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Reliability of Alumina Ceramics. 2: Effect of Processing

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Abstract

In this paper an attempt is made to review the mechanical properties that can currently be achieved with the best alumina powders available by utilizing a series of simple processing strategies and surface preparation techniques. The processing techniques include cold-isostatic pressing, slip casting, settling and centrifuging as well as hot-isostatic pressing of as-sintered alumina samples. The surface preparation techniques include grinding and polishing as well as annealing and grinding in the green state. While the fracture strength can be increased from 460 MPa for cold-isostatic pressing to 925 MPa for a centrifuged and cast alumina slip, the Weibull modulus remained between 8 and 12 for most processing strategies. © 1997 Elsevier Science Limited. All rights reserved.

Auszug

Diese Arbeit beschreibt den Versuch, die mechanischen Eigenschaften, die mit den besten derzeit verfügbaren Aluminiumoxidpulvern erreicht werden können, zu dokumentieren. Dazu werden einfache Herstellungsstrategien und verschiedene Oberflächenbearbeitungen verwendet. Zu den verwendeten Herstellungsmethoden zählen kalt-isostatisches Pressen, Schlickergießen, Sedimentieren und Zentrifugieren wie auch heiß-isostatisches Pressen von vorgesinterten Proben. Die Oberflächen wurden durch Schleifen, Polieren, Tempern und Bearbeitungen des Grünkörpers verändert. Die Bruchfestigkeit kann von 460 MPa von kalt-isostatisch gepreßten Proben auf 925 MPa von zentrifugierten und abgegossenen Schlickern gesteigert werden, während der Weibullmodul für alle Herstellungsmethoden unverändert zwischen 8 und 12 blieb.

1 Introduction

The purpose of the present paper is to expand on salient aspects of the reliability tree proposed by de With.[1] We will focus on two issues: (1) processing (in particular afforded by advanced consolidation techniques and hot-isostatic pressing) and (2) machining.

The reliability of Al_2O_3 is mainly determined by two factors: fracture strength and Weibull modulus. These are primarily governed by the processing technology, especially the consolidation technique. Several research groups[2–6] have shown that the fracture strength of alumina-based ceramics can be enhanced by colloidal consolidation techniques. In his fundamental review, Lange[4] showed that several flaw size distributions (soft agglomerates, hard agglomerates, organic inclusions) can be eliminated by an improved colloidal processing technology. These improved technologies result in an increased fracture strength. A question that is not considered by Lange is whether the variability of strength can be reduced by colloidal processing technologies. Recently, Huisman *et al.*[6] reported a Weibull modulus *m* of 24 for alumina by consolidating a concentrated slurry by centrifuging.

Another possibility to eliminate flaws in a ceramic material is achieved by a hot-isostatic pressing step after sintering, in order to eliminate porous regions produced by agglomerates or burn-out of organic inclusions. Kellet and Lange[7] have demonstrated the potential of this method with an alumina–zirconia composite. They were able to eliminate large, artificial pores with a hot-isostatic pressing treatment. The question arises whether this method has the potential to improve the Weibull modulus of pure alumina significantly.

A further technological aspect that has a modifying influence on the mechanical properties of brittle ceramic materials like alumina is the surface preparation.[8–10] Tressler *et al.*[8] were among

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the first to investigate the influence of surface preparation on the fracture strength of alumina with varying grain size. They found a strength reduction with increasing surface machining grit size. In connection with the surface preparation there are two basic questions. In which way does the surface preparation modify the flaw size distribution responsible for failure and what is the role of residual stresses introduced by the machining process?

In contrast to our previous paper on the reliability of Al_2O_3 as affected by grain size,[11] which noted more fundamental aspects of fracture, we focus in the present paper on the technological aspects. Different consolidation techniques were compared concerning their influence on the mechanical properties, especially fracture strength and Weibull modulus. The specimens were prepared either by cold-isostatic pressing or by slip casting. In a second consolidation route, the slurries were classified by sedimentation of a dilute suspension or by centrifuging a dispersed slip with a high solid loading in order to separate agglomerates. The influence of hot-isostatic pressing after sintering on the mechanical properties was also investigated. The relationship between surface preparation and mechanical properties was determined with as-sintered, ground and polished specimens and with specimens ground in the green state.

