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Characteristics of precursor film in the wetting of Zr-based alloys on ZrC substrate at 1253 K

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ABSTRACT

The study of precursor film is crucial to understanding the mechanism of triple line moving for the wetting at high temperatures, however, it remains incomplete. In this work, the wetting of ZrC by Zr-based alloys ($Zr_{50}Cu_{50}$, $Zr_{50}Cu_{40}Al_{10}$ and $Zr_{55}Cu_{30}Al_{10}Ni_5$) was studied by a modified sessile drop method at 1253 K. A transition of the precursor film was developed by the addition of constituent elements in the system. Based on the results of a series of wetting systems, the formation condition of the precursor film was proposed, i.e., the occurrence of the precursor film needs to satisfy that active composition has a strong affinity with substrate, and can remain relatively inert. The spreading dynamics were analyzed, which indicated that the wetting with a precursor film was a typical characteristic of adsorption wetting.

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1. Introduction

Wetting of solid surfaces at room temperature has been studied extensively both theoretically and experimentally [1]. A very thin precursor film, usually a single molecular layer propagating ahead of the nominal contact line, is a crucial part in hydrodynamic hypothesis for perfect wetting [2]. Meanwhile, most of these studies have been for the simple liquids and avoid the case in which wetting is accompanied by reactions, dissolution and adsorption, as an inert system. However, the precursor film for metal/metal and metal/ceramic systems at high temperature is more complex, and it usually takes place in a mesoscopic scale [3,4]. Due to the apparent differences between both, the basic underlying mechanisms of the formation of the precursor film at high temperature are poorly understood.

Zr-based alloys have a strong glass-forming ability, and thus it has attracted considerable attention in the preparation of bulk metallic glasses (BMGs). However, a catastrophic failure on one dominant shear band after elastic deformation limits their applications [5]. To improve their plasticity, great efforts have been devoted to the development of the BMG matrix composites. The second phase in the glass matrix can act as a "crack-stopper" by adding impediments to the shear band propagation, and thus considerable plasticity has been achieved [6]. The interfacial chemistry between molten alloys and the second phase is crucial under these circumstances, and it not only influences the alloys' glass-forming ability but also determines the comprehensive performance of composites directly. On the other hand, the occurrence of the precursor film in a metal–ceramic system should satisfy some conditions, as proposed by Xian [4]. One especially important condition is that the molten alloy should contain active elements, such as Zr, Ti, Nb, Hf, etc., and thus Zr-based alloys were preferred to further explore the formation conditions of the precursor film in this study.

The purpose of this study is to reveal the mechanism behind the phenomenon of the precursor film by evaluation of the wettability, reactivity and spreading dynamics between ZrC and liquid Zr-based alloys.

2. Experimental

The $Zr_{55}Cu_{30}Al_{10}Ni_5$, $Zr_{50}Cu_{40}Al_{10}$ and $Zr_{50}Cu_{50}$ alloys were prepared by arc-melting high-purity metals, Zr (99.8 wt.%), Al (99.99 wt.%), Ni (99.998 wt.%), and Cu (99.999 wt.%), in a Ti-gettered argon (99.999%) atmosphere and then suction-casting into a copper mold, as described previously [7]. The polycrystalline ZrC substrates with the dimensions of $15 \times 18 \times 5$ mm^3 were prepared by high temperature (2473 K) sintering of commercial ZrC powders (>98 wt.% purity) in a vacuum

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of 1.13×10^{-3} Pa for 2 h under a uniaxial pressure of 30 MPa. The relative sintered density was ~95%, and the stoichiometry of the C/Zr was determined to be ~0.62 by calculating its lattice parameter from an X-ray diffraction (D/Max 2500 PC; Rigaku, Tokyo, Japan) analysis and then fitting the value to the available relationship between the lattice parameter of ZrC_x and its stoichiometry [8]. The surfaces were polished by diamond pastes down to 1 μm. The average surface roughness, as measured by DEKTAK 6M (Veeco Metrology Corp., Woodbury, NY) over a distance of 2 mm at a speed of 100 mm · s^{-1}, was ~42 nm. Before the wetting test, the polished ZrC substrate and the Zr-based alloy were ultrasonically cleaned three times in acetone.

