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Surface modification on wetting and vacuum brazing behavior of graphite using AgCu filler metal

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ABSTRACT

Joining of graphite materials is problematic primarily due to the poor wettability of non-active filler metal on these materials. In an attempt to overcome this problem, magnetron sputtering deposition of Cr film on graphite was performed to modify the surface of the graphite. The wetting and brazing of graphite are carried out using non-active AgCu eutectic filler metals after deposition. The results indicated that surface modification enables wetting and joining of graphite with non-active filler. Cr film reacted with graphite forming Cr-C interfacial reaction layer, which resulted in the decrease of contact angle. Reliable graphite/graphite joints were obtained at temperature from 1113 K to 1233 K for 10 min. The typical interfacial microstructure of the brazed joint is graphite/Cr-C layer/Ag(s,s) + Cu(s,s) eutectic phase/Cr-C layer/graphite. The optimal shear strength of the joint was 13.6 MPa when the brazing parameters were 1173 K for 10 min.

1. Introduction

The application of graphite materials in nuclear energy, such as a moderator and neutron reflector in more than 100 nuclear power plants and plutonium production reactors, has been expanding by the virtue of their excellent properties for good electricity and thermal conductivity, high melting point, superior thermal impact resistance and chemical stability [1–3]. Therefore, the joining technology of graphite is increasingly valued by researchers for the purpose of extending their applications.

Brazing, as its simplicity, convenience and economy, has attracted special interest to join the graphite [4–6]. Some researchers demonstrated that most non-active filler metals were unsuitable for joining graphite due to their poor wettability on the carbon materials [4,7,8]. In order to solve this problem, both the direct brazing and indirect brazing have been widely used to promote the wettability. Direct brazing requires adding active element, such as Ti or Cr, into filler metals. The active element reacted with carbon materials or composites to form a metal-like layer to improve the wettability of molten filler on the substrates, as many article reviewed [6,9–11]. However, Some researchers [4,12–14] pointed out that the addition of active elements increased the melting point, reduced the fluidity and embrittled the alloy with detrimental effect on joint integrity due to the excessive amount of intermetallic compounds. Furthermore, the uniformity of filler cannot be guaranteed when raw powder was added after the milling process. It would affect the stability of the joint performance. Comparatively, surface modification treatment used in indirect brazing can usually reduce the residual stress of the joint [15–17] and maintain stability of brazing performance. A. Koltsov et al. [18] study the effect of thin carbon layers on the oxide for idea of improving wetting in a carbide-forming metal/oxide system by carbon coating. Song et al. [19] indicated that the Mo_2C coatings greatly improve the wettability of graphite by copper and that copper could be infiltrated into Mo_2C-coated graphite matrix without external pressure. Casalegno et al. [20] studied the effect of surface modification using W, Mo and Cr on the C/C composites surface to improve its wettability by copper. They pointed out Cu wetting on C/C composites was improved and direct joining of C/C composites to Cu was feasible without any filler metal, and the best results in terms of wettability of molten copper on C/C have been achieved by modifying C/C composites with Cr. Thus, the use of surface modification with electroless plating or metallization on the faying surfaces could dramatically improve wettability on substrate by liquid non-active filler metals and make the brazing process more feasible.

In this work, the “activation” of graphite surface was obtained by using magnetron sputtering deposition of element Cr, in order to join graphite avoiding the use of active metal brazing alloy. Magnetron sputtering deposition method can control the thickness of the film. Then the wettability and brazing behavior of deposited graphite were investigated using AgCu non-active eutectic filler metal under vacuum. The corresponding microstructure characteristic and evolution of Cr-

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Table 1
Technical information about sputtering parameters used for Cr film deposition.

Steps	Time/s	Bias voltage/V	Current/A	Vacuum degree/Torr
1	900	500	0.5	3.5×10^{-5}
2	900	60	6	

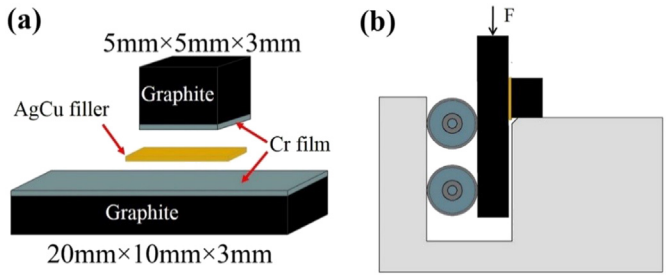

Fig. 1. Schematic diagram of (a) assembling brazing parts and (b) shear test experiment.

deposited graphite/AgCu/Cr-deposited graphite joint was analyzed in detail.

2. Experimental procedures

The graphite with porosity of about 15 vol% was supplied by Dong Xin Carbon Co., Ltd. Sichuan, China, and the average shear strength is 30 MPa. The surfaces were first ground flat by SiC paper of grade P180, P600 and P1200 and polished by 2.5 μm diamond paste followed by ultrasonically cleaned with acetone in order to remove the impurities of the surface. Prior to wetting and brazing experiment, the samples were bombarded with Ar ions for 15 min and chromium film with thickness of 0.5 μm was deposited by magnetron sputtering using closed filed unbalanced magnetron sputter ion plating equipment. The sputtering parameters were shown in Table 1.

