they will have the same fracture energy and the crack should immediately kink out of the porous interface. Nevertheless, practically, it is noted that in the porous interlayers, there exists an interaction effect between the homogeneously distributed pores.[16,17] Taking this pore interaction effect into account, Clegg et al.[11] have proposed that the fracture energy of the porous interlayer, R_i, can be related to the fracture energy of the ligament of the Al_2O_3 matrix in the porous interlayer, R_{lig}, by the expression

$$R_i = R_{\text{lig}}\left(1 - V_p\right) \tag{3}$$

where V_p is the volume fraction of pores in the porous interlayers. Putting Eq. (3) into (2), Clegg et al. proposed that the criterion for crack deflection should be

$$\frac{R_i}{R_m\left(1 - V_p\right)} < 0.57 \tag{4}$$

The fracture energy ratio, R_i/R_m, for the various volume fraction of porosity in the porous interlayers to the dense layers, were computed and summarized in Table 3. The results are plotted in Fig. 3 to compare with previous works in the literature and also Eq. (4) as proposed by Clegg et al. The figure show that Eq. (4) provides a good prediction on the deflection criteria of cracks in layered systems. It is also shows that the



(a) (b) (c) (d) (e)

Fig. 2. Crack deflection of the layered systems for different volume fraction porosity in the porous interlayers, (a) 30.2 vol.%, (b) 39.8 vol.%, (c) 48.7 vol.%, (d) 57.6 vol.%, (e) 65.2 vol.%.

present results are in good agreement with that obtained by Clegg et al. on Al_2O_3[11] and that by Blanks et al. on SiC.[2]

3.3. Optimum configuration in layered systems

In the previous sections, the fabrication of a layered system with porous interlayer and the verification of the crack deflection criteria have been discussed. In the present section, the mechanical performance of the overall layered system with the different configuration is investigated. The fracture energies of the layered systems with different volume fraction of porosity in the porous interlayers were quantified by four point bending tests. Fig. 4 shows the load-displacement curve for the layered system (60% porosity in interlayer) and that of the monolithic alumina sample. It can be seen that with effective crack deflection, the component possesses higher fracture toughness. Fig. 5 shows the results of the fracture energies of the layered systems with various volume fraction of porosity in the porous interlayers. It can be further deduced that, in general, the layered systems without crack deflection during the bending test possessed a lower value of fracture energy compared to that with crack deflection occurring in the system. In fact, such system even possessed lower fracture energy value than that of the monolithic sample due to the introduction of porosity in the component. On the other hand, it is also noted that for systems with crack deflection mechanism operating, i.e., the layered systems with 39.8, 48.7, 57.6 and 65.2% of porous volume fraction in the porous interlayers, the fracture energy increases as the amount of porosity increases in the porous interlayer. However, when the porosity is further increased to 65.2%, the fracture energy of the system decreased again. Hence, there exists an optimum amount of porosity in the porous interlayers, or an optimum value of R_i/R_m, to achieve the best fracture toughness for such systems. This is attributed to the fact that as the amount of porosity in the porous interlayers increases, despite the promotion of crack deflection mechanism, the overall mechanical strength of the component has been weakened. This strength weakening effect from the porosity hence becomes a competitive factor to the toughening mechanism effect resulting from crack deflection. Eventually, as the porosity in the porous interlayers exceeds a certain threshold where the component becomes too weak, the overall fracture energy of the system will deteriorate. This phenomenon can also be verified by estimating the total energy absorbed from that contributed by crack deflection and that contributed by porosity, using the experimental

Table 3
Fracture energy ratios between the dense and porous interlayers for layered systems with different amount of porosity volume fraction in the porous interlayers

Porosity V_p (%)	R_i/R_m
30.2	0.60
39.8	0.29
48.7	0.18
57.6	0.13
65.2	0.09

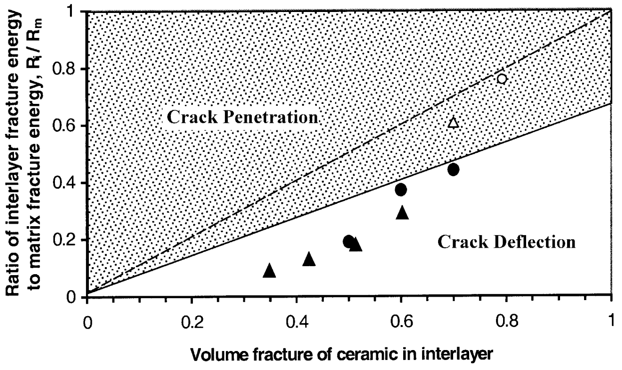


Fig. 3. Relationship between relative fracture energy between the dense and adjacent porous layers and the porosity in the porous interlayers. Circle symbols represent data from Clegg et al., and delta symbols are from present work. Filled symbols indicate cracked deflection is observed.

