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Joining of AlN Ceramic Bonded Carbon with AlN and W[†]

CHEN Weiwu*, MIYAMOTO Yoshinari**

Abstract

To obtain light and tough materials with high thermal conductivity, ceramic bonded carbon (CBC) composites consisting of carbon particles and thin ceramic boundaries were developed. The AlN ceramic bonded carbon (AlN/CBC, 20vol% AlN) could be densified by spark plasma sintering or hot-pressing, and exhibited light weight ($\sim 2.34 \text{ Mg/m}^3$), high bending strength ($\sim 100 \text{ MPa}$), and high thermal conductivity ($\sim 170 \text{ W/mK}$). AlN/CBC could also be joined strongly with ceramic (AlN) and some metals (W) due to its unique microstructure. These results suggest that the AlN/CBCs have large potential to be used as structural components and heat sinks for electrical and plasma facing devices.

KEY WORDS: (Ceramic bonded carbon), (Joining), (Aluminum nitride), (Tungsten)

1. Introduction

The thermal management system is a critical need in electronic, medical, and energy industries, especially for high power semiconductor devices, high speed X-ray target and plasma facing components, and others[1-3]. The system is regarded as releasing thermal energy, to be lightweight, to have a small differences of thermal expansion (CTE) in device materials, and a high strain tolerance under thermal and/or mechanical stress conditions.

Carbon/graphite materials have advantages of lightweight ($1.8\text{-}2.2 \text{ Mg/m}^3$), high corrosion resistance, thermal conductivity, and low CTE value[4-5]. However, bulk carbon materials are weak and difficult to join with other materials[6], making them unsuitable for thermal management applications.

Recently, we proposed a novel carbon-based composite, called ceramic bonded carbon (CBC), as shown in Fig. 1[7-8]. CBCs have a unique microstructure consisting of carbon particles and ceramic boundaries. In CBCs, the ceramic network bonds carbon particles together and provides high strength and other functional properties as required. Moreover, CBCs can be joined with other ceramics and metals because of the unique microstructure.

To realize this CBC concept, a process combining gelcasting and pressure sintering has been developed. In this report, the fabrication and properties of AlN/CBC (20 vol%) were examined. The joining of AlN/CBC to AlN and W were also examined. Joining mechanisms of AlN/CBC with AlN and W respectively were discussed, as well.

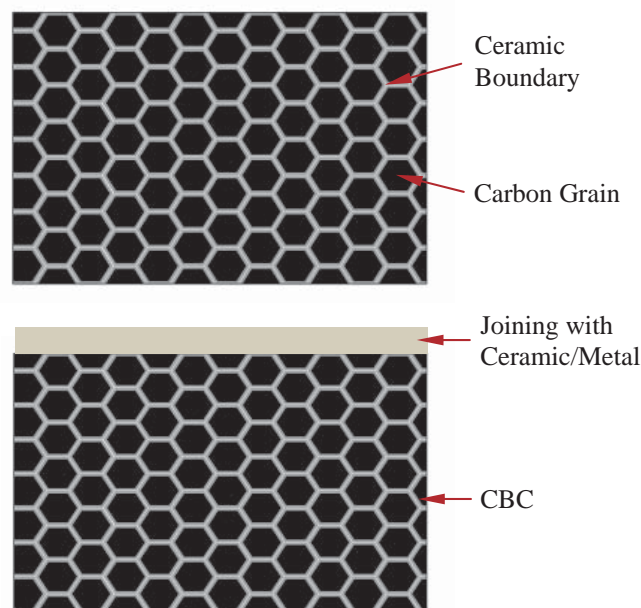


Fig.1 Concept of Ceramic Bonded Carbon (CBC).

2. Experimental

2.1 Fabrication of AlN/CBC

Carbon powders made from meso phase pitch carbon by a graphitisation step at 2500°C (Toyo Tanso Co. Ltd.) and AlN powders (Tokuyama Co. Ltd.) containing 5 wt% Y_2O_3 as a sintering additive were used as starting materials.

