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High-temperature fracture of hot-pressed AlN ceramics

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The fracture behaviour of AlN ceramics has been investigated as a function of porosity and temperature. The material was produced by hot-pressing and contained about 1.8 wt % impurities. Only slight anisotropy due to the hot-pressing procedure was present as observed by the ultrasonic pulse-echo technique. For the completely dense material, Young's moduli for directions parallel and perpendicular to the hot-pressing direction are 324 and 342 GPa, respectively. The fracture toughness and strength of the same material are 2.7 MPa m$^{1/2}$ and 310 MPa, respectively, both values being for fracture planes parallel to the hot-pressing axis. The fracture toughness value decreases by about 10% up to 1500 K, while the strength decreases by about 20%. All three properties show a strong decrease with increasing porosity.

1. Introduction

It has been reported previously [1, 2] that the mechanical properties of aluminium nitride (AlN) ceramics have been the subject of little investigation, in spite of the wide interest in nitrogen ceramics in general. Some strength and elastic data have been presented in the early literature, but values for the fracture toughness had not been reported until a recent publication [3] gave an indication. All these data, however, are quite useful for a number of applications where AlN offers advantages over other nitride (or oxide) ceramics. In this paper the mechanical properties of commercially available hot-pressed AlN ceramics are investigated as a function of porosity and temperature. A preliminary report has been given previously [2].

2. Experimental details

Several hot-pressed AlN billets with various densities were obtained from a commercial source*. The impurity content was analysed by spectrochemical analysis. X-ray diffraction was used to detect the presence of other phases.

The density, ρ, was determined for the various billets using weight and size information of rectangular pieces. For the denser materials, Archimedes method was also used. The theoretical density was assumed to be 3.26 g cm^{-3} [4].

For directions parallel (||) and perpendicular ($\perp$) to the hot-pressing direction, the longitudinal wave velocity, v_l, and the shear wave velocity, v_s, were determined at 10 and 20 MHz, respectively, using the pulse-echo technique [5]†. Young's modulus, E, was calculated from ρ, v_l and v_s. No correction was made for attenuation. The estimated standard deviation in E is about 5 GPa.

Specimens of dimensions 1 × 3 × 15 mm^3 were sawn from the various billets. The strength, σ_f of these specimens was measured in a three-point bend apparatus using a span of 12 mm. An average of five specimens was used for each material resulting in a mean sample standard deviation of 12%. The cross-head speed of the testing machine‡ was 0.1 mm min^{-1}. The measurement of fracture toughness, K_{Ic}, was done in the same way using the same specimen size. This small type of specimen makes efficient use of the material available while retaining reliability and accuracy [6]. Pre-cracking was done by Vickers hardness indentation

*Cerac/Pure.
†Panametrics 5223.
‡Overload Dynamics S200.

TABLE I Characteristics of AlN as used in this work

Material	ρ (%)	ρ (g cm^{-3})	d (μm)
1	66.1	2.16	3.0
2	79.4	2.58	10.0
3	94.6	3.09	1.5
4	100	3.28	2.0

Impurities (wt %): C 0.9%, Fe 0.2%, W 0.1%, Si 0.05% Ni 0.03%, Mo 0.02%.
Nitrogen content: 32.5%
Oxygen content: 0.5% (estimated).

(2 N load) just below the notch root on both sides of the specimens. The value of the compliance factor, Y, was calculated according to the method of Brown and Srawley [7]. Five specimens were used for each material resulting in a mean sample standard deviation of 10%. At room temperature, both σ_f and K_{Ic} measurements were carried out in air. At higher temperatures, the σ_f and K_{Ic} measurements were performed in an N_2 gas atmosphere containing about 200 ppmV H_2O using an all-ceramic set-up and a Pt resistance furnace.

Scanning electron micrographs were taken of a fracture surface for each type of material, after covering the surface with a thin gold layer. The grain size, d, was estimated from these fractographs using Mendelson's method [8].