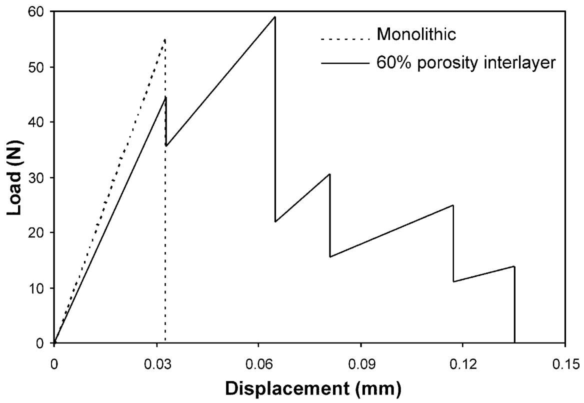


Fig. 4. Load–displacement curve of a layered system with 60% porosity interlayer and that of the monolithic sample.

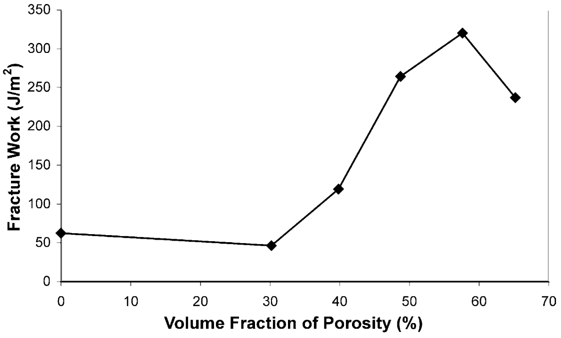


Fig. 5. Fracture energy of various layered systems as a function of volume fraction porosity in the porous interlayers.

Table 4
Total energy absorbed estimation in various layered systems

Porosity V_p (%)	Crack length L (mm)	Factor increase due to crack deflection (L/L_0)	Crack deflection energy E_d (J/m^2)	Energy reduction due to porosity E_l (J/m^2)	Energy absorbed E_a (J/m^2)
Dense	3.0	1.00	–	–	62.3
30.2	3.2	1.07	66.5	11.1	55.4
39.8	4.1	1.37	85.1	20.0	65.1
48.7	5.8	1.93	120.5	23.2	97.3
57.6	9.1	3.03	189.0	24.5	164.5
65.2	9.1	3.03	189.0	25.7	163.3

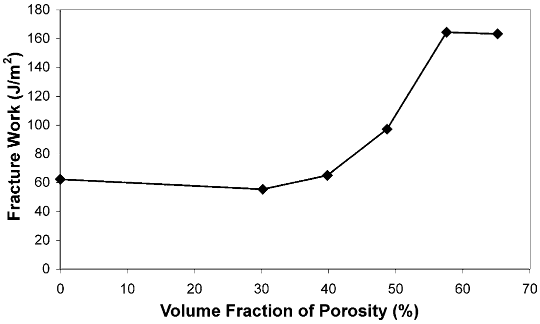


Fig. 6. Final energy absorbed in the layered systems as a function of interlayer porosity estimated after considering the effect from crack deflection length and porosity.

results obtained. The averaged crack deflection length for various porosity laminates were first estimated using the SEM micrographs, and the factor increase compared to that without crack deflection can then be computed. Using results from Table 2, the reduction in energy due to porosity for each porosity laminate system can also be evaluated by considering the amount of porous volume fraction in each system. The final energy absorbed for the various systems was calculated and presented in Table 4 and Fig. 6. It can be seen that the energy variation trend estimated is in good agreement with that from the experimental determined fracture energy values. With no crack deflection, the 30.2% interlayer porosity sample experienced a decrease in fracture energy compared to the monolithic sample. However, as crack deflection started to occur, the enhancement in energy from longer crack path has resulting in an overall improvement in system's fracture energy. Nevertheless, the increasing trend stops after 57.6 porosity interlayer system, as beyond this point, the amount of crack deflection shown almost no increase. As a result, the reduction of the energy due to higher porosity took over in significance, resulted in subsequent decrease in overall fracture energy level. It should be noted that despite the observation of a decrease in the fracture energy of the layered system from 57.6% to 65.2% of interlayer porosity, the fracture energy of the system at 65.2% interlayer porosity is still much higher than that of the monolithic sample (400% higher). As a result, in general, it can be concluded that crack deflection mechanism significantly enhances the fracture toughness of the component.

It is also noted in the process that the porous interlayer systems studied in the present work are different from that introduced by Clegg et al.[1]; as in the present systems, there exists two contributing mechanisms, namely, crack deflection and porosity. Although the porosity has shown to be fracture energy deteriorating factor, it should be noted that it could be an essential parameter when strength to weight ratio is concerned.

4. Conclusions

In the present work, the addition of PMMA in raw ceramic powders is found to be an easy and effective way to generate uniform porosities in porous ceramic materials. Ceramic layered systems with interlayers of different porosities were successfully fabricated. Theoretical models on crack deflection criteria for layered systems reported in the literature were studied and compared with the present experimental results. It is found that pore interaction effect in the porous interlayers cannot be neglected. It is also shown that an increase of porosity in the porous interlayers promotes crack deflection, and hence the fracture toughness of the system. However, as the porosity in the porous interlayers increases beyond a critical volume fraction, the overall system will be weakened due to the large amount of porosity introduced and finally result in a decrease in the fracture toughness. Despite that, it is noted that systems that promote crack deflection will possess higher fracture toughness than that of the monolithic sample.

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