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Toughening in very high pressure sintered diamond–alumina composite

T. NOMA, A. SAWAOKA
Research Laboratory of Engineering Materials, Tokyo Institute of Technology, Nagatsuta, Midori, Yokohama 227, Japan

Alumina ceramics and dispersed diamond were sintered at a temperature and pressure where the diamond phase is stable. The fracture toughness of the composite is remarkably increased after heat-treatments in vacuum. The transformation from diamond into graphite accompanied this phenomenon. The effects of the fraction of diamond and the heat-treatment temperature are investigated.

1. Introduction

A great deal of effort has been spent in order to toughen ceramics, which are ssentially brittle. One of the mechanisms applied is microcrack toughening which was first considered by Glucklich and Cohen [1]. Claussen first reported the toughening of alumina related to the transformation from tetragonal to monoclinic of dispersed zirconia particles [2] and Evans and Heuer applied fracture mechanics to interpret this phenomenon in terms of transformation toughening [3]. On the other hand, Kreher and Pompe considered microcrack toughening for this sytem on the basis of a volume change of about 6% during the transformation, acting as the driving force of nucleating microcracks in the matrix [4].

There are still some problems concerning the definition of the microcrack toughening mechanism. One is that the "microcrack" has not been clearly observed [5]. As the electron ray transmissivity of zirconia is much lower than that of alumina, it is very difficult to observe the microstructure by transmission electron microscopy (TEM). With a view to observing the microcrack more clearly, it is useful to disperse a second-phase particle which has a similar electron ray transmissivity to matrix alumina. In the present investigation, carbon is used as the second-phase particle and the volume change during transformation from diamond to graphite is utilized as the driving force of microcracking [6]. In addition, it should be noted that a large volume increase of about 50% during transformation paves the way for the reduction of the dispersed phase fraction in the matrix.

Diamond-dispersed alumina composite was sintered at a temperature and pressure at which the diamond phase is stable and then heat-treated in vacuum in order to transform the diamond into graphite. The Vickers hardness and fracture toughness are examined.

2. Experimental procedure

Sintering was performed under very high pressure using a slide-type cubic anvil apparatus [7]. Synthetic diamond* with an average grain size of 0.8 μm and alumina† with a size of 1.0 μm, were mixed with ethyl alcohol in an alumina mortar. Then 6 vol% camphor was added as a binder, and the composite was pressed at 1 GPa to form tablets 7.0 mm high and 7.1 mm diameter. These tablets were heated at 0.1 Pa and 500° C for 2 h and placed in high pressure cells (see Fig. 1). The pyrophyllite cube and tube were heated at 500° C in air for 1 h, and the carbon heater was treated at 0.1 Pa and 1000° C for 2 h before use. The mixtures were sintered at 6 GPa and 1300° C for 1 h. Tablets 6 mm high and 6.2 mm diameter were recovered. In addition, some of the composites were heat-treated at 0.1 Pa and 1100° C or 1300° C for 6 h.

*General Electric, USA.
†Meller, USA.

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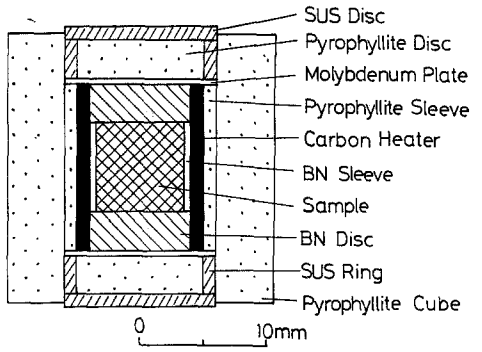

Figure 1 Cross-section of the high pressure sample assembly.

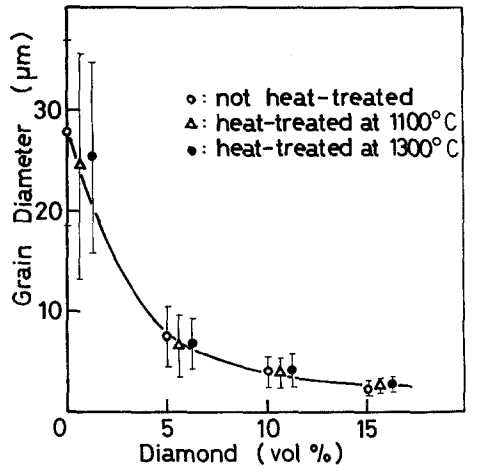

Figure 3 Grain size of alumina.

