

EFFECT OF DIPPING SOLDERING ON THICKNESS AND INTERMETALLIC COMPOUND MORPHOLOGY BETWEEN Sn-3.0Cu LEAD-FREE SOLDER AND COPPER SUBSTRATE

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Abstract

The thickness and morphology of intermetallic compounds formed between Sn-3.0Cu lead-free solder and the copper substrate were investigated. In the experiment, the dipping soldering was performed at 350°C using Sn-3.0Cu lead-free solder for various dipping time lengths of 5, 10, 20, 50, 75 and 100s, respectively. The results indicated that η -Cu₆Sn₅ intermetallic phase was found to exist between the lead-free solder and the copper substrate. The formation of an ε -Cu₃Sn particles on the η -Cu₆Sn₅ surface and grain boundary was found only when the substrate was soldered for 75 and 100s. The long soldering time and formation of ε -Cu₃Sn particles resulted in the morphology of the η -Cu₆Sn₅ IMC changed from scallop shape to more round. Nevertheless, the thickness and morphology of intermetallic compounds was also found to be influenced by the diffusion controlled process.

Keywords: Dipping soldering, Sn-3.0Cu solder, Intermetallic compound (IMC), Morphology.

Introduction

Traditional tin-lead (Sn-Pb) solder is commonly used as an interconnector in electronic packaging and electronic devices because of low-melting temperature, and good mechanical properties (Liu *et al.*, 2000). It could be performed with several soldering methods including reflow, wave, and hand soldering (Forstén *et al.*, 2000). The low-melting temperature of the solder reduced thermal damage to the electronic components (Zhao *et al.*, 2019). However, the toxicity of Pb in the solder alloy has

pushed development of lead-free alternatives. A widely used alternative is Sn-base solder, and vigorous effort has been underway in recent years to develop lead-free solders for high-temperature applications. A successful solder should also satisfy factors such as reliability, lifetime, cost effectiveness, and recyclability as well as the primary goal of a non-toxic solder (Gayle *et al.*, 2001).

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Overall, binary alloys are simple system for lead-free solder manufacturing, such as the Sn-Ag, Sn-Sb, and Sn-Cu (Gayle *et al.*, 2001; El-bediwi and El-bahay, 2004; Zhao *et al.*, 2019). The Sn-5.0 Ag solder has a melting point of 220.6°C and the solder displays a high elastic modulus and good solderability as compared with Sn-Pb eutectic solder alloy. However, the use of Ag, a precious metal, makes the cost relatively high (El-bediwi and El-bahay, 2004). Tomlinson and Bryan (Tomlinson and Bryan, 1986), used wave soldering with high-temperature antimonial solder containing up to 4.0-10.0% Sb at 300°C. The fracture through the intermetallic layer or via Sn-Sb precipitates in the solder. This was due to the high hardness value of the intermetallic compound (IMC) layer. Another useful system is Sn-Cu solder alloy which is one of the main lead-free solders materials used in electronics due to its excellent performance and low cost (Zhao *et al.*, 2019). Investigating reflow soldering of Sn-2.0Cu solder on Cu substrate at 360°C showed good wetting properties. In addition, a larger η -Cu₆Sn₅ phase was found in the solder matrix. However, the ϵ -Cu₃Sn phase has not been found in the samples (Zhao *et al.*, 2009). It is similar to the Sn-3.0Cu/Cu joint reflow soldered at 260°C, for 15 min. The failure of the joint samples mainly occurred in the IMC layer (Zou and Zhang, 2010).

Thus, a deeper knowledge of the formation and growth of interfacial IMCs is important to developing reliable solder joints (Bang *et al.*, 2019). In addition, the growth of IMC grains between Cu and solder interface is mainly determined by diffusion phenomenon including lattice diffusion, grain boundary diffusion and surface diffusion. (Yung *et al.*, 2018; Zhao *et al.*, 2018) Based on literature reviews, numerous studies regarding the reflow and wave soldering of the Sn-based solders at moderate and high temperatures were disclosed. However, research on dip soldering of Sn-3.0Cu solder used as a substrate in electronic packaging at high temperature has been limited. Consequently, this study investigates the effect of soldering on the thickness and the IMC morphology between solder and copper substrate by the dip soldering process at high temperatures of 350°C.