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To prepare the AlN/CBC green body by gelcasting 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 in a volume ratio) were then sequentially added to the premix and mixed for 3 minutes in a high-speed (2000 rpm) centrifugal mixer to form a 65 vol% slurry. The mixed slurry was cast into a plastic mold and then dried at 80°C to form a solid body via the monomer-polymer transition. After demolding, the dried green compacts were heated to 700°C under vacuum to burn out the gel binder. The green bodies were then loaded into graphite dies for spark plasma sintering (SPS) under a pressure of 30 MPa at 1900°C in vacuum for 5 minutes or hot-pressing (HP) in a N₂ atmosphere under a pressure of 30 MPa at 1950°C for 1 hour.

2.2 Joining AlN/CBC with AlN

For joining AlN/CBC to AlN, AlN powders were stacked on the top of an AlN/CBC green body and subjected to hot-pressing (HP) in a N₂ atmosphere under a pressure of 30 MPa at 1950°C for 1 hour at a heating rate of 10 °C/min.

2.3 Joining AlN/CBC with W

For joining AlN/CBC to W, firstly, the sintered AlN/CBC disk (Φ 25mm) was finished with a #80 SiC paper and then loaded into 25-mm diameter graphite dies. To form a 0.5 mm-thick W layer, 4.8 g of W powder was stacked onto the AlN/CBC disk, and then sintered in vacuum at 1700°C at a heating rate of 100°C/min and a uniaxial pressure of 30 MPa for 5 minutes by SPS.

2.4 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 identify crystalline phases of sintered AlN/CBC. 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, 3×2~4×20 mm³, were prepared to measure the three-point bending strength using a table-top universal tester (EZ-Test Type L, Shimadzu) with a span of 15 mm at room temperature. The speed of the crosshead displacement was 0.5 mm/min. The thermal conductivity of sintered pellets was measured by the laser-flash method (TC-7000, ULVAC-RIKO).

3. Results and Discussion

3.1 Fabrication and properties of AlN ceramic bonded carbon

By using a gelcasting method, an AlN/CBC green body with a unique microstructure containing separated carbon particles and a continuous AlN boundary layer can be obtained. **Figure 2a** shows the microstructure of

sintered AlN/CBC that has the unique microstructure containing carbon particles of 15 μ m in average size and an AlN boundary layer of 0.5-3 μ m in thickness. The fractured surface of AlN/CBC after strength measurements was observed under SEM, as seen in **Fig. 2b**. Almost all carbon particles are broken to some extent, showing a typical trans-granular mode. This indicates a strong bonding at boundaries. The measured density, bending strength, and thermal conductivity of AlN/CBC are listed in **Table 1**. AlN/CBC showed a density of 2.32 g/cm³ (98% of theoretical density), bending strength of 100 MPa, and thermal conductivity of 170 W/mK. The higher strength and thermal conductivity of AlN/CBC is attributed to its higher density and reinforcement of the AlN boundary network.

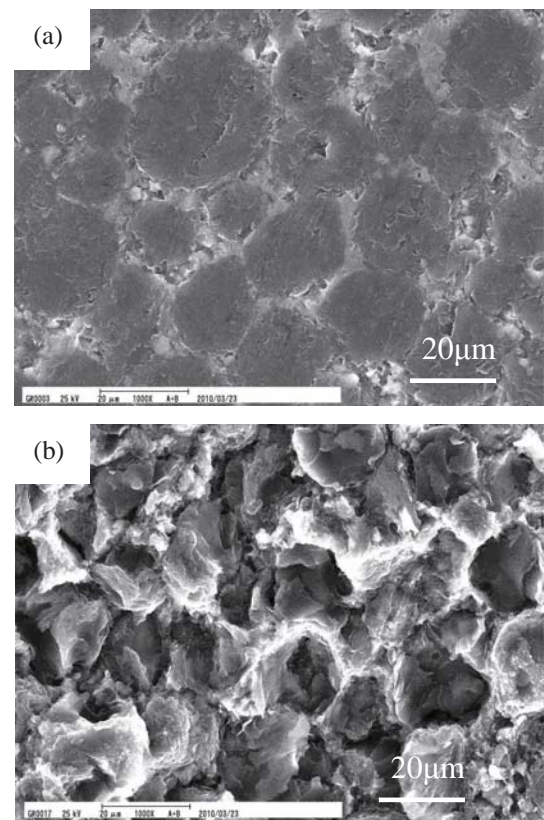


Fig.2 Microstructures of AlN/CBC (a) polished surface; (b) fractured surface.

