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Wetting mechanism of Sn to $Zr_{50.7}Cu_{28}Ni_9Al_{12.3}$ bulk metallic glass assisted by ultrasonic treatment

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ABSTRACT

In this work, pure Sn was used to wet $Zr_{50.7}Cu_{28}Ni_9Al_{12.3}$ bulk metallic glasses (BMGs) assisted by ultrasonic treatment. Without ultrasonic treatment, pure Sn showed a non-wetting condition to BMG. Ultrasonic vibration facilitated the wetting of Sn to BMG. Prior to ultrasonication for 30 s, only physical adsorption formed at the Sn/BMG interface. Increasing ultrasonic time led to the alteration of the bond at the Sn/BMG interface from point contact to local surface contact, and to diffusion layer. Two bonding modes of order–order and order–disorder were discovered at the Sn/BMG interface. Cu content was higher than the other elements near the bonding interface. Longer diffusion distances of Sn into the BMG were obtained at high ultrasonic power, high temperature and large dip depth.

1. Introduction

Bulk metallic glasses (BMGs) have been applied as structural components [1], such as amorphous motors and electrical transformers because of their good corrosion resistances, high strengths and hardness, and good soft magnetic property [2,3]. However, one of the reasons limiting the extensive application of BMGs was their product sizes [4]. In addition, BMGs always need to be joined with other crystalline alloys in engineering applications [2]. Therefore, techniques to join BMG/BMG or BMG/crystalline alloys have attracted extensive attention [5–12].

BMGs can become brittle due to crystallization when heated past the crystallization temperature. Crystallization results in the reduction in mechanical properties [5]. Therefore, the joining temperatures should be maintained below the glass transition temperature of BMGs. Several studies have attempted to use various techniques to join BMG/BMG or BMG/crystalline alloys. Kawamura et al. [6] used a spark welding method to join $Zr_{55}Al_{10}Ni_5Cu_{30}$ BMGs and successfully avoided crystallization in the joint. The results showed that the tensile strength of the obtained joints was equal to that of the parent BMG. Kim et al. [7] used electron beam welding to join $Zr_{41}Be_{23}Ti_{14}Cu_{12}Ni_{10}$ BMG plate to Ni metal and reported that crystallization was observed in the joint when the electron beam was too close to the crystalline metal. Moreover, Ohno et al. [8] successfully joined $Pd_{40}Ni_{40}P_{20}/Pd_{40}Cu_{30}P_{20}Ni_{10}$, and $Zr_{55}Cu_{30}Al_{10}Ni_5$ and $Zr_{41}Be_{23}Ti_{14}Cu_{12}Ni_{10}$ and found no crystallization at the interface. Their work also showed that BMGs could be successfully joined at temperatures approximately below 50 K from the glass transition temperature.

Ultrasonic-assisted soldering is a fluxless method that can be operated in air [13,14]. Ultrasonic vibration could improve the original wetting condition at the solder/substrate interface [15,16]. Thus, ultrasonic-assisted soldering could be used to join some hard-to-wet materials. Moreover, ultrasonic-assisted soldering could realize the bonding through low-temperature eutectic phase of solder/substrate [16–19]. Therefore, ultrasonic-assisted soldering serves as a promising method to join the BMGs below their crystallization temperatures. Few papers on the joining of BMGs assisted by ultrasonic treatment have been reported. Using Sn-based filler metals, Ji et al. [20] joined Fe-based BMGs with Al sheet and found $FeZn_{13}$ at the filler metal/BMG interface. The results showed that the initial interfacial wetting and microstructure refinement were improved by ultrasonic vibrations. Tamura et al. [21] wetted a Zr-based BMG using a Sn-Cu-Ni solder assisted by ultrasonic vibrations and they reported that the wetting mainly depended on the collapse of cavitation bubbles.

This work investigated the wetting behavior of Sn to BMG. This work is referential to the joining of BMGs in the furthre. Low-melting-point Sn was chosen to avoid the crystallization of the BMG. The bonding mechanisms at the Sn/BMG were examined. The diffusion distance of Sn into the BMG was also investigated.

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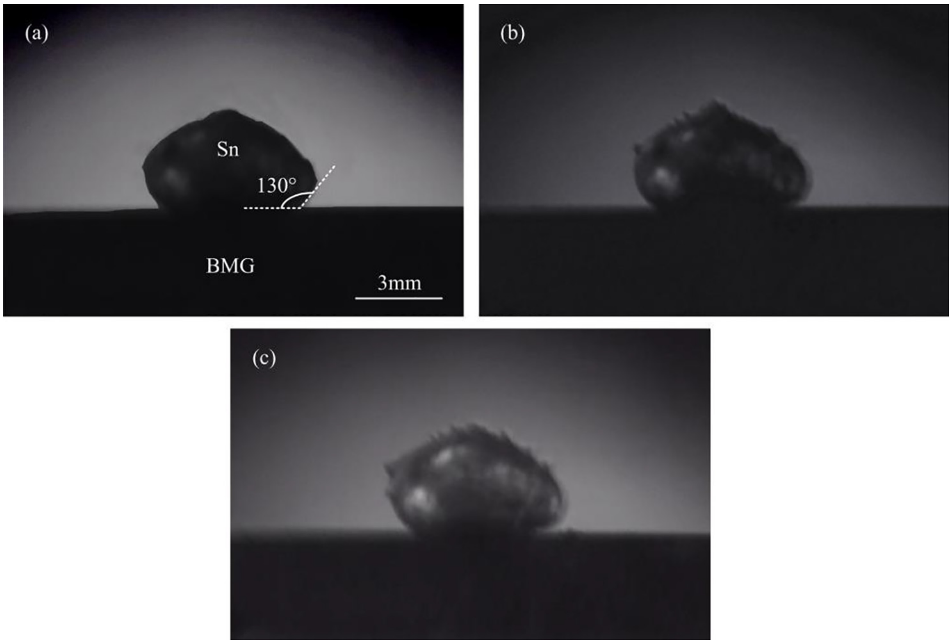

