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In situ observation on the formation of intermetallics compounds at the interface of liquid Al/solid Ni

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ABSTRACT

The growth of intermetallic compounds (IMCs) in liquid Al were observed on Ni substrate through synchrotron radiation real-time imaging technology. The results provided direct experimental evidence of the diffusion sequence of solid Ni and the growth mechanisms of the IMC layer. Through the snapped images, it was found that the Al_3Ni_2 layered phase formed first in cooling stage, then the formation of irregular Al_3Ni phase through peritectic reaction. The growth of Al_3Ni_2 layer was mainly controlled by diffusion.

Reaction mechanisms and growth behaviors of the interactions joining dissimilar metals are vital for the practical application. Due to the excellent performance in creep strength and oxidation resistance, the interfacial reactions in various Al-Ni intermediate phases have been studied extensively [1–3]. Among them, most of researches have been focused on the reaction between solid Al/solid Ni interface, while less attention was paid to the growth of Al_3Ni and Al_3Ni_2 phases at the liquid Al/solid Ni interface, which is a key clue to further understand liquid-metal infiltration, hot dipping coating, diffusion brazing and transient liquid phase bonding (TLPB) [4,5]. In previous work, it is widely accepted that Al_3Ni_2 phase and Al_3Ni phase would be found at the interface after solidification with the remnants of Al. However, there is no consistent conclusion on formation sequence and growth behavior of IMCs. For diffusion soldered Ni/Al/Ni interconnections, it is concluded that the primary phase is formed during dissolution of solid Ni, which could be Al_3Ni phase [5,6] or Al_3Ni_2 phase [7]. For Immersion test, the primary phase is found to be Al_3Ni phase followed by Al_3Ni_2 phase during dissolution of solid Ni [8,9]. For Al/Ni diffusion couple, Al_3Ni_2 phase is considered as the primary phase formed during holding, followed by Al_3Ni phase formed during holding [10] or during cooling [11]. Therefore, to clarify the formation sequence and growth behavior of IMCs during heating, holding and cooling, a further study should be conducted on the microstructural evolution at interface of liquid Al/solid Ni.

Previous studies were done by analyzing the solidified microstructure after quenching. Through this method, it was impossible to discover the overall microstructural evolution. Recently, synchrotron radiation real-time imaging technology was well used to in situ observe the growth behavior of IMC at liquid Sn/solid Cu interface [12–14]. In this study, this approach is first time utilized for in situ observation on microstructural evolution at the interface of liquid Al/solid Ni. The growth behavior of Al_3Ni_2 layered phase was analyzed.

Pure Ni (99.99% pure) and Al (99.999% pure) plates with the dimensions of 10 mm × 10 mm were prepared for the synchrotron observation. The thickness of the sheets was 0.5 mm. The Ni and Al plates were polished and cleaned ultrasonically, then fixed end to end within Al_2O_3 ceramic plates. The thickness of the ceramic plates is 0.4 mm. Assembled sample is placed in a specially designed furnace with two windows passed by the x-ray. Sample was heated to 850 °C at heating rate of 20 °C/s with an accuracy of ±1 °C, held for 1 h and then natural cooling in the furnace. Synchrotron radiation experiment was carried out on BL13W1 beamline at Shanghai Synchrotron Radiation Facility, China (SSRF). The main surface of the sample was set perpendicular to the incident monochromatic X-ray beam with energy of 26 keV, and the interface of the Al/Ni plates was adjusted in the center of the field of view. The time-sequenced images were captured by a charge coupled device (CCD) with a good resolution ratio of 3.25 μm per pixel. The exposure time was 0.5 s and the distance between the sample and CCD camera was 85 cm. The schematic diagram of synchrotron radiation experimental configuration was shown in Fig. 1(a). The cross-sectional morphologies and chemical composition of the sample after solidification was detected by scanning electron microscopy (SEM) equipped