Table 1 Properties of AlN/CBC and its joints with AlN and W

Samples	Sintering method	Density (g/cm ³)	Relative density	Strength (MPa)	Thermal conductivity (W/mK)
AlN/CBC	SPS	2.34	98%	100	170
AlN/CBC	HP	2.30	96%	112	170
AlN/CBC -AlN	HP	-	-	158	-
AlN/CBC -W	SPS	-	-	170	-

To understand the bonding mechanism between AlN boundaries and carbon particles, the fractured surface was further observed under a high magnification, as shown in **Fig. 3a**. AlN grains are 1~3 μm in size and some of them obviously penetrate into carbon particles, indicating a strong mechanical bonding. A boundary layer of AlN stopping crack propagation was also noticed, as denoted by an arrow. Because no reaction was seen between AlN and carbon by XRD analysis, the bonding mechanism of AlN boundaries and carbon particles is mainly attributed to a mechanical anchor or interlock effect. How the continuous AlN network can reinforce the composite is schematically illustrated in **Fig.3b**. During fracture, a tough AlN network can inhibit and deflect the crack propagation. In case of a weak interface, a crack will propagate along the interface between the carbon particles and the AlN layer, while with a strong interface bonding, as in AlN/CBC, the crack will propagate through the carbon particles.

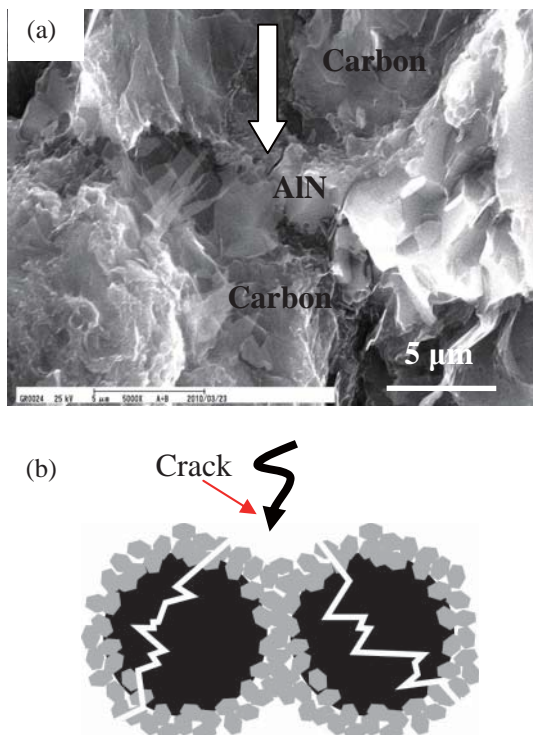


Fig.3 (a) AlN boundaries stop the propagation of a crack denoted by an arrow; (b) Schematic of reinforcing mechanism.

The thermal conductivity of AlN/CBC composites can be considered mainly as a contribution of phonon propagation, as described by the equation of $\lambda = C_p v L$ (C_p is specific heat of the bulk materials, v is the sound velocity, and L is the mean-free-path of phonons.)[9-10]. In AlN/CBCs, L is a varying parameter containing L_{carbon} and L_{AlN} . L_{carbon} may not change much before and after sintering, but L_{AlN} will become larger with the growth of AlN grains. In addition, in AlN/CBCs the phonon propagation will be degraded largely due to scatterings from pores, defects and interfaces. Because of the

presence of AlN, CBC can be highly densified, with the porosity and interface area decreasing with an increase in sintering temperature. Thus, the measured thermal conductivity of AlN/CBC is higher than most of the graphite bulks that contain about 10% porosity.

3.2 Joining AlN/CBC with AlN

Compared with SPS, hot-pressing provides a more homogenous temperature in large samples, even for composites of electrical conductive and insulating layers. In this work, $\Phi 50\text{mm}$ plates composed of the top AlN layer joined on the AlN/CBC were prepared by hot-pressing. The bending strength and thermal conductivity of AlN/CBC were measured, as listed in Table 1. The joining interface of AlN-AlN/CBC was analyzed.

