

## Wetting and Interfacial Reaction Characteristics of Sn-1.2Ag-0.5Cu-xIn Quaternary Solder Alloys

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Through the use of a wetting balance technique, the wetting characteristics of Sn-1.2Ag-0.5Cu-xIn quaternary solder alloys with respect to the In content and soldering temperature were investigated to validate the applicability of compositions with a low Ag content as solder material. It was found that a small addition (0.4–0.6 wt.%) of In significantly improved the wetting properties of the Sn-1.2Ag-0.5Cu-xIn composition at soldering temperatures ranging from 230 °C to 240 °C due to the excellent wetting property of In. In an observation of the interfacial reaction, it was found that the added In element did not participate in the interfacial reaction with a Cu or Ni pad, unlike in the Sn-Ag-Cu-In case, which has a high In content. The package or board-side IMC layers in Sn-1.2Ag-0.5Cu-0.4In joints were thinner than those of Sn-3.0Ag-0.5Cu, especially after aging.

**Keywords:** alloys, soldering, interfaces, wetting, scanning electron microscopy (SEM)

### 1. INTRODUCTION

During the past decade, Sn-3.0~4.0(wt.%)Ag-0.5~1.0Cu Pb-free alloys have been used as a common solder material in the electronics industry as replacements for Pb-based compositions. Therefore, extensive studies of their processes and/or the reliability issues related to their compositions have been reported [1-18]. However, an increase in the number of Pb-free alloy choices has been noted recently in electronics packaging research, including Sn-Ag-Cu alloys with a low Ag (less than 2.5 %) content, due to concerns such as the high raw material cost of alloys and their poor drop/shock performance [18-22].

In recent studies in the area of surface mount technology (SMT), the peak temperature for reflow soldering is mostly maintained at less than 240 °C to 245 °C to improve the board-level reliability and to suppress thermal degradation [28]. Thus, relatively low reflow temperature regions of 230 °C to 240 °C are frequently detected in high-density modules during actual reflow soldering processes. Considering this, the wettability of solder alloys should be evaluated as a function of the reflow temperature, as the wettability characteristics at the low reflow temperature range of 230 °C to 240 °C may

be substantial.

Generally, low-Ag solder alloys are relatively inferior in terms of their wettability characteristics compared to near-eutectic Sn-Ag-Cu alloys [23]. This has been the main bottleneck delaying their adoption in various industries. A reduction of the Ag content in the Sn-Ag-Cu ternary composition widens the pasty range and strongly affects the wettability of the solder in the normal soldering temperature range of 240 °C to 250 °C.

Despite wettability being one of the most important properties of solders, there are relatively few studies on enhancing the wettability of low-Ag solder alloys. Wang *et al.* indicated that wetting angles and wetting forces could be enhanced with a suitable addition of rare earth metals [24,25]. However, the times to buoyancy (corrected to a zero value i.e., the zero crossover times) showed a decrease, and the wettability with respect to the soldering temperature was not evaluated. Moreover, earlier research has shown that an addition of indium (In) is very effective for enhancing the zero crossover time as well as the wetting angle and the wetting force [26,27]. However, the amount added should be minimized in an effort to diminish the raw material cost while maintaining the wettability.

This work provides the results of improved wettability acquired in low-Ag and In-containing Sn-1.2Ag-0.5Cu-x(< 1 %). In quaternary compositions as a function of the In con-

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tent at 0.2 wt.% intervals and as a function of the soldering temperature. Microstructural peculiarities and interfacial reactions are also considered. The optimized amount of In added with respect to the wettability is also suggested.

## 2. EXPERIMENTAL PROCEDURES

A wettability test was conducted with several solder alloys using a wetting balance tester (SP-2, Malcom) [23-26,29-31]. Because this technique requires only a modest amount of time, the wetting balance method is one of the most industry-friendly examinations for solderability testing. The zero crossover time and wetting force at 2 s were measured for normally immersed Cu coupons with a width of 3 mm that were coated with a water-soluble (WS) type flux (WF6063M, Senju Metal). The immersion rate, depth, and time were 5 mm/s, 2 mm, and 10 s, respectively. To assess the solderability, the temperature of the solder bath was set to 250 °C, 240 °C, and 230 °C in three cases. Because the peak temperature of specific regions to be soldered may be less than 240 °C during an actual instance of reflow soldering, the wetting characteristics within the relatively low soldering temperature range of 230 °C to 240 °C could be considered as substantially important information.

