

Densification and properties of AlN ceramic bonded carbon

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Abstract

To obtain light and tough materials with high thermal conductivity, AlN ceramic bonded carbon (AlN/CBC) composites were fabricated at temperatures from 1600 to 1900 °C in a short period of 5 min by the spark plasma sintering technique. All AlN/CBCs (20 vol% AlN) have unique microstructures containing carbon particles of 15 μm in average size and continuous AlN boundary layers of 0.5–3 μm in thickness. With an increase in sintering temperature, AlN grains grow and anchor into carbon particles, resulting in the formation of a tight bonding layer. The AlN/CBC sintered at 1900 °C exhibited a light weight (2.34 g/cm³), high bending strength (100 MPa), and high thermal conductivity (170 W/mK). © 2011 Elsevier Ltd. All rights reserved.

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1. Introduction

Due to increased performance in a wide range of engineered products ranging from computer processors to high power semiconductor devices, there is a critical need for improved thermal management systems.^{1,2} These systems must not only quickly release thermal energy, but also must be smaller and lighter, as well as have high strain tolerance under thermal and/or mechanical shock conditions.

Carbon/graphite materials have the advantages of being lightweight (1.8–2.2 g/cm³), having high corrosion resistance and thermal conductivity, and having a similar coefficient of thermal expansion (CTE) to semiconductor devices.^{3,4} However, because bulk carbon materials have a lower strength and are difficult to join with other materials,⁵ applications for thermal management are limited.

Ceramic bonded carbon (CBC) is a novel carbon-based composite proposed recently.⁶ This material has a unique microstructure consisting of carbon particles and ceramic boundary layers, as schematically illustrated in Fig. 1. In CBCs, the ceramic network bonds carbon particles together and provides high strength, high oxidation resistance, and other functional properties as required. In addition, CBCs can be easily joined with the same or other ceramics, and thus can be

fabricated into a high heat dissipative, insulated substrate for electronic devices.

To realize this CBC concept, a process combining gelcasting and spark plasma sintering (SPS) method has also been developed.⁶ The AlN/CBC prepared using this method showed a higher strength and a higher thermal conductivity compared to conventional AlN/carbon (AlN/C) materials made by a ball-milling method. However, the density of obtained AlN/CBC is still low (92% theoretical density), and some issues such as how the AlN bonding layer in the AlN/CBC affects the mechanical and thermal properties still need to be investigated.

In the present study, the densification and microstructure development of AlN/CBCs (containing 20 vol% AlN) SPS sintered at different temperatures from 1600 °C to 1900 °C were observed systematically and analyzed in order to understand the densification behavior and obtain highly dense samples. Effects of the AlN network layer and the sintering temperature on mechanical and thermal properties of CBCs were investigated as well.

2. Experimental procedure

2.1. Starting materials

The starting materials included a meso phase graphite powder made from meso phase pitch carbon by a graphitization step at 2500 °C (Toyo Tanso Co. Ltd.) and AlN powder (Tokuyama

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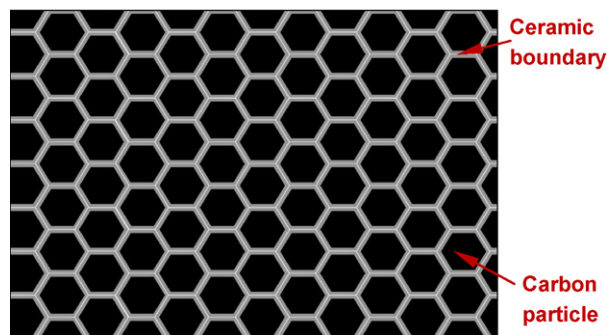


Fig. 1. A concept of ceramic bonded carbon (CBC).

Co. Ltd.) that contained 5 wt% Y_2O_3 as a sintering additive. The characters of starting powders can be found in our previous paper.⁶

2.2. Sample preparation

To prepare the AlN/CBC green body by the gel-casting method, acrylamide (AM) as the monomer and methylenebisacrylamide (MBAM) as the cross-linker were first dissolved in 1-propanol to form a premix at a weight ratio of 8(AM):1(MBAM):45 (1-propanol). The AlN and carbon powders (20:80 volume ratio of the solid) were then added sequentially to the premix for 3 min in a high-speed (2000 rpm) centrifugal mixer (AR-250, Thinky Co. Ltd.) to form a 65 vol% slurry. The mixed slurry was cast into a plastic mold and then heated at 80 °C to form a solid body via the monomer-polymer transition. After de-molding, the dried green body was heated at 700 °C under vacuum to burn out the gel binder. The green body was then loaded into a 25-mm diameter graphite die.

