Effect of the nickel coated precipitated calcium carbonate addition on microstructure, phase and wettability of Sn-9Zn solder

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Abstract. This paper presents the effect of nickel-coated precipitated calcium carbonate (Ni-coated PCC) on the wettability, microstructure and phase change of Sn-9Zn solder. The microstructure and phase analysis of the material plays a very important role as it determines the physical and mechanical behavior of materials. Both Sn-9Zn and Ni-coated PCC/Sn-9Zn produced almost similar microstructure, which contain needle-like, dark phase Zn-rich phase and distributed evenly in beta-Sn rich matrix. However, compared to pure Sn-9Zn, the addition of Ni-coated PCC produce finer size of Zn rich phase needle-like structure. The presence of new phases of nickel tin and calcium carbonate compounds was also found with the addition of Ni-coated PCC. This believed to help in refining the size of Zn-rich needle structure, while retaining the original phases of Sn and Zn of the Sn-9Zn solder, Furthermore, Ni-coated PCC/Sn-9Zn have a smaller contact angle compared to pure Sn-9Zn. This can be explained that, Ni-coated PCC reinforced particle was the interfacial active element and decrease the interfacial energy between the solid and liquid. The microstructure and phase analysis of the material plays a very important role as it determines the wettability behavior of the material.

1 Introduction

The tin-lead (Sn-Pb) solders are one of the most utilized solders in history due to its low melting temperature, eutectic melting characteristics, good mechanical properties and low cost of raw materials [1]. However, Pb was identified as one of the harmful elements that posed serious toxicity threat to the environment and humans. This had lead the search to find alternative candidate to replace the Sn-Pb solders. Tin-based alloys such as tin-copper (Sn-Cu), tin-silver (Sn-Ag), tin-bismuth (Sn-Bi) and tin-zinc (Sn-Zn) were globally investigated to be used as the new Pb-free solder. The Sn-9Zn alloy was reported to show very promising properties such as excellent mechanical properties and also melts at low melting temperature of 199 °C [2]. This allows the use of this solder in the existing production lines that was tuned for Sn-Pb solders without major modifications. Besides that, at its eutectic composition, this alloy also melts at single melting temperature which offers good advantage during solidification process [3].

However, continues development of new soldering material was necessary order to produce more advanced electronic devices. One key requirement of new soldering materials is ensuring the high reliability in various challenging operating environments, including harsh and corrosive environment. Since the last decade, it was widely discovered that the presence of Zn in Sn-9Zn significantly contributed to the low corrosion resistance of this alloy [4]. Since the solubility limit of Zn in Sn is low, no new compound was produced. This allows Zn to preferentially dissolved from the alloy during interaction with moisture or aggressive ions during applications [5]. To solve this issue, various methods have been implemented including micro-alloying process with several alloying elements [6, 7], with successful rate varied to several extent. The inclusion of ceramic materials to produce composite solders has received a lot of attention because of its ability to improve the reliability of solders, mostly through the microstructure refinement.

Ceramic materials such as graphene, reduced graphene oxide, alumina and nanoparticles were widely investigated. Recently precipitated calcium carbonate (PCC) has attracted the attention of researcher as the shape, size and texture of his material can be easily be tailored during synthesis [8]. Yet, the full application of composite solders was still plagued by agglomeration issues of the reinforcement [9-11]. Stark contrast in its density compared to solder introduce complication in production processes, and the properties of the composite solder [12]. This leads to severe phase segregation and non-uniform
dispersion during the reflow soldering [13]. To solve this problem, bridging between the reinforcement and the solder matrix should be made. Nickel (Ni) coating is seen as a possible bridging material suitable to be used in composite solder as Ni is widely known to produce intermetallic compound with Sn. Besides that, the use of Ni coating on silicon carbide was found to enhance the Sn-Ag-Cu solder performance [11]. However, the combination of Ni-coated PCC looks promising to improve Sn-9Zn alloy as soldering materials, but not yet to be explored. In this study, we attempt to investigate the effect of adding 0.5 wt % Ni-coated PCC on the microstructure, phase and wetting angle of Sn-9Zn solder.

2 Experimental Procedure

Calcium carbonate (CaCO3), hydrated nickel chloride (NiCl2·H2O), and hypophosphite (NaH2PO2) were weighed accordingly to 1:1:1 ratio, and added into water with the temperature maintained at 80 °C and stirred at 400 rounds per minute (rpm). Next, the mixture stirred for 2 hours. Once cooled, the solution needs to be filtered to obtain the Ni-coated PCC. The residue was dried in the oven overnight at 70 °C. Lastly, the dried residue was crushed into a fine powder using a mortar and pestle.

In this study, Sn-9Zn solder was used as the solder base matrix material. To fabricate the Sn-9Zn-Ni-PCC composite solder, 0.5 wt. % of Ni-PCC particles were added into the Sn-9Zn solder and heated in a graphite crucible at 550 °C, under inert atmosphere. The molten composite solder was cast and air-cooled to room temperature.

