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INVESTIGATION OF THE EFFECT OF MICROSTRUCTURE ON THE R-CURVE BEHAVIOR OF METAL-CERAMIC COMPOSITES

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ABSTRACT

An investigation was made into the effect of microstructure on the peak toughness and shape of the crack growth resistance curves for two ceramic-metal composites. An Al_2O_3/Al composite formed by Reactive Metal Penetration was used along with an AlN/Al composite formed using a reactive infiltration technique. The results indicate that the toughness increases with an increase in the volume fraction of the metal phase for a particular composite composition, and the peak toughness and shape of the R-Curve also depend on the composite microstructure and metal composition.

I. INTRODUCTION

The inclusion of a metallic phase into a ceramic matrix has been widely investigated as a method for improving the toughness of ceramic materials for use in structural applications. [1] A variety of methods have been used to fabricate ductile phase reinforced composites including squeeze casting of molten metal into a porous preform [2], directed metal oxidation [3], and liquid phase sintering [4]. One characteristic of these composites is that they exhibit R-Curve behavior, which implies that their resistance to crack propagation increases with increasing crack length. This behavior results from metal ligaments bridging the crack in its wake. A closing force is applied on the crack due to these ligaments, which results in an increased toughness. As the crack grows, the number of bridging ligaments increases until an equilibrium bridging length is reached at which point the toughness reaches a plateau [5].

Recent theoretical and experimental work has shown that the shape of the R-Curve may have a significant effect upon the reliability of tough ceramics. In particular, for materials with the same maximum fracture toughness, the material with a more steeply rising R-Curve may be more reliable [6-8]. Thus, it is important to investigate the effect of microstructure on both the plateau toughness and the shape of the R-Curve.

Flinn et al. have conducted a considerable amount of work in modeling the toughness behavior of ceramic-metal composites and have developed a model for the total increase in fracture resistance due to bridging metal ligaments. This model uses the metal reinforcements size, volume fraction, yield strength, work hardening coefficient, ductility, and extent of debonding at the interface to

describe the toughening affects of metal ligaments bridging a crack [5,9-10]. This model, like most others, has been concerned either with regularly aligned metal ligaments and with the average size of microstructural parameters and their effect upon the maximum toughness [10-11]. For experimental investigation of 3-D interpenetrating networks of metal and ceramic, average microstructures and models specifically developed for aligned systems have been used. The use of these models needs to be critically examined. Specifically, the effects of not only the average microstructure on the shape of the R-Curve, but also the effect of the distributions in the size of the metal on the R-Curve needs to be investigated.

This work is a preliminary investigation of the R-Curve behavior of two metal-ceramic composite systems; Al_2O_3/Al and AlN/Al. It primarily focuses on the effect of volume fraction, and to a lesser extent variations in microstructure, on the R-Curve and peak toughness of these systems. The next section deals with the materials systems used and the processing of the composites. Sample preparation and the mechanical test procedures used are discussed in Section III. The results are presented and discussed in Section IV and finally the important conclusions are summarized in Section V.

II. MATERIAL SYSTEMS AND PROCESSING

Both the systems used in this investigation have continuous interconnected metal-ceramic microstructures. The first system investigated was an Al_2O_3/Al composite formed using a novel Reactive Metal Penetration (RMP) technique [12-14]. The other system used was a composite consisting of AlN/Al which was fabricated using a reactive infiltration technique [15]. These systems were used because of the potential ability to control their microstructures, that is to control the volume fraction of the metal phase and the size and size distribution of the metal ligaments. In addition, because of the different processing routes, the strength of the interface between the metal and the ceramic is expected to be different.

In the RMP technique controlled aluminum content Al_2O_3/Al composites were fabricated in two steps: first ceramic preforms were cast using a powder with a prescribed alumina:silica ratio and densified, and then the dense ceramic preforms were reacted with molten aluminum. The ceramic preforms were prepared by slip casting a water-based dispersion containing 25 weight percent solids, 0.1 weight percent dispersant (Darvan C, R.T. Vanderbilt, Norwalk, CT), and up to 1.0 weight percent of an acrylic wax emulsion binder (Rhoplex B60A, Rohm and Haas, Philadelphia). To control the aluminum content of the reactively-formed composite, the silica:alumina ratio of the preform was varied from 2:3 to 2:1. Slips were prepared by dispersing the appropriate ceramic powders in water using a magnetic stir plate. Rectangular slabs approximately 6 to 8 mm thick were fabricated by slip casting for 30 minutes in a plaster of Paris mold. Casts were removed from the mold and dried under ambient conditions for 24 hours. To produce dense ceramic preforms for reactive metal penetration, the slabs were sintered at 1350°C for 3 hours in air.

