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Microstructural evolution and growth behavior of intermetallic compounds at the liquid Al/solid Fe interface by synchrotron X-ray radiography

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ABSTRACT

The growth of Inter-Metallic Compounds (IMCs) with dissolution was investigated on the liquid Al/solid Fe interconnection by using synchrotron radiation real-time imaging technology. At the initial stage of holding at 850 °C, a layer of the η-Fe_2Al_5 phase formed at the interface, and tongue-like η grew into α-Fe. The tongue-like η phase had a stronger (001) texture than the layered phase. The coalescence of tongue-like η started at the bottom, followed by the dissolution of IMCs, which leads to a further increase in its growth rate. During solidification stage, the dotted, needle-like and flake θ-$FeAl_3$ phases were sequentially formed.

1. Introduction

The dissimilar joints between solid Fe (α) and liquid Al (L) are widely studied in many manufacturing processes including aluminizing and bi-metals fabrication to obtain a good oxidation and corrosion resistant coating [1–3]. The protective coating is composed of an aluminum topcoat, a thin layer of θ-$FeAl_3$ with polyhedral morphology, and a thick layer of η-Fe_2Al_5 with a specific tongue-like structure [4–6]. At the same time, precipitated needle-like and flake θ phases are found in the topcoat layer. There are many differing opinions on the formation mechanism and growth behavior of θ phase adhered to η phase. For example, Bouché et al. thought the growth behavior followed a parabolic law [7], while Bouayad et al. believed that it was approximately linear [8]. Rezaei et al. proposed that IMCs formation was controlled by dissolution and precipitation of IMCs at 750 °C < T < 900 °C [9], while Chen et al. suggested that reaction diffusion and precipitation was dominant at 700 °C and only precipitation happens at 900 °C [10]. As shown, there was no agreement on the formation mechanism of the θ phase so far. And more remarkable, the above formation mechanisms were studied by analyzing the solidified microstructure after quenching, which was impossible to reveal the whole picture of microstructure evolution. Therefore, the formation of θ phase with different morphologies should be investigated by more effective methods.

For η phase, a parabolic time dependence of average thickness was reported [11,12]. Due to high vacancies concentration along the c-axis of orthorhombic η phase and stress field caused by the α → η transformation, the diffusion of Al atoms was enhanced along [001] direction, which led to a tong-like η phase preferential aligned in [001] direction [13–15]. Additionally, the effect of temperature and alloy elements, especially Si, on the morphology of the η phase was studied previously [16–18]. The deformation of α phase, tongues to planar growth transition of η phase and porosity formation in the η phase were in situ observed by X-ray absorption microtomography [19]. Apparently, a thick and brittle IMC layer would deteriorate the interfacial mechanical properties. At the liquid Al/solid Fe interface, the thickness of IMC layer increased with reaction time at low temperatures, but the dissolution at high temperatures could reduce the thickness to some extent [20]. Chen et al. considered that the increased solubility at high temperatures resulted in the dissolution of η phase, further caused the growth of η phase deviating from parabolic law [10]. While, the detailed information of the dissolution effect on the η phase was not shown. Dybkov et al. believed that the dissolution accelerated the growth rate of η phase and proposed a model describing the growth with simultaneous dissolution based on some assumptions [20]. However, the experimental data points during the early stage had a deviation from the calculated result. Therefore, the effect of dissolution on the growth behavior of η phase should be discussed in detail.

It is worth to note that conventional methods do not provide direct and detailed information on the formation and growth of IMCs. Recently, it has been proven that the X-ray synchrotron radiation imaging technology is a more effective approach in the investigation of the growth behavior for IMCs and dendrites at the liquid/solid

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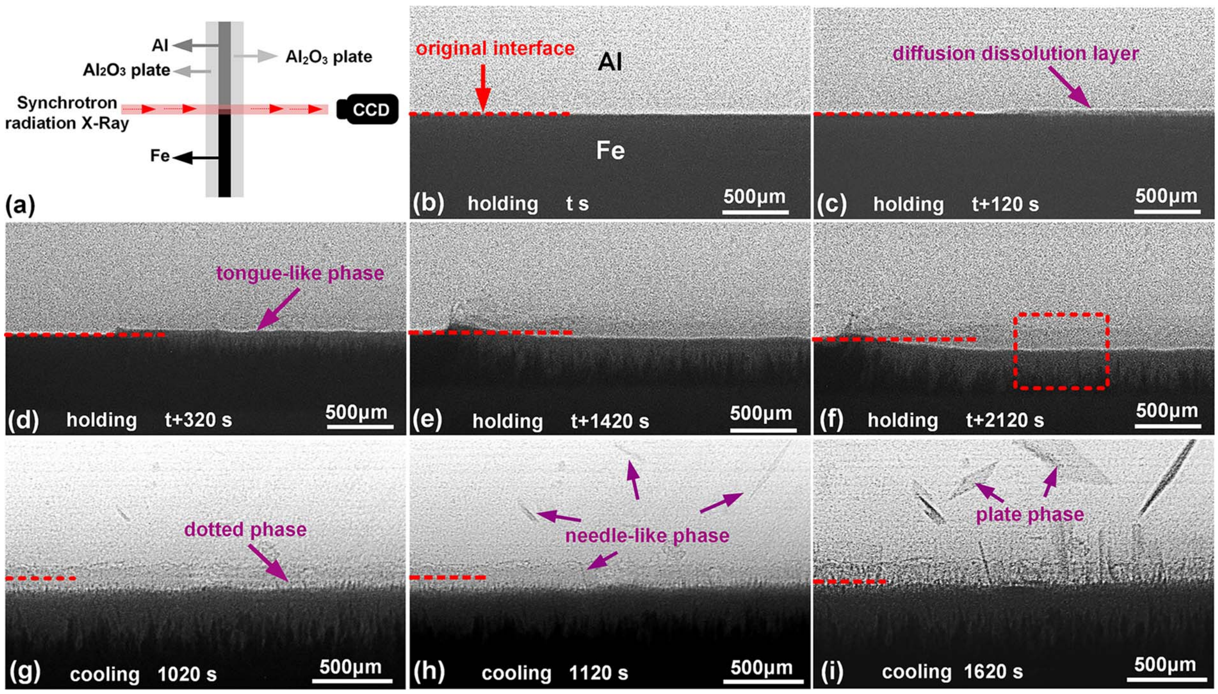

