Journal of Alloys and Compounds 586 (2014) 80–86

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jalcom

Effects of direct current on the wetting behavior and interfacial morphology between molten Sn and Cu substrate

Yan Gu, Ping Shen *, Nan-Nan Yang, Kang-Zhan Cao

Key Laboratory of Automobile Materials (Ministry of Education), Department of Materials Science and Engineering, Jilin University, No. 5988 Renmin Street, Changchun 130025, People's Republic of China

ARTICLE INFO

Article history:
Received 8 August 2013
Received in revised form 1 October 2013
Accepted 3 October 2013
Available online 12 October 2013

Keywords:
Intermetallics
Interfaces
Microstructure

ABSTRACT

The effect of applying a direct electric current on the wetting behavior of molten Sn on Cu substrates at a nominal temperature of 510 K was investigated using a sessile drop method. The final stable contact angles were 37 ± 5° without employing a direct current (DC) while they decreased from 29 ± 3° to 16 ± 2° when the current increased from 2.5 A to 7.5 A. The current polarity does not have a noticeable effect on the wetting behavior but on interfacial morphology. Cross-sectional microstructure observations revealed that applying a current promoted the dissolution of the Cu substrate in molten Sn and the effect was enhanced with increasing current intensity. An unusual morphology with Cu_3Sn being the principal phase and Cu_6Sn_5 being the secondary phase was observed under a relatively large current intensity, particularly for the case of electrons flowing from the Cu substrate to the molten Sn side. Joule heat-induced Marangoni convection in the liquid droplet and electromigration are likely to play significant roles in determining the wettability and interfacial microstructure under the application of a direct electric current.

1. Introduction

Wettability of solids by liquids is of vital importance in many technical applications such as soldering, brazing, plasma spraying and composite fabrication [1–3]. So far, it has been well realized that the wettability is strongly affected by many factors including temperature, atmosphere, alloying elements, and substrate surface structure and roughness [4]. In order to improve the wettablity, common methods such as raising temperature, adding alloying elements and coating the substrate surface with wettable metal layers, are usually adopted. However, in some special situations, these methods might be either ineffective or restrained since they usually bring about other unfavorable effects such as aggravation of interfacial reaction, increase in cost and deterioration of interfacial properties due to excessive growth of intermetallic compounds (IMCs) [5–7]. Therefore, scientists and technicians have never stopped pursuing more effective ways to improve the wettability.

In recent years, the applications of external powers such as electric field and ultrasonic vibration have been widely concerned as a promising way to improve the wettability. For example, electrowetting, which adjusts the wettability on a dielectric layer through an electro-capillary effect [8,9], has been extensively utilized in the fields of micro-fluidic operation, chip laboratory and variable-focus liquid lens. The principle of the electrowetting on dielectric (EWOD) is quite simple. When a voltage is applied between a liquid and a dielectric layer, the accumulation of surface charge on the dielectric layer will cause the Coulomb repulsion and change the free energy of interface, thus affecting the wettability. As far as a metal–metal system is concerned, however, both the melt and the substrate are good conductors. The rapid flow of electrons under a voltage fails to yield a substantial charge accumulation at the interfacial layer, implying that the effect of EWOD will not appear in the metal–metal systems.

Nevertheless, recent studies on the effect of applying a direct current on the interfacial reactions in metal–metal systems have given an implication for the regulation of the wettablity. Chen et al. [10–12] found that the density and polarity of current could influence the solid–solid interfacial reaction in Sn–Ni and Sn–Ag systems and suggested that the change in diffusion rates due to electromigration should be responsible for it. Zhao et al. [13,14] compared the dissolution of solid Ni in liquid Al with and without a DC application and reported that the application of DC significantly decreased the activation energy of dissolution. Moreover, the current polarity had a noticeable effect on the dissolution behavior. Since wetting is closely related to the dissolution and reaction events in metal–metal systems [15], it is reasonable for us to infer that the application of DC may influence the wetting behavior in the conductive metal–metal systems as well. However, very little work has been conducted on this issue even though the

---

* Corresponding author. Tel.: +86 431 85095326; fax: +86 431 85094699.
E-mail address: shenping@jlu.edu.cn (P. Shen).

0925-8388/$ - see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.jallcom.2013.10.021

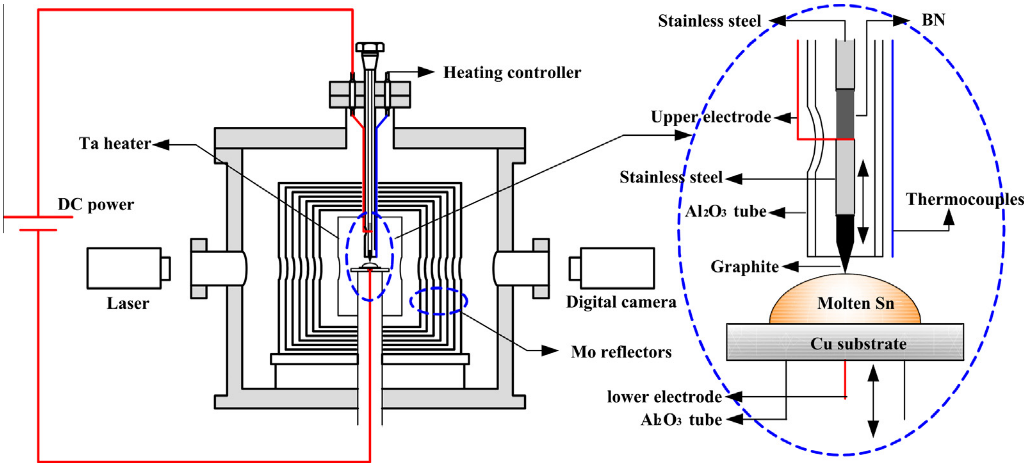

Fig. 1. Schematic illustration of direct current-coupled sessile drop apparatus.