Al_2O_3/Al composites were prepared by reacting the dense preform with aluminum pellets (99.9%, Johnson Matthey, Ward Hill, MA) equal in mass to the preform. Reactions were carried out at 1100°C for 12 hours in an alumina tube furnace under ultra high purity argon flowing at 1 liter per minute. To minimize the oxygen content in the furnace atmosphere, the argon was passed over titanium chips at 700°C before entering the alumina tube ("gettered argon").

In this technique the composite is formed by the reduction of SiO_2, in the dense preform, by molten aluminum following this reaction:

$$(4/3X + Y)\ Al + Al_2O_3 + X\ SiO_2 = (1 + 2/3X)\ Al_2O_3 + X\ Si + y\ Al \qquad (1)$$

The metal content in the final composite can be controlled by changing the silica:alumina ratio in the initial preform. This is a near net shape fabrication technique with the composite having the same size and shape as the initial alumina-silica preform. There is also some indication that the scale of the microstructure of the composite can be tailored by controlling the scale of the initial preform, i.e. by controlling the size of the silica and alumina components in the preform. The final metal composition consists of Al with some dissolved Si. The majority of the excess Si diffuses out of the composite into the Al source.

The other composite system investigated is an AlN-Al composite formed by the reactive infiltration of Al into a porous AlN preform [15]. In this technique, AlN is doped with B_4C and then heated to temperatures greater than 1700°C for a short time to allow the B_4C to react with the AlN. The preform is then heated to 1300°C in an O_2 free environment, in contact with molten Al. The B_4C enhances the wetting characteristics of Al on AlN and allows the Al to infiltrate into the porous preform. An investigation of the reaction mechanism will be presented in a future publication. The volume fraction of the metal in the composite can be controlled by varying the preform density, and the scale of the metal ligaments can be controlled by using AlN powders with different particles sizes and size distributions.

In this study AlN powders with two different particle size distributions were used. The powders used were Stark K (Hermann C. Starck Berlin, Laufenburg, Germany) and C-Axis (C-Axis Technology (Canada) LTD.), which will be referred to as K and C respectively throughout the remainder of this work. The particle size distributions of the two powders are shown in Figure 1. Both powders have similar mean particle sizes but very different particle size distributions. K has a much wider size distribution than C. One area of investigation in this work is the effect of the microstructure size distribution on the toughness and R-Curve of these composites.

The preforms used in this investigation were formed by pressing the B_4C doped AlN powders in a 38mm square die to ~35MPa. This resulted in AlN preforms of ~50% theoretical density. In order to obtain higher densities, the preforms were subsequently isostatically pressed to ~275 MPa. Iso-pressing produced preforms with ~60% theoretical density. Specimens with 60% theoretical density were fabricated from both the C and K powders and 50% theoretical dense samples were formed using only the K powders. These samples allowed an investigation of the effects of particle size distribution and volume fraction on the toughness and R-Curve behavior. After pressing the binder was burnt out be heating to 220°C at a rate of 0.25°C/min. This was followed by sintering in a N_2 atmosphere at 1750°C for 30 minutes and finally infiltration with molten Al, at 1300°C for 2 hours, in an environment of "gettered argon". Figure 2 shows optical photographs of representative microstructures for both the Al_2O_3/Al and AlN/Al composites.

III. MECHANICAL TESTING

In order to prepare samples for mechanical testing, first all of the excess metal was ground off and the specimens were machined flat and square using a diamond grinding wheel. Samples for compact tension experiments were then polished to a 1μm finish to observe the crack during testing. Holes were drilled in the samples and they were notched to a particular a/w according to ASTM 399 [17].