Materials and Methods

In this research, Sn-3.0Cu wt% commercial lead-free solder, a high temperature solder, from “Koki Products Co., Ltd.” was used. The substrate was pure copper foil (99.99% Cu JIS H3100 grade) with a thickness of 0.2 mm and cut to 10×20 mm size. The surface was cleaned in an HCl solution, followed by rinsing with ethanol and then dipped

into flux. The flux was of Ultracore’s RC-15SH RMA (15%) type. Prior to soldering, the solder wire was melted in a stainless steel mold using a hot plate. The dip soldering process was carried out at 350°C for 5, 10, 20, 50, 75, and 100 s, and cooled at ambient temperature. (Handwerker, 2002). The post-soldered samples were subsequently etched with ethanol to remove remaining flux.

To observe the morphology and grain size of IMC (in top-view of IMC), specimens were etched by dipping in a 10% nitric acid solution to remove the solder on the top of IMC. The etched specimens were subsequently cleaned with ethanol to remove residual nitric acid. To examine the interfacial microstructure and IMC thickness (in cross-section of specimens), the post-soldered specimens were individually mounted onto resin and their microstructures were determined. In addition, the specimens were polished using a series of SiC grit paper of sizes 600, 1,000 and 2,000, followed by Al₂O₃ with sizes of 0.3 and 0.1 μ m. The polished specimens were then Ar-ion milled (Hitachi, IM 4,000 Plus). It was operated at 4.0 kV acceleration voltage, 1.5 kV discharge voltage, 70° ion beam irradiation angle, 0.1 cm³/min gas flow, and 20-minute milling time. The Jeol JSM-5800LV scanning electron microscopy (SEM) was used to verify the morphology and intermetallic phases. The chemical composition of the phases was determined using the Oxford Instruments X-Max energy dispersive spectroscopy (EDS). An X-ray diffractometer (Panalytical, Aeris 600W) was used to identify the crystalline structure. The top-view and cross-section of the intermetallic compounds were identified based on the 2000x SEM images. The grain size numbers (G) according to ASTM E 112-13 standard and the thicknesses of the intermetallic compounds were identified using Olympus Stream Image Analysis software, respectively.

Results and Discussion

Microstructure and surface roundness of the substrates

The microstructure and roughness profile of the copper substrate is illustrated in Figure 1. The grain size numbers (G) of the substrates were determined using the ASTM E112-10 intercept method. The G value can be calculated by (Standard, 1996):

$$G = (6.643856 \log \overline{N}_L) - 3.288 \quad (1)$$

Where G is the grain size number and $\overline{N}_L$ is the number of grain intersections per unit length.

The substrate exhibited a G value of 8.40 for the surface microstructure and was classified as fine grain structure. The surface roughness the substrate was measured in terms of arithmetic mean roughness (R_a). In Figure 1, the 3D surface and roughness profile of substrate with examination length of $256 \times 257 \mu\text{m}$ is shown, and the R_a of the substrate was $0.182 \mu\text{m}$.

IMC thickness

A nucleus of intermetallic phase quickly forms between the substrate and the solder during the soldering reaction. The $\eta\text{-Cu}_6\text{Sn}_5$ phase formation between solder and substrate of each soldering samples is shown in Figure 2. Voids were frequently found at the $\text{Cu}/\varepsilon\text{-Cu}_3\text{Sn}$ interface during extended thermal soldering. This was due to a larger diffusion flux of Cu than that of Sn resulting in vacancy formation (Chia *et al.*, 2016; Chen, 2020). However, Kirkendall effects combined with impurities within the substrate may cause voids that mainly located at $\text{Cu}/\varepsilon\text{-Cu}_3\text{Sn}$ interface. Larger voids located at $\eta\text{-Cu}_6\text{Sn}_5$ were formed during thermal processes as well (Chen, 2020). In this research, the formation of voids is governed by mechanisms of diffusion throughout the soldering time. For instance, for the

soldering time from 50 to 100 s, the density of voids was considerably high.

