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Reaction bonded aluminum oxide composites containing cubic boron nitride

A. Shonhiwa [a], M. Herrmann [b], I. Sigalas [c,*], N. Coville [a]

[a] DST/NRF Centre of Excellence in Strong Materials, School of Chemistry, University of the Witwatersrand, Johannesburg, South Africa
[b] Fraunhofer-Institute of Ceramic Technologies and Systems (IKTS), Germany
[c] DST/NRF Centre of Excellence in Strong Materials, School of Chemical and Metallurgical Engineering,
University of the Witwatersrand, Private Bag 3, Wits, Johannesburg 2050, South Africa

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Abstract

The properties of alumina can be improved by incorporating second phases like zirconia or carbides. It has also been reported that reaction bonded aluminum oxide (RBAO) process can be used as a host matrix for large scale second phase reinforcement particles without causing harmful residual stresses normally encountered with shrinking matrix materials. The potential of using cubic boron nitride in reinforcing alumina has not yet been reported. This work reports some improvements in mechanical properties of alumina achieved by incorporating cubic boron nitride particles in a reaction bonded aluminum matrix. Attrition milled aluminum, alumina and cubic boron nitride powders were heat treated to 800 °C in air followed by sintering to 1300 °C in argon. Sintered samples were found to have better density, hardness and fracture toughness compared to conventionally sintered samples of same composition.

Keywords: B. Composites; D. Al_2O_3; D. RBAO; Cubic boron nitride

1. Introduction

Reaction bonded aluminum oxide (RBAO) process is a technique used to produce dense alumina ceramics. In this process Al/Al_2O_3 mixtures are heat treated in air in such a way that all the aluminum metal gets converted into nanometer-sized Al_2O_3 crystals which subsequently are sintered and bond the primary Al_2O_3 particles [1] to form a dense body. One big advantage of this technique is the possibility of near net shaping resulting from partial compensation of the sintering shrinkage by an expansion associated with the oxidation of aluminum to alumina. It also has potential to be used as a host matrix for large scale second phase particles, e.g. platelets and fibres, without causing harmful stresses during sintering [1–4].

One of the well-known mechanisms of improving mechanical properties of alumina-based materials is the incorporation of submicrometer-sized particles in a matrix. For Al_2O_3/SiC composites Niihara [5] reported that room temperature increases in toughness from 3.5 to 5 MPa $m^{1/2}$. Scheppokat et al. [6] tested TiC and TiN as candidates for particle reinforcement of RBAO. Ceramic/metal/TiN composites were obtained which achieved a hardness of 20.4 GPa and flexural strength of 350 MPa.

Another possible candidate for reinforcing alumina matrix is cubic boron nitride. Cubic boron nitride (cBN) is the second hardest known material after diamond and is used intensively in the cutting tool industry when machining ferrous alloys. Conventional cBN-based composites available commercially as cutting materials contain a ceramic or metallic binder which facilitates sintering and optimize cutting performance. Usually metals of groups 4, 5 and 6 of the periodic table and/or other metallic elements, such as aluminum, cobalt and nickel are used to activate sintering [7,8].

The purpose of this work was twofold. Firstly to explore the possibility of incorporating cubic boron nitride as a second phase in an alumina matrix and then to compare reaction bonded aluminum oxide process and conventional sintering as two alternatives for incorporating cubic boron nitride in an alumina matrix. Since cubic boron nitride cannot withstand oxidizing conditions needed to fully convert aluminum into

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* Corresponding author. Tel.: +27 11 717 7502; fax: +27 11 717 7545.
E-mail address: iakovos.sigalas@wits.ac.za (I. Sigalas).

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alumina and sinter it to full density in the reaction bonded aluminum oxide process a two-step firing process was used. The first step meant for oxidizing aluminum into alumina was done in oxidizing environment followed by the sintering step in an inert atmosphere.

2. Experimental

The following raw materials were used: Al from Saarchem (RSA) with an assay purity of 99.8%, α-alumina (AKP 50) from Sumitomo, Japan with a purity of 99.99% and particle size in the range 0.1–0.3 μm and cubic boron nitride from Element Six (Pty) Ltd. (RSA) with particle size in the 2–5 μm range. The compositions of the mixtures are listed in Table 1. Notation for the samples contain a letter A (for conventionally sintered samples) and B (for reaction bonded samples) followed by a number denoting the percentage by volume of cubic boron nitride added. Samples for conventional sintering were prepared by dispersing weighed quantities of powder in acetone with 1.5% stearic acid as binder followed by drying and sieving. For reaction bonded samples, powders were prepared by attrition milling weighed quantities of Al, cBN and Al_2O_3 in an attrition mill fitted with a 750 ml Al_2O_3 vessel and Al_2O_3 blades. The milling was done using cyclohexane as solvent and 2 mm diameter Al_2O_3 balls at a speed of 600 rpm for 8 h. After rotovap drying, the powders were sieved through 38 μm sieve to break agglomerates.

