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Effects of post-hot isostatic pressing on mechanical properties of presintered alumina ceramics

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Effects of post-hot isostatic pressing (post-HIP) on the elastic properties, strength and fracture toughness of different commercial alumina-based ceramics was investigated. The materials were presintered ceramics with alumina contents of 94, 97 and 99%. HIP was performed using a Mo or graphite furnace in a wide temperature range to establish regimes which allowed attainment of the best combination of mechanical properties, e.g. ultimate bending strength, Weibull's modulus, fracture toughness and modulus of elasticity. The results are discussed in relation to microstructure development.

1. Introduction

The mechanical properties of different alumina-based ceramics can be strongly affected by postsintering heat treatment because the mechanical behaviour is critically dependent on the microstructure of the ceramics [1, 2]. Hot isostatic pressing (HIP) proved to be a rather effective method for improvement of the ceramic microstructure. Post-HIP, e.g. hot isostatic pressing of presintered ceramics, results in elimination of large pores, healing of crack-like grain boundary defects, increased densification controlled by diffusion processes and in the mutual rearrangement (accommodation) of adjacent grains [3].

Post-HIP effects on densification and the resulting properties of alumina-based ceramics depends on many variable, such as temperature, pressure, duration, gas atmosphere of the HIP cycle, composition and technological prehistory of the ceramics. It has been shown [3] that high temperature–short time HIP is effective in increasing the strength of alumina ceramics. However, an increase in post-HIP temperature may result in substantial grain growth due to grain boundary diffusion processes in alumina ceramics [4]. The HIP treatment of alumina in Ar or oxidizing gas atmospheres results in improvement of the average bending strength from 470 to ca. 690 MPa [5]. There was little difference in strength regardless of whether an oxidizing or an Ar gas atmosphere was used, i.e. the HIP atmosphere had little effect on the mechanical properties. The operative post-HIP temperature is supposed to be dependent on the grain boundary composition in ceramics because of the temperature dependence of the rheological behaviour of grain boundary phases and because of some effects in the gas atmosphere–boundary phase interaction at high temperatures. Therefore, an optimum has to be in the post-HIP route for certain alumina-based ceramics. This paper describes the results of investigation of the effect of some post-HIP variables on the mechanical behaviour of different alumina-based ceramics.

2. Experimental procedure

Experiments were performed with the following ceramic materials supplied by MOH-9 (Pty) Ltd: slip cast and sintered M94 (94% Al_2O_3), M97 (97% Al_2O_3), M99 (99% Al_2O_3), and pressed and sintered M99P (99% Al_2O_3) series.

All the materials were HIP treated without encapsulation using commercial HIP equipment, (ABRA HIRP 25/70-200-2000). The HIP apparatus atmosphere was Ar. The regimes of the post-HIP are summarized in Table I.

The specimens were HIP treated under a pressure of 177–190 MPa, the cooling rate after exposure at the maximal cycle temperature was in the 5–15 °C min^{-1} range. The geometry of specimens for mechanical testing were $3 \times 5 \times 45$ mm^3. Both specimens without a notch and with a thin side-edge notch were tested.

The bending strength test was conducted by loading of unnotched specimens, using a stiff three-point bending adjustment at a span of 30 mm, on a testing machine (UTS-100). The crosshead speed was 0.5 mm min^{-1}. As with all brittle materials, the fracture strength of ceramics are governed by the most severe flaw and may be hence treated by Weibull statistics. A common simplification is the two parameter Weibull's function in the form

$$P(S) = 1 - \exp[-V(S/S_0)^m] \quad (1)$$

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TABLE I Post-HIP conditions

Series	HIP temperature (°C)	Exposure (min)	Material of furnace
$M94_0$	Non-HIP		
$M94_1$	1200	60	Mo
$M94_2$	1350	60	Mo
$M94_3$	1400	60	Mo
$M94_4$	1400	90	Mo
$M97_0$	Non-HIP		
$M97_1$	1435	90	Mo
$M97_2$	1550	60	Graphite
$M99_0$	Non-HIP		
$M99_1$	1435	90	Mo
$M99_2$	1550	60	Graphite
$M99P_0$	Non-HIP		
$M99P_1$	1350	60	Mo
$M99P_2$	1400	60	Mo
$M99P_3$	1440	60	Mo

where $P(S)$ is the probability function, V is the volume under tensile stress, S_0 is a normalizing factor and m is a material parameter (Weibull's modulus). The cumulative probability, $P(S)$, for a set of specimens, n, was experimentally obtained from

$$P(S) = i/(n+1) \tag{2}$$

where i is the number of specimens that fail at or below stress S. A sample size of >10 specimens is adequate for estimation of Weibull's modulus using Equation 2.