To evaluate the effects of the In content on the melting temperature/range of Sn-1.2Ag-0.5Cu-xIn quaternary compositions, four solder compositions of Sn-1.2Ag-0.5Cu-0.8In, Sn-1.2Ag-0.5Cu-0.6In, Sn-1.2Ag-0.5Cu-0.4In, and Sn-1.2Ag-0.5Cu-0.2In were prepared and characterized using differential scanning calorimetry (DSC). During the DSC test, the heating rate was 10 °C/min and the maximum temperature was set to 250 °C.

The quaternary solders were first turned into balls 450  $\mu\text{m}$  in diameter and were reflowed on a CSP package. Next, the bumped CSP packages were surface-mounted on a rigid printed circuit board (PCB). The surface finishes of the pads on which the solder balls were located were Au/Ni for the packages and organic solderability preservative (OSP) for the Cu metallization boards. A WS-type flux material was used for the process of solder ball bumping, and Sn-3.0Ag-0.5Cu solder paste (M705-GRN360-K2-V, Senju Metal) was used for the surface mounting of the CSP packages. The reflow oven (1809UL, Heller) used during the bumping and surface mounting was set to 242 °C as the peak temperature.

After the SMT process, an in-depth study of the interfacial reaction was performed. The cross-section of the solder joints including the solder/pad interface was analyzed via an electron probe microanalysis (EPMA) (EPMA-1600, Shimadzu Corporation) and by a scanning electron microscope (SEM) equipped with an energy dispersive spectroscope (EDS) for a metallurgical analysis. In order to take clear-cut SEM images at the interfaces, the solder joints were etched with a  $\text{CH}_3\text{OH}$ -4(vol.%)  $\text{HNO}_3$ -1HCl solution.

## 3. RESULTS AND DISCUSSION

Figure 1 shows the characteristics of the zero crossover time ( $T_0$ ) as a function of the solder composition and soldering temperature. For comparison, Sn-3.0Ag-0.5Cu, Sn-1.2Ag-0.5Cu, and Sn-1.0Ag-0.5Cu ternary alloys were also measured. As the soldering temperature increased, the  $T_0$  values measured in all samples decreased. In the Sn-xAg-0.5Cu ternary system, as the Ag content decreased, the  $T_0$  value increased [23]. As a result, the 1.0 and 1.2Ag cases showed significantly deficient wetting properties in the temperature range of 230 °C to 240 °C. These results indicate that the relatively low peak temperature condition of 230 °C to 240 °C using Sn-1.0~1.2Ag-0.5Cu alloys may result in a low yield in the industrial reflow soldering process. However, with the addition of a proper amount of In into the quaternary solder composition with 1.2 % Ag, the  $T_0$  values were reduced in general, especially in the range of 230 °C to 240 °C. Considering the zero crossover time, the optimum amount of In additive was observed to be 0.6 %, indicating the lowest  $T_0$  value. In the best case, the quaternary alloy presented a lower  $T_0$  value than that of a representative Pb-free composition, Sn-3.0Ag-0.5Cu, in the entire temperature range. When the In content was excessive or deficient with regard to the best value, the  $T_0$  values clearly increased. As a result, the Sn-1.2Ag-0.5Cu-0.8In and Sn-1.2Ag-0.5Cu-0.2In alloys exhibited strikingly high  $T_0$  values in the temperature range of 230 °C to 240 °C.

Figure 2 indicates the wetting force as the test continued to 2 s ( $F_{2\text{sec}}$ ) with respect to the solder composition and temperature. As the soldering temperature decreased, the  $F_{2\text{sec}}$  values of all of the samples decreased, especially in the range of 230 °C to 240 °C. In the Sn-xAg-0.5Cu ternary system, the  $F_{2\text{sec}}$  values decreased with a decrease in the Ag content. In the Sn-1.0Ag-0.5Cu composition, the wetting force after 2 s