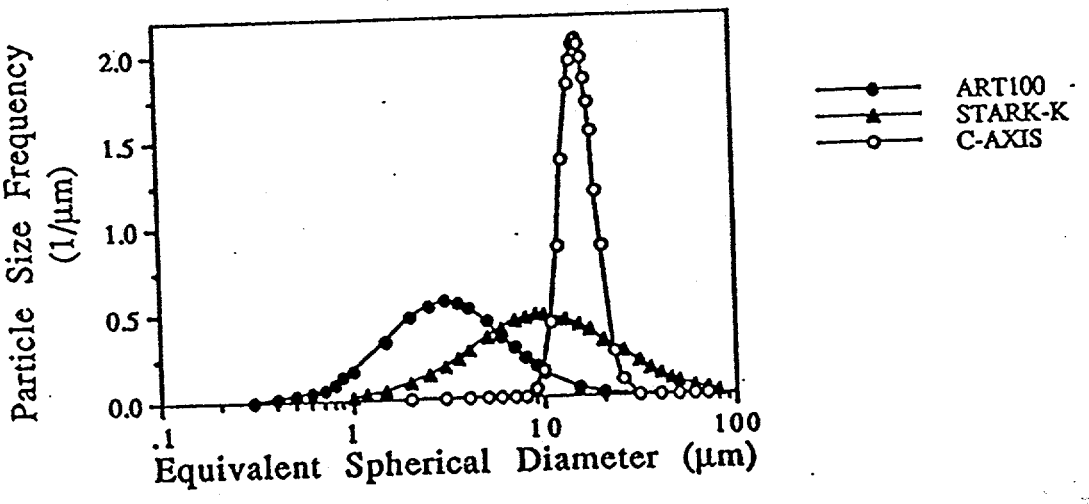
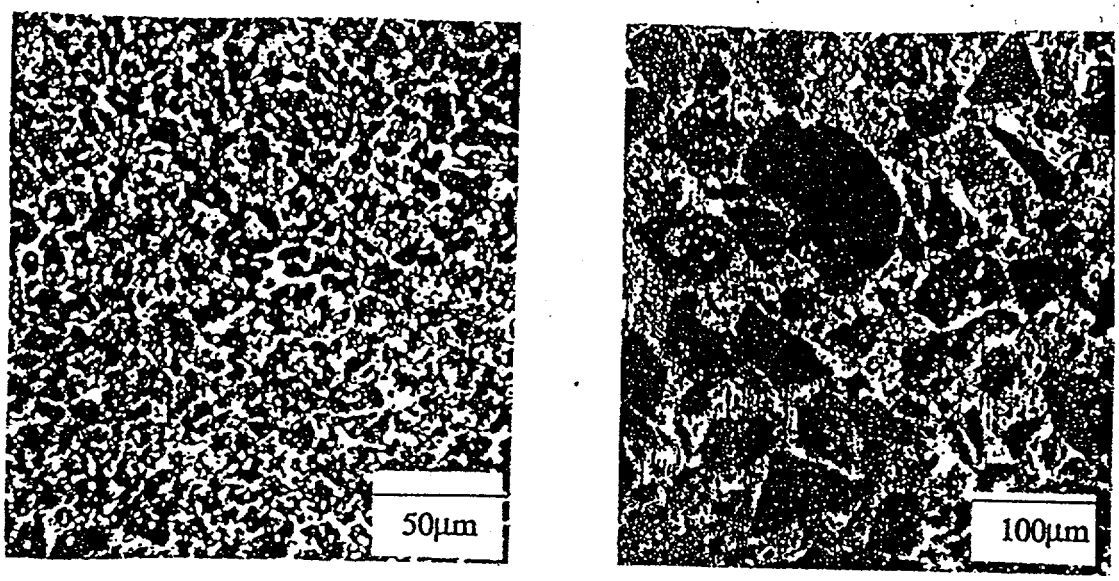


Figure 1. Particle size distributions for the C-Axis and Stark K AlN powders. [16]



(a) (b)

Figure 2. Optical photographs showing representative microstructures of (a) an Al_2O_3/Al composite sample and (b) an AlN/Al composite sample.

R-Curves were obtained by conducting compact tension (CT) experiments using a servo-hydraulic machine# under crack mouth opening displacement control, and the crack length was measured in-situ using an optical stereo microscope*. An organic based dye penetrant was used to facilitate crack observation. The load and crack length were measured at several points during the test. The toughness was calculated according to ASTM 399.

In addition to the crack growth resistance, work of rupture (WOR) was also measured for these materials[18]. The WOR was measured on at least 5 samples for each of the composites. Prior to notching the specimens, the modulus of the samples was measured using an ultrasonic method. This technique mechanically induces a vibration in the sample and then monitors the samples natural vibrational frequency, which can be related to its modulus (Grindo-Sonic MK4X by J.W. Lemmens, Inc.) [19].