2 Experimental Procedure

In the present investigation two commercial, ultrafine Al_2O_3 powders were used (Taimei DAR; Condea Ceralox HPA 0·5). Bars (4 mm × 5 mm × 40 mm) and discs (radius 12·5 mm, height 4 mm) were fabricated by uniaxial pressing of as-received powders at 50 MPa in a steel die followed by cold-isostatic pressing (CIP) at 800 MPa. Green densities of 55% of the theoretical density were attained by this procedure. The green bars and discs produced with the Taimei DAR powder were sintered in air at temperatures of 1350°C (dwell time 2 h) to produce a fine-grained material and 1600°C (dwell time 15 or 96 h) in order to get a coarse-grained material. Discs from the HPA 0·5 powder were sintered at a temperature of 1450°C (dwell time 0·5 h).

Hot-isostatic pressing was carried out with the CIPed, fine-grained material (Taimei DAR) at 1500°C and an argon gas pressure of 180 MPa (dwell time under this condition, 20 min).

For the consolidation by slip casting (SC), the powders were dispersed with a commercial dispersant (Dolapix CE 64, Tschimmer & Schwarz) in deionized water by ultrasonic agitation. The slurries were prepared with a solid loading of 40 vol%. The use of the dispersant leads to a shift of the isoelectric point of alumina from pH = 9 into the acid region. Therefore the pH value was adjusted to 9·5 with NH_4OH to assure processing in a region of high repulsive forces. After de-airing, discs were produced by casting the slurry in plastic rings, which were placed on a gypsum plate.

Experiments concerning the separation of agglomerates from the slurry via centrifuging were carried out with the HPA 0·5 powder. After centrifuging in a glass container with different rotational speeds, the slurry was separated from the sedimented layer (solid content decreased to about 30 vol%). De-airing was followed by slip casting discs (C/SC: centrifuged/slip cast) as described above.

Another possibility for slurry classification was proposed by Aksay *et al.*[12] A slurry with a solid loading of 5 vol% was prepared without dispersant at a pH value of 2–3 by ultrasonic agitation. The dilute slurry was allowed to settle under gravity. After separating the classified slurry from the sediment, the slurry was flocced (pH = 8–9) and then centrifuged in order to increase the solid content. After centrifuging the slurry was dispersed again. With this procedure a slurry with a solid content of about 30 vol% was produced. After de-airing, discs were fabricated (S/SC: sedimentation/slip casting) as described above.

Discs produced by slip casting with the Taimei DAR powder and the HPA 0·5 powder were sintered at 1350°C (dwell time 1 h) and at 1400°C (dwell time 0·5 h), respectively. This sintering schedule had been adapted to the different powders and consolidation techniques in order to produce a fine-grained material with a final density (measured by Archimedes' method) of at least 99%.

The influence of residual stresses and surface damage introduced by machining was evaluated by annealing one batch of bars (Taimei DAR, grain size (G) = 1·7 μm) at 1200°C for 1 h prior to strength testing. This time–temperature regime had been found to be most suited to remedy detrimental machining defects.[13]

Three different surface preparation methods were investigated. The following abbreviations are used:

(1) 'AS' for as-sintered specimen;
(2) 'G/P' for ground and polished specimen (after sintering);
(3) 'GG' for the specimens that were ground in the green state.

Green state surface preparation (GG) was carried out with SiC grinding paper with different grit sizes (46, 15 and 6 μm). The sintered specimens (G/P) were ground with a rotating diamond disc (grit size 107 μm) followed by grinding the prospective tensile surface with a 15 μm disc and polishing by hand to a 3 μm finish.

Uniaxial strength testing was carried out with a four-point bending test device with articulating fixtures and rotating load pins, with span lengths of 10 mm and 20 mm and specimen dimensions 3 mm × 4 mm × 30 mm. For biaxial strength testing a 'flat punch on three balls' testing device was used with an effective contact radius b = 1·8 mm, a support radius a = 8 mm, specimen radius 11 mm and a specimen thickness of t = 2·5 mm. The biaxial loading device has the advantage that edges cannot control fracture. This is important for investigation of the influence of surface preparation on strength and strength distribution and for the strength testing of green bodies. The nominal stress for the 'flat punch on three balls' test was calculated according to Marshall.[14] Variability of strength values was analysed corresponding to the two-parameter Weibull approach[15] using 20 to 30 specimens. The loading rate (crosshead speed) in our experiments was 1 mm s^{-1}. Fracture occurs within 3–10 s.