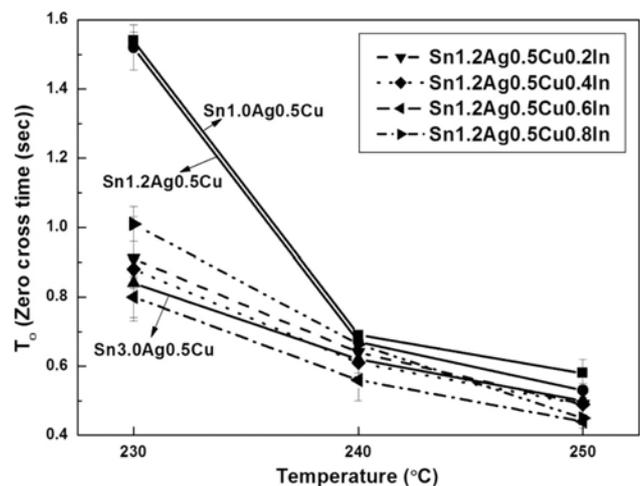


Fig. 1. Average zero cross time values as a function of soldering temperature for several Sn-Ag-Cu-(In) alloys.

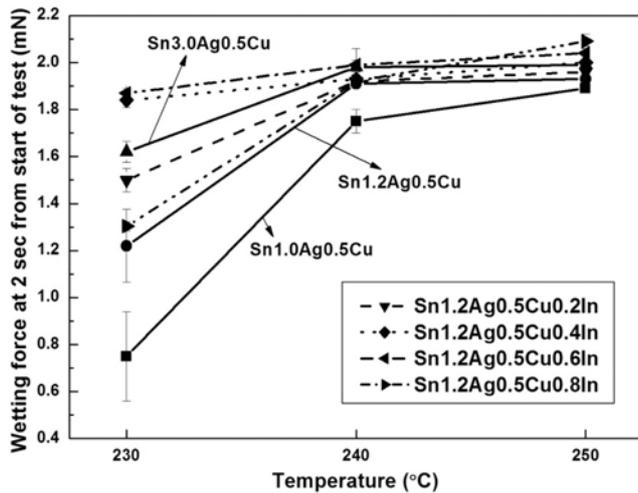


Fig. 2. Average wetting force values at 2 s as a function of soldering temperature for several Sn-Ag-Cu(-In) alloys.

was clearly and significantly reduced in comparison with Sn-3.0Ag-0.5Cu. Moreover, the decrease was more dominant within the solder temperature range of 230 °C to 240 °C. However, in the quaternary solder composition with added In, generally enhanced  $F_{2\text{sec}}$  values were observed. This is especially apparent when the In at 0.6 % was added; the  $F_{2\text{sec}}$  value at 230 °C to 240 °C became maximized in this case. Consequently, the Sn-1.2Ag-0.5Cu-0.4~0.6In composition had outstandingly high wetting force values that exceeded those of Sn-3.0Ag-0.5Cu in the temperature range of 230 °C to 240 °C. According to the results of Figs. 1 and 2, the Ag and In contents were minimized to 1.2 and 0.4% to 0.6%, respectively, with a definite increase in their wettability characteristics. Although the addition of a suitable amount of rare earth metal (REM) has been shown to improve the wetting properties in Sn-Ag-Cu alloys, the improvement is not as dramatic as the present results [24,25].

The melting behaviors of the measured alloys, including the fabricated Sn-1.2Ag-0.5Cu-xIn quaternary alloys, are indicated in Figure 3. The Sn-3.0Ag-0.5Cu alloy, which is near the ternary eutectic composition, showed what was close to a single endothermic peak. The Sn-1.2Ag-0.5Cu-0.2~0.8In alloys exhibited two peaks of bimodal shapes similar to those of the Sn-1.0~1.2Ag-0.5Cu alloys. The lower peak refers to the solidus temperature, whereas the other refers to the liquidus temperature. The increase in the In content shifted the entire endothermic curve to a lower temperature in principle. However, the In addition did not outstandingly reduce the gap between two endothermic peak points; that is, it did not increase the pasty range significantly. These results indicate that compositions made with 0.4 % to 0.8 % In in the Sn-1.2Ag-0.5Cu-xIn composition are still far from the quaternary eutectic composition. To approach the Sn-Ag-Cu-In quaternary eutectic composition of ~196 °C, an In

