Fabrication of SiC Ceramic Bonded Carbon and Its Joining with W†

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Abstract

To obtain light and tough ceramic bonded carbon (CBC) materials with high thermal conductivity SiC Ceramic bonded carbon (SiC/CBC) is developed using Si_3N_4 and carbon powders (30:70 in volume ratio) and sintering by spark plasma sintering at temperatures of 1900 ℃. The SiC/CBC were highly dense (98% theoretical density), lightweight (2.42 Mg/m^3), and had both relatively high bending strength and thermal conductivity (150 MPa and 138 W/mK, respectively). Moreover, the SiC/CBC could be joined tightly with W. The joints of SiC/CBC and W may be used as heat-sinks for W-based plasma facing components. The joining strength between W and SiC/CBC was 33 MPa when measured by a tensile test and 90 MPa by a bending test.

KEY WORDS: (Silicon carbide), (Ceramic bonded carbon), (Joining), (Tungsten)

1. Introduction

Tungsten (W) is extensively used for X-ray targets, discharge lamp electrodes, and high temperature armor because of its high melting point, high strength, and other unique physical/chemical properties. It is also a prime candidate for plasma facing components of fusion reactors because of its low sputtering yield, tritium retention, and good thermal properties. [1-2] But W is heavy, and difficult to machine or weld. W coatings on light carbon materials have been tried in order to realize good heat load resistance and steady state operation at high temperatures in the present plasma confinement devices. [3]

Carbon/graphite materials have advantages of lightweight (1.8-2.2 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 heat-sink in W-based plasma facing components

Recently, we proposed a novel carbon-based composite, called ceramic bonded carbon (CBC).[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 were also proved to be easily joined with ceramics and metals due to the ceramic network structure.[9-10] Therefore, compared with conventional W-coated carbon components, W-clad CBC may have superior reliability because of its high strength and thermal conductivity, as well as the controllable thickness of W.

In this work, Si_3N_4 and carbon powders were fabricated into SiC/CBCs by combining gelcasting and spark plasma sintering. The joining of SiC/CBC to W was also examined. The properties of SiC/CBC and joining strength of SiC/CBC-W joint were characterized by three-point bending and tensile tests.

2. Experimental

2.1 Fabrication of SiC/CBC

The starting materials included carbon powder made from the meso phase pitch carbon by a graphitization step at 2500 ℃ (Toyo Tanso Co. Ltd.) and Si_3N_4 powder (UBE-10, UBE. Co. Ltd.), which contained 6 wt% Al_2O_3 and 3 wt% Y_2O_3 as sintering additives.

To prepare the SiC/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-proponol). The Si3N4 and carbon powders (30:70 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 62 vol% slurry. The mixed slurry was cast into a plastic mold and then dried at 80℃ to form a solid body via the monomer-polymer transition. After demolding, the dried green compacts were heated to 700℃ under vacuum to remove the gel binder. The green bodies were then loaded into graphite dies for spark plasma sintering

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(SPS) under a pressure of 30 MPa at 1900℃ in vacuum for 5 minutes.

2.2 Joining SiC/CBC with W

To study the joining strength and mechanism of the W-clad SiC/CBC, two clad samples were prepared. One is the normal type of W clad SiC/CBC as shown in **Fig. 1a**. The other one is a sandwiched type of SiC/CBC-W-SiC/CBC, as shown in **Fig.1b**. In both samples, the thickness of the W layer was controlled to around 0.5 mm.

During sample preparation, SiC/CBC disks were finished using a #80 SiC paper and then placed into a 25-mm diameter graphite die. To form a 0.5-mm thick W layer, 5 g of W powder was stacked onto the SiC/CBC disk, and then sintered in vacuum at 1700℃ at a heating rate of 100℃/min and a uniaxial pressure of 30 MPa for 5 minutes by SPS.

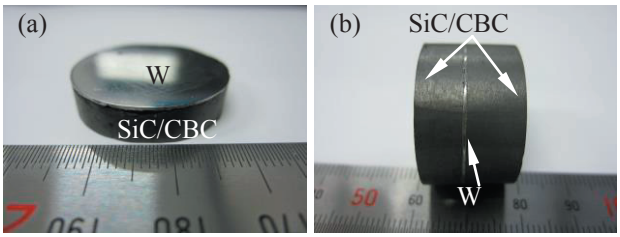

Fig.1 (a) A W cladding SiC/CBC sample; (b) a SiC/CBC-W-SiC/CBC joint.

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-3530 M, JEOL) was used to identify crystalline phases of the cladded samples. To characterize the joining properties between W and SiC/CBC, rectangular bars (3×2~4×20 mm^3) for the three-point bending test, and dog-bone shaped bars, (8 ×1.5×15 mm^3) for the tensile test were prepared and then measured using a Table-Top Universal Tester (EZ-Test Type S, Shimadzu) at room temperature. The speed of the crosshead displacement was 0.5 mm/min. At least three samples were used for each measurement. The thermal conductivity of SiC/CBC pellets was measured by the laser-flash method (TC-7000, ULVAC-RIKO).