Whose elemental composition was subsequently determined using the EDS analysis and illustrated in Figure 3, and presents the chemical composition of IMC at the various soldering time intervals. The findings indicated that Cu contents of the $\eta\text{-Cu}_6\text{Sn}_5$ phase range from 36.44 wt.% to 37.68 wt.%. The elemental content of the IMC phases was consistent with the results of previous research (Hu *et al.*, 2012; Dele-Afolabi *et al.*, 2020). XRD analysis was shown in Figure 4. The lattice structure, lattice parameters of $\alpha\text{-Cu}$, $\beta\text{-Sn}$ and intermetallic phases were shown in Table 1. However, $\varepsilon\text{-Cu}_3\text{Sn}$ was found in solder matrix as shown in Figure 5. The small $\varepsilon\text{-Cu}_3\text{Sn}$ phases distributed in a matrix of Sn-rich phase.

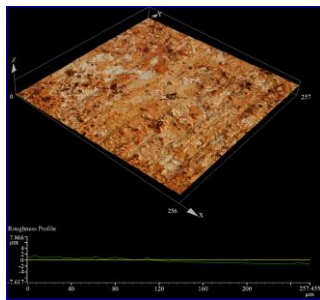


Figure 1. 3D Microstructure and roughness profile of copper substrate.

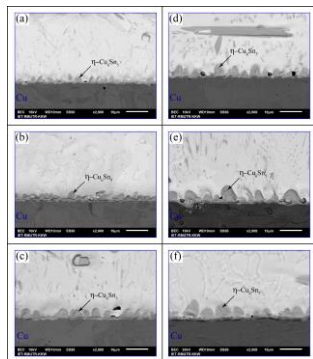


Figure 2. Intermetallic compounds formation between solder and copper substrate; (a) 5 s (b) 10 s, (c) 20 s, (d) 50 s, (e) 75 s, and (f) 100 s.

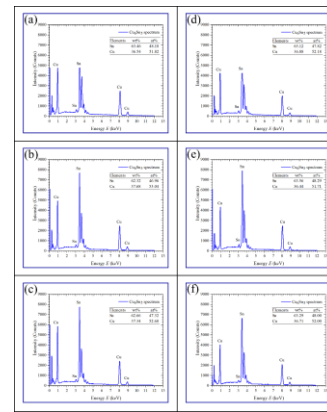


Figure 3. EDS pattern of the $\eta\text{-Cu}_6\text{Sn}_5$; (a) 5 s (b) 10 s, (c) 20 s, (d) 50 s, (e) 75 s, and (f) 100 s.

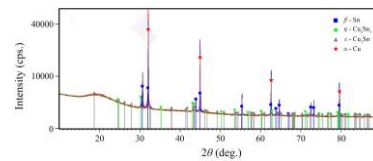


Figure 4. XRD patterns with soldering of 100 s.

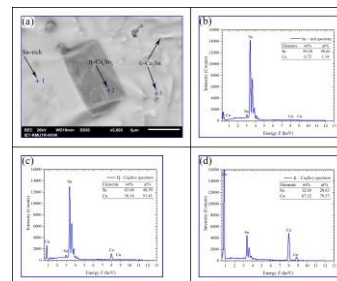


Figure 5. IMCs in solder matrix; (a) SEM image, (b) Sn-rich phase, (c) $\eta\text{-Cu}_6\text{Sn}_5$, and (d) $\varepsilon\text{-Cu}_3\text{Sn}$.

Table 1. Results of XRD analyses.