The wetting experiment was performed in a high vacuum (~2.0×10^{-4} Pa) using a modified sessile-drop method, similar to that described elsewhere [9]. The most significant characteristic of this modified technique is that the substrate and the alloy were separately located before the testing. The alloy sample could be transferred to the ZrC substrate surface when the desired testing temperature and vacuum were reached, and thus enabling the full monitoring of the wetting process and simultaneously eliminating the pre-interaction of the molten alloy with the substrate. Contact angles were directly measured from the captured drop profiles using drop analysis software (the error for the measurement of contact angle is 0.5°). The cross-sectional microstructures were observed using a scanning electron microscope (JSM-5310, Japan) equipped with an energy dispersive spectrometer. The phases in the precursor film were identified by X-ray microdiffraction (D8 Discover with GADDS, Bruker AXS, Germany) using an 800 μm beam diameter.

3. Results and discussion

Fig. 1 shows the variation of contact angle with time for Zr-based alloys on the ZrC surfaces at 1253 K. Obviously, the wettability was improved by introduction of the constituent elements. The initial contact angles θ_i are 93°, 65° and 46°, and the final contact angles θ_e are 14°, 2° and 0°, for $Zr_{50}Cu_{50}$, $Zr_{50}Cu_{40}Al_{10}$ and $Zr_{55}Cu_{30}Al_{10}Ni_5$ alloys, respectively. The spreading of molten metals appears at first in a quick then slow trend in a monotonic continuous process, but no incubation stage (the incubation stage reported by Xu et al. [10,11]) was observed in the whole spreading process. This difference may be ascribed to the experimental method, and they used the traditional sessile drop method in their study, i.e., the alloy and substrate were heated together before the wetting test, and thus the effect of pre-interaction in the further wetting test cannot be neglected.

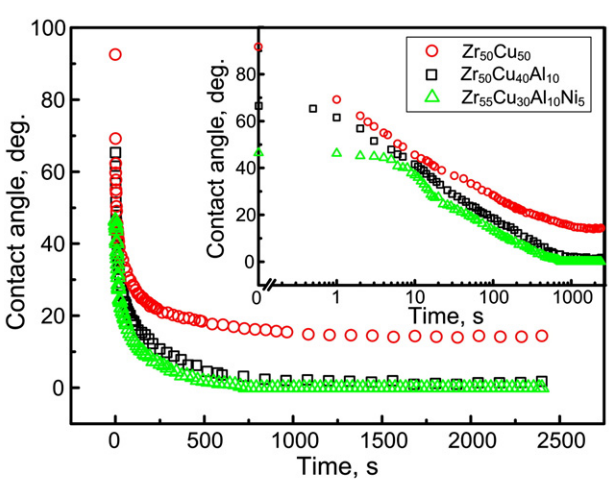

Fig. 1. Variation in the contact angle with time for molten Zr-based alloys on ZrC substrates during the isothermal wetting at 1253 K, upper-right insets used a logarithmic timescale.

Fig. 2 (a)–(c) show the top-view macrographs of solidified drops, the metallic drop of almost the same weight exhibits different features. The precursor film emerged in $Zr_{50}Cu_{40}Al_{10}$/ZrC and $Zr_{55}Cu_{30}Al_{10}Ni_5$/ZrC, rather than $Zr_{50}Cu_{50}$/ZrC. Further, the width of the precursor film increased with the introduction of Al and Ni. The cross-sectional microstructure of $Zr_{55}Cu_{30}Al_{10}Ni_5$/ZrC indicates the weak reaction between liquid and ZrC due to the absence of an evident reaction layer at the liquid–solid interface for even dissolution (Fig. 2 (d)). X-ray microdiffraction patterns at the surface of the precursor film indicate that the thin film is mainly composed of Cu–Zr intermetallics (Zr_2Cu and Zr_7Cu_{10}). Here, we noted that the wetting of ZrC by Zr (the equilibrium contact angle is ~11° [12] at 2183 K) was much better than ZrC by Cu (~127° [13] at 1373 K). Therefore, an apparent conclusion can be derived: the precursor film in the Zr-based alloy/ZrC system should have some relationship with the active element in molten metals, i.e., Zr element.