3. Results and discussion
3.1. Material characteristics
Table I presents some characteristics of the various materials. All the ceramics have similar grain size at increasing density, except the 79% dense material (no. 2). For the most dense material (no. 4) the observed density is slightly higher than the theoretical density of pure AlN (3.26 g cm^{-3}), probably owing to the impurities present. X-ray diffraction revealed only the pattern of the hexagonal AlN phase. This was also true for the other materials.

Fig. 1 presents the Young's moduli, E. A sharp increase with density is observed. Moreoever, there is a small difference between $E(\|)$ and $E(\perp)$ indicating some preference in grain orientation due to the hot-pressing procedure. At zero porosity, $E(\perp)$ is 342 GPa and $E(\|)$ is 324 GPa. Taylor and Lenie [4], using the bend test, reported a value of 345 GPa for $E(\perp)$ of 98% dense hot-pressed AlN. Gogotsi [9], also using the bend test, gave a value of 234 GPa for 97.2% sintered material. Recently, Boch *et al.* [3] also investigated the dependence of Young's modulus on porosity. They reported a value of 315 GPa, probably corresponding to $E(\|)$, for the fully dense material, using a resonance method. In view of the fact that the different experimental values for $E(\perp)$ and $E(\|)$ agree reasonably well (Table II), we conclude that the measurement of Gogotski [9] seriously underestimate the value of E.

The porosity dependence of the Young's moduli as determined in this investigation is definitely non-linear. On the other hand, the results of Boch *et al.* [3] can be described by a linear relation up

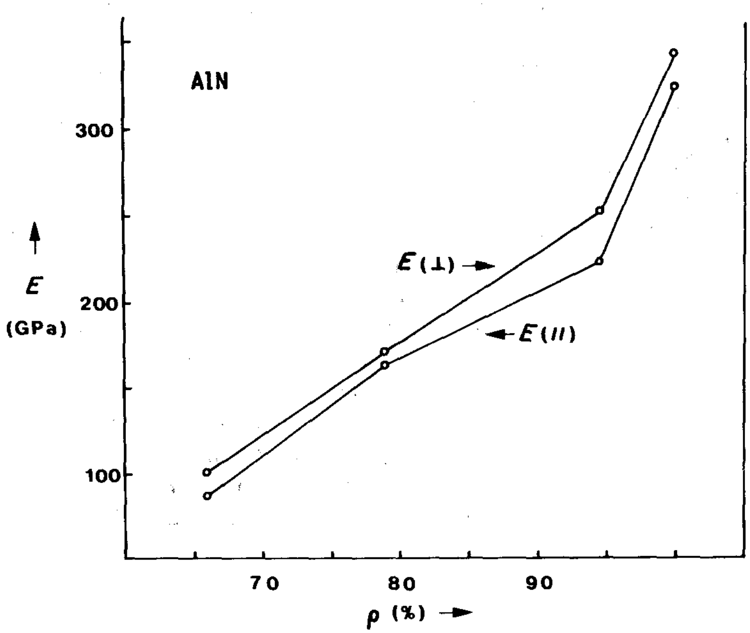


Figure 1 Young's modulus of AlN at room temperature or 300 K for directions parallel, $E(\|)$ and perpendicular, $E(\perp)$, to the hot-pressing axis as a function of density.

TABLE II Comparison of the elastic properties of AlN

ρ (g cm^{-3})	Porosity, P	d (μm)	E (GPa)	Remarks	Reference
3.20	0.02	5	345 (300 K) 317 (1300 K) 276 (1700 K)	⊥, bend test	[4]
314	0.028	–	234	bend test	[9]
3.26	0	9	315	‖?, resonance	[3]
3.28	0	2	342	⊥, pulse-echo	Present work
3.28	0	2	324	‖, pulse-echo	Present work

to a porosity, P, of 0.2; $E = 315\ (1 - 3.22P)$. This discrepancy is not understood.