The relative density of the composites was measured using the Archimedean method. They were lapped with 0.25 μm diamond paste and the Vickers hardness measured with a load of 10 kg. Evaluation of the fracture toughness was done using the Vickers indentation method [8] with a load of 30 kg. Powder X-ray diffraction of the composite was examined. SEM observation of the fracture surfaces was also done.

3. Results and discussion

According to the X-ray diffraction investigation, about 15% diamond was transformed into graphite during the high pressure sintering. After the heat-treatment, the ratio of the transformation was about 75 and 85% for 1100 and 1300° C, respectively. The ratio can be increased by a higher temperature and longer duration of heat-treatment.

A typical fracture surface of the composites observed by SEM is shown in Fig. 2. The diamond composition of both samples is 15 vol %. Sample a was heat-treated at 1300° C and sample b was not heat-treated. Intergranular fracture was observed in both samples. Diamond particles are well dispersed in the alumina matrix, and no noticeable reaction can be observed between diamond and alumina during the high pressure sintering. However, the transformed graphite cannot be seen in this micrograph.

The grain size of alumina measured from micrograph is shown in Fig. 3. The size is remarkably decreased with diamond dispersion. However, it is not increased during heat-treatment at either 1100 or 1300° C. The grain size of the diamond is not increased and remains at the original size.

The relative density of the composite is shown in Fig. 4. The ideal density is calculated on the basis of a diamond–alumina system. The density of the heat-treated composites at 1100 and 1300° C are in the range 89 to 93%. This is interpreted as being caused by the transformation of dispersed diamond into graphite. The Vickers hardness of the composites is shown in Fig. 5. The hardness decreases with increasing volume fraction of diamond in the range 10 to 15 vol %.

The results of the Vickers indentation measurements are shown in Fig. 6. Relative fracture tough-

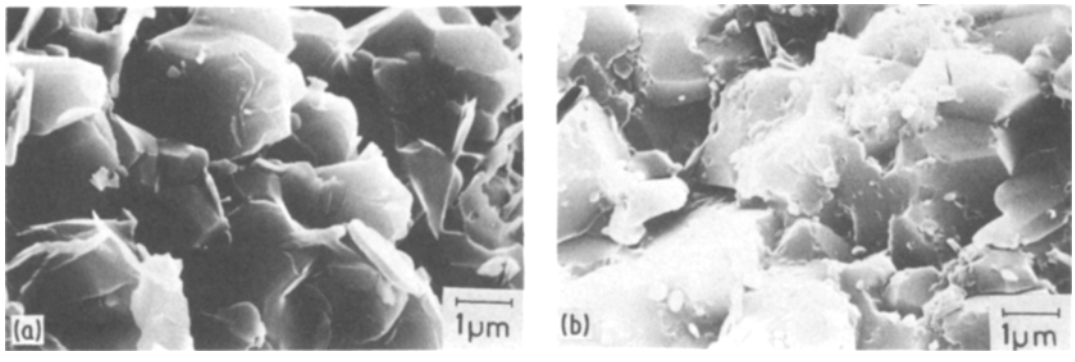

Figure 2 A typical fracture surface of the composites.

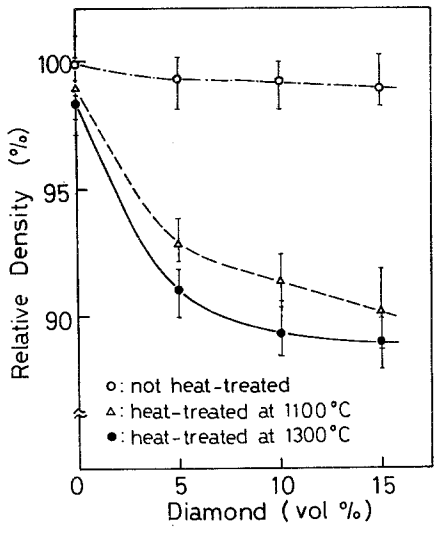

Figure 4 Relative density of the samples.

