Al2O3-Ni Composites with High Strength and Fracture Toughness

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Al_2O_3–Ni Composites with High Strength and Fracture Toughness

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Al_2O_3–Ni composites were prepared by the reactive hot pressing of Al and NiO. The composites had a two-phase, interpenetrating microstructure and contained ~35 vol% Ni. They exhibited an impressively high combination of strength and toughness at room temperature; the four-point bending strength was in excess of 600 MPa with a fracture toughness of more than 12 MPa·m$^{1/2}$. Examination of fracture surfaces showed that Ni ligaments underwent ductile deformation during fracture. SEM analysis revealed knife-edged Ni ligaments with a limited amount of debonding around their periphery (i.e., at the Ni–Al_2O_3 interface), indicating a strong Ni–Al_2O_3 bond.

I. Introduction

Al_2O_3–Ni composites have been prepared by several methods, including sol–gel processing,[1,2] pressureless sintering,[3,4] reduction of $NiAl_2O_4$,[5] and reactive hot pressing.[6,7] Ceramic–Ni composites are attractive because of the potential for high strength and toughness at room temperature combined with resistance to corrosion and oxidation at high temperature. To date, the mechanical performance of Al_2O_3–Ni composites has been significantly below that reported for other ceramic–metal composites.[1–9] For example, Al_2O_3–Al composites produced by reactive metal penetration of porous mullite preforms contained 38 vol% Al and had 315 MPa strength with 10.6 MPa·m$^{1/2}$ toughness.[9] The strength and toughness of Al_2O_3–Ni composites prepared by reactive hot pressing were reported to be 250 MPa with 6.4 MPa·m$^{1/2}$ while comparable materials prepared by conventional sintering had less than 200 MPa strength and 2.5 MPa·m$^{1/2}$ toughness.[3,6] Other reports have not included strength values, presumably because of unacceptably low values; toughness values ranging from 4 to 8 MPa·m$^{1/2}$ were reported.[1,2,5,7] The poor mechanical performance was attributed to a lack of adhesion at the metal–ceramic interface and porosity.[1–7] Indeed, it has been suggested that the formation of a small amount of $NiAl_2O_4$ at the Ni–Al_2O_3 interface is necessary to promote good adhesion.[10]

The reaction of Al with oxides has been used to prepare several different composites including Al_2O_3–Ni.[7,11,12] Composites can be formed by a displacement reaction between NiO and molten Al following reaction (1):

$$2Al + 3NiO \rightarrow Al_2O_3 + 3Ni \tag{1}$$

Reaction (1) is thermodynamically favorable and it generates a great deal of heat, $\Delta G_{rxn} = -890$ kJ at 1000°C.[13] If the reaction is not properly moderated, the heat generated can lead to a thermal avalanche condition known as combustion synthesis or self-propagating high-temperature synthesis (SHS).[14] Reaction control is possible if the heat input to the system is limited or if inert diluents such as Al_2O_3 are added to the reactants.

This note describes the processing and a preliminary investigation of the room-temperature mechanical properties of Al_2O_3–Ni composites prepared by reactive hot pressing.

II. Experimental Procedure

Nickel oxide (NiO, 99%, Alfa/Aesar) and Al (99.5% Alfa/Aesar) were combined in the molar ratio given by reaction (1). The powders were mixed with methanol and then attrition milled (Model 01-HD, Union Process, Akron, OH) in a polymer-coated container at 600 rpm for 1 h using ZrO_2 media. After drying, the powder was passed through a 35 mesh sieve. Next, the powder was loaded into a 63.5 mm diameter graphite die that had been spray-coated with BN. The die was inserted into the hot-press chamber, which was then evacuated to 10^{-2} torr and backfilled with UHP argon (repeated three times). Next, the assembly was heated at 1°C/min. When the die temperature reached 1450°C, a pressure of 34.5 MPa (5000 psi) was applied. The furnace was held at 1450°C for 30 min, and then cooled using a controlled ramp of 20°C/min. The pressure was released when the die temperature was less than 300°C. After removal from the die, the composite density was determined using the Archimedes technique with water as the saturation/suspension medium. Hot-pressed billets were cut into bend bars (3 mm × 4 mm × 45 mm) for analysis of mechanical properties. Strength was measured in four-point bending, modulus was determined by a pulse–echo technique, and fracture toughness was measured using single-edge v-notched beams fractured in four-point loading. The composite hardness was measured using Vickers indentation with loads of 78.4 and 98.0 N; the reported value is the average of 10 indentations at each load. The microstructure and fracture surfaces were observed using scanning electron microscopy and composite phase analysis was accomplished by X-ray diffraction analysis.