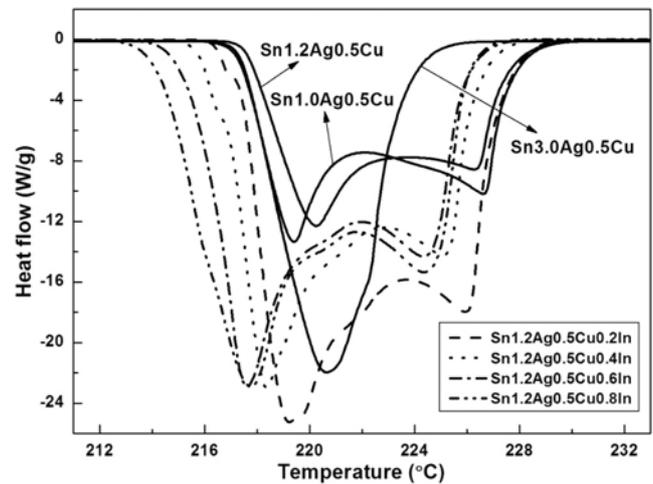


Fig. 3. DSC endothermic curves of several Sn-Ag-Cu(-In) alloys during heating.

addition of 8 % to 10 % is necessary [32].

Combining the above results, we can conclude that In additions at small amounts (0.4 % to 0.6 %) not only decrease the solidus and liquidus temperatures but also enhance the wetting properties, especially at low soldering temperatures in a range of 230 °C to 240 °C. Considering the equivalent soldering temperatures of 230 °C to 250 °C, the low liquidus temperature of the Sn-3.0Ag-0.5Cu ternary composition is evidently the primary cause of the relatively superior wettability. However, the slight decrease in the liquidus temperature in the quaternary composition cannot explain the excellent wettability in Sn-1.2Ag-0.5Cu-0.4~0.6In. Therefore, the excellent wetting properties obtained when a proper amount of In is added may be due to the inherent properties of In itself, such as its surface tension property.

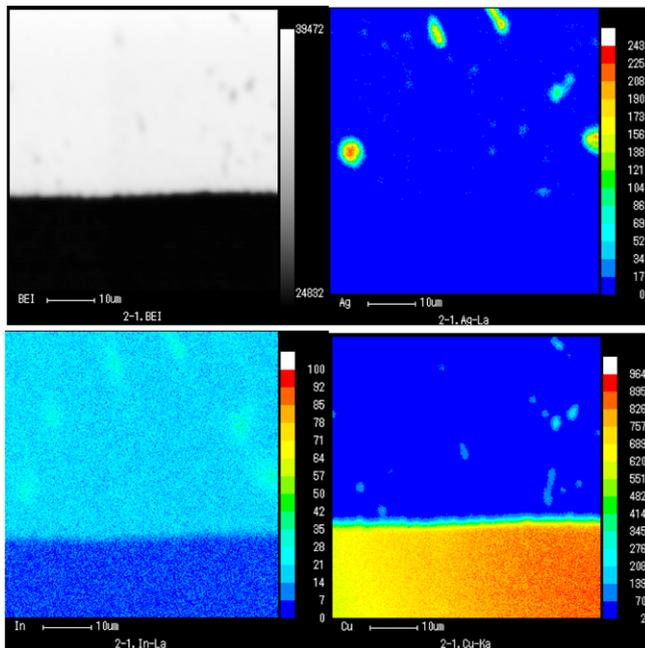
The enhancement in  $T_0$  and  $F_{2\text{sec}}$  in relation to the wetting properties was found to be mainly attributed to the fastest wetting rate and relatively high wetting force depending on the In content. The wetting force value measured in the wetting balance technique can be represented as follows [24]:

$$F_w = \gamma_{LV} P \cos \theta - \rho V g \quad (1)$$

Here,  $F_w$  is the wetting force,  $\gamma_{LV}$  is the surface tension of the solder in contact with the flux,  $P$  is the perimeter of the Cu coupon,  $\theta$  is the wetting angle,  $\rho$  is the density of the solder,  $V$  is the immersed volume, and  $g$  is the gravity acceleration constant. Between the two Sn-Ag-Cu alloys, the density of Sn-1.0Ag-0.5Cu is estimated to be lower than that of Sn-3.0Ag-0.5Cu due to the higher density value of Ag. The lower density may serve to increase the wetting force according to Eq. 1. Nevertheless, the lower surface tension and  $\cos \theta$  values in Sn-1.0Ag-0.5Cu decrease the wetting force more dominantly [33]; consequently, a lower wetting force value is obtained. Among the Sn-Ag-Cu-In and Sn-3.0Ag-0.5Cu

