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Effect of inclusions on size of surface flaws in glass–crystal composites

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Indentation fracture studies were conducted on three sodium borosilicate glasses containing a dispersed phase of alumina inclusions with different degrees of thermal expansion mismatch between the glass matrices and the alumina. The alumina inclusions were found to cause a significant decrease in the size of the indentation cracks compared to those in the glass. This effect was greatest at the higher values of indentation load, which resulted in cracks of dimensions of sufficient size that their propagation was impeded by the tougher alumina dispersions. The fracture toughness for the composite samples calculated from the indentation data showed a significant increase with increasing crack size. For the smallest cracks in these composites, the value for fracture toughness was well below the value obtained in an earlier study by the single-edge notch-beam technique. The fracture toughness for the larger crack sizes which interacted with the alumina dispersions showed excellent agreement with the notch-beam data. The residual stresses due to the thermal expansion mismatch appeared to lead to a slight increase in the mean crack size regardless of the direction of thermal expansion mismatch.

1. Introduction

Composites consisting of a glass matrix with a dispersed crystalline phase can exhibit property values superior to those of the glass matrix alone. This was demonstrated by experimental data for fracture energy [1], fracture toughness [2, 3], strength [1, 4], elastic moduli [5, 6] and thermal conductivity/diffusivity [7].

The improvement in strength obtained by dispersing a crystalline phase in a glass matrix has been attributed to the associated increases in elastic moduli [6], increases in fracture energy due to crack pinning [8, 9] and increases in fracture toughness due to crack deflection [10]. Internal stresses due to mismatches in the coefficients of thermal expansion of the glass and the crystalline dispersed phase were also thought to play a role [11].

Decreases in the size of the surface (Griffith) cracks in the glass matrix due to the presence of the crystalline dispersions was suggested to act as an additional mechanism of strengthening [5]. Such surface cracks would be created by surface preparation or during inadvertent surface damage. Based on this concept of limitation of flaw size, a theory was developed for the dispersion-strengthening of brittle matrices, appropriate for crack sizes of the order of the mean interparticle spacing (mean free path) within the matrix between the crystalline dispersions. The theory was supported by experimental data for hot-pressed composites of a sodium borosilicate glass containing alumina dispersions over a range of values of the mean free path. In view of the observed dependence [12, 13] of the strength of crystallized glasses on crystallite size, the

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concept of dispersion-strengthening by flaw size limitations is possibly appropriate to the general class of glass–ceramic materials as well. Direct experimental evidence for the flaw-size limiting effect of the crystallites was presented recently [14] for surface flaws introduced by the indentation-fracture method in a series of cordierite glass–ceramics.

The purpose of the present study was to use the indentation fracture method to investigate the possible flaw-limiting effect of crystalline dispersions in glass matrices.

2. Experimental details

2.1. Materials

Samples of three glass matrices with alumina inclusions, hot-pressed at 730° C, identical to those used for earlier investigations and described in detail elsewhere [2, 7] were selected for this study. The alumina inclusions were in the form of spheres with a radius of $25 \pm 7\,\mu m$ and ranged in volume fraction from 0 to ~35 vol %. The glass matrices consisted of sodium borosilicate glasses which contained 75 mol % SiO_2 with molar ratios of B_2O_3/Na_2O of 0.2, 0.67, and 1.3, for convenience hereafter referred to as 0.2, 0.67, and 1.3 glasses respectively. These glass compositions were chosen to yield coefficients of thermal expansion which differed from the corresponding value for the alumina by $+2.7$, $+0.7$, and $-3.7 \times 10^{-6}(°C)^{-1}$ respectively. These thermal expansion mismatches would result in internal stress of different sign and magnitude on cooling the composite from a stress-free condition, and could have an effect on the size of the flaws.

