

THE ROLE OF CRYSTALLIZATION OF AN INTERGRANULAR GLASSY PHASE IN DETERMINING GRAIN BOUNDARY RESIDUAL STRESSES IN DEBASED ALUMINAS

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ABSTRACT

The influence of microstructure on the crack resistance (R-curve) behavior of a commercial debased alumina containing large amounts of glassy phase (28 vol %) has been studied using the Indentation-Strength test. The effect of two microstructural variables, viz. grain size and the nature of the intergranular second phase (glassy or crystalline) has been evaluated. Crystallization of the intergranular glass was carried out in order to generate residual stresses at the grain boundaries, which have been shown to enhance R-curve behavior in ceramic materials. Enhancement of the R-curve behavior was observed with the increase in grain size. However, no effect of the nature of the intergranular second phase on the R-curve behavior, in small and large grain materials, was observed. The results from characterization of these materials using various analytical techniques is presented, together with possible explanations for the observed effects.

INTRODUCTION

Recently several researchers [1 to 4] have reported considerable increase in toughness of debased (liquid-phase-sintered) aluminas, containing 10 to 30 vol % intergranular glass, by crystallization of the glass via simple heat-treatments. However, it should be noted that the toughness measurements in the above studies were performed at single crack length values, whereas recent work has shown that many alumina ceramics show increase in toughness with crack length, i.e. crack resistance or R-curve behavior [5 to 10]. The effect of crack resistance has been attributed to the phenomenon of microstructural grain-localized bridging of the crack, in the wake of its tip, and is very sensitive to the microstructure of the material [5, 11]. In the present study we have set out to evaluate the effect of grain size and crystallization of the intergranular glass on the mechanical properties (R-curve behavior) of these materials over a wide range of crack lengths.

The grains bridging the crack, in non-cubic polycrystalline ceramics, have been postulated to be clamped in the matrix by compressive residual stresses generated during cooling from processing temperatures, due to thermal expansion anisotropy present in these materials [12]. These residual stresses play an important role in the bridging phenomenon. The grain size of the polycrystalline ceramic is also known to influence R-curve behavior of these materials [7,13].

Indentation-strength [7, 8] is a very convenient way of monitoring R-curve behavior in ceramic materials. Bending over of the fracture stress versus indentation load curve at the low indentation load end is a direct consequence of R-curve behavior exhibited by the material, and gives rise to a region where the fracture stress is independent of the indentation load (and hence crack size) [15, 16]. An important implication of R-curve behavior, therefore, is that it suggests a degree of flaw tolerance, which is very useful in terms of engineering design. R-curve behavior also has great significance in the wear properties of materials, since this is governed by fracture characteristics at low flaw sizes. More recently it has been postulated that R-curve behavior increases the Weibull modulus of ceramic materials which exhibit such behavior [17,

18].

The purpose of this study was to determine the separate and combined influence(s) of grain size and second phase crystallinity on the R-curve behavior of debased ceramics. The material chosen was Coors AD85 alumina containing about 28 vol % glass. This material was subjected to carefully designed heat-treatments so as to increase the grain size and increase residual stresses at the grain boundaries by crystallization of the intergranular glass. It was envisaged that in doing so, it would be possible to enhance R-curve behavior in these materials.

EXPERIMENTAL

About 300 samples of AD85 alumina in the form of disks (25mm dia x 3mm) were obtained from Coors Ceramic Company. A series of heat-treatments was carefully devised in order to produce four sets of samples of differing microstructures. Tables I and II show the details of the heat-treatments and the resulting microstructures respectively. The denotation S or L refers to small or large grain size respectively, and C or G refers to crystalline or glassy second phase respectively.

Table I Various heat-treatments, AD85 subjected to.

<u>Material</u>	<u>Heat-treatments</u>	<u>Purpose</u>
AD85-S-G	a) As-received	-
AD85-S-C	a) 1400°C for 6 hours, quenched b) 1150°C for 130 hours	Homogenize intergranular glass Crystallize intergranular glass
AD85-L-G	a) 1550°C for 250 hours	Increase grain size
AD85-L-C	b) 1200°C for 130 hours	Crystallize the intergranular glass

Table II Microstructural aspects of AD85 after the heat-treatments.