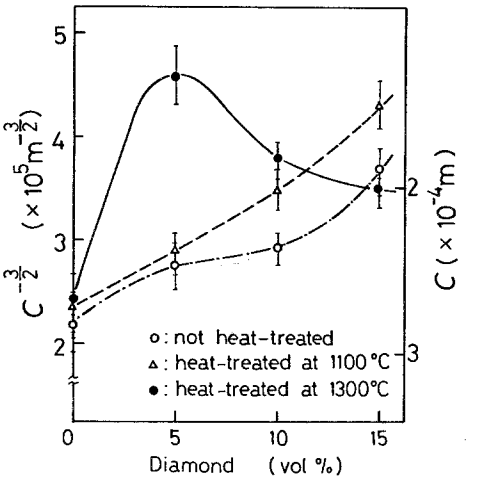

Figure 6 Fracture toughness of the composites.

ness is evaluated from the value $C^{-3/2}$ according to the relation given by Lawn *et al.* [8].

$$(K_{IC}/Ha^{1/2})(H/E)^{1/2} = 0.028(C/a)^{-3/2}$$

where K_{IC}, H, E, a and C are the fracture toughness, the hardness, Young's modulus, the half-length of the impression diagonal, and the indentation crack length, respectively. For the composites heat-treated at 1300° C, the maximum toughness was found at 5 vol% diamond composition. For composites heat-treated at 1100° C, the fracture toughness is increased slightly in comparison with those not heat-treated in the range of volume fraction of diamond up to 15 vol%.

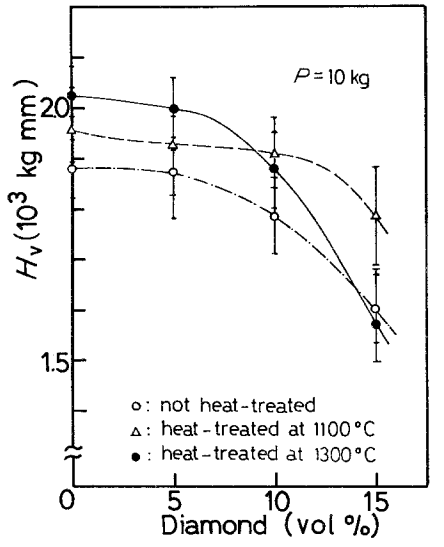

Figure 5 Vickers hardness of the composites.

In terms of microcrack toughening, the relation between the fracture toughness and the volume fraction second phase for 1300° C heat-treated composites is similar to the results for the zirconia–alumina system found by Claussen [2], although the dispersed material is completely different. In addition, it is suitable to consider that the microcrack toughening is dominant in this composite heat-treated at 1300° C according to the analysis performed by Kreher and Pompe [4]. However, as no quantitative analysis for the relation between microcracking and the fracture toughness has been done, the ratio of microcrack toughening to the whole mechanism cannot be evaluated.

On the other hand, for heat-treated composites at 1100° C, the maximum fracture toughness in microcrack toughening [4] is not achieved up to 15 vol% diamond. It is thus difficult to estimate whether microcrack toughening or another mechanism, e.g. deflection toughening, is dominant in the composite heat-treated at 1100° C. This difference in fracture toughness behaviour due to the heat-treatment temperature cannot be interpreted by the ratio of the diamond–graphite transformation, because the magnitude of the induced stresses during the transformation calculated according to Selsing [9] are similar, e.g. for radial compressive stress, 3.2 and 3.5 GPa for 1100 and 1300° C, respectively; i.e. the condition for stress-induced microcracking [10] is almost equivalent. Thus there should be some microstructural alternation beside the diamond–graphite transformation, e.g. creep deformation of the matrix.

4. Conclusion

The fracture toughness of a diamond–alumina composite, which is sintered at a temperature and pressure at which the diamond phase is stable, is increased by heat-treatment in vacuum. The degree of toughening can be controlled by either a dispersed diamond fraction or the heat-treatment temperature. The dominant toughening mechanism is supposed to be microcrack toughening which results from the increasing transformation of dispersed diamond into graphite during heat-treatment.

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