2.2. Introduction of surface flaws

Cracks were introduced at room temperature in diamond-polished surfaces by means of the hardness indentation method using a Vickers indenter with loads ranging from 0.98 to 19.6 N. Following polishing, the specimens were annealed at 300° C for 4 h in order to reduce or eliminate surface stresses introduced by the polishing action. For each composite sample at least ten indentations were made for each value of indentation load. The locations of the indentations were randomly distributed on the surface. The crack size was defined as the distance between the centre of the indentation and the crack tip, which yields the value of the radius of the median crack [15]. The sizes of the cracks were measured directly with the optical system of the hardness indenter with 5 min after making the indentation. Photomicrographs of typical indentations for purposes of illustration were made by scanning electron microscopy. Values for the fracture toughness were calculated according to the expression given by Niihara *et al.* [16] for the median crack configuration. Because of the lack of a theory for the indentation-fracture behaviour of composite materials, it was assumed that the composites behaved as a continuum. Accordingly, the fracture toughness was calculated using experimental values for Young's modulus [2] and the mean values for the experimental data for the dimensions of the indentations and surface cracks.

3. Results, discussion and conclusions

Fig. 1 shows typical indentations in the 0.67 glass with 6.7 vol % alumina dispersions. The indentation shown in Fig. 1a obtained at a load of 9.8 N is in a region free of alumina inclusions, with the crack propagating unimpeded in the glass matrix. Fig. 1b shows an indentation at a load of 9.8 N with a crack arrested at the glass–alumina interface. Fig. 1c shows an indentation in the glass matrix which resulted from a load of 19.6 N with a crack which propagated towards and through an alumina inclusion. Fig. 1d shows an indentation at a load of 4.9 N made within an alumina particle near the interface, with two cracks which propagated across the interface into the glass matrix.

In general, crack propagation into and/or through an alumina particle from an indentation in the glass matrix was found to be most prevalent for those particles which at the surface exhibited a diameter significantly smaller than the mean particle size. This effect is expected for two reasons. The propagation of an indentation crack through a small particle is expected to occur more readily than through a larger particle. Also, a particle with a small intersection at the specimen surface can represent a thin remnant of a larger particle nearly totally removed during surface preparation. Crack propagation through such a thin "cap" is expected to be controlled primarily by the crack propagation in the underlying glass matrix. It should be noted that indentations within the inclusions occasionally