effect of electric power on the solidified microstructure in various alloy systems has been widely reported. To our knowledge, only Xu et al. [16] have recently studied the wetting behavior of molten Bi on a Cu substrate under the effect of DC and reported that the application of the current could significantly improve the wettability, while the change in the current polarity did not exert a noticeable influence. This result is interesting. However, it is worthwhile to point out that there are some deficiencies in their experiments. For example, the Cu electrode was used which easily dissolved in liquid Bi at elevated temperatures as demonstrated in their work, and thus the wetting result was blemished. Moreover, their explanations for the mechanism of the wetting improvement using an electromagnetic theory is doubtable since the electromagnetic force is very small under the maximum current of 2 A in their work, as compared with the surface tension of the liquid, which seems unable to drive the wetting.

In this study, we investigated the wetting behavior using a DC-coupled sessile drop method and examined the interfacial microstructures for molten Sn on the Cu substrates under different current conditions with a primary purpose to explore the influences of DC on the wettability and interfacial microstructural evolution.

2. Material and methods

A DC-coupled sessile drop apparatus was set up in our laboratory. Fig. 1 shows the schematic illustration of the apparatus. The electric transmission device consists of a DC power, a BN insulating connector, two stainless-steel poles, a high-purity graphite (⩾99.99%) electrode with a sharp-pointed head and some pure iron (Fe) wires as connecting leads. The BN connector isolated the upper and lower stainless-steel poles and prevented the current flowing out of the upper stainless-steel pole. A pure iron wire in a diameter of 0.5 mm connected the lower stainless-steel pole through a hole in the Al_2O_3 tube, and the other one connected the substrate to introduce current. The selection of graphite as the upper electrode is to avoid its wetting, dissolution and reaction with molten Sn. Both the graphite electrode and the Cu substrate can move up and down to ensure the permanent contact of the electrode with the molten Sn drop during its spreading. A thermocouple was fixed at the side wall of the Al_2O_3 tube and its measurement tip was about 6 mm over the molten drop to promptly record and regulate temperature due to current-induced heat effect through a proportional, integral and derivative (PID) program controller. The temperature was carefully calibrated using high-purity metals of Sn (⩾99.999%), Bi (⩾99.99%) and Al (⩾99.999%) at a heating rate of 2 K/min in prior to the wetting test.

High-purity (⩾99.999%) Cu plates in dimensions of 20 × 20 × 2 mm^3 were used as substrates. They were mechanically polished to a mirror surface with roughness (Ra) of a few nanometers using diamond pastes. The Sn specimens in a purity of 99.999% were cut into cubic pieces weighing about 0.2 g.

Before the wetting experiment, both the Cu substrate and the Sn specimen were immersed in acetone and ultrasonically cleaned. The chamber was first evacuated to about 5×10^{-4} Pa and then heated at a rate of 20 K/min to the experimental temperature (510 K if not specifically indicated) to avoid or reduce the preinteractions between Sn and Cu in this contact heating mode. The Sn specimen was melted during heating or isothermal holding but did not spread in the initial several minutes after wetting due to the presence of oxide films at both the Sn drop and Cu substrate surfaces, which prevented their intimate contact. After the Sn specimen formed a droplet, the sharp-pointed graphite electrode was lowered down to insert into the molten Sn drop. As the interfacial reaction initiated, the oxide film was broken and molten Sn then began to spread. The moment of the onset of the spreading was defined as zero time in this work. Then we immediately switched on the DC power with a predetermined intensity and polarity, and took photos at a maximum speed of 2 frames per second using a digital camera. Contact angles were calculated from the captured photographs using a professional drop shape analysis program.

After the experiments, the solidified samples were cold-embedded in resin, cut perpendicular to the interface and then polished. Microstructures were observed using a scanning electron microscope (SEM, Evo18, Carl Zeiss, Germany) equipped with an energy dispersive spectrometer (EDS).

3. Results

For the sake of convenient description of current polarity, here we define the current direction from the molten Sn drop to the Cu substrate as positive and the opposite direction as negative. Fig. 2 shows the contact angle variations as a function of time under different current conditions. The initial contact angles were between 95° and 105°, which were obviously influenced by the surface oxidation of both the Sn drop and the Cu substrate [17]. However, the contact angle decreased rapidly with increasing time. The spreading rate was faster and the final contact angle smaller with the

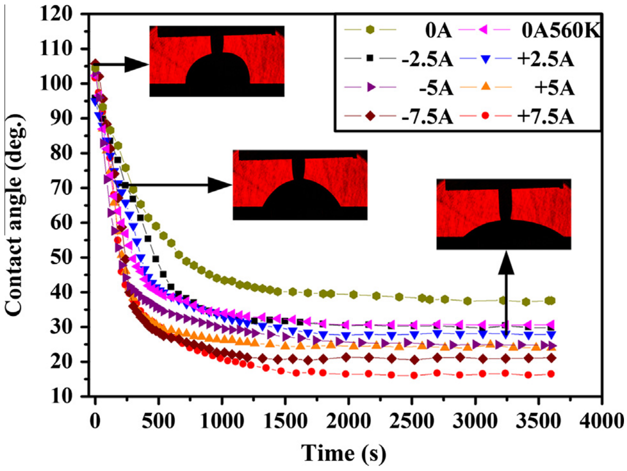

Fig. 2. Variations in contact angle with time under different current intensities and polarities. (The nominal temperature was 510 K, if not specially indicated.)