The commercial AgCu eutectic foil with 100 mg was mechanical pressured into 3 mm × 3 mm cube for wetting experiments. The melting point of the filler is 1053 K. The wetting experiments were performed through the sessile drop method under a vacuum better than 3×10^{-3} Pa. The system was heated to 1013 K at the rate of 36 K/min and held for 10 min to obtain a homogeneous heated sample. Then the temperature was raised to 1323 K at the rate of 5 K/min consecutively, furnace-cooled down to room temperature finally. The wetting process was photographed by a digital camera at regular temperature interval of 1 K. After the wetting experiment, the droplet profiles were analyzed by drop-analysis software to calculate the contact angle.

In the preparation of brazing experiment, the sandwich deposited graphite/AgCu/deposited graphite system was assembled shown in Fig. 1(a). The assemblies were heated to 1013 K at the rate of 20 K/min and remained for 10 min, then raised to brazing temperature at a rate of 10 K/min and held for 10 min, cooled down to 573 K at a rate of 5 K/min finally. During brazing, the vacuum degree of furnace was less than 3.0×10^{-3} Pa and a pressure of 60 KPa was applied on each assembly to ensure proper contact.

The microstructure of wetting and brazing samples were characterized by scanning electron microscope (SEM, MERLIN Compact, ZEISS) equipped with energy dispersive spectrum analysis (EDS, OCTANE PLUS, EDAX), and the phases were evaluated by X-ray diffraction analysis (XRD, D8-ADVANCE) with the sample etched off by chloroazotic acid. The room temperature shear strength was tested for five times by a universal testing machine (Instron 5967) at a constant speed of 0.5 mm/min and the average value was taken. The schematic diagram of shear test was shown in Fig. 1(b).

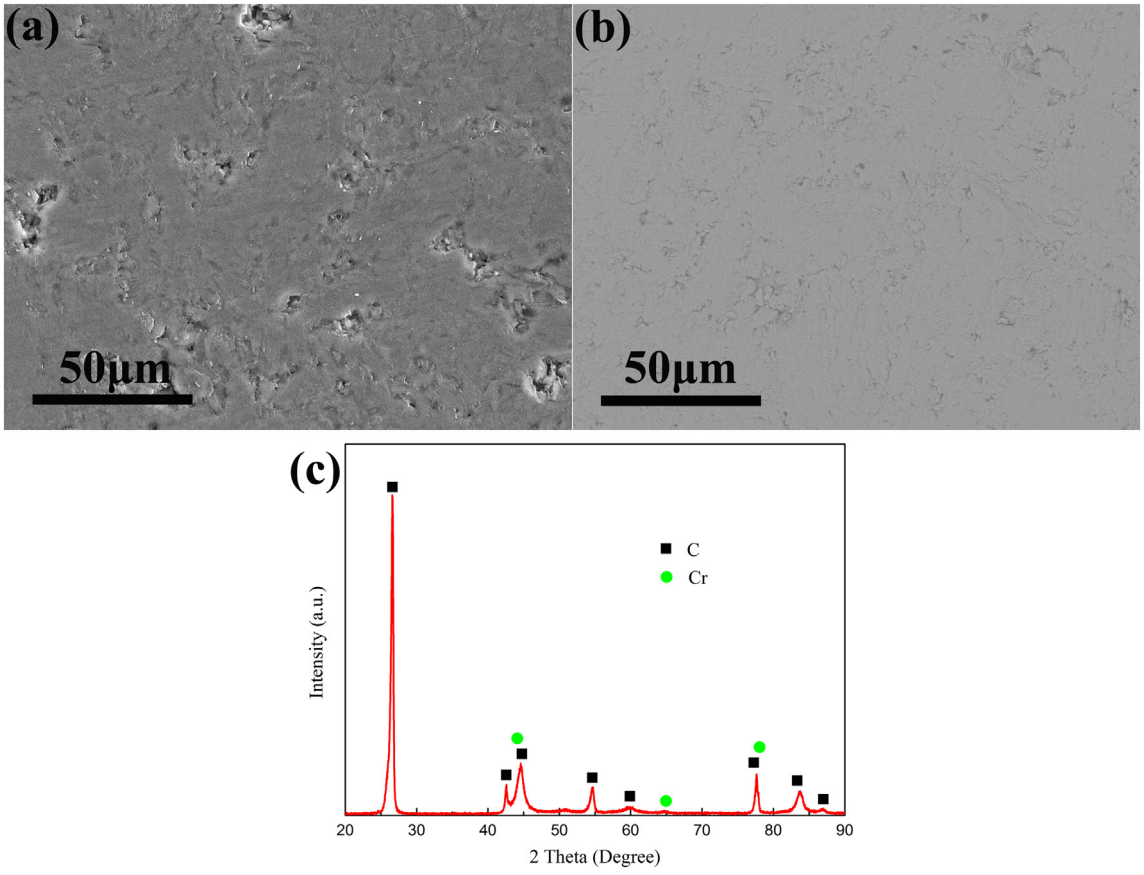

Fig. 2. (a) BSE image of base graphite, (b) and (c) BSE image and XRD pattern of as-deposited Cr film, respectively.