Study on coarsening of Ag_3Sn intermetallic compound in the Fe-modified Sn–1Ag–0.5Cu solder alloys

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ABSTRACT

The present study focuses on coarsening of Ag_3Sn intermetallic compound in the Sn–1Ag–0.5Cu and Fe-modified Sn–1Ag–0.5Cu solder alloy. The investigations showed that the Ag_3Sn intermetallics coarsened rapidly in the Sn–1Ag–0.5Cu solder alloy whereas the Ag_3Sn intermetallics were found to be quite stable in the Fe-modified Sn–1Ag–0.5Cu solder alloy. The lattice strain in the Ag_3Sn intermetallics and the blocking effect on Ag diffusivity in Sn matrix suggested the possible mechanisms for the coarsening suppression of the Ag_3Sn intermetallics in the Fe-modified solder alloy.

1. Introduction

The process resulting changes in solder microstructure over time is important as it will cause mechanical properties to evolve, often to the detriment of joint reliability [1–3]. Microelectronic solders based on Sn–Ag or Sn–Ag–Cu are generally exposed to microstructural coarsening during service or storage [4,5]. This microstructural coarsening causes continuous evolution of creep, fracture, and thermo-mechanical fatigue properties of the solder-joint over time, and thereby influences the long-term reliability of microelectronic packages [4,6–8]. Several attempts [9–11] have been made in the past to address the problem of microstructural coarsening in Sn–Ag–Cu solder during isothermal aging. It has been found that Ag_3Sn intermetallic compound (IMC) particles in the microstructure undergoes Ostwald ripening. The Cu_6Sn_5 IMC particles coarsen even more rapidly [4,12,13]. We would like to add here that the fraction volume of Cu_6Sn_5 is much smaller than that of Ag_3Sn [4,13,14]. The effect of Cu_6Sn_5 coarsening to the evolution of mechanical properties over time is small compared to that of Ag_3Sn. Hence, the impact of microstructural coarsening in both Sn–Ag and Sn–Ag–Cu alloys is dominated by the coarsening kinetics of Ag_3Sn IMC. Sabri et al. [15] investigated the effect of aging on the microstructural evolution and mechanical properties in Sn–1Ag–0.5Cu and Sn–3Ag–0.5Cu solder alloys. The results showed that the Ag_3Sn IMC particles became notably coarsened after aging which in turn degraded their mechanical properties. Maleki et al. [16] reported that a considerable coarsening of Ag_3Sn IMC particles occurs in the Sn–4Ag–0.5Cu solder alloy during ageing and therefore the mechanical behavior of solder shows a continuous reduction in the yield resistance. The impact of pure La on the microstructure and mechanical properties of Sn–3Ag–0.5Cu alloy at high temperatures was studied by Sadiq et al. [17]. According to them, the addition of La refined the microstructure, and reduced the size of particles of Ag_3Sn IMC up to 40% for the as-cast samples, and drastically reduced the coarsening rate of these particles by up to 70% with no changes in the particle spacing, which in turn improved the mechanical properties of Sn–3Ag–0.5Cu alloy.

Similarly Tsao and others [18] studied the effects of Al_2O_3 nanoparticles on the microstructure and microhardness of the Sn–3.5Ag–0.5Cu composite solder ball grid array joints on Sn/Cu pads, and pointed out that the average size of Ag_3Sn particles decreases significantly in the composite solder matrix. Zhao and co-workers [19] reported that Bi-bearing Sn–3Ag–0.5Cu exhibited relatively stable mechanical properties with aging which was attributed to the strengthening effect of Bi. Hammad et al. [20] pointed out that the addition of Ni to the Sn–0.5Ag–0.7Cu alloy results in microstructural refinement, uniform distribution of Ag_3Sn and $(Cu, Ni)_6Sn_5$ IMC particles, and small primary β-Sn grains, as well as the mechanical strength and microhardness were apparently

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enhanced. Sabri et al. [21] observed that small additions of Al to Sn–1Ag–0.5Cu solder alloy suppressed the coarsening of Ag_3Sn IMC, thus improving the grain structure stability of the bulk solder. This result changes the bulk solder into stable mechanical properties during aging. The effects of adding small amount of Zn and conducting an aging treatment at elevated temperatures on the microstructure and tensile properties of Sn–1Ag–0.5Cu solder alloy were investigated by Hamada et al. [22]. The results revealed that addition of Zn inhibited the growth of Ag_3Sn IMC and suppressed the decline in the flow stress after the aging treatment. In our previous study [23,24], we investigated the effects of Fe addition on the microstructure and tensile properties of Sn–1Ag–0.5Cu solder alloy and conducted an aging treatment at elevated temperatures. The results revealed that the addition of Fe increased the elastic compliance and the plastic energy dissipation ability of the solder alloy. Moreover, the addition of Fe effectively enhanced and stabilized the mechanical properties of the aged Sn–1Ag–0.5Cu solder alloy. This observation was made because of the coarsening suppression of the Ag_3Sn intermetallics. Therefore, the present paper further sheds light on the possible mechanisms for the coarsening suppression of the Ag_3Sn intermetallics.