alloys, the lower density of Sn-Ag-Cu-In is estimated to increase the wetting force, whereas the slightly lower surface tension of Sn-Ag-Cu-In alloys may minutely cause a decrease in the wetting force [27,33]. However, the lower final wetting angle measured in Sn-Ag-Cu-In effectively serves to increase the wetting force [27]. Consequently, the higher wetting force values of the Sn-Ag-Cu-In quaternary alloys shown in Figure 2 were found to be mainly influenced by the enhanced wetting force in addition to the faster wetting rate, especially at the low soldering temperature range of 230 °C to 240 °C. In content levels exceeding 0.6 % and 0.8 % did not enhance the wetting properties. Too much In additive will clearly reduce the surface tension of the molten quaternary solder alloys or increase the density and contact angle [27,33,34], resulting in a decrease in the wetting force according to Eq. 1. For example, the isothermal surface tension and density values of Sn-3.13Ag-0.74Cu + x (< 1 %) In alloys at 250 °C have a wavy shape with respect to the In content [27]. Therefore, the highest wetting force value can be attained from the optimum combination of the surface tension, density, and contact angle with molten quaternary alloys. In the future, a more detailed discussion and numerical consideration using CALPHAD and thermodynamic calculation is necessary.

Figure 4 shows EPMA mapping images at the Sn-1.2Ag-0.5Cu-0.4In/Cu interface as a function of the element characteristics after the SMT process. The phase of the intermetallic compound (IMC) layer at the board side was identified

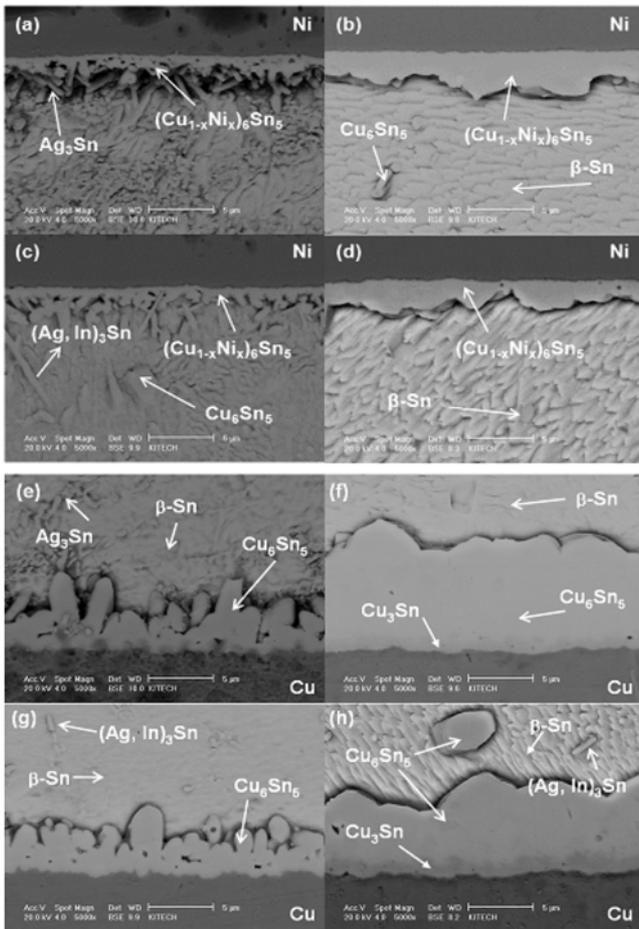


**Fig. 4.** EPMA mapping images at the Sn-1.2Ag-0.5Cu-0.4In/Cu interface as a function of element: micrographs indicate backscatter electron image (BEI), Ag mapping image, In mapping image, and Cu mapping image, respectively.

as  $\text{Cu}_6\text{Sn}_5$ . EPMA analysis showed that a trace of In element was not detected on the package and board-side interfacial IMC layers but was detected where Ag was detected in the solder matrix. In previous research with respect to Sn-3.5Ag-0.5Cu-9In composition having a high In content, it was reported that the added In participates in the formation of an IMC-like Sn-Cu-Ni-In or Sn-In-Au on Au/NiP pad finish [35]. Thus, the Sn-1.2Ag-0.5Cu-xIn alloys having a low In content in this study showed different interfacial reaction characteristics in comparison with Sn-3.5Ag-0.5Cu-9In, which has a high In content. Consequently, it was found via an EDS analysis that most of the added In element exists as  $\text{Ag}_3(\text{Sn}, \text{In})$  phases in the microstructure [35,36]. When a microstructural investigation of the Sn-3.5Ag-3In composition was performed, In was detected in Sn dendrites or Sn phases within fine eutectics and in the  $\text{Ag}_3(\text{Sn}, \text{In})$  phases [36]. Thus, this result differed slightly from the microstructural results of the Sn-3.5Ag-3In composition. The behavior of the In element is unlikely to exert any detrimental effect on the mechanical drop/shock reliability in that additional precipitates in a matrix or reaction layer at the interface did not form [19,37,38].