Fig. 1. Wetting condition of liquid Sn to $Zr_{50.7}Cu_{28}Ni_9Al_{12.3}$ at (a) 300 °C, (b) 350 °C and (c) 400 °C.

2. Experimental

2.1. Substrates and filler metals

The substrates were $Zr_{50.7}Cu_{28}Ni_9Al_{12.3}$ BMGs with a thickness of 3 mm. The BMGs were provided by Huang et al. [22] of Harbin Institute of Technology. The detailed fabrication of the BMGs was reported in their work. The $Zr_{50.7}Cu_{28}Ni_9Al_{12.3}$ BMG has superior mechanical properties such as high hardness, excellent wear resistance, high strength and elastic limit. The glass-transition and crystallization temperatures of the substrate were tested using a differential scanning calorimeter (DSC, Netzsch-STA449F3) under Ar atmosphere and heating speed of 10 °C/min. The phases of the substrate were tested using a D/max-RB X-ray diffraction (XRD) using CuK_α radiation. The tube voltage and current were 40 kV and 40 mA, respectively. Pure Sn was chosen as the filler metal. Pure Sn has a melting point of approximately 231 °C. The contact angle of Sn to $Zr_{50.7}Cu_{28}Ni_9Al_{12.3}$ was tested. A wetting angle tester VAF-300 was used under the testing temperature of 300–400 °C and pressure of 5×10^{-3} Pa. The contact angle of liquid Sn to $Zr_{50.7}Cu_{28}Ni_9Al_{12.3}$ was 130° at 300 °C, which showed a typical non-wetting condition (Fig. 1a). The contact angles at 350–400 °C showed similar non-wetting conditions (Fig. 1b and c).

2.2. Wetting experiment

The substrates had dimensions of 10 mm × 5 mm × 3 mm during the wetting experiment. Prior to experiment, the surfaces of the substrates were polished using 200 # emery papers and ultrasonically cleaned in acetone for 2 min. The schematic of the wetting experiment is illustrated in Fig. 2. The filler metal was put into a self-designed mold (Fig. 3) and heated to predetermined temperature. The substrate was immersed into the filler metal, and ultrasonic vibration was turned on. The ultrasonic vibration system was operated at a frequency of 20 kHz and pressure of 0.2 MPa. The system had maximum rated power of 1000 W. Three modes of power output, namely, 1/3 Pm (Mode I), 2/3 Pm (Mode II) and Pm (Mode III) were used. Ultrasonic vibration time varied from 5 s to 3600 s. The temperature ranged from 300 °C to 400 °C.

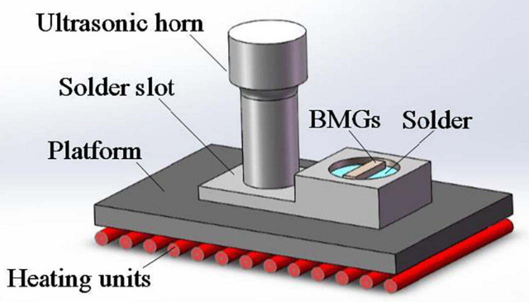

Fig. 2. Schematic of the wetting experiment.

2.3. Microstructure and mechanical property test

The microstructure of the metallographic samples was observed using a scanning electron microscope (SEM, FEI-Quanta 200) equipped with an energy dispersive X-ray spectroscopy (EDS) system after standard polish procedure. The microstructure of the bonding interface was also investigated by a high-resolution transmission electron microscope (HRTEM) (FEI, TecnaiG2F30) equipped with a Super-X EDS system. The TEM samples were prepared by a focused ion beam (FEI, Helios NanoLab 600i). G200 nano-indentation instrument was used to measure the hardness at the bonding interface. The testing distance was 20 μm, testing force was 0.01 N, and dwell time was 10 s.

3. Results

3.1. Microstructure of the substrate

The microstructure of the $Zr_{50.7}Cu_{28}Ni_9Al_{12.3}$ BMG is shown in Fig. 3. The bright-field TEM image of $Zr_{50.7}Cu_{28}Ni_9Al_{12.3}$ BMG in Fig. 3a presented a homogeneous composition. The magnified image of the $Zr_{50.7}Cu_{28}Ni_9Al_{12.3}$ BMG is shown in Fig. 3b. The microstructure showed a typical glassy structure. No crystalline phases could be found. The diffraction pattern of the $Zr_{50.7}Cu_{28}Ni_9Al_{12.3}$ BMG showed a dispersive circle, indicating a typical amorphous state. The XRD pattern of $Zr_{50.7}Cu_{28}Ni_9Al_{12.3}$ is shown in Fig. 3c. The diffraction pattern had a broad, disperse peak without any sharp diffraction peak. The result