

Effect of Heat Treatment on Crack-Resistance Curves in a Liquid-Phase-Sintered Alumina

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The effects of heat treatment on the R-curve (crack-resistance) behavior of a commercial liquid-phase-sintered (LPS) alumina have been studied using the indentation-strength test. An enhancement of the R-curve characteristic of this LPS alumina is obtained by a treatment that increases the scale of the microstructure. The enhanced R-curve characteristic leads to the desirable property of flaw tolerance, albeit at the expense of a diminished strength at small crack sizes. The implications of these findings are discussed with reference to processing and design strategy. [Key words: alumina, sintering, cracks, mechanical properties, strength.]

SEVERAL workers have reported that the toughness of liquid-phase-sintered (LPS) aluminas can be improved by suitable heat treatments.¹⁻⁴ Those workers attributed the changes to modification of residual thermal expansion mismatch stresses or to crystallization of the amorphous intergranular phase. Such claims deserve detailed attention because they open up the prospect of tailoring mechanical properties via simple heat treatments.

Implicit in these previous studies, however, is an assumption which is now known to be restrictive; i.e., that the "toughness" is a single-valued material quantity. Recently it has been shown, using indentation-strength⁵⁻⁷ and double-cantilever-beam⁸⁻¹⁰ techniques, that the toughness of alumina (and other) ceramics is not generally single-valued, but tends to increase with increasing crack size (*R*-curve behavior). The extent of the increase is found to depend critically on the microstructure, with the grain size and the nature of the intergranular phase the apparent controlling parameters.

The form of the *R* curve has significant implications for structural applications.¹¹ In particular, flaw tolerance becomes an important design factor. One

interesting feature of the alumina data collected by Cook *et al.*⁵⁻⁷ is the tendency for the *R* curves to cross each other, corresponding to an inverse relationship between large-scale and small-scale toughness values (a result of special consequence to wear resistance¹²). Hence, in evaluating the significance of toughness "improvements," it is important to specify the crack size range over which measurements are made. In this context, we note that the previous studies¹⁻⁴ of the effect of heat treatment on toughness were generally made at "large" crack sizes: i.e., large with respect to the scale of the microstructure. A complete assessment of the changes in mechanical properties requires a determination of the entire *R* curve.

Accordingly, the aim of the present study was to investigate the effects of microstructural changes resulting from simple heat treatments on the *R*-curve behavior of a LPS alumina. We use the indentation-strength technique because of its special usefulness in the investigation of *R*-curve characteristics at small as well as large crack sizes.⁵

EXPERIMENTAL PROCEDURE

A commercial LPS alumina[†] containing ≈10 wt% (≈18 vol%) intergranular second phase was chosen for the study. The samples were provided as disks, 25 mm in diameter and 2 mm thick, suitable for biaxial flexure testing.

Heat treatments (HT) of the as-received material were conducted in air using a MoSi₂ resistance furnace according to the schedules in Table I. The aims of the heat treatments were to (1) vitrify the second phase without changing the grain size (HT-1), (2) recrystallize the intergranular phase without changing the grain size (HT-2), and (3) increase the grain size with a controlled (vitrified) in-

tergranular phase (HT-3). A heating and cooling rate of 250°C/h was used for all firings.

The following specimen characteristics were determined: (1) the degree of crystallinity and composition of the intergranular phase, using transmission electron microscopy (TEM) and energy-dispersive X-ray microanalysis (X-ray EDS); (2) grain size, using scanning electron microscopy (SEM) with a lineal intercept method;¹³ and (3) density, using the Archimedes method. The specimen characteristics resulting from the heat treatments are included in Table I.

The prospective tensile face of each specimen was diamond polished to a 1-μm finish prior to mechanical testing. Most of the disks were indented at the face centers with a Vickers diamond pyramid at contact loads of 2 to 300 N. Indentations were made through a piece of carbon paper to mark the contact sites. Some specimens were left unindented as controls. All indentations were made in air and the samples allowed to stand for 10 min. The biaxial strength tests were made using a flat circular punch, 4 mm in diameter, on three-point support, 20 mm in diameter.¹⁴ A small drop of silicone oil was placed on the indentations prior to testing, and failure times were kept below 20 ms to minimize effects from static fatigue. Strength values were calculated from the breaking loads and specimen dimensions using thin-plate and beam formulas.^{14,15} Care was taken to examine all specimens after they fractured to verify the contact site as the origin of failure. Unsuccessful breaks were incorporated into the data pool for unindented controls.