Phase	Crystal Structure	Space group (No.)	Lattice parameters (Å)
α -Cu	FCC	Fm-3m (225)	a = b = c = 3.6080
β -Sn	BCT	I41/amd (141)	a = 5.8320, b = 5.8320, c = 3.1820
η -Cu ₆ Sn ₅	Hexagonal	P63mc(186)	a = 4.1900 b = 4.1900, c = 5.0860
ϵ -Cu ₃ Sn	Orthorhombic	Pmmn (59)	a = 4.3200, b = 5.9000, c = 4.7400

The formation of the IMC will consume the substrate elements which have dissolved in the solder, this allows more substrate material to diffuse into the molten solder. The IMC grain will gradually cover the surface of the substrate and decrease the reaction rate (Zhang, 2015). The thickness of η -Cu₆Sn₅ layer found at the interface was measured after dip soldering. As illustrated in Figure 6, and Equation (2) the thickness of a particular η -Cu₆Sn₅ layer (h) was measured by summing the area of the η -Cu₆Sn₅ layer (A_m) which was then divided by the measured length (L).

$$h = \frac{A_m}{L} \quad (2)$$

Figure 7 show the relation between η -Cu₆Sn₅ thickness and soldering time interval. It was found that the thickness of IMC is increased with increasing soldering time. The relationship between η -Cu₆Sn₅ thickness and soldering time as follows (Bang *et al.*, 2019; Tang *et al.*, 2019; Li *et al.*, 2020):

$$h = kt^n \quad (3)$$

$$\ln h = \ln k + n \ln t \quad (4)$$

Where h is the thickness of η -Cu₆Sn₅, k is a growth constant, t is the dipping time and n is the growth exponent of the IMC layer

The linear plot with Equations (4) for the experimental data show in Figure 8. The results show that the n value of η -Cu₆Sn₅ with the soldering time range 5-50 s were equaled to 0.1033, and then after 75 s the n value was about 0.6575. The growth rate of the intermetallic layer can be determined by the n value of the IMC layer. The relationship between the n value and reaction controlled could be classified as “grain boundary diffusion-controlled” for n = 1/3, “volume diffusion-controlled” for n = 1/2 or 0.5, and “chemical reaction-controlled” for n = 1 (Lee and Kim, 2002; Park and Arróyave, 2012; Yao *et al.*, 2016). Thus, in this research the growth of intermetallic layer is governed by both mechanisms of grain boundary diffusion and volume diffusion and throughout the soldering. The

n value of 0.6575 for the longer soldering time, a phenomenon which was attributable to the growth rate governed by volume diffusion controls. The experimental results were compared to the previous research as presented in Table 2. The n value is consistent with other reports on Sn and Sn-3.5Ag

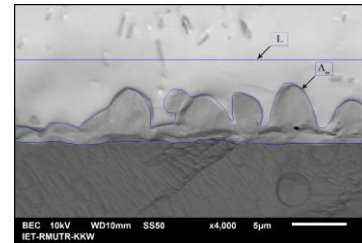
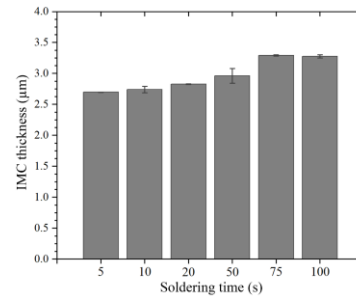
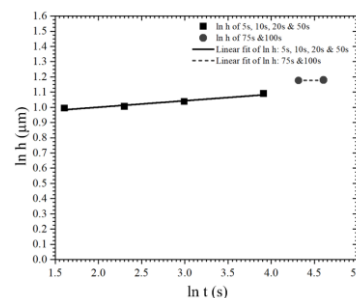
**Figure 6. Thickness measurement of η -Cu₆Sn₅ layer soldered at 20 s.****Figure 7. Relationship of η -Cu₆Sn₅ thickness and soldering time interval.****Figure 8. Linear plotted results of η -Cu₆Sn₅ thickness and soldering time.**

Table 2. Comparison of n values of this work and previous research.