The hot-pressed AlN/CBC has a similar density, bending strength and thermal conductivity compared with the SPS treated AlN/CBC. Moreover, a joint of AlN (0.7 mm thick) and AlN/CBC (1.8 mm thick) showed an improved bending strength in comparison with the monolithic AlN/CBC, reaching 158 MPa. During the bending test, the AlN layer faced the tensile plane of a test bar, as illustrated in **Fig.4a**. But there was no large difference in strength when the AlN/CBC faced the tensile plane. The load-displacement responses of the monolithic AlN/CBC and the joint are illustrated in **Fig. 4b**. The joint showed a typical response of linear elasticity up to fracture, similar to the monolithic AlN/CBC. Assuming that a crack initiates from the AlN layer, the flexural response of the joint may mean that the crack in the AlN layer propagates readily into the AlN/CBC layer. No failure occurs along the interface of the AlN layer and AlN/CBC. Therefore, the bonding between the AlN layer and AlN/CBC is considered fairly strong[11].

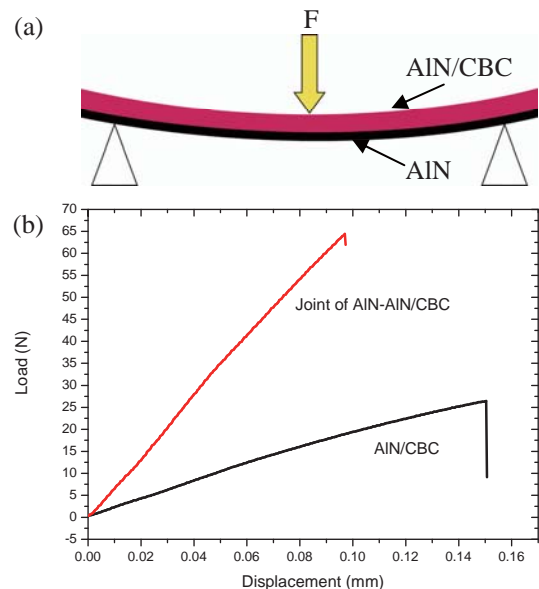


Fig.4 (a) Schematic illustration of measuring method; (b) Load-displacement curves of a monolithic AlN/CBC and an AlN/CBC-AlN joint.

The cross-section of the joint is shown in **Fig. 5a**. Both layers are tightly bonded, and no delamination is observed even after a bending test. AlN grains of the AlN layer are directly sintered with AlN grains at the surface of the AlN/CBC layer. The fractured surface of the joint is shown in **Fig. 5b**. The AlN layer shows a typical inter-granular fracture mode, whereas the AlN/CBC layer shows a trans-granular fracture mode. In the interface, both AlN grains belonging to the AlN layer and AlN/CBC are bonded together. This further indicates that the joining of the monolithic AlN and AlN/CBC layer is very strong due to the direct AlN-AlN sintering at their interface.

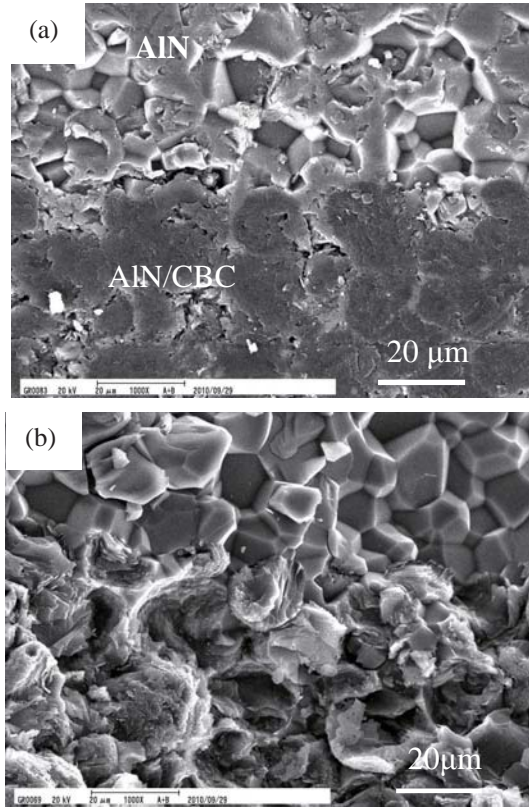


Fig.5 Interface microstructures of AlN/CBC-AlN joint (a) polished surface; (b) fractured surface.