RESULTS

Figure 1 plots the results of the mechanical tests of the various heat-treated

Table I. Heat Treatments Used for Liquid-Phase-Sintered Alumina and the Resulting Material Characteristics

Material	Anneal temp. (°C)	Time (h)	Grain size (μm)	Density (Mg·m ⁻³)	Second phase*
As-received			4.2	3.61	A/C
HT-1	1600	8	4.8	3.63	A
HT-2	1600	8	4.8	3.64	C
	1200	48			
HT-3	1600	196	24.2	3.60	A

*A is amorphous and C is crystalline.

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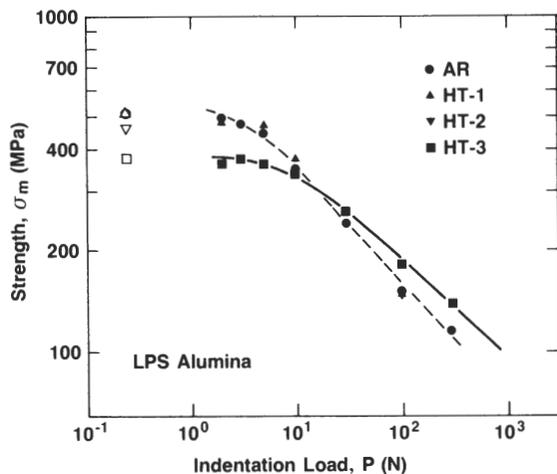


Fig. 1. Results of the indentation–strength tests for alumina specimens subjected to the heat treatments in Table I. Open symbols at left represent strength values for specimens that broke from natural flaws. Curves through data are fits to as-received (dashed line) and HT-3 (solid line) materials; note data for HT-1 and HT-2 materials are indistinguishable from those of the as-received.

aluminas, inert strength, σ_m , versus indentation load, P . Each point on the plot represents the mean of at least 10 specimens per load. The standard deviation for each point is $\approx 11\%$. (Error bars are omitted from the plot for clarity.) From the fracture-mechanics analyses based on a crack-interface bridging model,^{16–18} the flattening of the response may be interpreted as an enhancement of the R -curve characteristic. The curves through the data in the figure are best fits to the as-received (dashed line) and HT-3 (solid line) materials, respectively, from such analyses. The results in Fig. 1, in conjunction with

the microstructural characteristics illustrated in the SEM photograph in Fig. 2, enable us to deduce the effect of the heat treatments on the mechanical behavior.

The as-received material has a grain size of $\approx 4 \mu\text{m}$, as shown in Fig. 2(A). The individual grains are surrounded by a continuous second phase, which is partially crystalline in form. All the classical features of a LPS material, such as wetting of grains, pockets of amorphous phase, and faceted grain structures, are evident. Microanalysis indicates that the intergranular phase consists primarily of silicates of calcia, magnesia, and alumina.

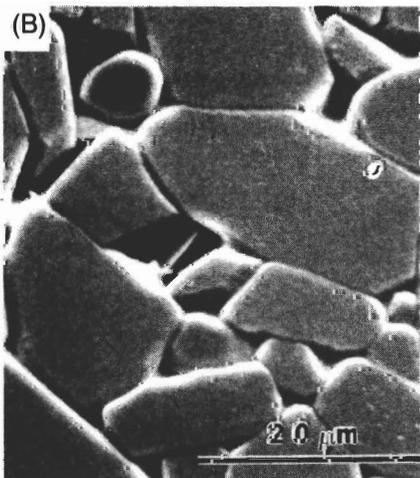
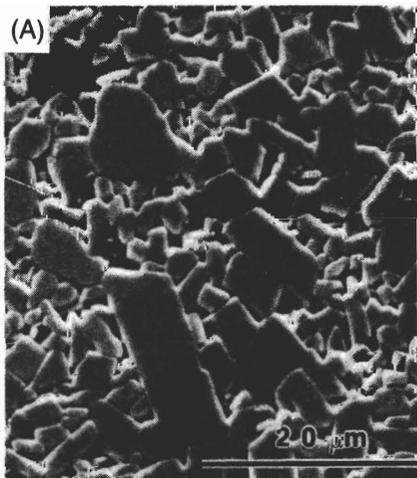