Solder	Soldering method	Soldering time (s)	Temperature (°C)	IMC type	n	Ref.
Sn-3.0Cu	Dipping	5-50	350	η -Cu ₆ Sn ₅	0.1033	This work
		75-100			0.6575	
Sn	Dipping	5-60	300	η -Cu ₆ Sn ₅	0.1000	(Faizan, 2015)
		60-360			0.6700	
Sn-3.5Ag	Dipping	5-60	300	η -Cu ₆ Sn ₅	0.1000	
		60-360			0.6300	
Sn-2.0Ag	Reflow	1-240	350	Ni ₃ Sn ₄	0.34800	(Taneja, 2018)
			300	Ni ₃ Sn ₄	0.3590	
			250	Ni ₃ Sn ₄	0.3930	
			230	Ni ₃ Sn ₄	0.3990	
Sn-0.7Cu	Reflow	1-10	250	η -Cu ₆ Sn ₅	0.0700	(Mohd Salleh <i>et al.</i> , 2015)
		10-50			0.3100	
Sn-3.0Cu	Reflow	10-480	350	η -Cu ₆ Sn ₅	0.4078	(Mookam <i>et al.</i> , 2018)

solders (Faizan, 2015). The IMC layers initially grow rapidly due to the fast mass flux through the η -Cu₆Sn₅ grain boundary by dip soldering. After that, the growth rate of the IMC layers slows due to the loss of fast mass flux channels. As the soldering time increased (i.e. from 75 to 100 s), the n values increased to be greater than with Sn-3.5Ag, but lower than a pure Sn, and doubled in comparison to reflow soldering. Thus, a higher n value which indicates that more mass flux through the grain boundary dominates the increasing in the thickness of the η -Cu₆Sn₅ layer (Bang *et al.*, 2019).

The morphology should also be considered, indicating the uniformity of intermetallic layer, and it can be expressed as the Equation (5) (Tsukamoto *et al.*, 2011):

$$M = \frac{L_R}{L_l} \quad (5)$$

Where M is the morphology parameter, L_R is the perimeter of the real η -Cu₆Sn₅ layer, and L_l is the perimeter of the equivalent rectangular η -Cu₆Sn₅ layer.

A higher value of the morphology parameter means rougher and more uneven morphology of the IMC layers. By comparing the soldering results for 5, 10, 20, 50, 75 to 100 s, the average value of morphology parameters of the IMC layers equalled 0.685, 0.586, 0.567, 0.557, 0.415 and 0.364, respectively. The morphology parameter value of intermetallic compounds decreased when the soldering time increased. This result implied that the soldering was controlled by diffusion. However, differences have been reported for reflow soldering by Tsukamoto *et al.* (Tsukamoto *et al.*, 2011), the morphology of the IMC layers become rougher with increasing number of reflows. In addition, a more uniform intermetallic layer (lower morphology parameter) reduces the interfacial energy (Kanlayasiri and Sukpimai,

2016), and suppressed growth of the lateral IMC (Shang *et al.*, 2018). However, limitations of all the variables were estimated from the SEM cross-sectional micrographs may not be the real representative of the morphology parameter value.

Morphology and Grain size of IMC

The top-view of the IMC at the various soldering time intervals was observed using the SEM operated at 20 kV, as shown in Figure 9. The morphology of the IMC phase formed between solder joints was scallop-type and rough-type. It indicated that with short soldering time (5s), randomly distributed small and larger grain sizes were observed. This is due to the lower diffusion of Sn and Cu atoms at the interface and the subsequent formation of the IMC. After soldering time for 10, 20, and 50 s, the morphology becomes uniform with large grain size. The scallop grains presence was found to be positively correlated to the soldering times. Therefore, the round shape under the long dip time condition was better than the short dip times.

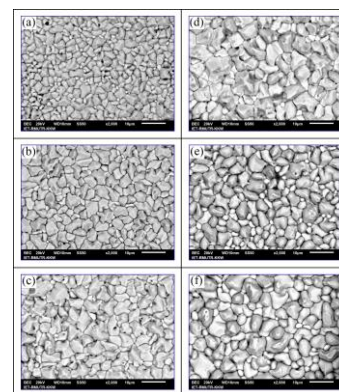


Figure 9. Top-view of the intermetallic compound at the various soldering time intervals; (a) 5 s (b) 10 s, (c) 20 s, (d) 50 s, (e) 75 s, and (f) 100 s.