3. Results and Discussion

3.1Fabrication and properties of SiC ceramic bonded carbon

Figure 2 shows the morphologies of carbon and Si_3N_4 powders. Both of them have a spherical morphology of approximately 15 μm and 0.5 μm in size, respectively. By using a gelcasting method, a Si_3N_4/CBC green body with a unique microstructure containing separated carbon particles and a continuous Si_3N_4 boundary layer can be obtained. During the sintering, the Si_3N_4 will be converted the more stable phase of SiC with excess carbon at high temperatures by the following reaction (1) from 1400℃[11]. And at the same time, the densification will be finished till 1900℃.

$$Si_3N_4\ (s) + 3C\ (s) \rightarrow 3SiC\ (s) + 2N_2\ (g) \qquad (1)$$

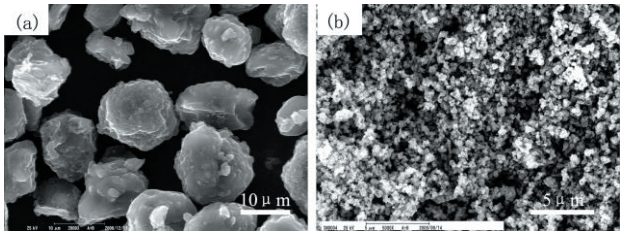

Fig.2 Morphology of starting powders for SiC/CBC: (a) carbon; (b) Si_3N_4.

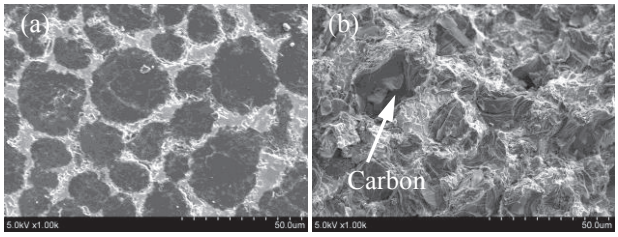

Fig.3 (a) Polished surfaces and (b) fractured surface of SiC/CBC. The fractured carbon particle is indicated by an arrow.

Figure 3a shows the microstructure of sintered SiC/CBC that has the unique microstructure containing carbon particles of 15 μm in average size and an SiC boundary layer of 0.5-3 μm in thickness. The fractured surface of SiC/CBC after strength measurements was observed under SEM, as seen in **Fig. 3b**. Almost all carbon particles are broken to some extent, showing a typical trans-granular mode. This indicates a strong bond at boundaries. The SiC/CBC shows a density of 2.42 g/cm^3 (98% of theoretical density), bending strength of 150 MPa, and thermal conductivity of 138 W/mK. The higher strength and thermal conductivity is attributed to its higher density and reinforcement of the SiC boundary network.

3.2 Joining SiC/CBC with W

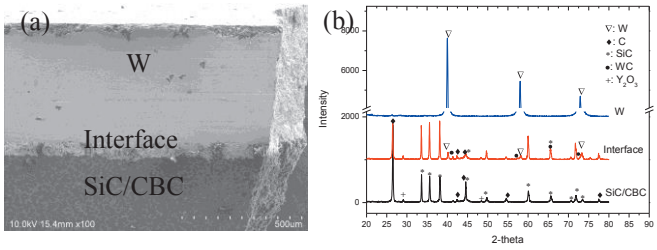

Fig.4 (a) Cross sectional view of the W clad with SiC/CBC; (b) XRD patterns of W, interface, and SiC/CBC layers.

A side-view of a W-clad SiC/CBC sample is shown in **Fig. 4a**. The W layer of around 0.5 mm in thickness is

clad tightly with SiC/CBC. No voids or cracks are observed at the interface. However, between W and SiC/CBC, an interlayer with a rough surface can be noticed. **Figure 4b** shows XRD patterns of the W layer, SiC/CBC, and the interface area. In the interface area, small WCx peaks are observed, but no W-Si compound is found.

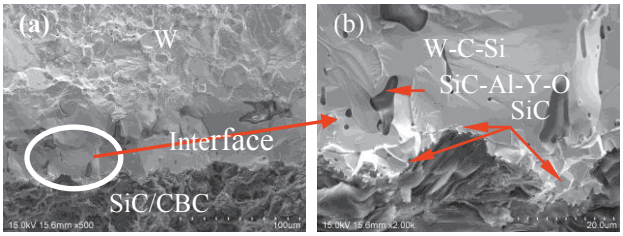

Fig.5 (a) Fractured surfaces of interface area of W clad SiC/CBC; (b) an enlarged image of interface layer consisting of mixed WCx and ceramic particles.