The produced composite solder was sent for phase and microstructure characterization. For phase analysis, X-ray Diffraction (XRD) model (Shimadzu XRD 6000) was used at 2θ of 20-90° and the phase peaks were matched using Xpert Highscore Plus software. For microstructure characterization, optical microscope (Olympus BX50) was used after mirror finished surface was obtained through common metallographic process. The wettability of the solder sample was analyzed by calculating the contact angle by using ImageJ software on the image of cross-sectioned samples. The solder sample was put in the center of the copper substrate and melted it through reflow oven.

3 Result and discussion

The optical microstructure of the Sn-9Zn and Ni-coated PCC/Sn-9Zn solders are shown in Fig. 1. Based on the optical micrograph it is shown that the microstructure of the Sn-9Zn (Figure 1a) solder consists a needle-shaped structure dispersed through the entire matrix of Sn. The formation of such microstructure is due to the low solubility of Zn in the Sn matrix [5]. Hence, the low solubility of Zn causes it to form a needle shaped structured and distribution evenly within the Sn matrix, without the formation of intermetallic compound. The additions of Ni-coated PCC, the needle-like Zn phase become finer (Fig. 1b). The addition of Ni-coated PCC particle able to refine the Zn grain by stimulating the grain nucleation through heterogeneous nucleation reaction [12]. The existence of Ni-coated PCC served as alternative site that allows molten solder to initiate solidification on its surface (Fig. 2). This decreased the undercooling temperature needed [14] and allow nucleus to stabilized at smaller critical size, compared to the pristine Sn-9Zn. Consequently, as the solidification progressed the grain size produced are smaller in the composite solder.

![Fig. 1. Optical microstructure of the (a) Sn-9Zn and (b) Ni-coated PCC/Sn-9Zn](image-url)
Fig. 2. Schematic diagram of solidification process for (a) Sn-9Zn and (b) Ni coated-PCC as nucleation sites for Sn-9Zn.

Fig. 3 shows the obtained XRD pattern for the pure Sn-9Zn and Ni-coated PCC/Sn-9Zn. For the Sn-9Zn, two phases were detected, identified as Sn (ICDD 00-001-0926) and Zn (ICDD 00-001-1238), strikingly similar with the research reported by Omac et al. [15], validating the result obtained. No intermetallic compound was detected for Sn-9Zn, consistent with the observation seen in microstructure analysis. Additional peak belonging to Ni₃Sn₄ (ICDD 03-065-4310) and PCC (ICDD 01-072-1937) were detected after the additions of Ni-coated PCC, along with the peaks for Sn and Zn. This indicates that Ni reacted with Sn and formation of Ni₃Sn₄. Peak intensity ratio of peaks of Sn (2θ = 32°) to Zn (2θ = 43°) is shown in Fig. 4. The Ni coated-PCC additions caused the peak intensity of Sn/Zn ratio reduced as shown in Fig. 4 and this indicates that amount of primary Zn was increased [16]. The existence of Ni coated-PCC reinforcement was found to reduce the diffusion of Sn and Zn during solidification, as explained by Billah et al. [16]. This might also contribute to the grain refinement of Zn phase seen.

Fig. 3. XRD analysis of the Sn-9Zn and Sn-9Zn + Ni-PCC.
The Sn-9Zn produces contact angle of 25 ± 2°, and reduced to 19.1° ± 2° with the addition Ni-coated PCC (Fig. 5). Reduction in contact angle indicates that better wettability was obtained for the Ni-coated PCC/Sn-9Zn possibly due to the decrease in interfacial energy between solid and liquid [17]. Improvement in wettability is commonly seen in composite solder which the presence of reinforcement segregated in the triple point zone altered the balance of forces in the system and reduce the interfacial energy [18]. Thus, it can be said that the additions of Ni-coated PCC improve the wettability of Sn-9Zn.

4 Conclusion

The present work reports the microstructure, phase and wettability changes in Sn-9Zn reinforced with Ni-coated PCC. The conclusions are drawn as follows:

i) Microstructure of the Ni-coated PCC/Sn-9Zn exhibited refined needle-like Zn structure as compared to the monolithic Sn-9Zn.

ii) XRD analysis shows the presence of new phase Ni₃Sn₄ and calcium carbonate for the Sn-9Zn with addition of Ni-PCC. The identification of Ni₃Sn₄ proved that Ni successfully reacted with Sn. The change in the relative peak intensity ratio of Sn/Zn also indicates that the presence of Ni-coated PCC served as diffusion barrier that also contributed the refinement of needle-like Zn.

iii) The Ni-coated PCC/Sn-9Zn had lower contact angle compared to the Sn-9Zn which shows better wettability.
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