This is because of the ripening process. The smaller IMC grains dissolve into solder and contribute to further growth of neighboring grains (Yu *et al.*, 2005).

The coarsening and growth of grains decreased the formation of IMC with increasing dip soldering time due to the dipping times effect on the diffusion, the increase of dipping time led to the increased diffusion of Sn and Cu atoms. However, as the soldering time increased (i.e. from 75 to 100 s), the grain morphology had significantly changed from uniform to random distribution. This was due to the IMC grains providing a relative barrier to each other on lateral expansion. Thus, the growth of IMC grain with the vertical length was faster growth rate than the lateral rate (Kunwar *et al.*, 2020). In addition, dipping for both 75 and 100 s causes a morphology transition from scallop-type to rough-typed. This can be explained by the limited supply of Cu atoms for growth of η -Cu₆Sn₅ IMC, so grain growth was impeded during the longer soldering times. In contrast, the Cu atoms from the substrate passed through the η -Cu₆Sn₅ layers and reacted with the Sn atoms from the solder, which resulted in the sufficient supply of Cu atoms for the formation of ϵ -Cu₃Sn particles. The small ϵ -Cu₃Sn particles were found on the η -Cu₆Sn₅ surface and grain boundary, as illustrated in Figure 10 and indicated by arrows. It is possible with longer soldering time, a new intermetallic phase (ϵ -Cu₃Sn) was formed at the grain boundary of η -Cu₆Sn₅ and on the surface of the η -Cu₆Sn₅ grains. The preceding η -Cu₆Sn₅ layer could be decomposed into ϵ -Cu₃Sn and Sn, the chemical reaction can be represented by η -Cu₆Sn₅ $\Rightarrow$ 2(ϵ -Cu₃Sn)+3Sn (Paul and Divinski, 2017). Thus, new ϵ -Cu₃Sn particles shield the growth of IMC by reducing the surface energy of IMC grain and blocking the annexation between the adjacent η -Cu₆Sn₅ grains. The Cu atoms in substrate can diffuse to the grain boundaries promoting the longitudinal growth of the IMC layer (Huang *et al.*, 2019). The IMC grains continued a longitudinal growth resulting in open channels between the rough-typed IMCs as show in Figure 11.

The grain size number based on the ASTM E112-10 standard was examined using Jeffries planimetric method or Jeffries via the Olympus Stream software. Figure 12, depicts the G value of IMC under the various time conditions. Results indicated that the random distribution of grain size with average grain size number for different soldering times of 5, 10, 20, 50, 75 and 100 seconds, given the G of 14.75, 14.53, 14.23, 14.13, 14.74 and 14.31, respectively (mean grain size = 2.05, 2.27, 2.57, 2.67, 2.06, 2.49 μ m). The growth of intermetallic compound of the short soldering times (5, 10, 20, 50 s) was lower than that of the

longer soldering times (75, 100 s). Among various dipping substrates, as soldering time increases, the G values of intermetallic grains were found to decreased with the increase in dipping time.

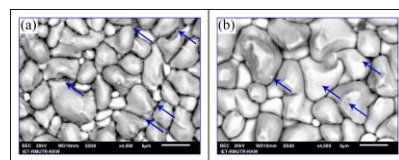


Figure 10. Top-view of the intermetallic compound at (a) 75 and (b) 100 s.

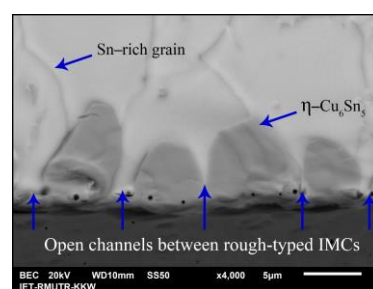


Figure 11. Open channels between the rough-typed IMCs.