A chevron notch was cut into bend bars to provide a sharp point from which the crack could initiate, and then grow stably. In order to determine the fracture toughness of the samples the strain energy release rate, G_{IC}, must first be calculated. G_{IC} is determined by dividing the area under the load displacement curve by the area of the fracture surface. G_{IC} can then be related to K_{IC} by Irwin's relation:

$$K_{IC} = (E' G_{IC})^{1/2} \qquad (2)$$

with $E' = E/(1-\nu^2)$, for plane strain, and a poisson's ratio of $\nu = 0.2$, was assumed for these systems. The work of rupture provides the maximum toughness of the composite which can then be compared to the peak toughness obtained using the compact tension specimens, to determine if steady state bridging has been achieved in the CT samples.

IV. RESULTS AND DISCUSSION

The results from the modulus measurements are shown in Figure 3. It is apparent that as the Al content decreases in the AlN samples, the modulus of the samples increases, and the modulus does not appear to depend on the size distribution of the AlN powder. The Al_2O_3/Al samples show a slightly higher modulus than the AlN samples, which is expected since the volume fraction of metal in those samples is only about 30%.

MTS Model No. 810 with a Microconsole 458.20 controller..
* Optical microscope Olympus SZH 10 Zoom Stereo Microscope.

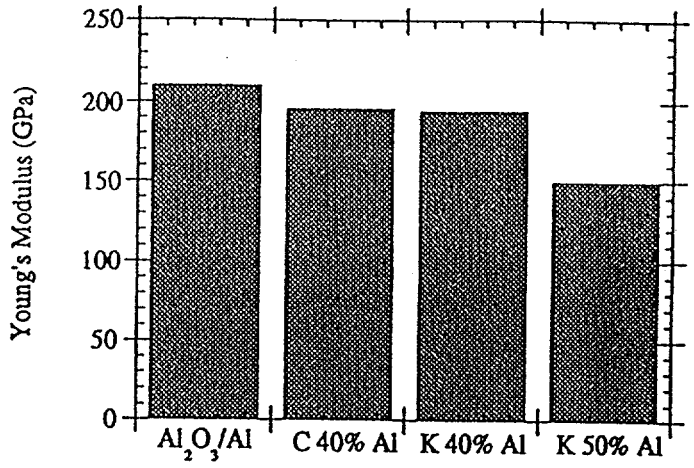


Figure 3. Results of Young's Modulus measurements.

Figure 4 presents the average values and the standard deviations in K_{IC} calculated from the WOR specimens using equation (2). As seen in previous studies the toughness of metal reinforced composites increases with the volume fraction of the metal phase [4,20-21]. For type K AlN/Al composites, the higher metal content samples showed a significantly higher toughness than the lower metal content composites, 15.2 $MPam^{1/2}$ and 9.5 $MPam^{1/2}$ respectively. The volume fraction of the metal phase is not the only parameter which affects the toughness of these composites. In the case of the Al_2O_3/Al composites their metal content is only ~30% but their toughness is approximately equal to the AlN samples with 40% metal. In addition to the difference in the volume fraction of the metal, the two types of composites have different ceramic phases, microstructures, and metal compositions. The difference in the ceramic phase was not expected to contribute significantly to the composites toughness. Therefore, the role of the microstructure and metal composition needs further investigation.

As shown in Figure 4 the Al_2O_3/Al specimens show a higher standard deviation in their toughness values then any of the AlN/Al specimens. This larger scatter in data can be attributed in part to large scale heterogeneities in the composite microstructure. These heterogeneities consisted of high metal content regions which may have affected the work of rupture values.

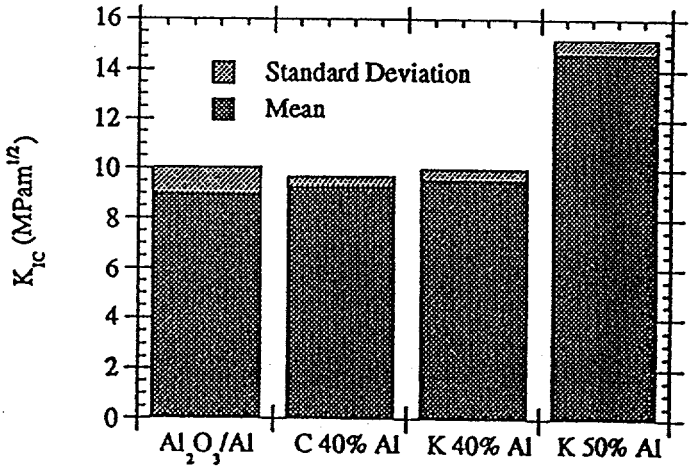


Figure 4. Peak toughness obtained by work of rupture.