After preparing the surfaces by grinding and polishing, the microstructure was revealed by thermal etching for 30 min at temperatures 20°C below the sintering temperature for the fine-grained material and 50°C below for the coarse-grained materials. The average grain diameter was measured using the linear intercept technique, using a prefactor of 1·56. Observation of polished samples in an optical microscope was used to characterize the frequency and size of surface defects as a function of processing technology. Scanning electron microscopy was used to examine the fracture surfaces and to identify the strength-determining defects.

3 Results

Both test geometries employed in this study (four-point bend bars loaded in a uniaxial stress state and discs loaded in biaxial flexure) were compared using cold-isostatically pressed alumina (Taimei DAR) with 1·7 μm grain size. No definite influence of the stress state on the Weibull modulus could be observed (m = 11·4 for uniaxial loading, 90% confidence band 8·3–14·1; m = 7·6 for biaxial loading, 90% confidence band 5·3–9·6). The mean strength of the discs tested under biaxial flexure (σ_B = 556 MPa) is comparable to the strength of bars tested under uniaxial loading conditions (σ_B = 564 MPa). The results can be rationalized[16] using a suitable fracture criterion.[17] The focus of this paper, however, should be on processing.

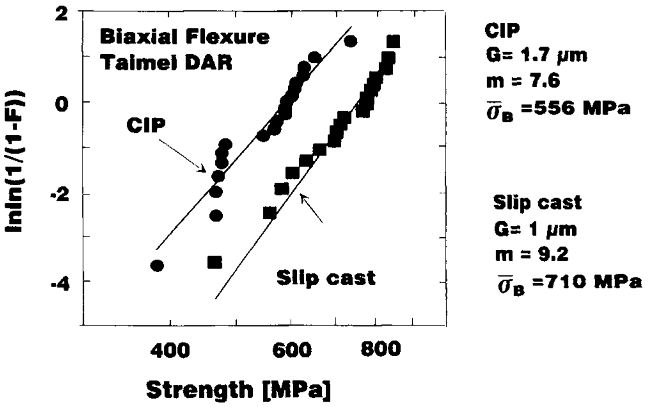

Fig. 1. Strength distributions of cold-isostatically pressed (CIP) and slip cast specimens.

Figure 1 compares the strength distributions tested in biaxial flexure of fine-grained alumina (Taimei DAR, G/P) consolidated by cold-isostatic pressing (CIP) or slip casting (SC). The Weibull modulus appears to be unaffected by the processing technology (m = 7·6 for CIP; m = 9·2 for SC). There is, however, a noteworthy increase of the fracture strength from σ_B = 556 MPa for the CIP specimen to 710 MPa for the SC specimen.

The potential of colloidal processing to increase fracture strength and hence to produce more reliable ceramics is proved with a series of tests with a small number of specimens with an Al_2O_3 powder containing agglomerates (HPA 0·5). The mean strength of the specimens fabricated by CIP was 460 MPa (Fig. 2). Slip casting enhanced the strength to 670 MPa. An advanced colloidal processing technology results in an additional strength increase. The sedimentation technology described in section 2 yielded specimens with a mean strength of 718 MPa. The highest mean strengths were achieved with specimens produced by centrifuging dispersed slurries. By varying the rotational speed of the centrifuge, the strength could be raised up to a mean strength of 925 MPa.

Figure 3 provides a series of representative photographs of polished surfaces fabricated with

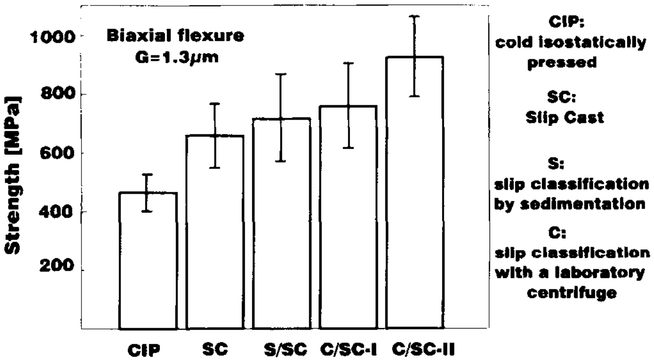

Fig. 2. Mean strength and standard deviation (error bars) depending on consolidation technique (testing performed in biaxial flexure).