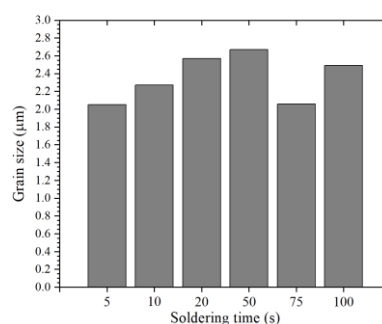


Figure 12. Grain size number at the various soldering time intervals.

Finally, to examine the distribution of grain sizes, an area of 3072 μ m² was analyzed as illustrated in Figure 13 and Figure 14. The comparison of grain size was represented by red, crimson, and grey colors. The colors of each figure were related to the grain size number. A majority of grains with the G value below 12 (5.6 μ m) was red. Crimson grain had a G value of 12-15, while grey had a G value greater than 15 (1.8 μ m).

Table 3 presents the relationship between the mean grain area and the number of grains obtained in this study, significant difference was observed in the morphologies of IMC grains formed during soldering. The IMC grain sizes were 2.05-2.67 μ m after soldering for 5-50 s. However, the grain size

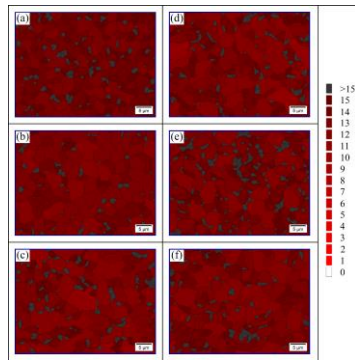


Figure 13. Distribution and grain size number at the various soldering time intervals; (a) 5 s (b) 10 s, (c) 20 s, (d) 50 s, (e) 75 s, and (f) 100 s.

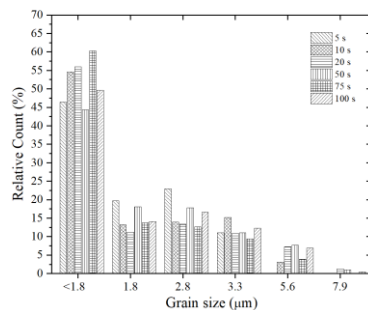


Figure 14. Distribution of grain size at the various soldering time intervals.

exhibited apparent change after long soldering time. The grain size decreased to 2.06 μm after soldering time for 75s. However, when reaction time increased to 100s the grain size was larger due to the decreased number of IMC grains (IMC microstructure coarsening). It meant that the thickness of the IMC layer increases (Park and Arroyave, 2010). It was clear that the thickness and morphology evaluations of the IMC from this research depend on the dip soldering reaction and subsequent diffusion mechanism control IMC between solder and substrate. As the soldering time

increased, the thickness of intermetallic compound increased, implying that the soldering was governed by diffusion controlled process.

Conclusions

This research has investigated the effect of dip soldering on thickness and morphology of IMC between Sn-3.0Cu lead-free solder and copper substrate. The dipping time significantly modified the thickness and morphology of IMC. $\eta\text{-Cu}_6\text{Sn}_5$ was found at the interface between the solder and the substrate. It was also found that the thickness of the intermetallic compound at the interface increased with increasing the dipping time. Furthermore, the formation of $\varepsilon\text{-Cu}_3\text{Sn}$ particles on the $\eta\text{-Cu}_6\text{Sn}_5$ surface and grain boundary was shown following longer soldering time lengths of 75 and 100 s. The morphology of the $\eta\text{-Cu}_6\text{Sn}_5$ changed from a scallop shape to more round, implying that the mechanisms of IMC changed from grain boundary diffusion to volume diffusion control. Thus, the diffusion control of soldering would be a fundamental phenomenon for controlling the thickness and the morphology of IMC within the soldering process.

Acknowledgments

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Conflicts of interest

The authors declare that there is no conflicts of interest regarding the publication of this article.

Table 3. The IMC grain attribute for various soldering time intervals.

Soldering time (s)	Grain size number (G)	Average grain size (μm)	Total Number of grains
5	14.75	2.05	554
10	14.53	2.27	447
20	14.23	2.57	375
50	14.13	2.67	298
75	14.74	2.06	554
100	14.31	2.49	391

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