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Joining of silicon carbide and graphite by spark plasma sintering

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Abstract

After placing SiC powder on an isotropic graphite substrate, the two materials were successfully joined by spark plasma sintering (SPS). The effect of an Al_2O_3–Y_2O_3 sintering aid on SiC during joining was studied. The tensile strength of the joints prepared at 1800 °C under 30 MPa for 5 min reached 18 MPa. The fracture occurred not at the interface, but at the graphite substrate. The joining mechanism of SiC/graphite is attributed to the following: after the SiC powder squeezes into the open pores of graphite during sintering, the sintered SiC produces a strong bond through the interface between SiC and graphite.

Keywords: A. Joining; Graphite; Silicon carbide; Spark plasma sintering

1. Introduction

Isotropic graphite materials are typically produced using the following process: after mixing filler coke with binder pitch, the mixture is cold isostatically pressed, baked at about 1000 °C, and then graphitized at about 3000 °C by the Acheson method [1,2]. The combination of cold isostatic pressing and graphitization results in high quality graphite with isotropic properties that is easy to machine. Large graphite blocks ranging in size from several tens of centimeters to several meters can be produced. The blocks are machined into a variety of products used in various industries, including crucibles, molds, heaters, electrodes, heating trays, and others. The material's high refractoriness, excellent thermal shock and chemical resistance, appropriate electric and thermal conductivities, self-lubrication, and light weight make it suitable for these applications. However, one limitation is its low strength, which is a result of its high porosity (9–25%) [2,3]. This high porosity is caused by the low carbon yield of the binder pitch after baking.

In order to further expand the applications of graphite, it is joined to or coated with strong and hard ceramics or tough metals. Forming metal/graphite joints by using brazing metals [4–7], or by solid state bonding has been studied [8–14]. However, only a few investigations on joining of ceramic/graphite, using brazing metal [15] and adhesives [16] have been reported in the literature. Moreover, most ceramic/graphite joints are prepared below 1400 °C, which reduces the working temperature of the joints. Many brazing metals are limited to low-temperature applications because of their low melting points and mismatched coefficients of thermal expansion (CTE) with graphite. In addition, few studies on joining of ceramic and graphite for higher temperature applications exist in the open literature. By directly joining ceramics to graphite without requiring brazing metals or adhesives, high temperature applications are possible.

On the other hand, SiC-coated graphite susceptors prepared by CVD are widely used for the epitaxial growth of Si, GaN and other compound semiconductors in the electronic, lighting, and laser industries. The SiC coating protects the semiconductor crystals from being contaminated by the carbon particles and gas emissions from the susceptors and prevent corrosion of graphite with process gasses at high temperature. In this case, the coating is only several tens of microns in thickness. By forming a strong joint of thicker and densier SiC with graphite would further improve the graphite's resistance to hot corrosion and wear, as well as its toughness.

Spark plasma sintering (SPS) is an efficient process to sinter ceramics and metals [17–21]. It is also used for joining of

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dissimilar ceramics or metals, and ceramic-to-metal combinations. For example, Kondo et al. reported that stacked powders of TiN and apatite were sintered and simultaneously joined by SPS [22]. ZrB_2/SiC joints [23] have also been fabricated.

In this study, the direct joining of SiC to graphite was carried out using SiC powders containing a sintering aid of Al_2O_3–Y_2O_3 by SPS. The effect of sintering aid on joining was investigated by analyzing the interface structure and its composition. The bonding strength was evaluated by tensile testing, and the joining mechanism was proposed.

2. Experimental procedure

Isotropic graphite (IG-12, Toyotanso Co., Ltd.) was selected because it has a similar coefficient of thermal expansion (CTE) (4.7×10^{-6}/K) to that of SiC (4.3×10^{-6}/K). The tensile strength of the graphite substrate is 28 MPa [2]. The bulk density and porosity are 1.78 Mg/m^3 and 21%, respectively. α-SiC powder (SER-A06, Shinano Electric Refining Co., Ltd.), with an average particle size of 0.6 μm, was mixed with Y_2O_3 (RU-P, Shin-Etsu Chemical Co., Ltd.) and Al_2O_3 (TM-DAR, Taimei Chemicals Co., Ltd.) as the sintering aid at 3 and 6 mass%, respectively. SiC powder (1.8 g) with or without sintering aid was placed on the top and bottom of a graphite substrate (∅25 × 4 mm) in a graphite mold (inner diameter: ∅25 mm).The graphite sheets (PF-50, Toyotanso Co., Ltd.) were placed between the joint and a graphite punch. The sintering was carried out at temperatures of 1800–2000 °C for 5 min under a pressure of 30 MPa in vacuum by SPS (SPS-1050, Sumitomo Coal Mining Co., Ltd.). Graphite spacers and punches were placed between the electrode and the mold. The joining temperature was determined by focusing a pyrometer into a 1-mm diameter hole made through the side of the mold.