Fig. 1. A schematic diagram of experimental configuration (a); Synchrotron radiation real-time images of microstructural evolution at liquid Al/solid Fe interface during holding (b–f) and during cooling (g–i) stages. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

interfaces comparing with conventional methods [21–23]. In this paper, synchrotron radiation imaging technology was utilized to in situ monitor the growth behavior of η and θ phases at the liquid Al/solid Fe interface, and to further understand the effect of dissolution on IMCs growth.

2. Experimental Methods

Pure Fe (99.99%) and Al (99.999%) sheets were pre-polished to fabricate Al/Fe interconnection. The dimensions of both Al and Fe sheets were 10 mm × 10 mm × 0.4 mm. Two pieces of Al_2O_3 ceramic sheets with a thickness of 0.5 mm were used to fix the interconnection. The synchrotron radiation experiment was carried out using the BL13W1 beam line at Shanghai synchrotron radiation facility, China. The imaging method was phase and absorption contrast imaging technique. A furnace with a window was vertically aligned with the direction of the X-rays, as shown in Fig. 1(a). In order to reach a homogenized condition, the sample was heated to 850 °C for 0.6 h during the holding period. The samples were then cooled in the furnace. A synchrotron X-ray beam with an energy of 26 keV was set for imaging, and a high speed CCD camera with a resolution of 3.25 μm/pixel and an exposure time of 0.5 s was used to record the images. The working distance between the detector and sample was 85 cm. The morphologies were examined by a scanning electron microscopy (SEM) equipped with an energy dispersive spectrometer (EDS) detector. For the EBSD analysis, a NOVA Nano SEM with electron backscatter diffraction (EBSD) detector was used to obtain the orientation information. The EBSD scans were conducted at 20 kV with a step size of 0.5 μm.

3. Results and Discussion

3.1. Microstructural Evolution at the L/α Interface

The synchrotron radiation real-time images of microstructural evolution at liquid Al/solid Fe interface were shown in Fig. 1 (b)–(i). The top gray zone was molten Al, the black zone at the bottom was solid Fe. The position of the initial interface was marked using the red dotted line. The interface was smooth at the early stage during holding, as shown in Fig. 1(b). After 120 s, solid Fe began to dissolve into liquid Al with the formation of a diffusion dissolution layer (Fig. 1(c)). In the situation of 320 s, an IMC layer formed at the L/α interface, which has a tongue-like morphology beneath the non-planar interface (Fig. 1(d)). After 1420 s, the position of interface moved downwards relative to the original position. As shown in Fig. 1(e)–(f), the downwards displacement of the interfaces increased with time when exceeding 2120 s. This is attributed to the dissolution of the top solid layer into liquid Al and growth of the tongue-like phase into solid Fe [19]. During cooling, a dotted phase firstly formed inside liquid Al above the interface, as shown in Fig. 1 (g). After 1120 s, needle-like phase emerged in a region inside Al melt further away from the interface. At the late stage of solidification, a part of the needle-like phase transformed into flake phase, as shown in Fig. 1(h)–(i).

The phase maps and point analysis of the interfacial IMCs after solidification examined by EDS and EBSD analysis are shown in Fig. 2. There are two kinds of IMCs found at the L/α interface. The electron backscatter patterns (EBSPs) and point component analysis were taken to identify the crystallographic structure of the IMCs as θ and η phases, as shown in Fig. 2(b)–(e). The tongue-like IMC was η-Fe_2Al_5 phase, and the dotted, needle-like, flake-like and fine IMCs were identified as θ-$FeAl_3$ phases. In the map, high-angle boundaries with misorientation angles ($\theta > 15°$) were presented as bold lines, and low-angle boundaries ($2° < \theta < 15°$) were shown as fine lines, as shown in Fig. 2(a). There was high density of low-angle boundaries as well as high-angle boundaries developed in the η phase, which was consistent with the misorientation angles in the previous literature [15]. The θ/α interface was identified from the phase map. The upper part of the η phase was depleted to form the θ phase, as indicated by the white arrows in Fig. 2(a).

3.2. Growth of η-Fe_2Al_5 Phase During Holding

3.2.1. Growth Behavior of η Phase Accompanied With Dissolution

Fig. 3 shows high-magnification in situ observations in the red