Preliminary crack growth experiments have been conducted and the results are shown in Figures 5 and 6 for the AlN/Al and Al_2O_3/Al composites respectively. The lines connecting the data points are only to assist the visualization of the results. The relative peak heights for the different samples compares well with the WOR data. For the AlN/Al (Fig.5) composites the plateau toughness for the K 50% Al sample is significantly higher that the K 40% Al sample. Note that the C 40% Al sample shows a slightly lower peak toughness than the K 40% Al sample. This would seem to indicate that the wider particle size distribution in the K powder improves the composite toughness. The 30% Al, Al_2O_3 sample (Fig.6) shows a peak toughness similar to that measured using WOR, and as was seen in the WOR data the peak toughness is similar to that of the C 40% Al and K 40% Al composites. As before the apparent enhancement in toughness for the Al_2O_3/Al samples is probably due to the differences in microstructure and the metal composition.

Both composite systems show steeply rising R-Curves, and the crack growth needed to achieve steady state bridging was so small that it was difficult to measure it accurately using the *in-situ* optical microscopy technique. In order to obtain the full crack growth resistance curve, more accurate crack length measurement techniques will be required. Preliminary evidence suggests that for the AlN samples (Fig.5) the higher metal content composite shows a larger increase in a/W to reach peak toughness. Also the R-Curve for the Al_2O_3/Al sample (Fig.6) may indicate that this type of composite requires a larger increase in a/W to reach peak toughness then the AlN/Al composites, even with a lower metal content.

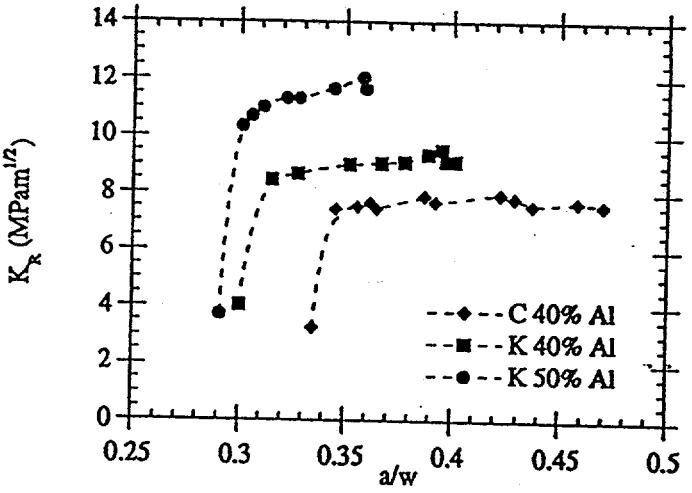


Figure 5. R-Curve for the AlN/Al composites.

