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The effect of annealing on fracture toughness, strength and microstructure of hot-pressed alumina

The effect of microstructure on fracture toughness of ceramics is not firmly established in spite of numerous investigations of the influence of both grain size [1-3] and porosity [3, 4]. In particular, reports on porosity are contradictory. Coppola and Bradt [4], using the work-of-fracture method of testing [5], found that up to 10% porosity did not affect toughness of hot-pressed alumina. Simpson [3] using cold-pressed and sintered alumina and a fracture mechanics technique (analytical notched beam test [6]), found that connected porosity reduced the fracture toughness. Only connected porosity was studied because the sintering process did not yield closed porosity until porosity levels dropped below 4%. In addition, the porosity distribution was frequently non-uniform. The work-of-fracture method was also used but some doubt was raised concerning the accuracy of this method when applied to high strength ceramics. Hence, in this work, an analytical technique has been used to determine the effect of closed porosity on fracture toughness of hot-pressed alumina.

Alcoa XA-16 alumina* powder was hot-pressed in graphite dies in vacuum at a pressure of 35 MN m^{-2} for 1 h at temperatures ranging from 1300 to 1450°C, yielding slabs about 30 × 25 × 6 mm^3. About 75% of the powder was treated with isopropyl alcohol prior to hot-pressing to remove surface adsorbed water carefully following the procedure outlined by Rossi and Fulrath [7]. Hot-pressing yielded final densities as shown in Fig. 1.

The hot-pressed slabs were cut into bars 25 × 6 × 3 mm^3 and either tested as-hot-pressed or after annealing in groups of two or three for 3 h at temperatures ranging from 1500 to 1750°C.

When the high density (> 96% T.D.), as-hot-pressed material was annealed, a reduction in density occurred accompanied by an increase in the porosity. The degree of this density reduction increased with annealing temperature and in some extreme cases caused blistering of the material due to the growth of a few very large pores. This increase in porosity did not occur when low density as-hot-pressed material was annealed; instead, a dense (98 to 99% T.D.) body was produced. Immersion of the as-hot-pressed material in dye penetrant indicated that the material whose density increased upon annealing had connected porosity in the as-hot-pressed

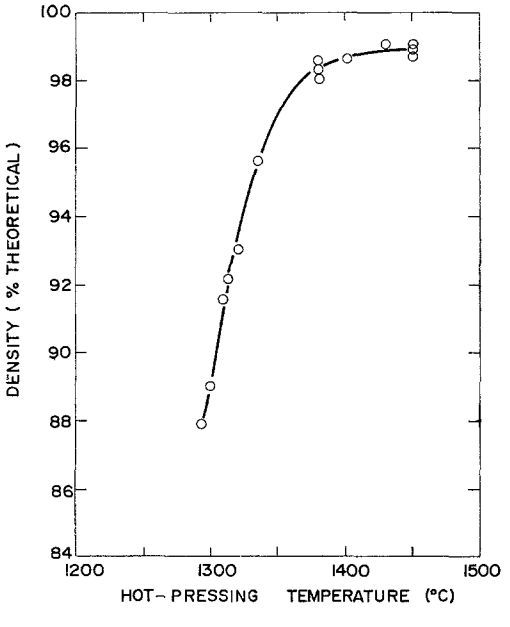

Figure 1 Final density as a function of hot-pressing temperature.

*Aluminum Co of America, Pittsburg, Pa. 15219, USA.

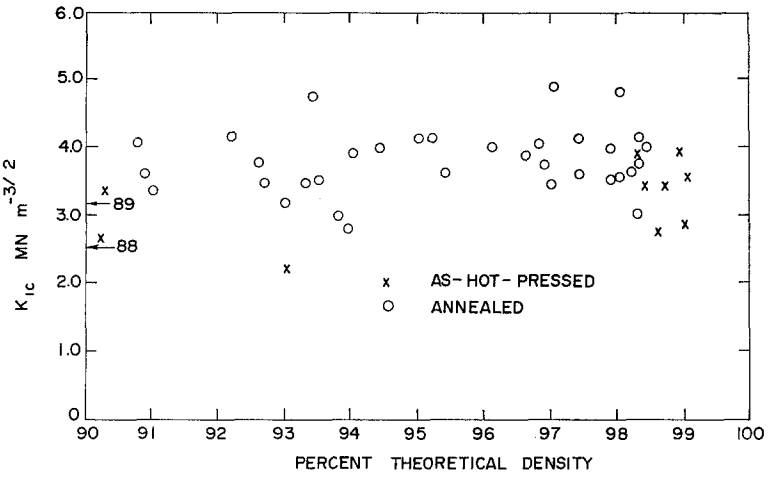

Figure 2 Fracture toughness versus porosity for hot-pressed and annealed alumina.