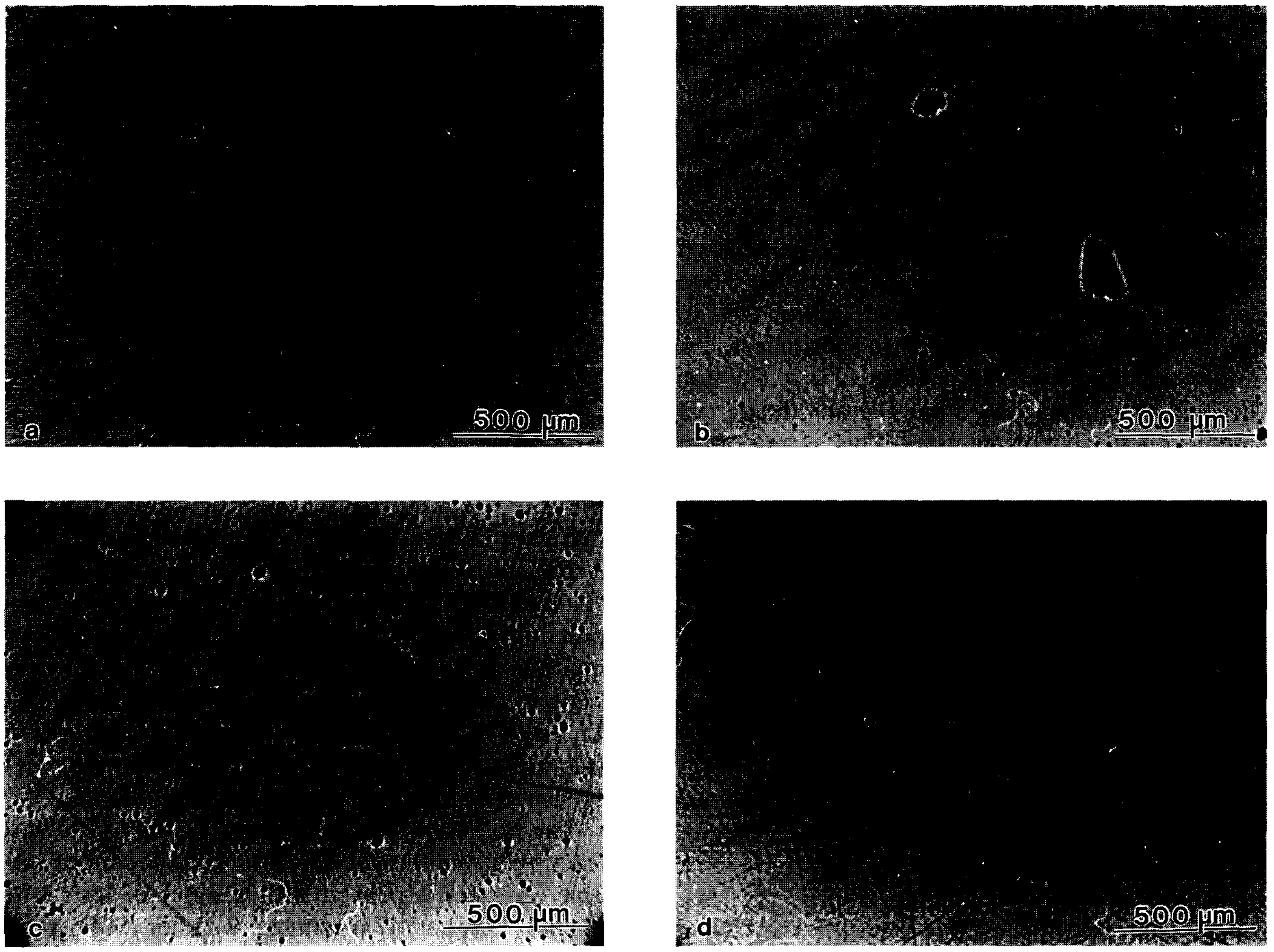

Fig. 3. Polished surfaces (optical microscopy) of specimens consolidated by different techniques: (a) Taimei DAR, cold-isostatically pressed; (b) HPA 0·5, cold-isostatically pressed, (c) HPA 0·5, slip cast; (d) HPA 0·5, classified, settled and slip cast.