<u>Material</u>	<u>Grain size</u>	<u>Intergranular phase</u>
AD85-S-G	3 μ m	Glassy
AD85-S-C	3 μ m	80% crystalline
AD85-L-G	18 μ m	Glassy
AD85-L-C	18 μ m	80% crystalline

Specimens for Transmission Electron Microscopy (TEM) were prepared from the above samples using a dimpler and then ion-beam milling until perforation. TEM investigation was performed on a Phillips EM 400T at an accelerating voltage of 120 keV. Chemical composition of the intergranular glass was determined using Scanning Transmission Electron Microscopy (STEM) and Energy Dispersive Spectroscopy of x-rays (EDS) on the same instrument. Samples were prepared for Scanning Electron Microscopy (SEM) by polishing sections to 1 μ m grade followed by thermal etching at 1500 °C for 15 minutes.

Mechanical testing of AD85-S-G, AD85-S-C, AD85-L-G and AD85-L-C was carried out as follows. About 50 disk samples of each were polished to 1 μ m grade on the prospective tensile side. A Vickers indentation was made at the center of the polished surface with loads varying from 2 to 300 N. Some samples were left unindented. The samples were broken in biaxial flexure using the 3-point support and punch fixture. Details of this particular method of mechanical testing have been described elsewhere [7].

RESULTS AND DISCUSSION

Table III shows the composition of the intergranular glass of AD85-S-G samples after homogenization heat-treatment, as determined by STEM and EDS. This is an average of many different spectra obtained from different regions of the sample. The compositions were observed to be fairly consistent, which implies that the glassy phase is homogeneous. The composition obtained agrees closely with that determined by Weiderhorn et al. [19] for AD85 with the same heat-treatment. Using this composition as the basis, the heat-treatment given in table I was devised. Figure 1 show SEM micrographs of AD85-S-G (grain size $3\mu\text{m}$) and AD85-L-G (grain size $18\mu\text{m}$). Figures 2 and 3 show TEM micrographs of AD85-S-G and AD85-S-C showing glassy and crystalline intergranular phases respectively. The grain size did not change appreciably during crystallization heat-treatment. The crystalline intergranular phase in AD85-S-C was observed to be mostly anorthite. With this composition it was not possible to achieve 100% crystallinity, thus pockets of residual glassy phase were observed at the triple points.

Table III Average composition of the intergranular glass in AD85.

<u>Oxide</u>	SiO ₂	Al ₂ O ₃	MgO	CaO	BaO
<u>Wt%</u>	56.5	27.5	2.1	8.6	5.3

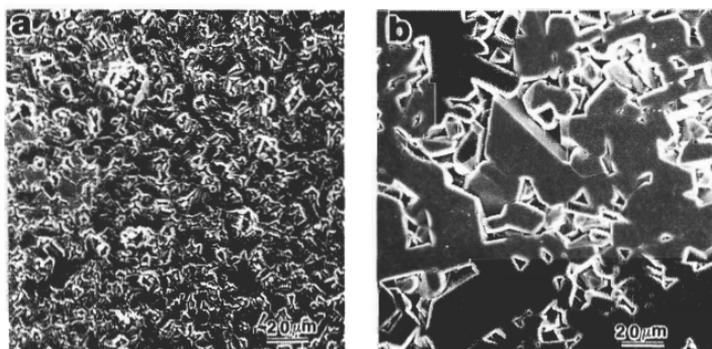


Figure 1 SEM Secondary electron images of polished and etched sections of AD85 aluminas a) AD85-S-G (fine grained material) , b) AD85-L-G (coarse grained material).

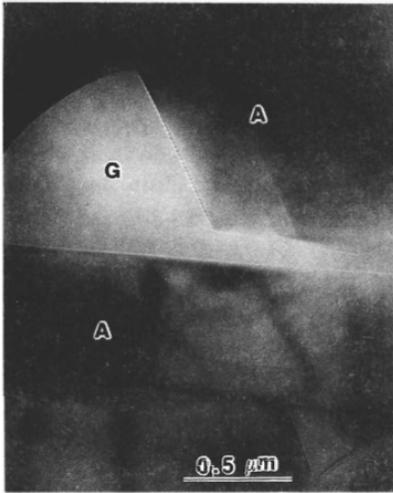


Figure 2 TEM bright field image of AD85-S-G showing intergranular glassy pockets (A-Alumina, G- Glass).