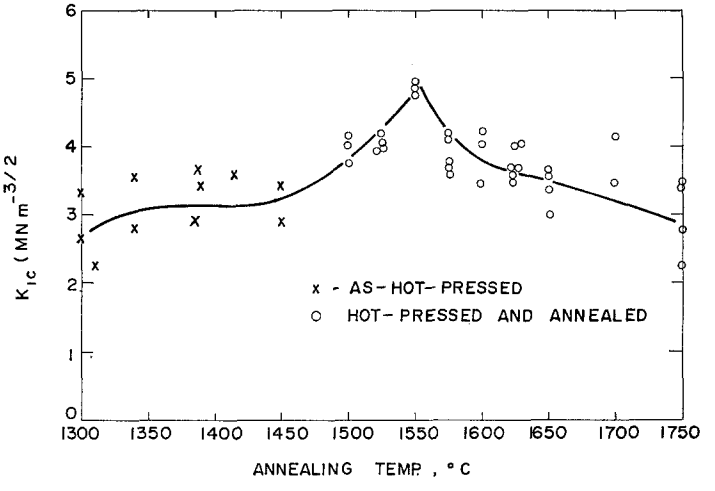

Figure 3 Fracture toughness versus heat-treatment temperature for hot-pressed and annealed alumina.

condition whereas the material which degraded had closed porosity. The additional porosity produced by annealing dense material was unconnected even for volume fractions greater than 10%.

The grain size of the as-hot-pressed material was about 0.5 μm while that for the annealed material ranged from 1.5 to 4 μm except for the specimens annealed at 1750°C, which had grain sizes of about 10 to 15 μm.

The porosity produced by annealing was both intergranular and intragranular and roughly spherical in shape. The mean pore size increased with annealing temperature and the porosity was uniformly distributed with respect to both size and location, except for the occasional large bubble in the higher temperature anneals.

Fracture toughness measurements were carried out using the notched beam technique in four point bending [6]. Some additional slabs were used for strength measurements (modulus of rupture) using four point bending.

Fracture toughness is plotted against porosity in Fig. 2. There is considerable scatter in the data. Each circle corresponds to two or three specimens which were annealed together after being cut from a given slab. The crosses represent two or more specimens tested in the as-hot-pressed condition. Variations in grain size are not thought to be responsible for the scatter in Fig. 2. The range of grain sizes is small (0.5 to 4.0 μm for most points), and earlier work suggests that fracture toughness is not very grain size sensitive in this range [3].

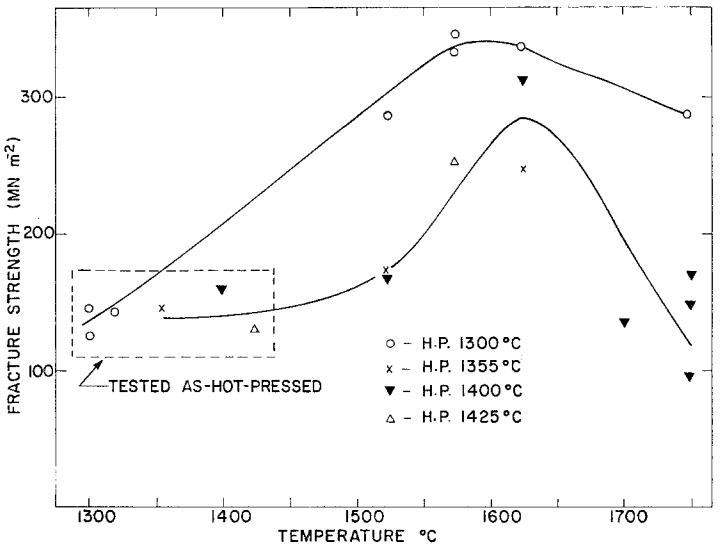

Figure 4 Fracture strength (modulus of rupture) versus heat-treatment temperature for hot-pressed and annealed alumina with approximate hot-pressing temperatures.

When the data is plotted against final heat-treatment temperature correlation is much better (Fig. 3). The hot-pressing temperature does not appear to affect the toughness of the as-hot-pressed material but annealing at moderate temperatures appears to maximize the toughness. Above 1550°C the toughness drops and the scatter increases. The toughness of the annealed material was not dependent on hot-pressing temperature. A similar trend with temperature is followed by the strength measurements, Fig. 4, except that material hot-pressed at 1300°C (connected porosity) is stronger when annealed than material hot-pressed at higher temperatures.