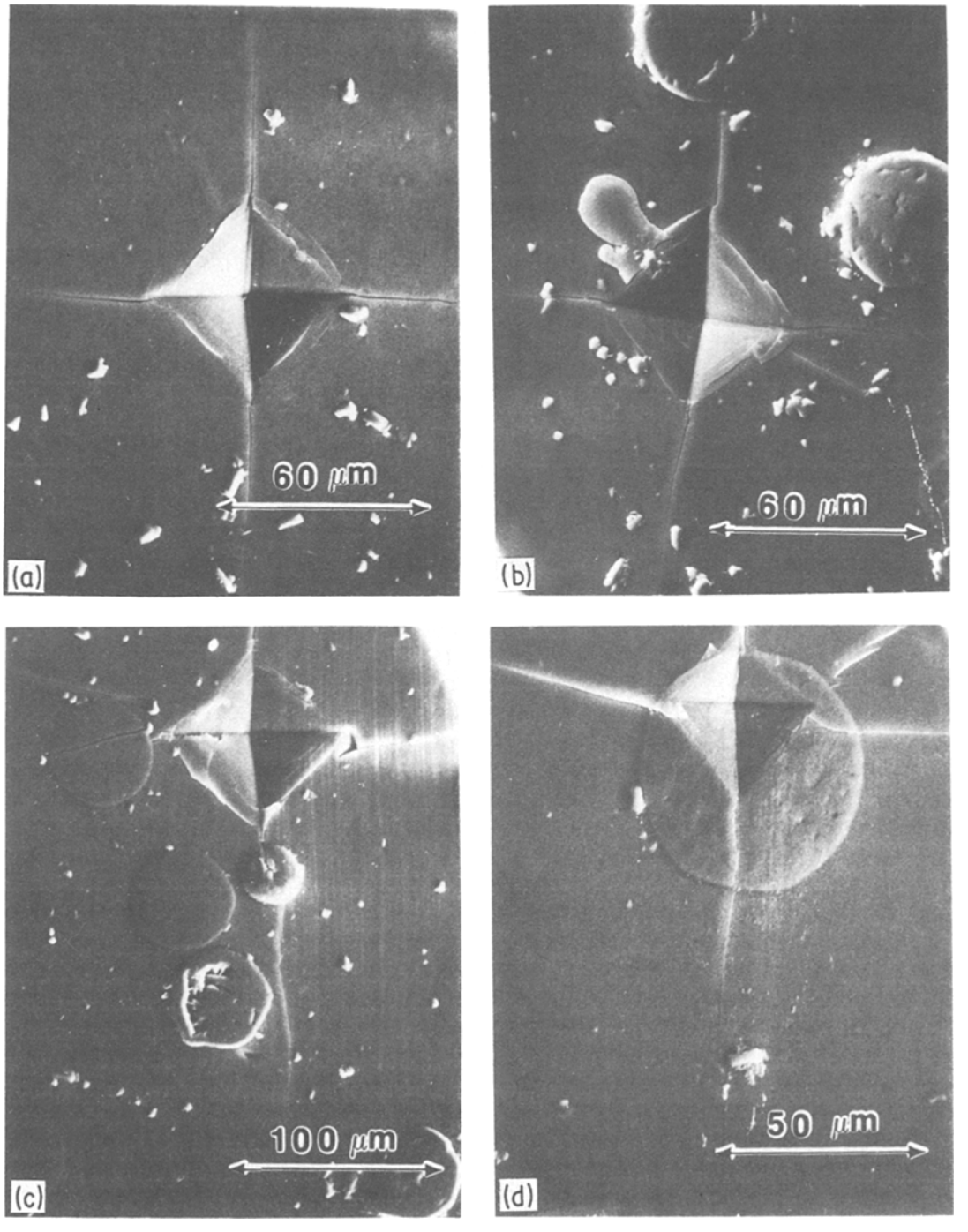
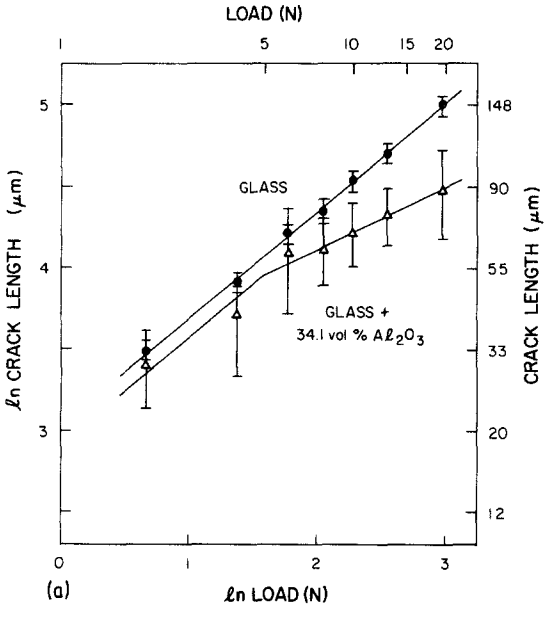
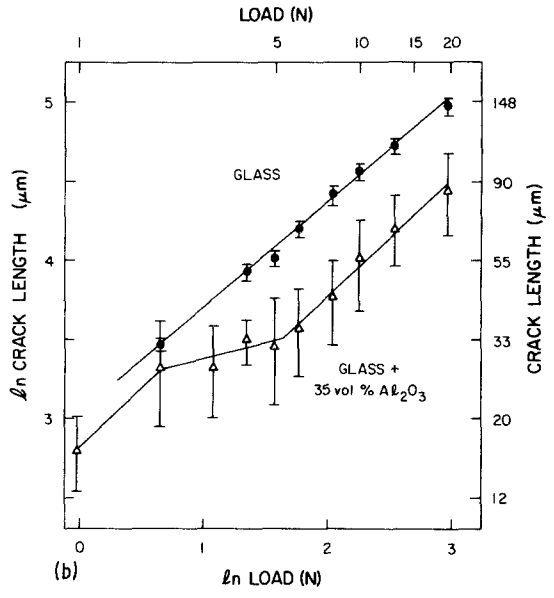


Figure 1 Indentation cracks in 0.67 glass with 6.7 vol % alumina dispersions made with a Vickers diamond indenter at loads of (a) 9.8 N; (b) 9.8 N; (c) 19.6 N and (d) 4.9 N.

resulted in deformation by crushing rather than by plastic flow. Such crushing is attributed to the presence of pores within some of the particles, evident in the scanning electron micrographs of Fig. 1. Such deformation by crushing on occasion also occurred in the glass matrix at the highest values of load, possibly due to the presence of an alumina particle located close to the surface underneath the site of indentation.