2. Experimental procedures

Solder alloys were prepared by melting pure ingots of Sn, Ag, Cu, and Fe in an induction furnace at more than 1000 °C for 40 min. Then, the molten alloys were mixed with pure liquid Sn in a melting furnace at 290–300 °C for 60 min. Subsequently, some portion of the molten alloys were cast to disk-shaped ingots and sent to a third-party laboratory (SGS) to verify the Fe element concentration. Chemical composition analyses were carried out to determine the exact composition of the cast ingots. The details of the chemical composition analysis are given in Refs. [23,24]. Then, the remaining portion of the molten alloys was poured into the stainless steel moulds which were preheated at 120–130 °C, and the moulds were naturally air-cooled to room temperature (25 °C). The moulds were disassembled, and the samples were removed and visually inspected to ensure that the surface of the parallel area was free of damage and voids. Microstructural analysis of the solder alloy was reported previously [16,19,21]. Scanning electron microscopy (SEM) with backscattered electron detection was used to examine the microstructures. Elemental compositions of the intermetallic compound (IMC) particles were measured by energy dispersive X-ray spectroscopy (EDX). More than 500 IMC particles were measured for each phase. 95% confidence intervals statistic was used to predict the range of concentration of Fe present in Ag_3Sn IMC particles. X-ray diffraction (XRD) was also carried out to determine the IMC phases. To obtain the microstructures, the solder samples were prepared by dicing, resin moulding, grinding, and polishing. The samples after grinding were mixed with four grades of SiC paper (#800, #1200, #2400, and #4000) and then mechanically polished with a diamond suspension (3 μm). Finally, the specimens were polished with a colloidal silica suspension (0.04 μm). In order to investigate further, the possible presence of Fe in the Ag_3Sn IMC particles, TEM process was used under an FEI Tecnai F30 electron microscope equipped with EDX.

3. Results and discussion

In our previous studies [15,21], we have shown that the Ag_3Sn IMC particles in the Sn–1Ag–0.5Cu (SAC105) solder alloy clearly undergoes Ostwald ripening during aging, and the primary β-Sn dendrites coarsen as a result. Ostwald ripening is defined as the growth of larger crystals by the dissolution of smaller crystals with a higher interfacial enthalpy than the larger crystals, In other words, it describes the growth of large particles by the dissolution of small particles [25]. The driving force for an Ostwald ripening process is the reduction in the total interface area between the matrix phase and the dispersed phase. The free energy of a particle changes as the particle size changes. This change causes a change in the solubility at the particle/matrix interface, which is governed by the Gibbs–Thomson effect. The effect is that the matrix region surrounding smaller particles contains more solute than the matrix region surrounding larger particles, which sets up solute concentration gradients in the direction of the smaller particles to the larger particles. The result is that larger particles grow with time and smaller particles shrink, even while the overall volume fraction of the particles remains constant [12,13].

The solubility of Ag in the liquid Sn at 250 °C was estimated by Thermo-Calc as 5.03 wt.%, whereas the solubility of Ag in the solid Sn at 200 °C and 150 °C was less than 0.052 wt.% [12]. Therefore, all excess Ag solute atoms above the solubility limit in the solid state precipitate as Ag_3Sn IMC particles when the solder alloy solidifies under equilibrium. However, we have seen that air-cooling after casting, all excess Ag atoms do not immediately precipitate due to the rapid cooling rate. Therefore, more Ag atoms remain in the Sn matrix than predicted by the equilibrium phase diagram. The as-cast microstructure of SAC105 solder alloy possesses Ag_3Sn particles in the size range of approximately 0.11–0.72 μm, as shown in

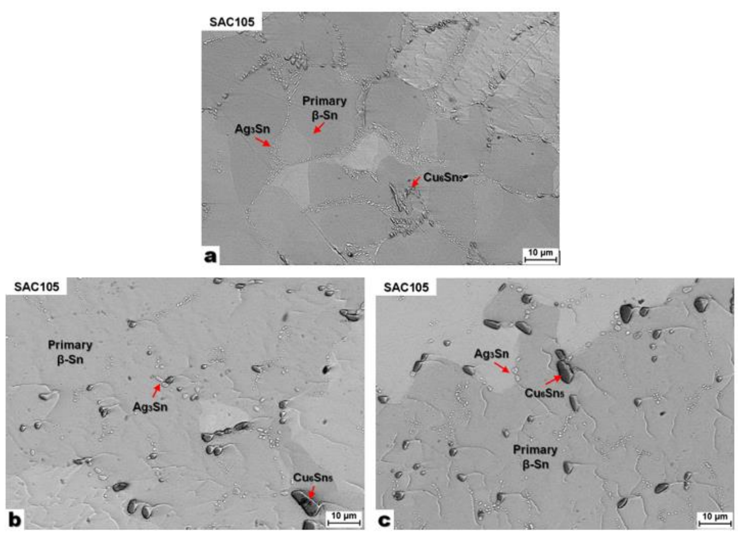

Fig. 1. SEM micrographs of SAC105 solder alloy (a) as-cast condition, (b) after 720 h of aging at 100 °C, and (c) after 24 h of aging at 180 °C.