3.2. Fracture toughness

Owing to limitations with regard to billet size, the fracture toughness, K_{Ic}, was measured only for fracture plane directions parallel to the hot-pressing axis. However, in view of the fact that E exhibits only little anisotropy, no great differences are expected, between toughness values for fracture plane directions parallel and perpendicular to the hot-pressing axis. The results for K_{Ic} are presented in Fig. 2. A sharp decrease with porosity can also be observed here. For the densest material the value measured is 2.7 MPa m$^{1/2}$. Accordingly the fracture energy, $\gamma = K_{\mathrm{Ic}}^2/2E$, is about 10 J m^{-2}; this value is much lower than that for Si_3N_4 ceramics which have a fracture energy of the order of 50 J m^{-2}. The fracture toughness, K_{Ic}, was also measured up to 1500 K. A slightly decreasing value is observed for all densities, except for the densest material (no. 4) which initially shows an increase and then a decrease. This increase is not understood. In view of this irregular behaviour an interpretation of the temperature dependence of K_{Ic} in terms of a simple Gilman model [15] would not be meaningful.

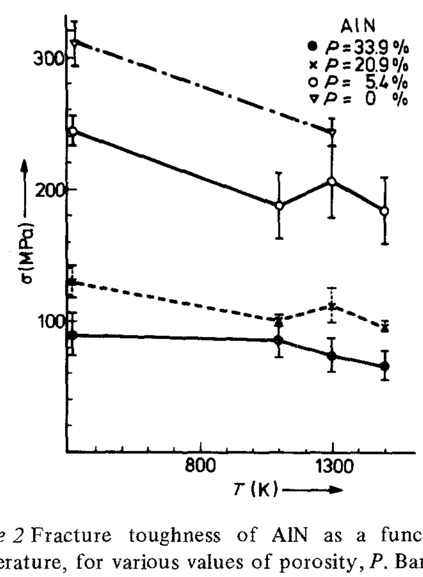


Figure 2 Fracture toughness of AlN as a function of temperature, for various values of porosity, P. Bars at the data points indicate sample standard deviation.

At room temperature the fracture mode is entirely intergranular at 66.1% density, is mixed at 79.1% density and mainly transgranular at the density values of 94.6% and 100%. At higher temperatures the fracture mode of the last two materials becomes mainly intergranular. This effect is shown in Fig. 3.

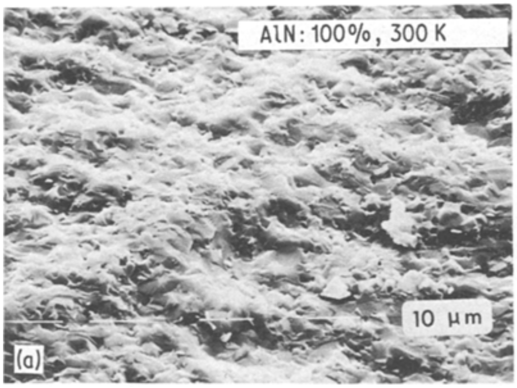
The data could be fitted to empricial equations such as $K_{\mathrm{Ic}}\ (T,P) = K'_{\mathrm{Ic}}\ (T) \exp(-\beta P)$, where β is a characteristic exponent. Owing to the irregular behaviour with temperature and the change in fracture mode, however, the interpretation of the constants in these equations would be rather difficult, if not impossible. Accordingly, the analysis has not been performed.

The only other determination for K_{Ic} found in the literature [3] is a value of 2.5 to 2.7 MPa m$^{1/2}$ for almost completely dense material with a grain size of about 9 μm. Close agreement with the present data can thus be observed.

3.3. Strength

For the strength measurements, again only specimens with a fracture plane parallel to the hot-pressing axis were used. The results are presented in Fig. 4.

At zero porosity, σ_{f} is about 310 MPa for as-sawn specimens with a centre line average roughness, $R_{\mathrm{a}} \sim 0.2\ \mu$m. This figure compares favourably with values reported earlier (Table III): $\sigma_{\mathrm{f}} = 265$ MPa [4] and $\sigma_{\mathrm{f}} = 275$ MPa [10] for hot-pressed AlN [4] and $\sigma_{\mathrm{f}} = 196$ MPa [9] and $\sigma_{\mathrm{f}} = 200$ and 250 MPa [10] and sintered material. Long and Foster [11] reported a strength value of about 80 MPa for an AlN ceramic that was not characterized any further. In view of the other results this material was probably highly porous.