Figs. 2a, b and c show double-logarithmic





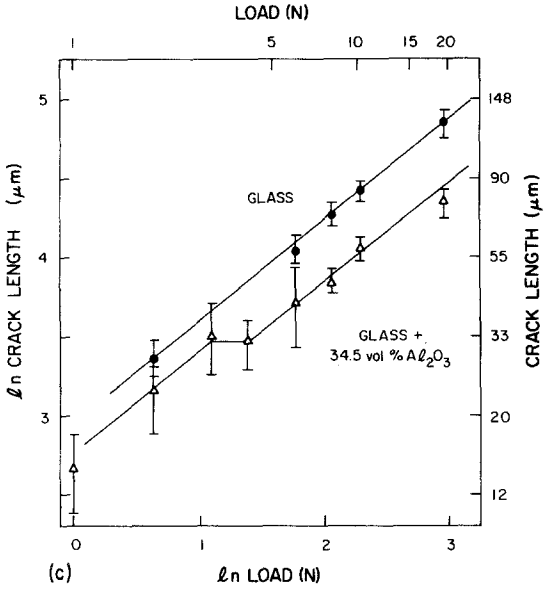
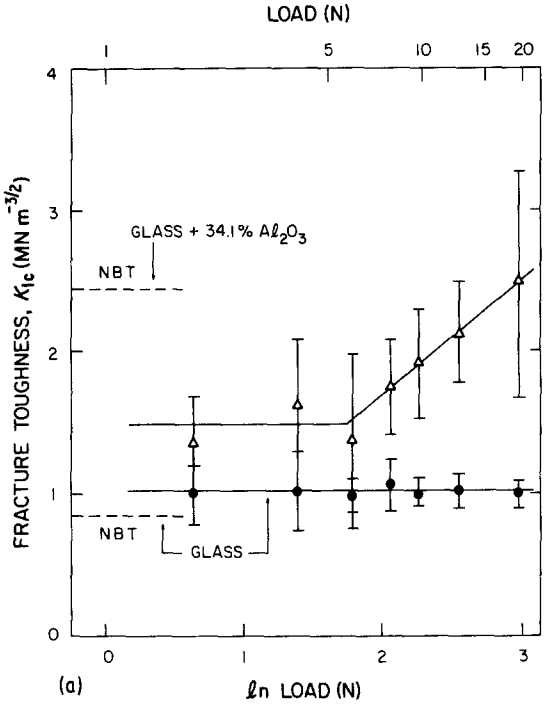
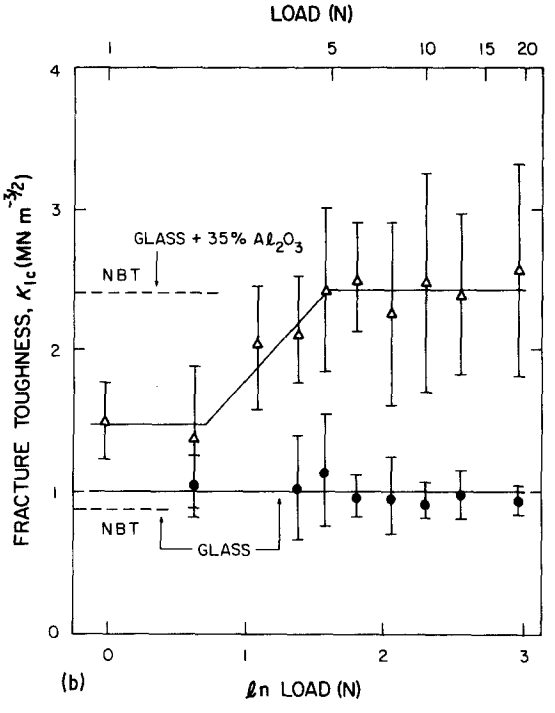


Figure 2 Dependence of surface crack size on indentation load for glass matrices with and without alumina inclusions: (a) 0.2 glass, (b) 0.67 glass, (c) 1.3 glass.

plots of the dependence of crack size on indentation load for the 0.2, 0.67 and 1.3 glass matrices, respectively, without and with about 35 vol % alumina. For all three sets of data for the glass matrices without inclusions the logarithm of the crack size depends linearly on the logarithm of the indentation load, with a slope near the value of 2/3 expected for median cracks [15].

