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Study on diffusion behavior and microstructural evolution of Al/Cu bimetal interface by synchrotron X-ray radiography

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ABSTRACT

Synchrotron X-ray radiography was used to in situ study the interface diffusion behavior and microstructural evolution during the melting and solidification of Al/Cu bimetal. During the solidification, the dendritic growth around the interface is mainly dominated by the variation of Cu concentration and thermal field. Four transition zones of solute profile around the interface were identified to be I (α-Al), II (Al + Al_2Cu), III (Al_2Cu) and IV (AlCu, Al_3Cu_4 and Al_2Cu_3), respectively. During the melting, the concentration variations of Al and Cu around the interface were quantitatively analyzed through the extraction of gray level from sequenced X-ray images. The diffusion coefficients of Cu in liquid Al were calculated from the known concentration variations by an inverse method based on Fick's second law. The diffusion coefficients of Cu in Al were found to follow linear Arrhenius equation dependencies with the pre-exponential factor of $2.83 \times 10^{-5}\ m^2\ s^{-1}$ and the activation energy of $96.0\ kJ\ mol^{-1}$ in a temperature range of 893–970 K.

1. Introduction

Bimetals have been widely applied in many industrial fields because of their excellent physical, chemical and mechanical properties that cannot be obtained from a single material [1,2]. Up to now, many preparation methods, such as roll boding [3,4], explosive cladding [5,6], extrusion cladding [7] and continuous casting [8–10], have been developed for bimetals. Among all the above mentioned methods, continuous casting is a promising technique with good metallurgical bonding, low cost and high efficiency. In the process of preparation of bimetals by continuous casting, one of the solid–liquid bonding technology, the metallurgical bonding process involving the formation of interfacial structure is a major interesting feature [11–13]. As we known, the knowledge of the diffusion kinetics is a basic and important factor to understand such important phenomena [14]. Much research has been done on this using conventional static analysis and mainly focused on the growth behavior of intermetallic compounds usually formed in the vicinity of the interface as a consequence of elements diffusion [15–18]. Among those studies, the effect of annealing time and temperature, magnetic field, trace elements on the growth kinetics of intermetallic compound layers were addressed by considering the atom diffusion. However, the dynamic analysis of diffusion behavior is needed for more in depth study compared with the conventional static study. Much studies [19–27] have shown that in situ and real time observation of crystal growth can now be achieved by applying synchrotron X-ray radiography with satisfying spatial and temporal resolution. This imaging technique can also be applied to uncover the dynamic interface diffusion process of bimetals.

In addition to the growth of intermetallic compounds, the evaluation of diffusion coefficients of elements is also important to understand the diffusion behavior [28,29]. Different methods have been used to evaluate the diffusion coefficients not only in solid metals, such as diffusion couple method with lathe sectioning (DCLS) [30], residual activity method (RC) [31] and microtome sectioning (MS) [32,33], but also in liquid metals, such as capillary reservoir technique (CR) [34], solid/liquid contacting technique (SLC) [35] and X-ray radiography [36]. Little work is reported to study the diffusion behavior and the evaluation of diffusion coefficients at liquid phase using the synchrotron X-ray radiography.

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In this study, we mainly study the dynamic diffusion behavior and microstructural evolution of Al/Cu bimetal interface using synchrotron X-ray radiography. The Al/Cu bimetal was selected because it is a typical bimetal material widely used in electronics industry as cables. The interface diffusion, dendritic growth and the formation of transition zones around the Al/Cu bimetal interface were quantitatively analyzed. Based on the extraction of gray level from sequenced X-ray images, the concentrations of Al and Cu around the interface were obtained. Moreover, the diffusion coefficients of Cu in Al were calculated from the known concentration by an inverse method based on Fick's second law, which is simplified by the difference expression.

2. Experimental procedure

Pure Al (99.99%) and pure Cu (99.99%) were well polished into thin samples with 10×10 mm^2 in surface area and 150 μm in thickness, respectively. A flat cross section of pure Cu sample was close contacted with that of the pure Al sample, and then sandwiched between two ceramic plates. The flat cross sections of the Al and Cu samples were carefully ground, fine polished and cleaned ultrasonically in acetone to modify the contact at the interface. A hollow Mica sheet was also placed between the two ceramic plates, and used to fix the Al/Cu sample. A special vacuum furnace was designed for melting the sample during the experiments. The sample was heated to 973 K with a heating rate of 20 K min^{-1}. At this temperature, Al was melted but Cu was still solid. After the sample held at 973 K for 20 min, the solidification experiment was carried out. The cooling rate was kept at 5 K min^{-1}. The temperature along the sample cell was measured by two embedded K-type thermocouples 2 cm away from each other, one close to the top-right and another close to the bottom-left of the sample cell. The temperatures mentioned in this study refer to an estimation of the temperature in the middle of the field view extrapolated from values on the two thermocouples. A small temperature gradient (0.63 K mm^{-1}) exists inevitably in this experiment, but it had little impact on the dynamic diffusion behavior of Al/Cu bimetal interface since the diffusion is sensitive to local temperature only.