Fracture toughness measurements were performed using the side-edge notched beam (SENB) configuration specimens. The notch was machined with a thin diamond wheel saw. The measured notch tip curvature radius was ca. 50 μm. The specimens were loaded with a crosshead speed of 0.1 mm min^{-1} in a three-point bending mode with a distance between supports of 20 mm. The depth of the notch was one-half the specimen width.

To determine the elastic characteristics of the materials, a resonance frequency method was applied. The specimens were in a long bar configuration of $3 \times 6 \times 50$ mm^3 size. Measurements were performed using special ultrasonic equipment in a longitudinal vibration mode. Frequency was varied in the range up to 200 kHz depending on the properties of the material being used. Sound velocity was estimated as

$$v_l = 2Lf_l \tag{3}$$

where L is the specimen length and f_l the resonance frequency for the first harmonic of the longitudinal mode. Young's modulus of elasticity was evaluated as follows

$$E = \rho v_l^2 \tag{4}$$

where ρ is the measured bulk density of the material. To measure ρ, Archimedes' water-replacement method was applied.

3. Results and discussion

It was established that the pecularities of the post-HIP effects on the properties of the investigated alumina ceramics are strongly dependent on composition.

3.1. M94 series

The colours and the surface view of the specimens were changed in varying degrees by HIP. The specimens which were yellow–white before HIP treatment were pure white afterwards. Some surface defects, such a small voids and pores, were revealed.

The values of bulk density, modulus of elasticity and mechanical properties of the specimens of the M94 series are given in Table II.

The density of the specimens went through a maximum at an HIP temperature of 1350 °C. An increase in the post-HIP cycle duration at 1400 °C did not affect the density. SEM investigations of the microstructure reveal that the large closed individual pores and the planar network of pores cannot be eliminated by post-HIP. Moreover, the data of the density measurements indicate that post-HIP at 1400 °C results in some enlargement of the pores' volumes. It can be supposed that density increases as a result of Ar gas penetration into the ceramic body from the HIP apparatus atmosphere, controlled by diffusion processes, through the grain boundary phases in the microstructure of the ceramics. To verify this speculation, the as-received and post-HIP specimens were annealed in an air furnace at ambient atmospheric pressure at 1200–1400 °C for 2 h. The specimens which underwent HIP treatment at 1350 or 1400 °C spontaneously "failed" in the furnace. This result is assumed to be due to the surplus internal pressure in the pores after cooling the specimens from the HIP temperature. The pressure is too high for this material to stand without failure during the reheating cycle in the air furnace. Thus, the maximal post-HIP temperature for the M94 series has to be < 1350 °C.

A density peak at 1350 °C can be attributed to competition between densification (controlled by the relative movement of adjacent grains) and the Ar penetration effect. Some grain growth was revealed after post-HIP at 1350 and 1400 °C, despite the HIP temperature being lower than the pre-sintering temperature.

Post-HIP increases Young's modulus, the effect being most significant after HIP at 1200 °C. The effect of HIP on Young's modulus has the same tendency as on

TABLE II Density, Young's modulus and mechanical properties for the M94 series

Series	Density (g cm^{-3})	Young's modulus (GPa)	Strength (MPa)	Weibull's modulus	Fracture toughness (MPa m$^{1/2}$)
$M94_0$	3.60	290	227.4	7.1	3.4
$M94_1$	3.66	330	305.2	13.3	3.7
$M94_2$	3.72	303	181.1	6.9	2.7
$M94_3$	3.69	308	170.6	6.5	2.6
$M94_4$	3.68	308	–	–	–