Figure 5a shows the fractured surface of the W clad SiC/CBC after the bending test. A reacted interlayer of around 50 μm in thickness is seen at the interface between W and SiC/CBC. The W layer contains faceted cube-shape grains with a size around 10 μm, while the interlayer shows a microstructure of dark particles embedded in a grey matrix. This interlayer was enlarged and analyzed further by EDS, as shown in **Fig. 5b**. The dark particles have an irregular shape, but a smooth surface, suggesting that their surfaces have been molten to some extent. They include Si, C, Al, O, Y, and a small amount of W. The grey matrix shows plate-like grain morphology. It contains W, C, and a small amount of Si. Considering the composition of SiC/CBC and the XRD analysis as shown in Fig. 4, the dark particles seem to consist of SiC and Al-Y-O glass. A small amount of W may exist in the glass. The grey matrix is assumed to be WCx containing a small amount of Si. In addition, at the edge of the interlayer close to the SiC/CBC, small faceted grains were observed. The EDS indicated that they were SiC grains. This SiC layer would prevent further reaction of W and SiC/CBC. The mixed interlayer of WCx and SiC-Al-Y-O may contribute mainly to clad W-SiC/CBC by the W-WCx bonding to the top W layer, and by the SiC-SiC bonding to the SiC/CBC.

3.3 Joining strength

Two methods and corresponding test samples were used to characterize joining strength between W and SiC/CBC, as shown in **Fig. 6**. The bending mode is a convenient method to know the joining strength at a bending condition, while the tensile mode can offer a joining strength at a tensile condition. To carry out the tensile test, dog-bone shape samples were prepared by electrical discharge machining (EDM), as shown in Fig. 6b.

The measured bending strength of the W-SiC/CBC clad is 90 MPa, lower than that of the SiC/CBC (150 MPa). The failure occurs in the SiC/CBC side, but very close to the interface. As shown in **Fig. 7a**,.the side view of a fractured sample indicates that a SiC/CBC layer around 60 μm in thickness is still bonded tightly to the W layer, and no cracks and delaminations are observed at the interface.

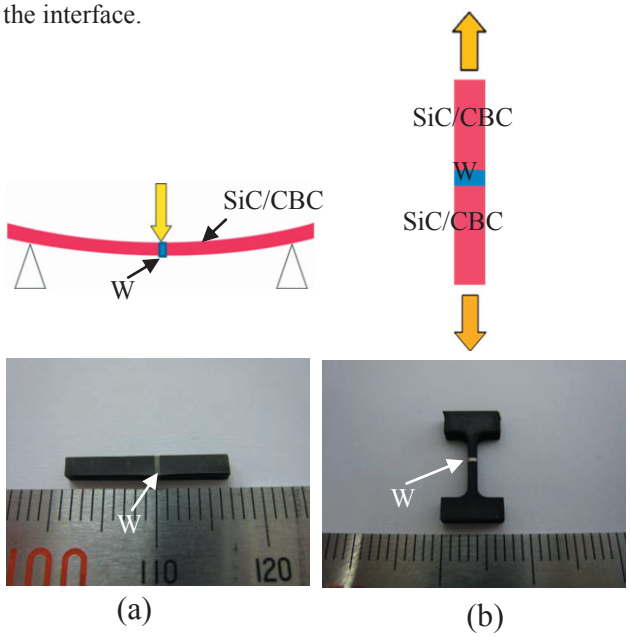

Fig.6 Methods and corresponding samples used to characterize the joining strength between W and SiC/CBC: (a) bending mode; (b) tensile mode.

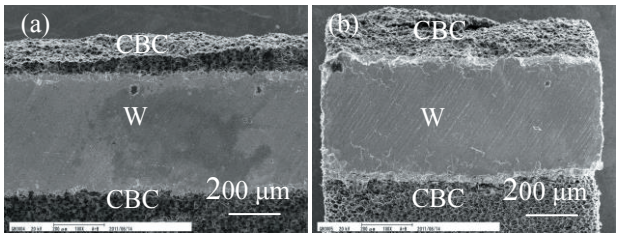

Fig.7 Side-view microstructures of fractured samples (a) after bending test; (b) after tensile test.

The measured tensile strength of the joint is 33 MPa, which is also lower than that of SiC/CBC (around 46 MPa). As shown in a side-view of fractured sample (**Fig.7b**), similar to the sample tested by bending mode-2, a SiC/CBC layer around 60 μm in thickness remains on the W layer.

These fractographies observed for W-clad SiC/CBC samples suggest that the adhesion between W and SiC/CBC is even stronger than the SiC/CBC. However, the measured bending strength and tensile strength were lower than that of the SiC/CBC. This decrease in strength might be attributed to the damage caused during the sample machining. Because the W and SiC/CBC have very different hardness and strengths, damage may easily be produced at their interface by grinding. The residual stress may also reduce the adhesion strength. Although W and SiC/CBC have the similar coefficient of thermal expansion (CTE) values, the interface reaction occurs and

new interfaces are formed during SPS. Moreover, W and SiC/CBC have very different elastic moduli, which may contribute to the additional stress as new interfaces form.

4. Conclusions

(1) SiC/CBC containing the SiC ceramic network bonding of carbon particles could be densified by SPS at 1900℃, and exhibited a lightweight of 2.42g/cm^3, high bending strength of 150 MPa, and high thermal conductivity of 138 W/mK.

(2) W can be directly clad with the SiC/CBC at 1700℃ by spark plasma sintering. A new reacted interface consisting of the mixed WCx and SiC-based ceramics formed between W and SiC/CBC. The measured adhesion strengths are 90 MPa and 33 MPa by the bending and tensile test, respectively.

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