For the glass matrices with alumina dispersions, as indicated by the data shown in Fig. 2, the crack size does not increase with increasing indentation load in a linear manner but is manifested by a discontinuity in slope at intermediate values of load. A similar discontinuity was observed also by Morena *et al.* [14] in the dependence of crack size on indentation load for a series of fully crystallized cordierite glass–ceramics. For the samples of this study, this discontinuity is thought to correspond to the size of cracks nucleated in the glass matrix for which the crack tips are arrested at the glass–alumina interface. The mean interparticle distance (mean free path) at 35 vol % alumina, as calculated by means of the expression of Fullman [17], for spherical inclusions with a radius of 25 μm is $\sim 62\ \mu$m. For the 0.67 glass with 35 vol % alumina, the discontinuity in slope occurs over a range of crack radii from ~ 30 to $35\ \mu$m with a crack diameter of twice this value, in excellent agreement with the value for the mean free path, and direct evidence of the existence of crack-pinning, also observed by Morena *et al.* [14]. It should be noted that the mechanism of crack-pinning, however, will be effective only for surface flaws introduced in the glass at values of indentation load or mechanically analogous mechanisms of surface damage, for which the flaws cannot penetrate into the tougher crystalline phase. For much higher values of indentation load at which cracks can penetrate a significant distance into or through the dispersions, their size is expected





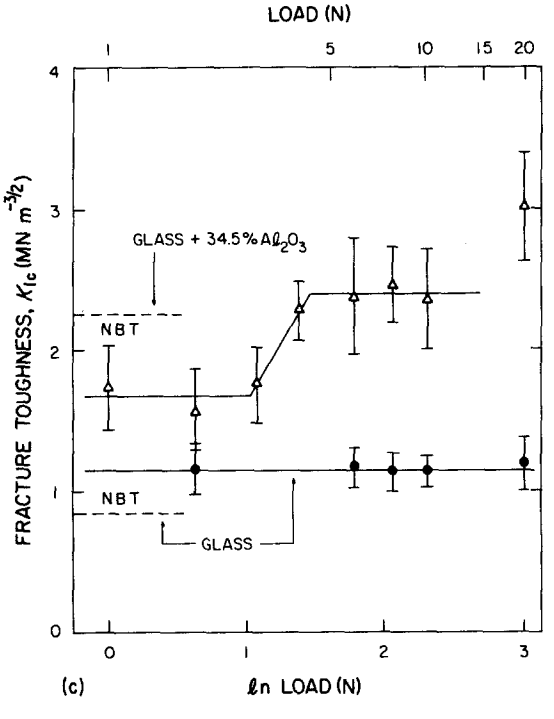


Figure 3 Indentation load dependence of fracture toughness of glass matrices with and without alumina inclusions: (a) 0.2 glass, (b) 0.67 glass, (c) 1.3 glass.

to be governed primarily by the toughness of the dispersed phase, rather than by pinning at the interface. The data for the crack size for intermediate volume fractions fell between those for 0 and 35 vol %. These data were not reported as they exhibited a less clear-cut trend than those for the composites shown in Fig. 2.

Figs. 3a, b and c show the values of the fracture toughness (K_{Ic}) for all three glass matrices without and with the ~ 35 vol % alumina, as a function of indentation load, calculated from the data for crack sizes in Fig. 2. For the glass matrices the fracture toughness is independent of load. In contrast, a significant increase in fracture toughness with increasing load (i.e. crack size) is observed for the glass matrices with the alumina dispersions; it occurs over the range of loads at which the data for the crack size as a function of indentation load show the discontinuity in slope as shown in Fig. 2. As discussed earlier, this effect arises because the larger cracks introduced at the higher values of load are more likely to interact with the much tougher alumina dispersions than the smaller cracks formed at the lower values of load, which are located primarily within the glass matrix.