Figure 5 shows the cross-sectional microstructures of the main solder joints using Sn-3.0Ag-0.5Cu and Sn-1.2Ag-0.5Cu-0.4In. The IMC structure at the solder/Ni interface after as-mounting or 1000 h aging at 150 °C was found to be the  $(\text{Cu}_{1-x}\text{Ni}_x)_6\text{Sn}_5$  phase by measuring it using EDS [39]. The Cu existing in the IMC layer resulted from the Cu provision from the solder composition. In addition, the IMC structure at the solder/Cu interface after aging for 1000 h was found to be the  $\text{Cu}_6\text{Sn}_5/\text{Cu}_3\text{Sn}$  phase. The package or board-side IMC layers in the Sn-1.2Ag-0.5Cu-0.4In joints were visibly thinner than those of Sn-3.0Ag-0.5Cu, especially after the aging process. Although the aging temperature was held constant at 150 °C, the effectiveness of the diffusion should be different as a function of the liquidus temperatures. The Sn-1.2Ag-0.5Cu-0.4In showed an increased liquidus temperature in comparison with Sn-3.0Ag-0.5Cu, as indicated in Figure 3. Therefore, a small diffusion coefficient (D) value for the Sn-1.2Ag-0.5Cu-0.4In composition may be feasible [23]. Given that thin IMC layers are also one of the most crucial factors for the enhancement of the mechanical drop/shock reliability [19,37,38,40], the interfacial microstructures of Sn-1.2Ag-0.5Cu-0.4In joints are highly favorable.

The performance of solder joints used in modern electronic products simultaneously demands thermal fatigue and mechanical drop/shock reliability [18]. It now appears clear that low-Ag content solder alloys improve the drop/shock performance over near-eutectic alloys such as Sn-3.0Ag-0.5Cu [19,37,41]. Because its microstructure is similar to that of low-Ag solder alloys, the findings of this study show that Sn-1.2Ag-0.5Cu-0.4In joints offer improved drop/shock



**Fig. 5.** Cross-sectional SEM images at the interface of solder joints ( $\times 5000$ ): Sn-3.0Ag-0.5Cu composition joints at the package-side interface after (a) as-reflow and (b) 1000 h aging at 150 °C, Sn-1.2Ag-0.5Cu-0.4In composition joints at the package-side interface after (c) as-reflow and (d) 1000 h aging at 150 °C, Sn-3.0Ag-0.5Cu composition joints at the board-side interface after (e) as-reflow and (f) 1000 h aging at 150 °C, Sn-1.2Ag-0.5Cu-0.4In composition joints at the board-side interface after (g) as-reflow and (h) 1000 h aging at 150 °C.

performance comparable to that of other low-Ag solder alloys [42]. The reduced thermal fatigue reliability in low-Ag content solder alloys can also be improved by a minor increase in the Cu content or with additional micro-alloying [8,18,43]. Therefore, Sn-1.2Ag-xCu-0.4In-based compositions can be considered as a strong candidate for low-Ag Pb-free solders.

#### 4. CONCLUSIONS

From experiments that assessed the In content and soldering temperature, it was found that the low-Ag quaternary Sn-1.2Ag-0.5Cu-0.4~0.6In composition showed outstandingly improved wettability characteristics in the low soldering temperature range of 230 °C to 240 °C. The added In element did not participate in the interfacial reaction with a Cu

or Ni pad after as-reflow or aging processes at 150 °C. The Sn-1.2Ag-0.5Cu-0.4In composition also demonstrated an excellent interfacial microstructure with respect to the intermetallic thickness at the obtained joint, especially after aging. Given that the addition content of In element is negligible, the increase in the raw material cost is also limited. Thus, Sn-1.2Ag-xCu-0.4In-based compositions can be considered as a strong candidate for low-Ag Pb-free solders.

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