Fig. 2. S.E.M. photographs of a liquid-phase-sintered alumina for (A) as-received material and (B) following heat treatment HT-3 designed to increase the grain size. These specimens were first polished, then thermally etched at 1500°C for 1 h in air to reveal grain structure. (Pockets of intergranular phase were removed by this preparation.) Note the sixfold increase in grain size in the latter material. Faceting of alumina grains is apparent in both micrographs.

Note from the dashed line in Fig. 1 that the $\sigma_m(P)$ response of this control material deviates only slightly from the $P^{-1/3}$ dependence appropriate to a single-valued toughness; i.e., the R -curve characteristic for this particular material is not pronounced.

Examine now the effects of changing only the degree of crystallinity of the intergranular phase on the mechanical behavior by referring to the data for the HT-1 and HT-2 materials. Analysis by TEM shows that the intergranular phases in these two materials are, respectively, completely amorphous and predominantly crystalline. However, the $\sigma_m(P)$ data points for HT-1 and HT-2 in Fig. 1 are virtually indistinguishable from the dashed-line fit for the as-received material. Thus, in these two cases, the effect of heat treatment on the R curve is insignificant.

The effect of increased grain size, on the other hand, is significant, as seen from the solid line fit for the HT-3 material in Fig. 1. The scale-up in grain size, from ≈ 4 to $\approx 24 \mu\text{m}$, is readily apparent from a comparison of the morphology for this material in Fig. 2(B) with the corresponding morphology for the as-received material in Fig. 2(A). The HT-3 material shows reduced strength at small indentation loads, with a distinctive plateau in the $\sigma_m(P)$ response in this region, and a countervailing increase in strength at large indentation loads. This third heat treatment has led to a noticeably stronger R -curve behavior.

DISCUSSION

The above results lead us to an important conclusion: *the toughness properties of ceramic materials can be modified by simple heat treatments.* For the alumina material studied here, the most significant modifications were achieved by a treatment that coarsened the microstructure (although the possibility of a contributing effect resulting from some subtle change in the grain-boundary toughness cannot be entirely discounted). More generally, this means that one may be able to adjust properties of as-received ceramic components before placement in service. In the present case, the desirable feature of flaw tolerance is obtained at the expense of a decreased strength in the region of small crack sizes (balanced somewhat by an increased strength at large crack sizes). An enhanced R -curve characteristic may not, however, always be beneficial, e.g., in applications in which maximum resistance to microfracture-controlled wear and erosion is a premium requirement.¹² We need also be aware that the toughness properties of ceramic components exposed to thermal cycles may change, for better or for worse (again, depending on the application), during service.

It is interesting to consider the findings here in the context of the previous studies,^{1–4} where no attempt was made to

define the operative region of crack size. Note that if our materials were to have been tested at a single, intermediate crack size, corresponding to an indentation load of ≈ 10 N, the change in R curve for HT-3 would have passed unnoticed. Alternatively, tests by two sets of experimenters operating at extremes of large and small crack sizes would have lead to totally opposite conclusions. The danger of toughness evaluations at a single crack size is that any such perceived improvement (or, indeed, degradation) may all too easily be misconstrued as universal.

This still leaves unanswered the question of the role of the heat treatment in relation to the underlying mechanism of the R curve. We alluded in the previous section to toughening by bridging.¹⁶⁻¹⁸ In this interpretation the increased crack resistance arising from the scale up of grain size may be attributed to an enhancement of frictional tractions associated with pull-out of grains bridging the interfacial walls in the wake of the crack tip. Residual stresses arising from thermal expansion mismatch¹⁻⁴ could play an important role in augmenting these frictional tractions by "clamping" the bridging grains into the alumina "matrix". A detailed description of this residual stress-induced friction process will be presented elsewhere.¹⁹ It

seems that, contrary to earlier suggestions,^{4,5} the degree of crystallinity is not necessarily the principal factor in determining the influence of such residual stresses on the R curve, at least in the type of LPS aluminas studied here.

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