Also, the precursor film in the Zr-based alloy/ZrC system is not surprising, and it is an equilibrium static film. However, the formation mechanism of the precursor film during spreading remains incomplete and controversial, such as rapid absorption then film overflow [4], evaporation–condensation mechanism [14,15] and diffusion mechanism [12]. The vapor pressures of Zr and Cu are so small (the vapor pressures of Zr and Cu are 9×10^{-14} Pa [16] and 4×10^{-2} Pa [16] respectively), and thus the evaporation–condensation mechanism can be excluded. On the other hand, the addition of Al element in the alloys induced the formation of the precursor film, but the main chemical composition of the precursor film was not Al. We noted that the atomic radius of Al was smaller than Zr and Cu. If the formation of the precursor film was related to the diffusion, the movement of Al in molten alloy would be prior to Zr and Cu, and thus the diffusion mechanism can also be excluded. On the other hand, Xian [4] considered that Al was a harmful element for the formation of the precursor film even with a small addition to the alloy, but here Al has an opposite role which may be due to the different thermodynamic properties of alloys. The good wettability of ZrC by Zr (as mentioned above) indicated that a strong affinity existed between ZrC and Zr at the liquid–solid interface, which also indicated that Zr might be an interface tensioactive element in the alloys and then adsorbed at the close of the triple line. Further, Xian [4] also observed the adsorbed film in the Sn–Zr/Sialon system. Therefore, the precursor film in Zr-based alloy/ZrC system should be an adsorbed film. However, the phases in the precursor film are mainly Zr–Cu intermetallics. It may be strange as to why the precursor film did not exist in the Zr–Cu binary alloy/ZrC system (i.e., $Zr_{50}Cu_{50}$/ZrC system). Hence, we should pay close attention to the condition of the formation of the precursor film, so as to provide some basic guidance for the spreading mechanism.

Table 1 shows the contact angles, width of the precursor film and some thermodynamic parameters for the wetting systems with a precursor film, where $\Delta G_r^0(1\ \text{mol})$ means the Gibbs free energy for 1 mol of a reactant participating in the reaction meanwhile ignoring the effect of activity on reaction, e.g., in the $Zr_{55}Cu_{30}Al_{10}Ni_5$ alloy/Al_2O_3 system, the possible reaction is $3/5Zr + 2/5\,Al_2O_3 \rightarrow 3/5ZrO_2 + 4/5\,Al$, and $\Delta G_r^0(1\ \text{mol}) = 3/5\Delta G_r^0(ZrO_2) - 2/5\Delta G_r^0(Al_2O_3) = -5.869\ \text{kJ} \cdot \text{mol}^{-1}$ at 1253 K ($\Delta G_r^0(ZrO_2)$ and $\Delta G_r^0(Al_2O_3)$ referred to in [20]). Based on the results in Table 1, the most positive value of $\Delta G_r^0(1\ \text{mol})$ is the $Zr_{55}Cu_{30}Al_{10}Ni_5$ alloy/ZrC system, and then the $Zr_{55}Cu_{30}Al_{10}Ni_5$ alloy/Al_2O_3 system. Meanwhile, the width of the precursor film decreased with the decrease of $\Delta G_r^0(1\ \text{mol})$ Therefore, a general conclusion can be roughly drawn; the precursor film appears in a weak reactive system and the width of the precursor film increased with the weakening of reactivity. Due to the complexity of the metal/ceramic system, the metal/metal system with the appearance of the precursor film was firstly considered. Xian [21] suggested that the formation of the precursor film should have the dissolution (in a very small amount) from the substrate metal into molten drop metal, and then form a solid-solution or new compounds at the

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