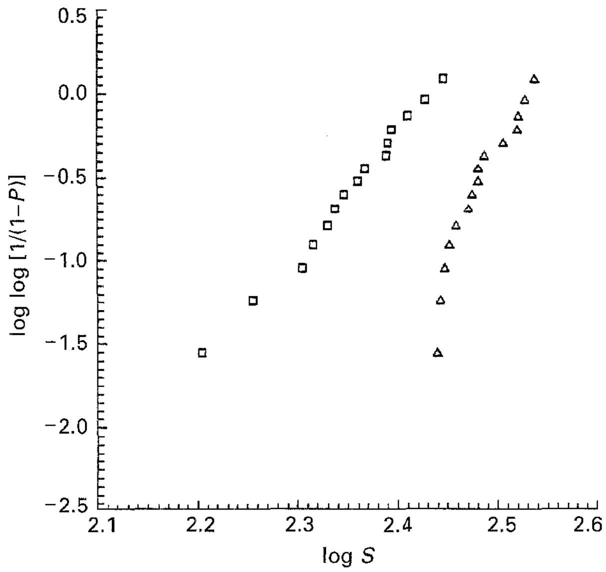

Figure 1 Cumulative probability versus fracture stress for the M94 series: □, non-HIP treatment; △, HIP treatment at 1200 °C.

TABLE III Density, Young's modulus and mechanical properties for the M97 series

Series	Density ($g\,cm^{-3}$)	Young's modulus (GPa)	Strength (MPa)	Weibull's modulus	Fracture toughness ($MPa\,m^{1/2}$)
$M97_0$	3.75	327	317.0	6.8	3.7
$M97_1$	3.84	341	338.2	12.8	4.1
$M97_2$	3.83	349	308.3	–	3.8

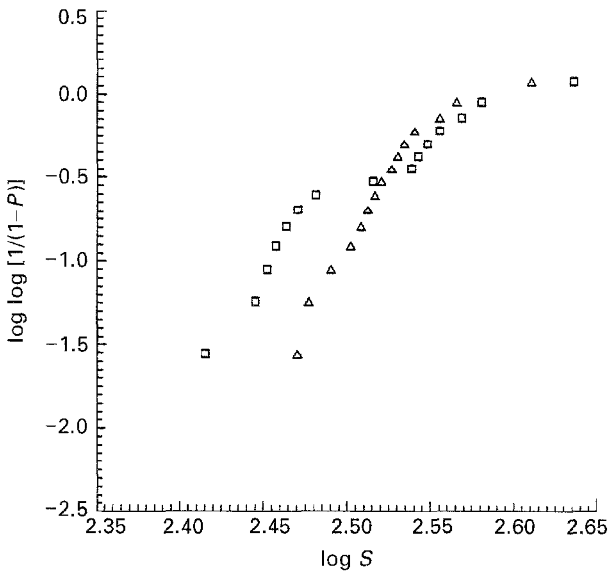

Figure 2 Cumulative probability versus fracture stress for the M97 series: □, non-HIP treatment; △, HIP treatment at 1435 °C.

the mechanical behaviour. All the values of the mechanical properties for the specimens HIP treated at 1200 °C exceed those for the non-treated specimens. Comparing the results obtained with well known strength data for other alumina-based ceramics, one can be persuaded that low temperature post-HIP allows improvement in the strength of 94% Al_2O_3 ceramics up to a value equal to 97% Al_2O_3 ceramics [1, 2].

The most significant effect is that of post-HIP at 1200 °C which strongly increases Weibull's modulus (Fig. 1) and, consequently, the uniformity of microstructure.

An increase in HIP temperature $\geqslant$ 1350 °C results in a decrease in the mechanical properties. The effect can be explained taking into account an assumption regarding Ar penetration into the pores. High internal tensile stress caused by retained Ar lowers the level of external applied stress which is able to fracture the ceramic body and, therefore, decreases the strength and fracture toughness.

3.2. M97 series

In contrast to HIP in a Mo furnace, the colour of the specimens post-HIP treated in a graphite furnace became grey. Table III summarizes the effect of HIP on the density and properties of M97 specimens.

The elastic and mechanical properties are improved by HIP in a Mo furnace. However, high temperature HIP using a graphite furnace decreases mechanical properties, despite increases in densification and Young's modulus. The C-containing atmosphere during the HIP cycle in a graphite furnace can negatively influence the mechanical properties as a result of C reactions with constituents of the material. In particular, C can react with silica impurities which have been detected in the M97 ceramics by energy dispersive X-ray (EDX) microanalysis. Such a type of interaction is a carbothermal reduction process which may result not only in silicon carbide formation along the grain boundaries, but also in silicon monooxide appearance in the gas atmosphere. As a result, grain boundary cracks can be formed.