The as-hot-pressed material by itself (crosses in Figs. 2 and 3) is in good agreement with the results of Coppola and Bradt, i.e. porosity has little effect on toughness and their fracture energy of 12.5 J m^{-2} converts to K_{IC} = 3.1 MN $m^{-3/2}$ in good agreement with our results. Subsequent changes in toughness following annealing is probably due to evolution of adsorbed surface impurities. It thus appears that the strong porosity dependence of toughness reported for cold-pressed and sintered material [3] was a result of the connectivity and non-uniform distribution of porosity in that material.

The problem of adsorbed gases on powder surfaces is a common one in hot-pressing and is often believed to prevent attainment of theoretical density [8]. Rossi and Fulrath considered water to be the main problem and patented a process [7] for removing adsorbed water by immersing the powder in a non-polar solvent such as isopropyl alcohol. This treatment was not effective in the present work and no difference in final products was observed between treated and untreated powder.

Rice [9] studied the hot-pressing behaviour of a number of alumina powders and observed some porosity production upon annealing, including blistering in extreme cases. Knudsen cell experiments suggested that a number of gaseous species were evolved at temperatures above 1400°C including OH, CO, CO_2 and mass number 34 which he suggested was H_2S. It seems reasonable to assume that the porosity produced by annealing dense hot-pressed specimens is caused by the evolution of such impurities. Because the material hot-pressed at 1300°C had connected porosity the gases were probably able to escape before densification was complete, hence new porosity was not produced by annealing this material.

The fracture mode was primarily intergranular for all specimens underlining the importance of surface impurities in this investigation. The fact that fracture toughness correlates better with annealing temperature than any microstructural feature suggests that the annealing treatment improves the bonding between grains possibly by increasing contact areas and eliminating impurities on the powder surfaces. The increase in strength (Fig. 4) with annealing temperature also supports this hypothesis. Rice [10] has drawn similar conclusions for MgO which also

exhibits a strength maximum. The reduction in fracture toughness above 1550°C could result from two factors. For the dense as-hot-pressed material, the high gas pressures generated internally during the higher temperature anneals could nucleate some microcracks and weaken the material by providing low energy crack paths. This may also have occurred to a lesser extent in the material hot-pressed at 1300°C if annealing at the higher temperatures caused the material to sinter up before all the impurities had escaped through the connected porosity. In fact, the densities of this material annealed at 1750°C were slightly lower than that for material annealed at 1550°C. In addition, the fracture toughness-grain size effect reported for cold-pressed and sintered material [3] could be taking effect as the grain size begins to increase rapidly. This effect (amounting to a 15% decrease in K_{IC} between grain sizes of 4 and 20 μm) is not sufficient to account for the entire decrease in fracture toughness above 1550°C, hence, both effects probably contribute. The variability of the above effects coupled with the location of the occasional large pore on the crack path probably accounts for the increased scatter in Fig. 3 above 1600°.

The strength after annealing of material hot-pressed at 1300° is consistently higher than that for material exhibiting pore growth (Fig. 4). Since the fracture energy behaviour for the two types of material are indistinguishable, the difference in strengths must be due to differences in flaw sizes. The porosity produced by annealing must contribute to a larger effective flaw size. The drastic reduction in strength after annealing at 1750° occurs in material with very large pores and is consistent with this assumption. The strength drop above 1550°C for material hot pressed at 1300°C is more moderate and likely reflects only a drop in fracture toughness.

In summary, the degree of connectivity and distribution of porosity is important in determining its effect on fracture toughness in alumina. For as-hot-pressed material the work-of-fracture results of Coppola and Bradt are confirmed. It is also suggested that surface impurities are an important factor in the determination of fracture toughness of alumina and should be considered in any investigation of microstructural effects.

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The effect of solidification microstructure on the corrosion behaviour of a grain-refined aluminium-copper alloy

The corrosion behaviour of a unidirectionally solidified binary aluminium-4.5 wt % copper alloy in an air-saturated saline environment has recently been investigated [1]. In the solutionized and solutionized-and-aged conditions, grain-boundary attack and intergranular pitting was observed similar to that occurring in corrosion of solutionized wrought alloys [2]. However, in the as-cast condition there was preferential attack of the copper-rich portion of the cored dendritic structure, specifically the α-phase containing more than 3.2 wt % copper. Potential measurements on homogeneous alloys indicated that the α-phase in this range is anodic relative to the θ-phase (Al_2Cu) and to α in the concentration range of 1.6 to 3.2 wt % copper. While α containing less than 1.6 wt % copper also behaves anodically, it did not occur in the examined microstructure of 4.5 wt % copper alloy.

In practice, cast aluminium alloys are grain-refined by innoculation and exhibit an as-cast