different powders and different consolidation technologies. In Figs 3(a) and (b) the CIP specimens produced with the Taimei DAR and the HPA 0·5 powders are shown. On the HPA 0·5 surface the size and frequency of large pores is higher than on the Taimei DAR specimen surface, an observation which correlates with the lower fracture strength of the specimen from the first powder. It is also possible to rank the strengths of the specimens produced with different consolidation techniques. The slip cast specimen [Fig. 3(c), HPA 0·5] exhibits surfaces with fewer defects than the CIP specimen, [Fig. 3(b), HPA 0·5]. A further reduction of size and frequency of large pores is visualized in Fig. 3(d), which shows a polished surface derived from a classified and settled HPA 0·5 powder.

Figure 4 illustrates the influence of sintering pressure on strength and Weibull modulus. The strength distributions (uniaxial tension, G/P) of the 1·7 μm alumina (Taimei DAR) and the same material subsequently hot-isostatically pressed at 1500°C at a gas pressure of 180 MPa (dwell time under this condition, 20 min) are compared. Hot pressing did not improve Weibull modulus, but rather led to a decrease in the mean fracture strength. Due to the higher hot-pressing temperature as compared with the sintering temperature, the average grain size increased from $G = 1{\cdot}7$ μm to $G = 12$ μm.

The next topic of the present investigation concerns the influence of machining on mechanical properties. Stresses introduced by grinding and polishing can be relaxed by annealing an alumina specimen at 1200°C for 1 h after machining (G/P). Figure 5 demonstrates that this treatment did not

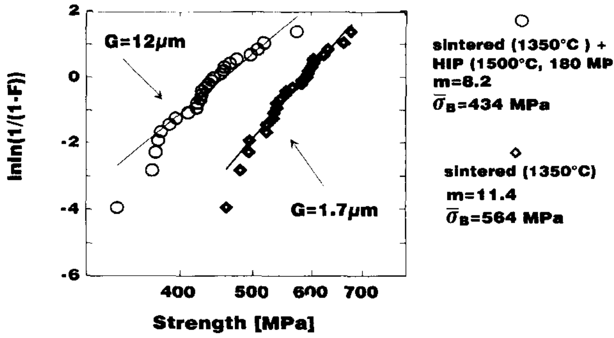

Fig. 4. Strength distributions of sintered specimens and sintered and then hot-isostatically pressed alumina.

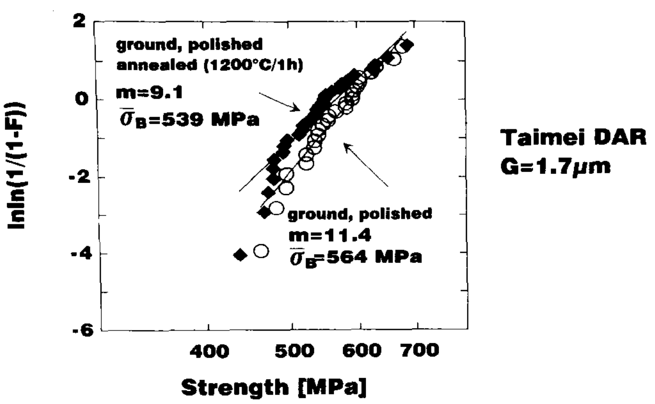

Fig. 5. Strength distributions of ground/polished specimens and ground/polished/annealed specimens.

alter mechanical properties significantly but rather had an adverse effect.

In Fig. 6 the strength distributions of G/P and AS samples for two different grain sizes are compared. For the 1·7 μm alumina strength is increased by grinding and polishing compared with the as-sintered specimen. The large-grained alumina, however, does not show a distinct difference in strength between G/P (σ_B = 326 MPa) and AS (σ_B = 300 MPa) samples. The Weibull modulus appears to be decreased by grinding and polishing. Fractography reveals that specimens with polished surfaces mainly fail from pores located at the surface or in the volume beneath the surface, while fracture in as-sintered samples is initiated either from edge flaws or from surface defects that join with pores beneath the surface.