The cumulative probability versus fracture stress plot is given in Fig. 2. These data allow the conclusion that HIP leads to healing of the most dangerous defects in the microstructure. SEM reveals that HIP did not result in noticeable grain growth in M97 ceramics. Thus, low temperature post-HIP treatment in a Mo furnace seems to be an effective procedure to improve the properties of 97% Al_2O_3 ceramics.

3.3. M99 series

Table IV summarizes the effect of post-HIP on density and properties of M99 series. The highest value of Young's modulus was achieved for post-HIP M99 ceramics. Fracture toughness increases whereas bending strength decreases as a result of post-HIP treatment. SEM and petrographic examinations reveal that the content of large grains in M99 ceramics' microstructures increases after HIP. Some grains having linear dimensions up to ca. 10 μm and above were detected. Increased grain size in HIP ceramics is supposed to be the reason for the lowered bending strengths.

On the other hand, the fracture toughness for alumina is greatly dependent on the processes in the

TABLE IV Density, Young's modulus and mechanical properties for M99 series

Series	Density ($g\,cm^{-3}$)	Young's modulus (GPa)	Strength (MPa)	Weibull's modulus	Fracture toughness ($MPa\,m^{1/2}$)
$M99_0$	3.99	394	361.3	7.6	4.1
$M99_1$	4.00	405	273.4	9.5	5.6
$M99_2$	3.97	401	304.7	–	4.8

wake zone near the crack tip. The crack bridging stress which prevents crack opening displacement can be strongly influenced by the HIP, affecting the microstructure of grain boundary regions, e.g. lowering micropore size etc. This is supposed to be a reason for different tendencies in strength and fracture toughness for the M99 series.

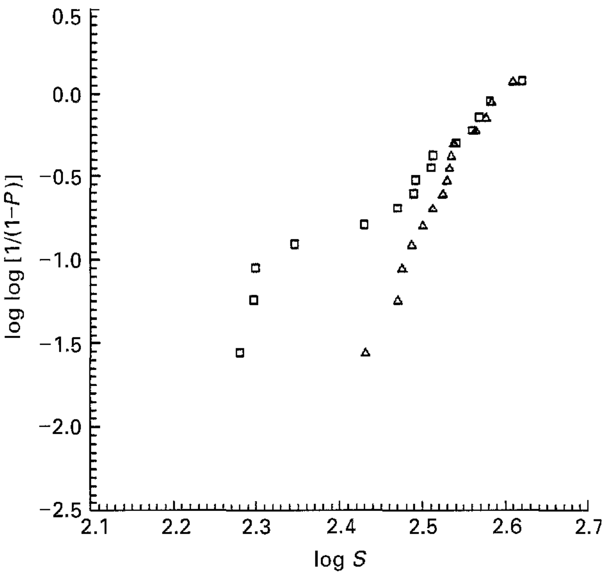

Figure 3 Cumulative probability versus fracture stress for the M99P series: □, non-HIP treatment; △, HIP treatment at 1400 °C.

3.4. M99P series

Contrary to the M94 series, the effect of post-HIP temperature on the properties of M99P ceramics is quite obvious. Table V gives the values of density, Young's modulus and mechanical properties of M99P ceramics.

An increase in HIP temperature increases Young's modulus, strength and fracture toughness. The data on strength distribution are most interesting. Fig. 3 shows the cumulative probability versus fracture stress plots. Obviously, the bimodal Weibull strength–cumulative probability is rather characteristic for non-HIP treated specimens, with different exponents, m, for lower and higher fracture stress regions. Such a situation indicates that two different populations of the fracture initiating defects exist in the microstructure of ceramics. Post-HIP eliminates some "weak links" which are responsible for the lowest fracture stress region. SEM reveals some chains of flat defects in non-HIP M99P ceramics (Fig. 4). Apparently, it can be supposed that these defects are mostly healing or disappearing during the post-HIP process resulting in transformation from a two- to one-parameter cumulative probability–fracture stress distribution.