3.3 Joining AlN/CBC with W

The side view of the polished joint of AlN/CBC-W is shown in **Fig. 6a**. The W layer, around 0.3 mm in thickness, is strongly bonded to AlN/CBC. No void was observed at the interface. **Figure 6b** shows XRD patterns of W layer, AlN/CBC, and the interface areas. In the AlN/CBC area, besides the peaks of graphite and AlN, small amounts of Y_2O_3 and $Al_2Y_4O_9$ were detected because the AlN in AlN/CBC contains 5wt% Y_2O_3 as a sintering additive. In the interface area, small WC peaks are observed besides other peaks appearing in W and AlN/CBC layers. No reaction occurs between W and AlN as previously reported[12].

The joint of AlN/CBC-W sample (0.3-mm thick W and 2.2-mm thick AlN/CBC layer) exhibits an average bending strength of 172 MPa, much higher than the

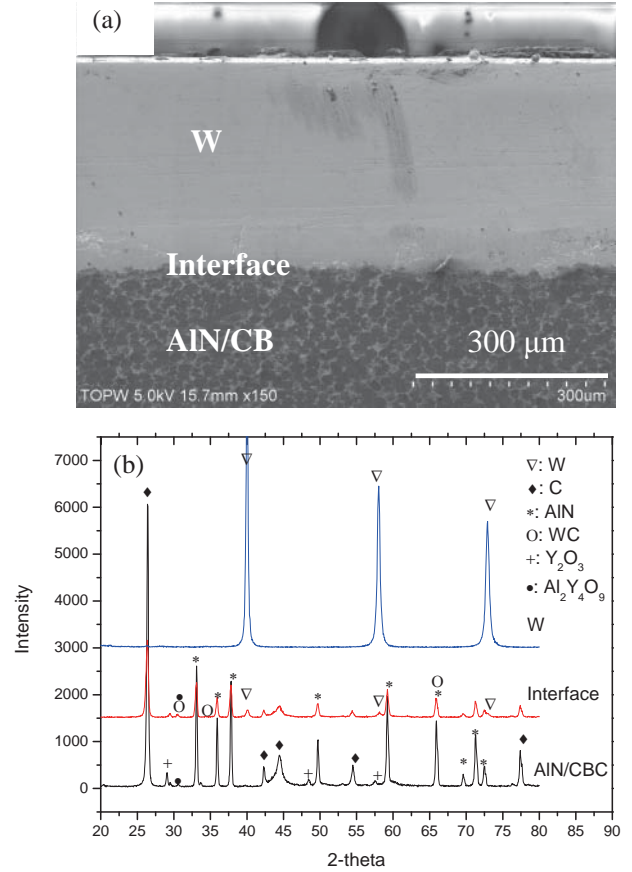


Fig. 6 (a) Cross sectional view of the AlN/CBC-W joint, (b) XRD patterns of W, interface, and AlN/CBC layers of the joint.

monolithic AlN/CBC. The W layer side was placed on the tensile side in bending test. Load-displacement response of AlN/CBC-W joint also shows a typical response of linear elasticity up to fracture, similar to that of monolithic AlN/CBC. Therefore, the bonding between W layer and AlN/CBC layer is also considered strong.

The fractured surface of the AlN/CBC-W joint was observed, as shown in **Fig. 7**. The phase compositions change gradually from W to the AlN/CBC side in the following sequence: W layer, WC/W₂C layer, ceramic layer, and AlN/CBC. The W layer contains grains with a size around 5-10 μm , whereas WC/W₂C grains have an average size around 30 μm . The thickness of the WC/W₂C layer is around 70 μm . The ceramic layer is around 10 μm in thickness, and tightly bonded with both WC/W₂C and CBC layers. EDS analysis shows that this new ceramic layer contains large size AlN grains and a Y-Al-O glass.

