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Wetting behavior and interfacial characteristics of In–Sn alloy on CuZr-based bulk metallic glass

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Abstract

The wettabilities of In–Sn alloy on $Cu_{40}Zr_{44}Al_8Ag_8$ BMG substrate were investigated using the sessile-drop method at different temperature. The result shows that the equilibrium contact angle decreased with increasing temperature. The interfacial reaction of the active Sn atoms in molten In–Sn alloy and the active Zr atoms in $Cu_{40}Zr_{44}Al_8Ag_8$ BMG caused crystallization reaction. Ion beam sputtering profiling in combination with AES technique was employed to investigate the Sn diffusion in $Cu_{40}Zr_{44}Al_8Ag_8$ BMG. Between 473 K and 673 K, the diffusion coefficients vary from 0.7×10^{-16} m²/s to 12.9×10^{-16} m²/s. It is concluded that the interfacial reaction is favorable to the crystallization and then the crystallization also promotes Sn diffusion.

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1. Introduction

The wetting behavior of liquid metals on solids is of fundamental scientific interest in understanding the development of liquid metal–solid bonding during brazing, soldering and composites [1–3]. The wetting of liquid metals on crystalline substrate has been studied extensively since 1900s [4,5]. However, few works were carried out on the wetting of liquid alloy on bulk metallic glasses (BMGs) substrate.

BMGs possess a number of desirable properties that are potentially of technological value for semiconducting devices and metallic components, both in thin film and bulk forms [6,7]. These desirable physical and chemical properties of BMGs materials derive directly from their unique atomic structure. However, the amorphous structure is thermodynamically metalstable, which is very sensitive to heat treatment [8]. Moreover, it is considerably difficult to investigate diffusion in BMGs since diffusion is very small and the annealing time and temperature are limited by the need to avoid crystallization. Cahn et al.

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[9] succeeded for the first time in measuring the self-diffusion of boron in $Fe_{40}Ni_{40}B_{20}$ by using secondary ion mass spectrometry. Different diffusion mechanisms in an amorphous alloy were proposed. For example, Ahmadzadeh and cantor [10] proposed the possibility of an interstitial diffusion mechanism in metallic glass. However, this model was generalized to a binary amorphous alloy like Ni–Zr.

Here we present the results of wetting behavior of molten In–Sn alloy on $Cu_{40}Zr_{44}Al_8Ag_8$ BMG substrate by the sessiledrop method. The effect of temperature on the interfacial reactions and contact angle is examined. Systematic investigations of Sn atom diffusion in $Cu_{40}Zr_{44}Al_8Ag_8$ BMG are reported in order to understand diffusion mechanisms.

2. Experimental procedure

The eutectic alloy composition (49.1Sn–50.9In wt%) was prepared by induction melting in high-purity argon atmosphere (99.99%). Then the alloy was cut into ~50 mg pieces. Cu₄₀Zr₄₄Al₈Ag₈ alloy was produced by arc melting under a Ti-gettered Ar atmosphere. The purity of all elements is above 99.5%. Cu₄₀Zr₄₄Al₈Ag₈ alloys were remelted in a quartz tube by induction melting, followed by casting into copper moulds with plate cavity of 2 mm in thickness. Cu₄₀Zr₄₄Al₈Ag₈ BMG plates was cut into small substrates of 20 mm × 20 mm and then polished. Before being subject to measurements, both substrates and pieces of In–Sn alloy were cleaned in acetone.

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Wetting examinations were performed by the sessile-drop method under vacuum (10^{-3} Pa) described in detail elsewhere [11]. In the experiment, various temperatures below glass transformation temperature ($T_g = 720 \text{ K}$) were used in order to avoid crystallization of BMG substrate. While the desired operation temperature was achieved, wetting angles were recorded photographically using back lighting at various times until an equilibrium angle could be recorded. After completion of the sessile-drop experiments, the solidified samples were sectioned to examine the interface between molten In-Sn alloy and Cu₄₀Zr₄₄Al₈Ag₈ BMG. Cross-sections were polished and examined using the high-resolution scanning electron microscopy (HRSEM). The chemical compositions of any interface compounds were determined using an attached energy diffraction spectrum (EDS). The unreacted molten alloy of the wetting experiment specimens was etched off with H2O2-acetic acid solution to reveal the intermetallic compounds formed at the interface. The exposed compound layer was further characterized using X-ray diffraction. Ion beam sputtering profiling in combination with AES technique was employed to investigate the Sn diffusion in Cu40Zr44Al8Ag8 BMG.

3. Results and discussion

Fig. 1 shows contact angels of molten In–Sn alloys on Cu₄₀Zr₄₄Al₈Ag₈ BMG at different temperature for 10 min. It is obvious that the contact angles depend largely on the processing temperature. The initial angle decreases significantly as the processing temperature. The final contact angle is 38° when heated to 473 K. As the temperature increases to 673 K, this angle is only 27.1°. By using the exponential equation, $y = A_1 \exp^{(x/t_1)} + y_0$, to fitting the experiment results, we found the contact angle curves tested under 523 K, 573 K, 623 K and 673 K fit exponential rule very well. However, the curve tested under 473 K is not well consistent with exponential rule. The



Fig. 1. Time dependence of contact angles of molten In–Sn alloy on $Cu_{40}Zr_{44}Al_8Ag_8$ BMG substrates at different temperatures.

spreading process can be classified into three stages when preformed at 473 K: the first is characterized by a slope where the contact angle decreases at a specific rate, the second presents a slighter slope and the third demonstrate that the value of θ is nearly constant.

Fig. 2 presents HRSEM micrographs of cross-section of the $In-Sn/Cu_{40}Zr_{44}Al_8Ag_8$ BMG examined at 523 K, 573 K, 623 K and 673 K. Apparently, a reactive layer formed in the interface of In-Sn/Cu-based BMG. EDS analysis shows that the layer consists of Sn, Zr, Cu and Al elements. XRD results further shows



Fig. 2. HRSEM micrographs for the interfaces between molten In–Sn and $Cu_{40}Zr_{44}Al_8Ag_8$ BMG substrate after spreading at (a) 523 K; (b) 573 K; (c) 623 K; (d) 673 K for 30 min.

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