Green machining is an attractive and economic possibility for the surface preparation of ceramics. In Fig. 7 the strength of GG specimens (G = 1·7 μm) is compared with the strength of AS and G/P samples. The strength of the specimen machined in the green state with the 46 μm and the 15 μm grit SiC paper is comparable to the strength of the as-sintered specimen (see Fig. 7), but is lower than the strength of the ground and polished samples. This latter value of σ_B = 564 MPa was surpassed by the alumina which had been ground in the green state with a grit size of 6 μm (σ_B = 599 MPa).

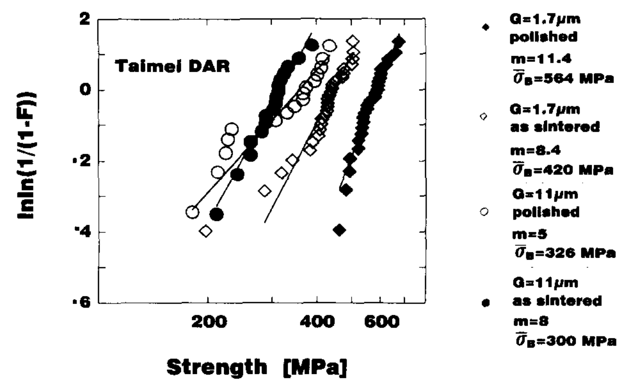

Fig. 6. Strength distributions of ground and polished specimens and as-sintered specimens evaluated in four-point bending.

4 Discussion

We were able to show that colloidal consolidation techniques result in higher strength compared with CIP and that the most refined technique can lead to fracture strength values higher than 900 MPa as measured in biaxial flexure. The Weibull modulus could not be increased by these techniques. This strength increase is attributed to the elimination of soft agglomerates.[4] This explanation is supported by micrographs of polished specimens (Fig. 3), which prove that the polished surface of the slip cast sample exhibits fewer voids (surface pits) than the CIP sample. In order to demonstrate the potential of the slip casting technology for the fabrication of high strength alumina, a powder containing rather many agglomerates (HPA 0·5, but not Taimei DAR) was investigated. A strength of 460 MPa was determined from sintering the cold-isostatically compressed as-received powder. With the slip casting technology a strength of 670 MPa could be attained. As shown by Aksay *et al.*,[12] a further strength increase could be realized by separating the coarser parts of the slurry by sedimentation. A simple sedimentation technique yielded a strength increase of 40 MPa. Increasing the sedimentation rate by centrifuging a concentrated, dispersed slurry resulted in mean strengths up to 925 MPa (more than doubling the strength obtained by the cold pressing route). This strength increase again correlates with the polished sample surfaces seen in the micrographs of Fig. 3. The CIP sample reveals large surface pits. The frequency and the dimensions of the pores are reduced for the slip cast specimen. The samples

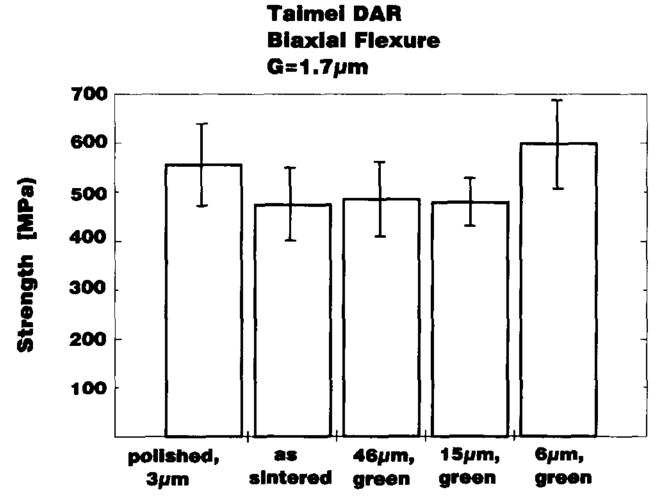

Fig. 7. Mean strength and standard deviation (error bars) as a function of surface preparation evaluated under biaxial flexure.

produced with the centrifuged slurry show only few small surface pits. The implication is that the mean size of the defects (pores due to soft or hard agglomerates) was reduced by the colloidal consolidation technique, but that the distribution of the defects and thus the Weibull modulus remain unaffected.