All powders were pressed into pellets using a stainless steel die. Reaction bonded samples (B0, B10 and B30) were heated in air at a rate of 3 °C min^{-1} to 500 °C followed by a hold for 5 h to maximize oxidation in the solid state. The temperature was then raised to 800 °C at a rate of 3 °C min^{-1} followed by cooling at a rate of 10 °C min^{-1}. At this stage almost all the aluminum was converted into alumina. This was then followed by hot pressing in argon or vacuum at 1300 °C for 2 h using a load of 80 MPa.

Conventionally sintered samples were hot pressed for comparison. Phase analysis was done using X-ray diffraction. Diffraction patterns were collected using a Bruker AXS D8 machine equipped with a primary beam Göbel mirror, a radial soller slit, a V Åntec-1 detector and using Cu Kα radiation (40 kV, 40 mA). Data were collected in the 2θ range 5–90° in 0.021 steps, using a standard scan speed with an equivalent counting time of 14.7 s per step.

Densities of the sintered samples were determined by the Archimedes' method in distilled water and expressed as a percentage of the theoretical value. The theoretical density in each case was calculated by applying the law of mixture to the volume fractions of the constituent phases in each powder.

Table 1
Composition of precursor materials studied

Sample	Composition weight (%)			Composition volume (%)		
	Al_2O_3	Al	BN	Al_2O_3	Al	BN
A0	100	0	0	100	–	–
A10	91.15	0	8.85	90	0	10
A30	72.74	0	27.26	70	0	30
B0	59.58	40.42	0	50	50	0
B10	53.40	36.23	10.38	45	45	10
B30	24.95	42.32	32.73	20	50	30

Mechanical properties (hardness and fracture toughness) were determined by the Vickers indentation technique using a load of 10 kg for 10 s. Fracture toughness was determined by measuring crack lengths produced during indentation according to the equation [9]

$$K_{\mathrm{IC}} = 0.0889\sqrt{(H_{\mathrm{v}}P)/(4l)}.$$

Microstructural features of sintered samples were examined using scanning electron microscopy.

3. Results

The densities of reaction bonded and conventionally pressed samples are given in Table 2. The density in the reaction bonded state are in the range of 56–60% of theoretical density and therefore higher than for conventionally sintered samples which are in the range of 53–57% of the theoretical density.

Figs. 1 and 2 show diffractograms for materials sintered at 1300 °C for 2 h. The reaction bonded materials and the conventional sintered materials consisted of α-alumina and cubic boron nitride. No residual Al was observed.

Table 2
Properties of samples sintered for 2 h in argon under a pressure of 80 MPa

Sample	Reacted density (%)	Sintered density (%)	Hardness (GPa)	Fracture toughness (MPa $m^{1/2}$)
A0	57.3	96.4	18.5 ± 0.9	2.2 ± 0.1
A10	54.5	93.8	18.8 ± 1.0	2.7 ± 0.1
A30	53.2	91.2	19.2 ± 1.1	3.1 ± 0.2
B0	61.0	97.0	18.6 ± 0.8	2.0 ± 0.1
B10	58.2	96.7	20.2 ± 1.2	3.3 ± 0.2
B30	55.5	95.8	22.2 ± 1.1	4.0 ± 0.1

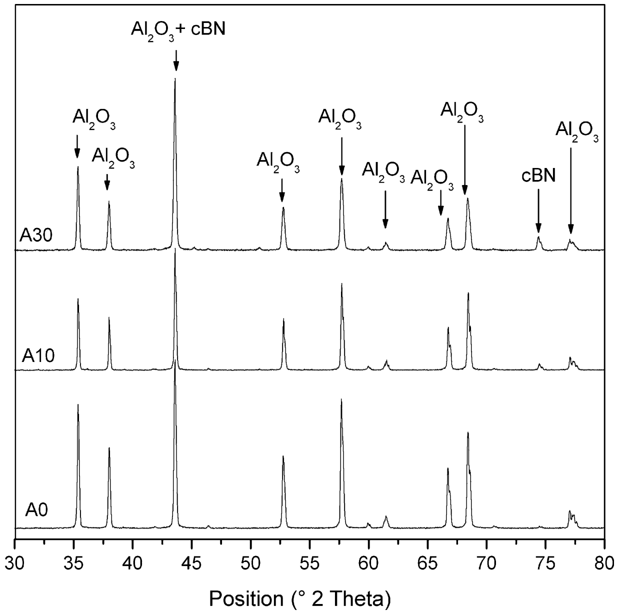

Fig. 1. X-ray diffractogram for conventionally sintered samples.