The experiments were carried out on beamline BL13W1 of Shanghai Synchrotron Radiation Facility (SSRF) in China. Phase and absorption contrast synchrotron X-ray radiography was used to investigate the interface evolution of Al/Cu bimetal. The main surface of the sample was perpendicular to the incident monochromatic X-ray beam with an energy of 18 keV. The interface of the Al/Cu sample was adjusted in the center of the field of view by altering the position of the sample holder. A YAG: Ce scintillator screen was used to convert the transmitted X-rays to visible light. The time-sequenced images were recorded by a fast read out-low noise charge coupled device (CCD) camera with 2048×2048 pixel array and a 14 bit dynamical range. The field of view at selected magnification used for all experiments was 6.0×4.1 mm^2 with a satisfying spatial resolution (pixel size 3.7×3.7 μm). The exposure time per frame is 1 s and the sample-to-detector distance is set at 30 cm. The schematic diagram of synchrotron radiation experimental setting is shown in Fig. 1. The electron probe microanalysis (EPMA) measurements were carried out post-mortem after the experiment at the synchrotron source to reveal the content of Cu in the interfacial transition zone of Al/Cu bimetal.

3. Results and discussion

Fig. 2 shows a sequence of in situ radiographs during the melting and solidification of Al/Cu bimetal. The bright region on the upper side is the Al sample, and the dark region on the lower side is the Cu sample. Fig. 2a–e shows the melting process of Al/Cu bimetal, the Al and Cu elements diffused mutually at the interface and formed a clear diffusion front. Fig. 2f–j shows the solidification of Al/Cu bimetal, the Al-rich side solidified in the form of Al–Cu alloy due to the diffusion of Cu element.

3.1. Gray level extraction and EPMA measurements

The concentration variation of Al and Cu can be reflected by the evolution of the gray level (related to X-ray transmitted intensity) during the melting and solidification. Based on X-ray image sequences at different stages of melting and solidification process, the gray level of each pixel along red vertical lines shown in Fig. 2 was read out by the image analyzer. The gray level was then plotted versus distance, as shown by blue curves in Fig. 2c, e, h, i. The curves peak appeared in the interface are due to the loose contact of partial area at interface resulting in the highest X-ray transmitted intensity. As denoted by arrows in Fig. 2c, e, h, i, inflection points (A–E) of the gray curves indicate an abrupt change of Cu or Al concentration. The inflection points A and B characterize the diffusion fronts at Al side and Cu side during the melting, respectively. The propagation of the diffusion fronts on the Al and Cu sides clearly starts with a periodic undulation which was formed in the process of cutting Al and Cu samples. That is, the microscopic cross sections were not completely flat although the samples were carefully ground and fine polished. The inflection point C corresponds to the front of dendrite where the gray level rises much due to the rejected solute of Cu. The inflection points D and E reveal the boundaries of the interfacial transition zones which will be discussed later. Since the X-ray transmittance in Al is higher than that in Cu, the gray level is proportional to the Al concentration in an Al–Cu system. As inferred from the gray level curves, the Al content gradually increases from the Cu-rich side

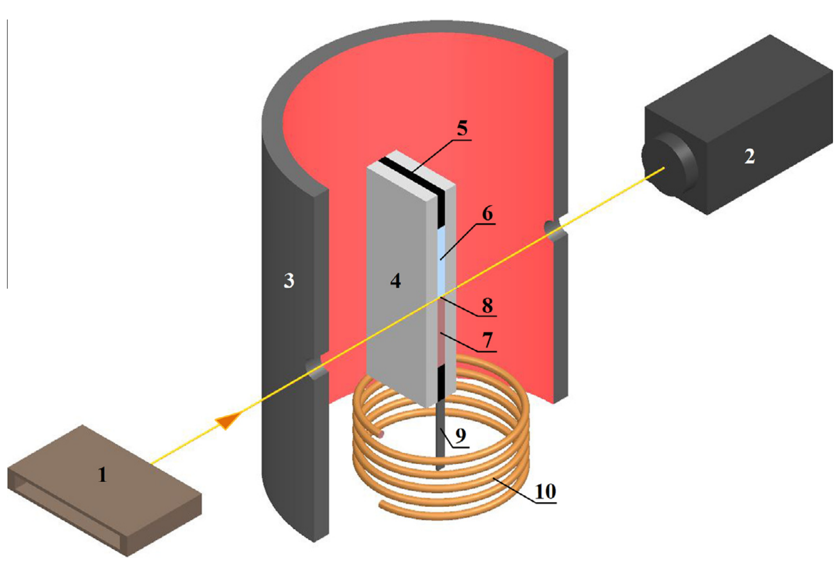

Fig. 1. Schematic of synchrotron radiation imaging experimental setting. 1: Synchrotron radiation X-ray source; 2: CCD camera; 3: vacuum furnace; 4: ceramic plate; 5: mica sheet; 6: pure Al sample; 7: pure Cu sample; 8: interface; 9: sample holder; 10: water-cooled copper pipes.