The sintering was carried out in vacuum at temperatures from 1600 to 1900 °C for 5 min at a heating rate of 100 °C/min and a pressure of 30 MPa using a Dr. 1050 spark plasma sintering (SPS) apparatus (Sumitomo Coal Co. Ltd., Japan). The graphite dies were covered with carbon felt to inhibit the thermal radiation, which should make the detected temperature close to the sample temperature.

2.3. Characterization

Microstructure characterization was carried out using a field emission scanning electron microscope (FE-SEM, ERA-8800, Elionix). X-ray diffraction (XRD, JDX-3530M, JEOL) was employed to characterize the phase development of sintered samples. The density of the sintered pellets was measured by the Archimedes method. The theoretical densities of graphite (2.16 g/cm³) and AlN (3.3 g/cm³) were used to calculate relative densities. Rectangular bars, 3 mm × 2 mm × 20 mm, finished with a 60 μm-diamond disk were used to measure the three-point bending strength using a Table-Top Universal Tester (EZ-Test Type S, Shimadzu) with a span of 15 mm at room temperature. The speed of the crosshead displacement was 0.5 mm/min. The

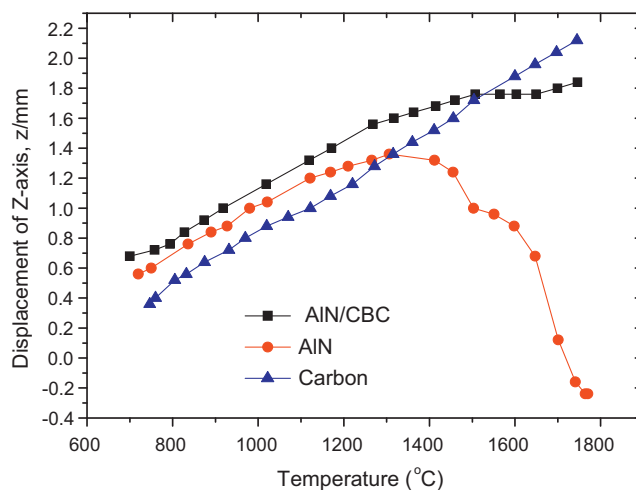


Fig. 2. Sintering curves of carbon, AlN/CBC and AlN samples in SPS.

thermal conductivity of sintered pellets was measured by the laser-flash method (TC-7000, ULVAC-RIKO).

3. Results and discussion

3.1. Densification of AlN/CBC

The densification of AlN/CBC was evaluated based on the displacement of a graphite punch rod in the Z-axis during sintering.⁷ For comparison, the densifications of monolithic carbon and AlN were also monitored. Fig. 2 illustrates recorded displacements of the punch rod for different specimens with increasing temperature up to 1750 °C. This recorded expansion is mainly contributed by the expansion of graphite punches due to the temperature evolutions, while the shrinkage can be attributed to the sample due to the densification. As shown, the monolithic carbon sample keeps expanding in the entire temperature range. After sintering at 1750 °C, the carbon sample was still in a powder form. On the contrary, an obvious shrinkage was observed in the AlN sample from around 1300 °C. It was fully densified at 1750 °C. Compared with the carbon and AlN

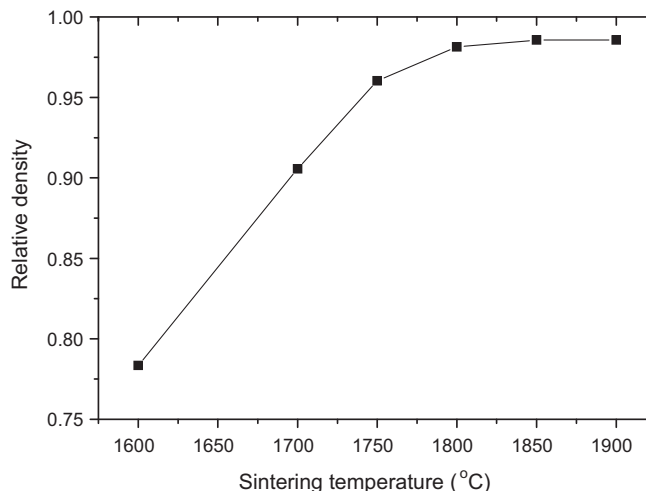


Fig. 3. Relative densities of AlN/CBCs as a function of sintering temperature.

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