Based on the above results, the phase composition and microstructure of AlN/CBC-W joint is illustrated in **Fig. 8**. During sintering of W powders there is accompanying grain growth, whereas there is no change in AlN/CBC because it has been pre-sintered at a higher temperature of 1900°C than the joining temperature (1700°C). Between the W and AlN/CBC layers, W will react with carbon and form WC/W₂C, but will not react

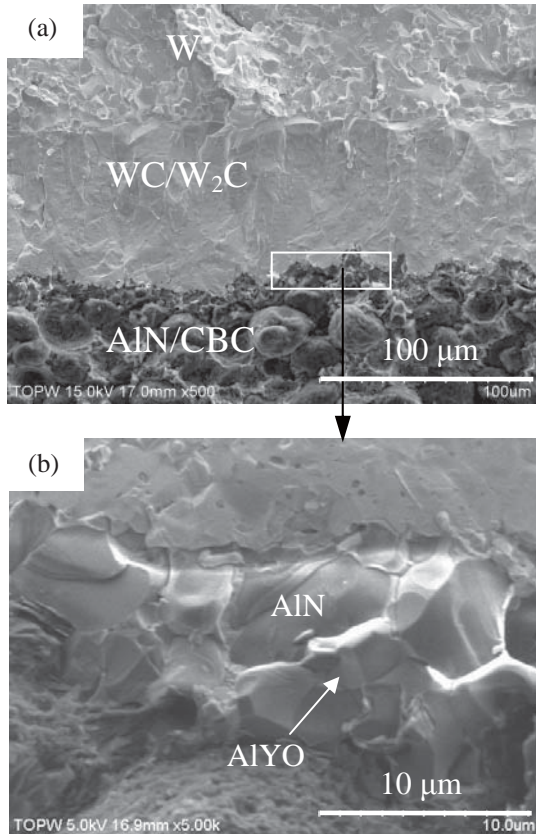


Fig.7 Fractured surface of a AlN/CBC-W joint (a) low magnification; (b) high magnification.

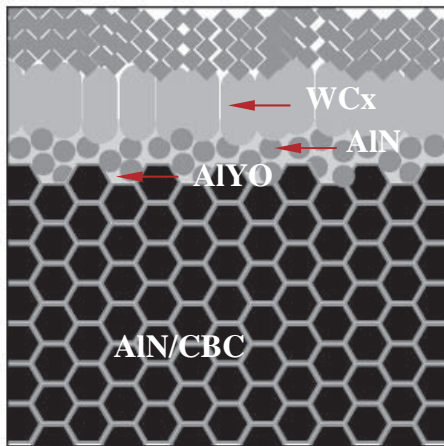


Fig. 8 Schematic illustration of the interface structure of AlN/CBC-W joint.

with AlN boundaries that cover carbon particles. The remaining AlN boundary layer that contains AlN and 5wt% Y_2O_3 may separate and aggregate from the W-C reaction layer gradually at high temperature, and then form a new ceramic layer. This newly formed ceramic layer is strongly connected with the AlN/CBC by the ceramic-ceramic bonding. On the other hand, the upper region of the ceramic layer, as illustrated in Fig. 8, is

bonded tightly with the WC/ W_2C layer probably due to the interlocking of the ceramic and WC/ W_2C grains at the interface. Therefore, the unique interface layer containing WC/ W_2C and ceramic layer results in the strong joining of W and AlN/CBC. The formation of this interface structure is related to the unique microstructure of AlN/CBC, which is composed of carbon particles and AlN ceramic boundaries.

4. Conclusions

We proposed a novel ceramic bonded carbon that exhibits not only high strength and high thermal conductivity, but also joining strongly with ceramics and metals. The results can be summarized as follows.

1. AlN/CBC containing the ceramic network bonding of carbon particles could be densified by SPS or hot-pressing, and exhibited a lightweight, high bending strength, and high thermal conductivity.
2. A monolithic plate composed of an AlN layer on top of AlN/CBC could be fabricated by sintering and joining in one step by hot pressing at 1950°C. Joining is very strong as a result of direct AlN-AlN sintering at the interface of AlN layer and AlN/CBC.
3. AlN/CBC could be also joined with W metal strongly by SPS. The interface consists of WC/ W_2C and ceramic layers resulting from the reaction between W and AlN/CBC. The formation of this unique interface is due to both the composition and structure of AlN/CBC.

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