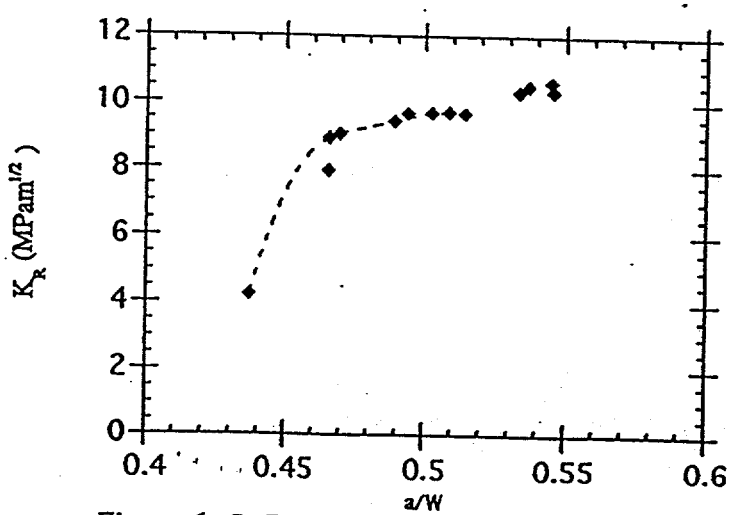


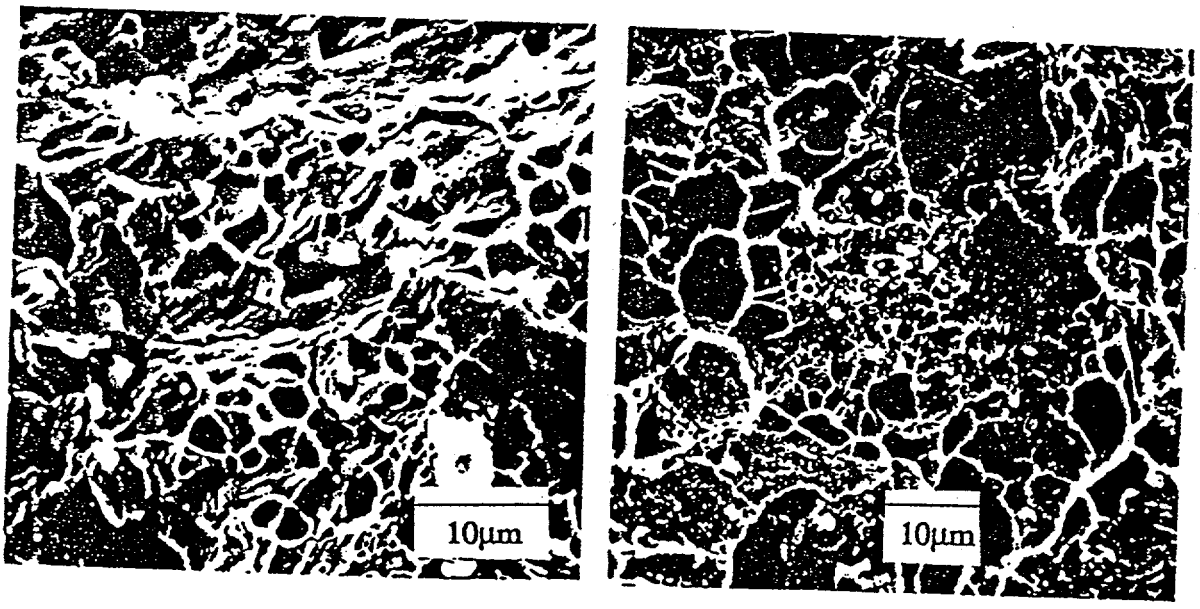
Figure 6. R-Curve for the Al_2O_3/Al composite.

The agreement between the toughness values measured using WOR specimens and the peak toughness measured using the compact tension specimens is not perfect. Some of this discrepancy is due to the difficulty in accurately measuring the length of the crack while it was growing. An organic based dye was used as a penetrant to show the cracks position more clearly, but the dye was difficult to observe and did not seem to penetrate fully to the tip of the crack. One possibility is that the bridging ligaments prevented the dye from reaching the crack tip. In addition, crack length measurements were made on one surface and we observed that in some cases, the crack growth was not planar. However, the trends in the measured peak toughness were the same for both the compact tension and work of rupture samples.

Figure 7 shows fracture surfaces of representative samples from the two systems. The metal ligaments shown in the Al_2O_3/Al system show little or no debonding between the metal ligand and the alumina matrix indicating that there is a strong bond between the metal and ceramic phases. The AlN/Al fracture surface shows slightly more debonding and shows greater stretching of the metal ligaments than the Al_2O_3/Al. The cause for this difference may be due to different processing techniques, and its role in governing the crack growth resistance of these composites needs further investigation.

V. CONCLUSIONS

The toughening behavior of two different ceramic-ceramic composite systems has been investigated. One system was an Al_2O_3/Al composite formed using the Reactive Metal Penetration technique which resulted in a composite with ~30v/o metal phase. The other system was formed by a reactive infiltration of Al into a AlN preform. In this system two different particle size distributions of AlN and two different preform densities were used. The results show that, in the AlN/Al system, as the volume fraction of the metal phase increases the toughness of the composite increases, an increase from approximately 9.5 to 15 $MPam^{1/2}$ for an increase in metal content from 40 to 50 volume percent. Preliminary results indicate composites with a wider microstructure size distribution have higher toughness than those with a narrower distribution, for the same volume fraction of metal. Other factors that control the toughness of metal reinforced ceramics are the strength of the interfacial bond and the metal composition. The two composites in this investigation provide systems in which these factors can be varied.



(a) (b)

Figure 7. Fracture surfaces of WOR specimens: (a) Al_2O_3/Al and (b) AlN/Al.

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