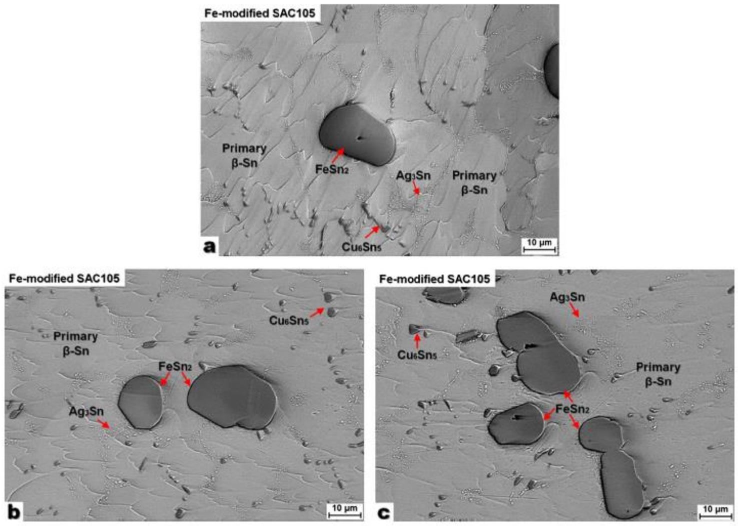

Fig. 2. SEM micrographs of SAC105 solder alloy (a) as-cast condition, (b) after 720 h of aging at 100 °C, and (c) after 24 h of aging at 180 °C.

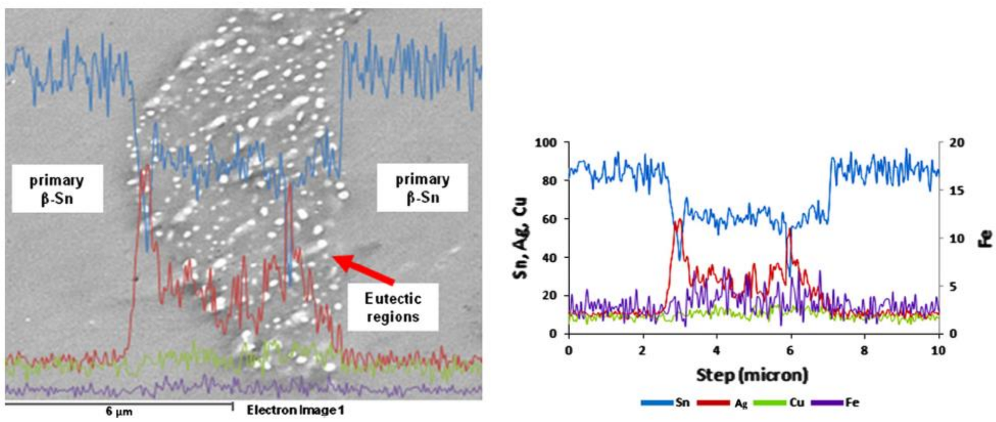

Fig. 3. Summary of line-scan EDS results across the eutectic region from the primary β-Sn, through the eutectic region, and into the primary β-Sn of Fe-modified Sn–1Ag–0.5 Cu solder alloy.

Fig. 1a [15,21,24]. Since, the diffusion of Ag atoms in Sn increases during aging [12,26], the Ag solute atoms migrate from the Sn matrix surrounding the smaller Ag_3Sn particles and diffuse into the Sn matrix toward the Sn matrix surrounding the larger Ag_3Sn particles based on the above-mentioned Gibbs–Thomson effect. This migration of Ag solute atoms causes an increase in the vacancy diffusion rate of the Ag_3Sn particles. Consequently, Ag atoms break from the Ag_3Sn lattice and diffuse along the Ag_3Sn/β-Sn interface into the Sn lattice towards the larger Ag_3Sn particles. This effect points to the growth of large Ag_3Sn particles by the dissolution of small Ag_3Sn particles. Therefore, the number of Ag_3Sn in the SAC105 solder alloy drastically decreases after aging compared with the as-cast microstructures due to the Ostwald ripening process (Fig. 1b and c) [15,21,24].

As an ongoing fundamental research activity [23,24], our group has mainly concentrated on the study of the microstructure and mechanical properties of the SAC105 solder alloy containing a minor addition of Fe. This work was prompted by our discovery of the dual response of the SAC105 solder alloy to a minor addition of Fe, resulting both in a microstructure coarsening suppression (Fig. 2) [24] and in stable mechanical properties with high temperature aging. It was because of the coarsening suppression of Ag_3Sn IMC particles. The possible mechanisms for the coarsening suppression of the Ag_3Sn IMC particles are discussed in the current study. There is no doubt that, microstructural results show a significant fraction of the Fe solute forms $FeSn_2$ phase, consistent with the limited solid solubility of Fe in primary β-Sn. However, in air cooling after casting, all excess Fe atoms do not immediately precipitate due to the rapid cooling rate. Therefore, more Fe atoms remain in the eutectic regions. Besides, Fe atoms may exist in the primary β-Sn. In other words, not only does Fe exist in $FeSn_2$ IMC (higher Fe concentration). The change in concentration of Sn, Ag,

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