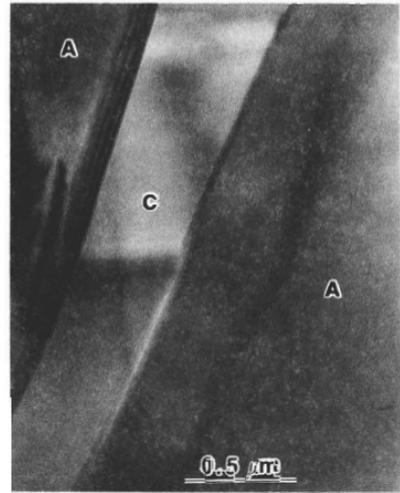


Figure 3 TEM bright field image of AD85-S-C showing crystalline intergranular phase (A-Alumina, C- Crystalline).

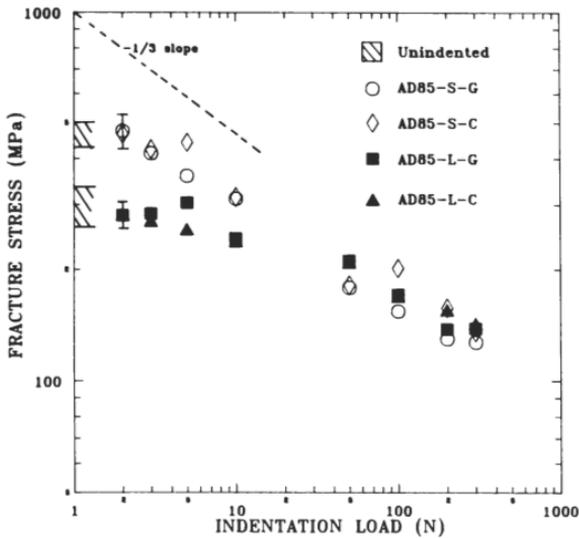


Figure 4 Plot of indentation load versus fracture stress for four different materials, derived from AD85. The error bars for all the data are shown in the left. The hatched region represents failures from natural flaws.

Figure 4 shows indentation load versus fracture stress for AD85-S-G, AD85-S-C, AD85-L-G, and AD85-L-C. It can be seen that all the data essentially falls on two graphs corresponding to 1) fine grained materials (AD85-S-G and AD85-S-C) and 2) coarse grained materials (AD85-L-G and AD85-L-C). It can be clearly seen that coarse grain aluminas show much more pronounced R-curve behavior than fine grain aluminas.

Because the curves for materials with glassy and crystalline intergranular phases show similar behavior, this indicates marginal or no effect of crystallization on the R-curve properties of these materials. The effect of grain size on the R-curve behavior, however, is much more marked, and this is in agreement with results obtained by Cook et al.[7] for single phase aluminas, and Bennison et al. [20] for debased aluminas. Given that grain-bridging processes must be dependent on the residual stresses in the grain boundary regions, the lack of effect of crystallization on the R-curve behavior is somewhat surprising.

Possible explanations for the lack of effect of crystallization of the intergranular glass are thought to be as follows:

1. Stress relaxation by residual glass
2. Fracture through the residual glass
3. Stress relaxation by high temperature deformation (twinning) of anorthite.

Work is currently underway to determine which of the above mechanisms (if any) are correct [21].

CONCLUSIONS

The major conclusions from the above study can be summarized as follows:

1. The effect of grain size on the R-curve behavior predominates.
2. Crystallization of the intergranular glass has relatively little or no effect on the R-curve behavior of AD85. Possible explanations attributing to the observed behavior have been described above.
3. An important conclusion can however be drawn from the observed behavior. Coors AD85 can be subjected to prolonged heat-treatment cycles up to 1200°C without having any significant effect on room temperature mechanical properties (pertaining to fast crack growth). This property of AD85 can be very useful in prolonged high temperature structural applications and in metallization applications imparting heat-treatment tolerance along with flaw tolerance.

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