III. Results and Discussion

The density of hot-pressed composites, determined by the Archimedes technique, was 5.52 g/cm^3. The composite was slightly Al_2O_3-rich compared with the composition predicted by reaction (1) because a small amount of Ni was squeezed out of the die during hot pressing. From the mass of Ni exuded from the die,

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Table I. Al_2O_3–Ni Composite Property Comparison

	Ni content (vol%)	Relative density (%)	Bend strength (MPa)	Fracture toughness ($MPa \cdot m^{1/2}$)	Hardness (GPa)	Young's modulus (GPa)
This study	35	97	613 ± 20	12.1 ± 0.2	5.4 ± 0.4	292
Tuan[3]	20	98	200	2.5		
Jones[6]	40	90	250	6.4	2	209

a composition of 35 vol% Ni and 65 vol% Al_2O_3 was estimated. The theoretical density based on this composition is 5.70 g/cm^3, giving a relative density of ~97%. This is in good agreement with SEM micrographs that show almost no porosity. X-ray diffraction analysis (data not included) identified Ni and α-Al_2O_3 in the composite.

The four-point bend strength of the composites was 613 ± 20 MPa (seven samples) and a toughness of 12.1 ± 0.2 $MPa \cdot m^{1/2}$ (three samples) was measured. The physical and mechanical property data are summarized in Table I. For comparison, the properties of composites prepared by pressureless sintering[3] and another reactive hot-pressing technique[6] are also included.

Composites produced for this study had a two-phase, interpenetrating microstructure. That is to say, both the Ni and the alumina phases were continuous through the microstructure, similar to other reactively formed ceramic–metal composites.[6,8,9,11,12] The continuity of the ceramic phase gives the composite a high Young's modulus (Table I) and a low thermal expansion coefficient.[15] Electrical conductivity measurements indicated that the Ni phase was also continuous across hot-pressed billets. Figure 1 shows the polished cross section of a composite after Vickers indentation. Typically, radial cracks extended directly away from the corners of an indentation. Cracks were not drawn preferentially to the ceramic–metal interface, as reported for other Al_2O_3–Ni composites.[2,3,6] Examination of crack paths shows evidence for extensive crack bridging. In low-toughness Al_2O_3–Ni composites, cracks generally travel along the metal–ceramic interface with very little crack bridging due to poor adhesion.[1–7,10] A strong bond at the ceramic–metal interface is required to promote energy absorption by ductile deformation during fracture.[10] Further evidence of a strong Al_2O_3–Ni bond in the present composites is given by examining the composite fracture surface. A typical fracture surface is shown in Fig. 2. Ni ligaments are bound strongly enough to the Al_2O_3 that knife edging occurs before failure. Only a small amount of debonding occurs at the periphery of Ni ligaments.

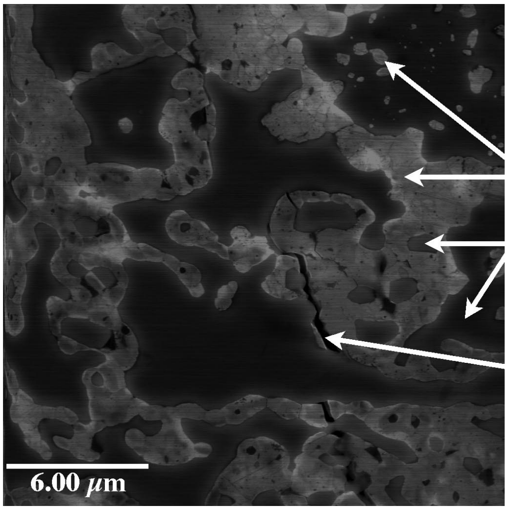

Fig. 1. SEM micrograph showing an indentation crack in an Al_2O_3–Ni composite.

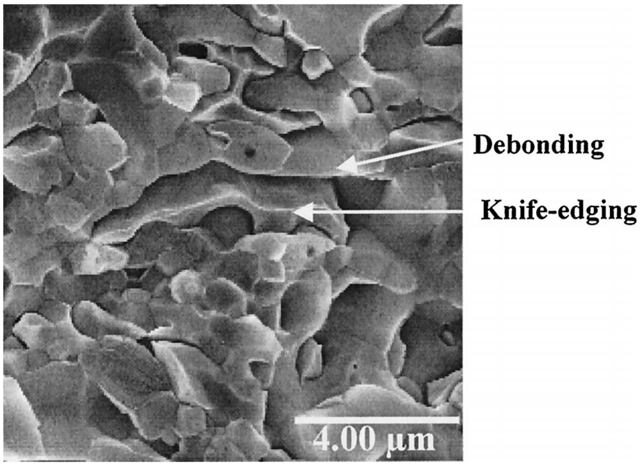

Fig. 2. SEM micrograph of the fracture surface of an Al_2O_3–Ni composite.

IV. Summary

Al_2O_3–Ni composites that contained ~35 vol% Ni were prepared by reactive hot pressing. The composite strength was over 600 MPa and the fracture toughness was in excess of 12 $MPa \cdot m^{1/2}$. Both are significantly higher than previously reported for other Al_2O_3–Ni composites. SEM analysis revealed a co-continuous microstructure. Indentation cracks propagated directly away from the corners of Vickers indentations, and they were not preferentially drawn to the Ni–Al_2O_3 interface. More importantly, this allowed the Ni ligaments to bridge cracks, and, in turn, increase the toughness of the composite. Observation of fracture surfaces showed knife edging of the Ni ligaments with a limited amount of debonding at the Ni–Al_2O_3 interface.

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