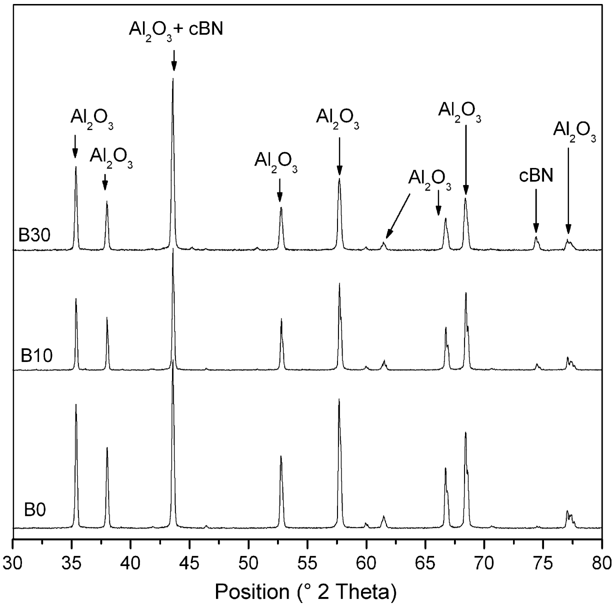

Fig. 2. X-ray diffractogram for reaction bonded samples.

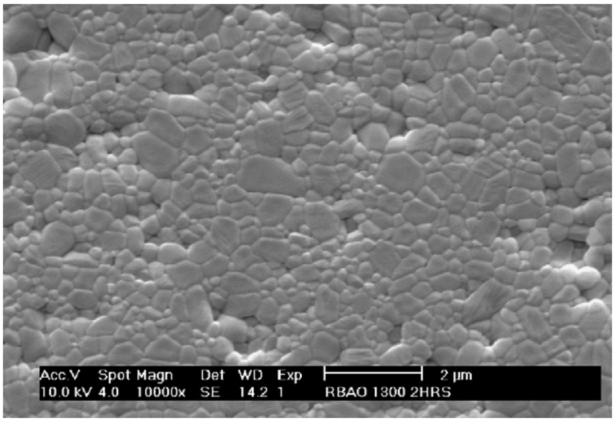

Fig. 3. Micrograph for reaction bonded sample B0 after thermal etching.

The densities after hot pressing are given in Table 2. In general reaction bonded materials showed after densification higher densities compared to their counterparts which were conventionally prepared. Highest degree of densification achieved was 97% TD for sample B0 (Fig. 3). This decreased gradually to 96.7 and 95.8% with increase in cBN content. This trend of decreasing densification with increase in cBN content was also observed in conventionally sintered samples and can be attributed to the fact that cBN behaves as a rigid inclusion during sintering.

Reaction bonded samples also show higher hardness and fracture toughness values compared to their conventionally sintered samples. Hardness and fracture toughness values for both samples improve with increase in cBN content. Improvements in hardness with cBN content can be attributed to the hardness of cBN which is much higher than that of alumina. Improvements in fracture toughness with increase in cBN are due to the effects of crack deflection and grain bridging by cBN grains as shown in Fig. 4. Crack deflection is due to the intrinsic stresses produced by the thermal mismatch of the two phases during cooling. This is caused by the different thermal expansion coefficients $7.0 \times 10^{-6}\,\mathrm{K}^{-1}$ for corundum and $5.4 \times 10^{-6}\,\mathrm{K}^{-1}$ for cubic boron nitride.

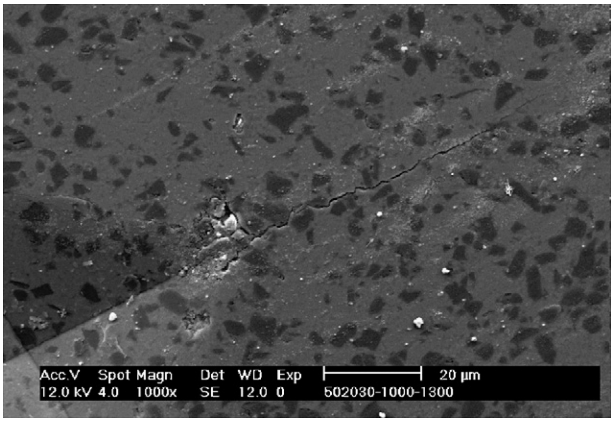

Fig. 4. Micrograph for reaction bonded sample B30 showing a crack deflection around cBN particles.

4. Conclusions

The mechanical properties of alumina were improved by the addition of cubic boron nitride. Hardness and fracture toughness of alumina were 22.2 GPa and 4.0 MPa m$^{1/2}$, respectively. Densification of the composites was better in the reaction bonded composites compared to conventional sintering of same compositions. For both reaction bonded and conventionally sintered samples densification decreased with increase in cubic boron nitride loading.

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