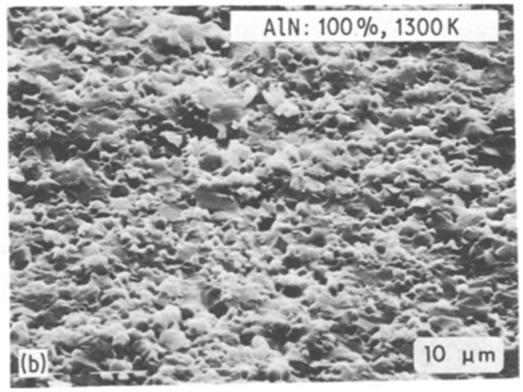


Figure 3 Fracture surface of the 100% dense AlN at room temperature (a) showing transgranular fracture and at 1300 K (b) showing intergranular fracture.

The effect of the quality of the surface condition on strength is demonstrated using 100% dense samples with a slightly rougher surface ($R_a \sim 1.1$ μm). In this case the resulting σ_f value decreases to 237 MPa. All these strength values are a little lower than the values generally reported for Si_3N_4.

In the recent work of Boch *et al.* [3] and Glandus *et al.* [1] a value of 380 MPa was reported for polished specimens of almost completely dense hot-pressed AlN. This relatively high value is due to the polishing of the specimens. High values (up to 500 MPa) were also reported by Schwetz *et al.* [12] again for polished specimens ($R_a < 0.5\ \mu$m) of sintered materials. In the latter work, however, the material generally contained a second phase and interlocked needle or platelet-like grains which were probably the cause of the high strength values reported.

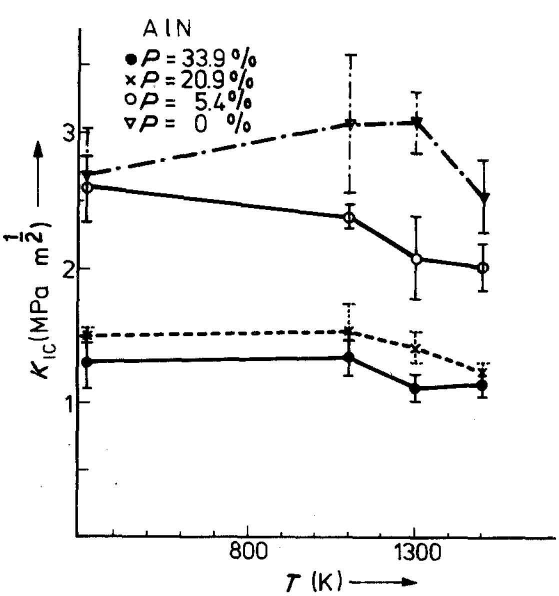


Figure 4 Strength of AlN as a function of temperature, for various samples of porosity, *P*. Bars at the data points indicate sample standard deviation.

The strength values show evidence of a sharp decrease with decreasing density. Boch *et al.* [3] reported a constant strength up to 10% porosity and a decrease at higher porosities. From the present result it is not quite clear whether they confirm this plateau or not.

For all materials the strength at 1500 K decreases by about 20% as compared with the room temperatue value. A decrease of about 50% is observed by Boch *et al.* [3] for their almost completely dense material, while Glandus *et al.* [1] indicate a decrease from 380 MPa at 300 K to 240 MPa at 1100 K and a constant value at temperatures up to 1500 K also for a fully dense material. Hence, there is only approximate agreement between the different experiments.

From the values for K_{Ic} and σ_f an estimate can be made of the size of the critical flaw a_c. Assuming semi-circular surface flaws, the value of Y in the equation $K_{Ic} = Y\sigma_f a_c^{1/2}$ is about 1.26 [13]. For the most dense material this results in a flaw size of about 50 μm. This is a fairly common value for ceramic materials.