The sintered graphite/SiC joints were ground and polished to a size of ∅25 × 6 mm, and then cut into pieces of 4 mm (W) × 4 mm(L) × 6 mm(H) for microstructure observation and strength measurement. Microstructural observation and elemental analysis of the graphite/SiC joints were carried out by scanning electron microscopy – energy dispersive X-ray spectroscopy (SEM-EDS, ERA-8800FE, ELIONIX Co., LTD). Crystalline phases were examined by XRD (Ultima IV, Rigaku Corporation). The strength of the joints was measured by the tensile test (EZ-L, SHIMADZU CORPORATION). Fig. 1 shows a schematic drawing of jigs used for measuring the strength. The top and bottom of the joints were adhered to stainless steel jigs using an epoxy resin adhesive (E-60HP, Henkel AG & Co. KGaA) at 80 °C for about 24 h. The cross head speed of the tensile test was 0.5 mm/min. Seven test bars with the size of 4 mm(W) × 4 mm(L) × 6 mm (H) were prepared and subjected to tensile testing.

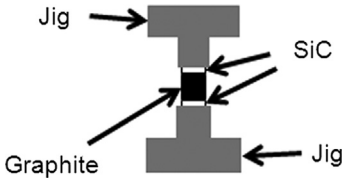

Fig. 1. Schematic of the jigs used for measuring the joining strength.

3. Results and discussion

Fig. 2 shows the interface structures of SiC/graphite joints. In the joints with sintering aid prepared at 1900 °C (Fig. 2a) and 2000 °C (Fig. 2b), graphite and SiC were tightly joined. No gaps and delaminations were observed. In contrast, in the joints without sintering aid prepared at 1900 °C (Fig. 2c) and 2000 °C (Fig. 2d), some gaps were observed at the interface. In addition, SiC ceramic parts without sintering aid had a

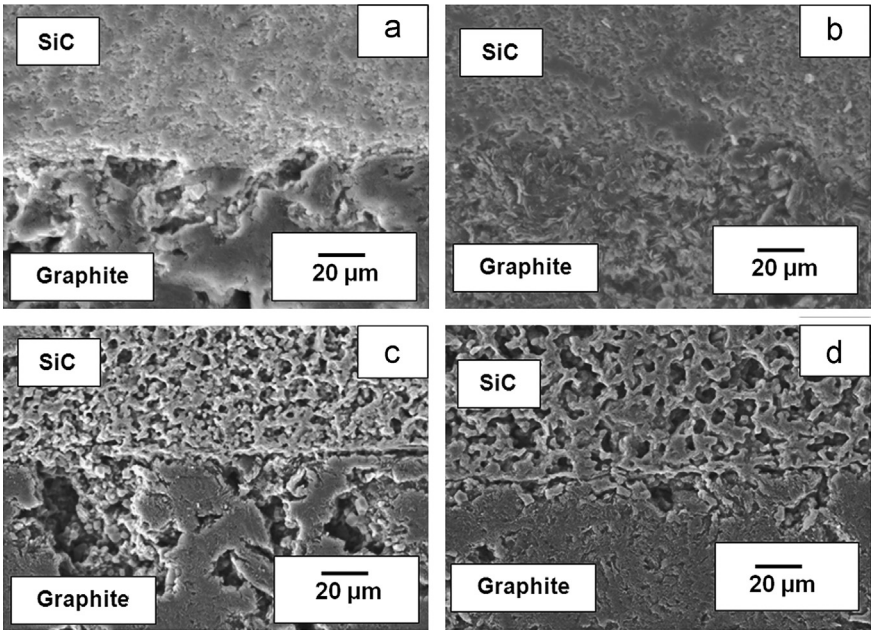

Fig. 2. Cross-sectional SEM images of SiC/graphite joints with sintering aid prepared at 1900 °C (a) and 2000 °C (b): without sintering aid prepared at 1900 °C (c) and (d) 2000 °C.