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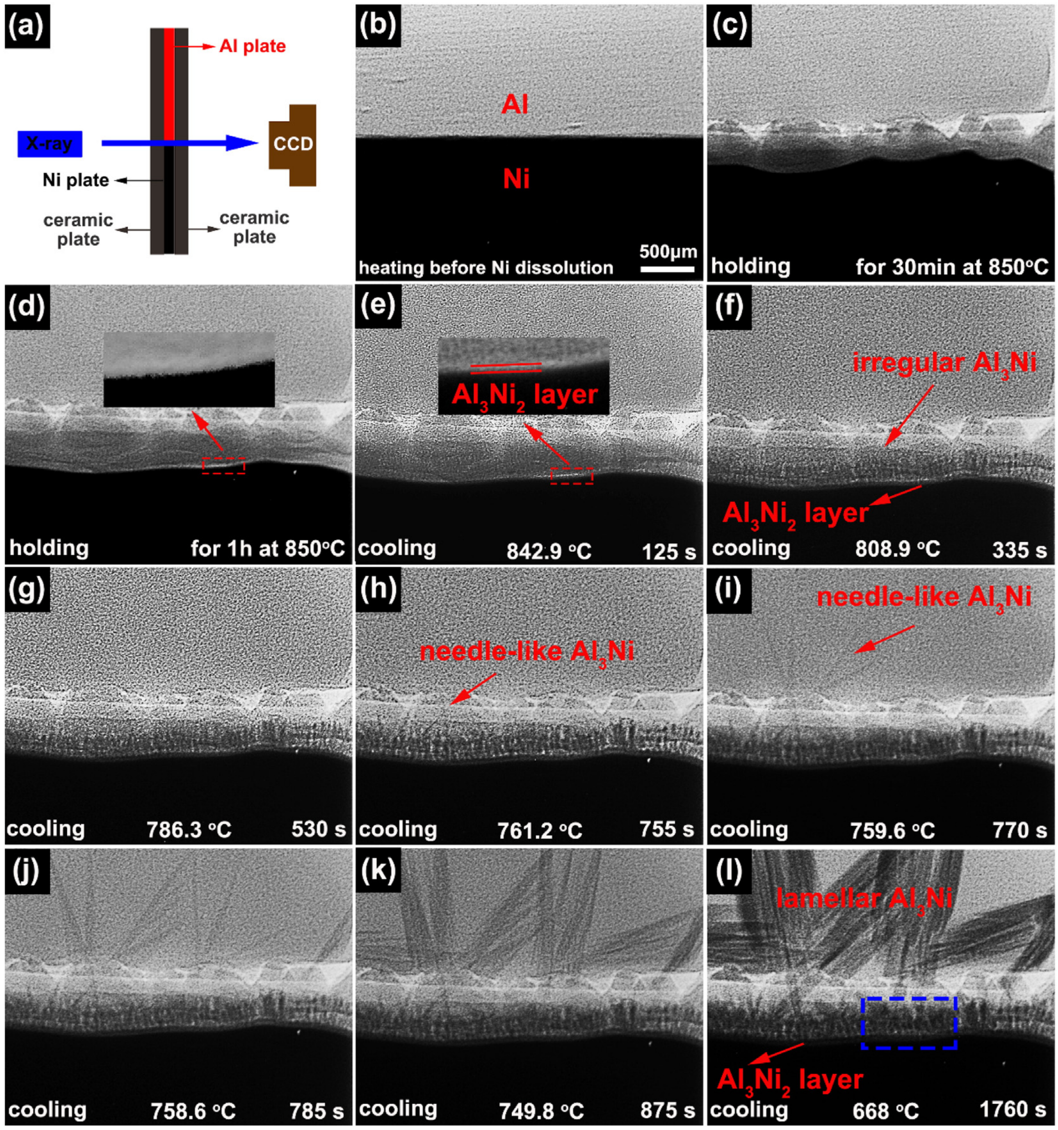

Fig. 1. (a) Schematic diagram of synchrotron radiation experimental configuration; (b–l) Synchrotron radiation images of the microstructural evolution at the interface of liquid Al/solid Ni.

with an energy dispersive spectrometer (EDS). The crystallographic structure of the phases was analyzed by X-ray diffraction (XRD) using monochromatic Cu-Ka radiation at 40 kV and 30 mA.

The synchrotron radiation images of the microstructural evolution at the interface of liquid Al/solid Ni during heating, holding and cooling are shown in Figs. 1(b)–(l). The solidification process has also been recorded as a video, which can be found in the supplemental material (Supplementary video 1). The top gray zone is molten Al, the black zone at the bottom is Ni substrate. During the early stage of heating process, the interface was smooth, as shown in Fig. 1(b). After held for 0.5 h (Fig. 1(c)), solid Ni dissolved into Al melt non-homogeneously. The interface was a wavelike morphology. After 1 h (Fig. 1(d)), no IMC was formed. The interface tended to be planar. During the early stage of cooling (Figs. 1(e)–(f)), a layered IMC formed at the interface first, followed by an irregular IMC. As time prolonged, the irregular IMC became coarser, accompanied by the growth of layered IMC (Fig. 1(g)). As shown in Figs. 1(h)–(i), needle-like IMC formed on or adjacent to the irregular IMC, and grew into molten Al. Subsequently, needle-like IMC extended into molten Al growing as lamellar phase in a short time and accompanying with the growth of interfacial IMCs (Figs. 1(j)–(l)).

The cross-sectional morphology of the interfacial IMCs in blue region in Fig. 1(l) was further analyzed after solidification under SEM and the image is shown in Fig. 2(a). As described above, there are two kinds of Al-Ni IMCs found at the interface. The thin continuous layered phase (point 1) and the irregular phase (point 2), are identified as Al_3Ni_2 phase and Al_3Ni phase through the analysis of EDS and XRD results (Fig. 2(b)–(e)). Also, lamellar Al_3Ni (point 3) and fine Al_3Ni phases (circled in white) are found next to irregular phase.

To further map the sequence of the IMCs at the interface, the sample was heated and cooled in the differential scanning calorimetry (DSC). DSC results are shown in Fig. 2(f). There is one endothermic peak during heating, which attributes to Al melting. The exothermic peak 1, peak 2 and peak 3 during cooling are attributed to Al_3Ni_2 phase precipitation, peritectic reaction and eutectic reaction respectively [15]. In short, there is no formation of IMCs at the interface during heating and holding, except for the dissolution of solid Ni into liquid Al. But in cooling sequence, layered phase first forms at the interface, referring to exothermic peak 1 in DSC curve, then irregular Al_3Ni phase by peritectic reaction. As time prolongs, proeutectic Al_3Ni phase on or adjacent to peritectic phase and eutectic phase in the molten Al are formed successively, accompanied by the growth of layered Al_3Ni_2 and peritectic Al_3Ni. Therefore, the irregular, lamellar and fine Al_3Ni phases are identified as peritectic, proeutectic and eutectic phases, respectively. The results are different from the previous works, as listed in Table 1, which may be related to the unsaturation of Ni in the liquid Al after holding for 1 h in this paper [16–19].