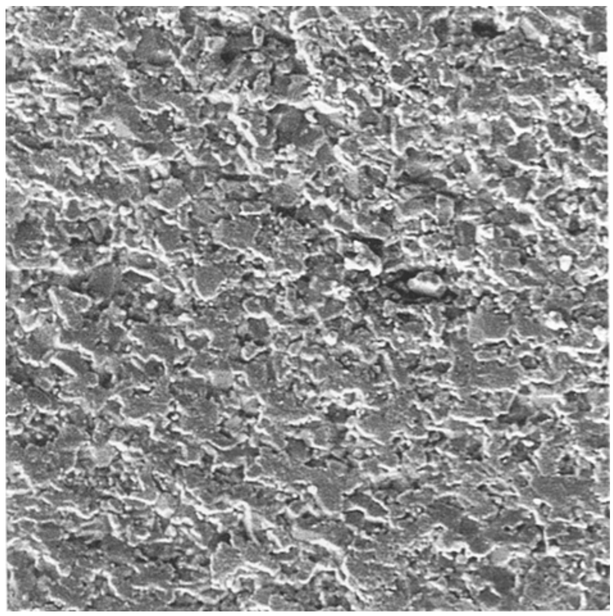

Figure 4 SEM micrograph of non-HIP treated M99P ceramics (1000 ×).

However, as compared with the cast/sintered M99 series, the properties of dry pressed/sintered M99P ceramics are lowered due to initial forming defects. Comparison of the data given in Tables IV and V indicates that post-HIP may only partially improve the microstructure of M99P ceramics. Perhaps, prolonged HIP exposure and/or higher temperatures are needed to obtain fully dense M99P ceramics with high mechanical properties.

TABLE V Density, Young's modulus and mechanical properties for the M99P series

Series	Density ($g\,cm^{-3}$)	Young's modulus (GPa)	Strength (MPa)	Weibull's modulus	Fracture toughness ($MPa\,m^{1/2}$)
$M99P_0$	3.85	360	302.2	4.0	3.6
$M99P_1$	3.93	373	331.3	8.5	4.1
$M99P_2$	3.94	380	337.4	9.6	4.1
$M99P_3$	3.94	398	338.1	8.3	4.4

4. Summary and conclusion

Experiments were performed with HIP without encapsulation of presintered alumina ceramics M94, M97, M99 and M99P series. An industrial HIP apparatus equipped with a Mo or graphite furnace was used. HIP was performed in an Ar gas atmosphere. The results of this study suggest that the post-HIP effect on properties is strongly influenced by the composition of the ceramics used.

1. M94 (94% Al_2O_3, cast) series: post-HIP at 1200 °C results in densification and in significant

improvement of the mechanical properties. An increase in the post-HIP temperature up to 1350 °C and above did not allow achievement of an admissible level of mechanical properties. It was speculated that the negative effect of high temperature treatment is due to Ar penetration from the HIP apparatus atmosphere into the pores of ceramic body. As a result, high internal stress develops.

2. M97 (97% Al_2O_3, cast) series: low temperature HIP in a Mo furnace is more effective than high temperature HIP in a graphite furnace. A twofold increase in Weibull's modulus is achieved as a result of post-HIP at 1435 °C. At the same time, Young's modulus, strength and fracture toughness values are also increased.

3. M99 (99% Al_2O_3, cast) series: low temperature HIP in a Mo furnace is preferable. Post-HIP significantly increases Young's modulus of elasticity, fracture toughness and Weibull's modulus. However, bending strength decreases due to grain growth during HIP. Different tendencies in the fracture toughness and bending strength are supposed to be a result of competing processes of grain growth and microstructure changes in the grain boundary regions.

4. M99P (99% Al_2O_3, dry pressed) series: post-HIP treatment increases remarkably the elastic and mechanical properties of the material. In addition post-HIP results in a significant improvement of the cumulative probability–fracture stress distribution from bimodal (non-HIP specimens) to Weibull's distribution with one exponent. This effect is apparently due to healing and/or disappearance of the network of flat defects in the microstructure. However, the mechanical properties of M99P ceramics are lowered due to initial forming defects, as compared with cast/sintered M99 ceramics. Perhaps, longer exposure and/or higher temperatures in the HIP cycle are needed to obtain fully dense M99P ceramics having high level mechanical properties.

Thus, the results obtained in this study suggest that post-HIP without encapsulation of presintered alumina ceramics allows significant improvement in their elastic and mechanical properties and, therefore, in service reliability.

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