The remark made in Section 3.2 regarding the fitting to empirical equations may also be made for strength.

4. Conclusion

The hot-pressed AlN investigated has only a slight anisotropy due to the hot-pressing technique. The Young's moduli for the completely dense material

TABLE III Comparison of strength data for AlN at room temperature

ρ (g cm^{-3})	Porosity, P	d (μm)	σ_f (MPa)	Remarks	Reference
3.20	0.02	5	265	hot-pressed, MOR	[4]
3.14	0.028	–	196	sintered, 4pb	[9]
–	–	–	80	sintered, MOR	[10]
–	0.02	9	380	hot-pressed, 3pb polished	[3]
–	0	–	380	sintered, 20 MPa N_2 3pb,	[1]
–	0.02	1	275	hot-pressed, 3pb	[10]
–	0.01–0.02*	1	200–250*	sintered, 3pb	[10]
3.0–3.4*	–	–	250–500*	sintered, 3pb polished, $R_a \sim 0.5\,\mu$m	[11]
3.28	–	2	310	hot-pressed, 3pb sawn, $R_a \sim 0.2\,\mu$m,	Present work
3.28	–	2	237	hot-pressed, 3pb, sawn, $R_a \sim 1.1\,\mu$m	Present work

*Dependent on sintering aid and sintering temperature.
Abbreviations: MOR, modulus of rupture, 3 (4) pb, three- or four-point bend.

measured parallel and perpendicular to the hot-pressing direction are 324 and 342 GPa, respectively. For the same material the strength and fracture toughness are about 300 MPa and 2.7 MPa m$^{1/2}$, respectively. The fracture toughness value decreases by about 10% up to 1500 K, while the strength is decreased by about 20%. All these properties show a strongly decreasing value with increasing porosity. In conclusion, the mechanical properties of these AlN ceramics are not outstanding, and they are only justified if other favourable properties such as corrosion resistance (see for example, [14]) prevail.

References

1. J. C. GLANDUS, J. L. BESSON and P. BOCH, *Sci. Ceram.* **11** (1981) 419.
2. G. DE WITH and N. HATTU, *ibid.* **11** (1981) 489.
3. P. BOCH, J. C. GLANDUS, J. JARRIGE, J. P. LECOMPTE and J. MERMAIN, *Ceramics Int.* **8** (1982) 34.
4. K. M. TAYLOR and C. LENIE, *J. Electrochem. Soc.* **107** (1960) 308.
5. H. F. POLLARD, "Sound Waves in Solids" (Pion Press, London, 1977) p. 25 and p. 145.
6. G. DE WITH and N. HATTU, *J. Mater. Sci.* **16** (1981) 841.
7. W. F. BROWN and J. E. SRAWLEY, ASTM-STP-40, ASTM, Philadelphia (1966).
8. M. I. MENDELSON, *J. Amer. Ceram. Soc.* **52** (1969) 443.
9. G. A. GOGOTSKI, *Ceramurgia Int.* **6** (1980) 31.
10. D. N. POLYBOYARINOV, M. R. GORDOVA, I. G. KUZNETSOVA, M. D. VERSHADSKAYA and V. G. ARETIKOV, *Inorg. Mat.* **15** (1979) 1616.
11. G. LONG and L. M. FOSTER, *J. Amer. Ceram. Soc.* **42** (1959) 53.
12. K. A. SCHWETZ, H. KNOCH and A. LIPP, "Nitrogen Ceramics 2", edited by F. C. Riley, in preparation.
13. G. K. BANSAL, *J. Amer. Ceram. Soc.* **59** (1976) 87.
14. R. A. SHARMA, R. A. MURIE and S. E. J. CAIRNS, *J. Electrochem. Soc.* **123** (1976) 1132.
15. R. L. STEWART and R. C. BRADT, *J. Amer. Ceram. Soc.* **63** (1980) 619.

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