In a related paper[11] it was shown that the CIP material failed mainly from volume pores situated just beneath the surface, with a minor defect population being associated with surface pits. In order to eliminate large pores an experiment was carried out in which the sintered samples were subjected to a subsequent hot-isostatic pressing treatment. This led to an elimination of the smaller pores with the resultant specimens appearing transparent. The larger pores, which are responsible for failure and determine the Weibull modulus, however, remained unaffected. This result is in accordance with the investigation of Kellet and Lange.[7] They observed that pore closure could be achieved for a fine-grained alumina/zirconia composite whereas the pores in pure alumina remained unchanged. It might be possible to achieve pore closure by an increased temperature for hot-isostatic pressing. As shown in Fig. 4, the strength of the hot-isostatically pressed samples is reduced when compared with the sintered samples. This strength degradation can be correlated with an increased average grain size,[11] which in our case leads to a reduction in strength. When optimization by hot-isostatic pressing is contemplated, a balance between pore closure and grain growth therefore is required. This result is in accordance with the investigation of Weiss *et al.*[18] They showed that the strength of alumina could be improved by hot-isostatic pressing at suitable conditions but they could not realize an improvement of the Weibull modulus.

An issue of considerable technological relevance concerns the influence of surface preparation on the mechanical properties of brittle ceramics. Our results discussed so far were obtained with ground and polished specimens. The strength may still be affected by residual stresses introduced during machining.[19] Annealing the specimen (1200°C, 1 h) in order to relax these stresses led to a slightly reduced strength and no variation of the Weibull modulus. The difference in characteristic strength amounts to 25 MPa. With regard to the confidence bands, no influence of the residual stresses on the macroscopic mechanical properties could be found.

The issue of surface preparation can further be elaborated on by a comparison between G/P and AS specimens. The fine-grained material shows a distinct strength increase if the surface is ground and polished, but the strength distribution remains unaffected. This can be compared to the work of Hessert and Eigenmann[10] and Frei and Grathwohl.[20] The Weibull modulus remained unchanged if comparing samples with ground and polished surfaces ($G = 1{\cdot}4\ \mu m$)[10] and if comparing samples having ultrasonically treated surfaces with ground and finally polished surfaces ($G = 1{\cdot}3\ \mu m$).[20]

The matter is slightly more complicated for the coarse-grained material, where several competing effects are active. Grinding and polishing leads to an increase in strength with some specimens due to an alleviation of edge flaws and introduction of a compressive surface stress.[19] At the same time, coarse-grained alumina is more susceptible to severe surface degradation and wear-induced microfracture associated with pull-out of large grains.[21] These grains in combination with neighbouring pores can lead to an additional failure population, reducing the strength of some of the specimens and in the last consequence changing the Weibull modulus. This low Weibull modulus of $m = 5$ is comparable to the lowest Weibull modulus of the largest grain size alumina of $m = 5{\cdot}8$, where abnormal grain growth and localized grain pull-out were suggested to lead to an additional failure origin.[11]

Grinding in the green state with a small grit SiC paper constitutes a simple, cheap and effective alternative for the preparation of high-strength alumina. It is interesting to note that the surface preparation with the 46 μm and 15 μm SiC grinding paper yielded no difference in strength compared with the as-sintered samples. A surprising result is that the strength of the specimens prepared with the 6 μm SiC paper showed a strength value somewhat higher than the samples which were ground and polished after sintering. This effect opens up new avenues for further studies, since green state grinding is much less understood than grinding and polishing of as-sintered specimens.

5 Conclusions

(1) The strength of alumina is enhanced by changing the consolidation technique from cold-isostatic pressing to colloidal techniques. The highest strength value obtained for a fine-grained alumina is 925 MPa.
(2) No dependence of the Weibull modulus on consolidation technique is observed.
(3) An alternative to grinding and polishing after sintering is grinding in the green state with a sufficiently fine grit size, yielding strength values comparable to surface machining techniques applied on as-sintered materials.

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