Fig. 3 also includes the data for the mean values for the fracture toughness of the same materials obtained by the single-edge notch-beam method in an earlier investigation [2]. At the highest values of indentation load, the mean values of the present data for the fracture toughness of the glass–alumina composite samples

agree very well with the corresponding notch-beam technique (NBT) values. At the lower values of indentation load, however, the present data for the fracture toughness fall well below the NBT data and more closely resemble those for the glass matrix, as expected from the data for crack size on indentation load. For the glass without alumina dispersions the fracture toughness is independent of load and appears to be somewhat higher than the NBT value, although the difference is not significant in view of the scatter in data.

The highly heterogeneous nature of the glass–alumina samples in terms of the large differences in hardness, elastic moduli and fracture toughness is manifested in the relatively large scatter in data for the crack size and fracture toughness. This large scatter prevents drawing any conclusions as to whether the internal stresses due to the thermal expansion mismatch have a significant effect on the crack size and associated fracture toughness. Nevertheless, it should be noted that a comparison of all three sets of data reveals that the relative reduction in crack size due to the alumina inclusions for the nearly stress-free 0.67 glass matrix composite appears to be somewhat greater than for the other two glass–alumina composites with the higher state of internal stress. It should be noted also that the region of load over which the slope of the plot of crack size against indentation load exhibits discontinuous behaviour differs for the three composite systems. Possibly, this effect as well may be related to the magnitude and sign of the residual stress state. In this respect, it should be noted that attempts to obtain data for the 0.2 glass–alumina systems for indentation loads in excess of 20 N were unsuccessful. These tests were conducted to see if the data on fracture toughness for loads greater than 20 N would level off to the value of the NBT. Invariably, these tests resulted in crushing rather than the formation of well-defined indentation-crack systems, which prevented acquisition of the required data. Possibly, such chip formation was due to the nature of the residual stress in that particular composite.

The observed scatter in the toughness data for the composite samples shown in Fig. 3 may be attributable at least in part to the assumption that the measured values of Young's modulus for the composite samples were appropriate to the calculation of fracture toughness from the indentation data. The measured data, however, represent the continuum elastic behaviour of the composite samples, which is not likely to be representative of the local elastic response near an indentation with a dimension near the order of the scale of the microstructure of the present samples. Taking this effect into account requires the development of theories of indentation fracture of composite samples with components which differ in their elastic, plastic and fracture behaviour.

A final remark is in order with regard to measured values of hardness. Because of the high relative difference in hardness of the glass and alumina phases a large scatter in hardness data would be expected. For the 0.2, 0.67 and 1.3 glass matrices the mean hardness values ranged from 450, 520 and 570 diamond-pyramid hardness (DPH) respectively, with corresponding values for standard deviation of about 5%. No significant dependence on load over the total load range was found. For the composites with ~ 35 vol % alumina the corresponding data were 627, 839 and 831, with coefficients of variation of 43, 29 and 35%, and maximum values of 735, 1218 and 1195, for the 0.2, 0.67 and 1.3 glass matrices respectively. These maximum values are well below the values of hardness of fine-grained, fully-dense polycrystalline alumina [18]. Of interest to note is that these maximum values of hardness were observed for those indentations on or near the interface, rather than those entirely within an alumina particle. Possibly, such high values at the interface are the result of the asymmetric distributions of the plastic deformation around the indenter. This could result in a shear force on the indenter perpendicular to its direction of motion, thereby limiting the depth of penetration. A theoretical analysis of this effect is required to check the validity of this hypothesis.

The results of this study are of general relevance to the fracture mechanics of continuous-matrix particulate-composite materials. Theories [8, 9, 10] for the fracture energy or toughness have generally considered mechanisms of toughening without associated changes in crack size. The theory [4] of dispersion-strengthening of brittle matrices is based on the reduction in the size of the cracks, without consideration of the accompanying changes in fracture

toughness. The results of this study indicate that decreases in flaw size and increases in fracture toughness occur simultaneously. Both these effects must be considered in assessments of the failure response of continuous-matrix particulate composites. In this respect, the observation that the toughness depends on crack size may well be most critical. For carefully prepared samples of such composites without major inadvertent surface damage, the failure-initiating flaw may well be located entirely within the low-toughness matrix. In that case, care should be taken in making failure predictions based on data for fracture toughness measured by a technique which relies on specimens with macrocracks such as those used in the NBT, or in double-torsion or short-rod methods. In this respect the indentation-fracture method, which allows measurement of the fracture toughness of relatively small cracks, should be regarded as particularly advantageous.

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