

# Role of Electrode Potential Difference between Lead-Free Solder and Copper Base Metal in Wetting

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The role of the electrochemical reaction between solder and copper base metal in soldering flux was investigated in relation to wetting. The research includes the contact polarization between solder and base metal in soldering flux of RA type. The difference of electrode potential between solder and base metal in soldering flux plays an important role in removing the oxide film by contact polarization. It is concluded that the electrode potential of Sn-based lead-free solders should be significantly lower than copper to obtain good wettability, because the accelerated anodic dissolution of tin by contacting with the copper base metal enhances the wettability by the removal of tin oxide which is one of the stable oxide on base metal and solder. Sn-Pb has the adequate electrode potential to be effectively dissolved when contacted by copper. On the other hand, the electrode potential of Sn-3.5Ag is very close to copper: the condition gives extremely small contact current. The addition of less noble elements that can lower the electrode potential is effective to enhance the wettability of Sn-3.5Ag. Sn-Zn solder has extremely low electrode potential than a copper base metal; the situation excessively accelerates the preferential anodic dissolution of zinc resulting in the no dissolution of tin. The addition of lead to Sn-3.5Ag solder lowered the electrode potential, which increased the potential difference between solder and copper base metal, as a result the contact current between them was increased. The improved wettability is confirmed by adding lead to Sn-3.5Ag; all lead added solders showed a larger spread area, *i.e.*, a smaller contact angle than Sn-3.5Ag after the spreading test. This work proposed the role of electrochemistry in wetting based on the potential difference between base metal and solder including the degree of contact corrosion current between them.

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## 1. Introduction

The research for conversion of ordinary Sn-Pb soldering to lead-free for the practical assembly of printed circuit boards is now widely conducted especially for consumer use electronic products.<sup>1)</sup> The Sn-Ag family alloys of Sn-3.5Ag and Sn-Ag-Cu solders have acceptable wettability and more excellent mechanical reliability than Sn-Zn system solders, therefore, the major lead-free solder project groups had recommended the Sn-Ag family solders as candidate alloys for practical use.<sup>2-5)</sup> However, the lead-free soldering has disadvantages in respect of wettability and in higher operation temperature range due to the characteristics of lead-free solders.<sup>1,6,7)</sup> Improving the wettability of lead-free solders is quite important for wide adoption of them in practical use. Many factors such as activating reagents in flux, solder composition and soldering condition can also influence the wettability.<sup>8,9)</sup> Especially, the role of soldering flux is important for the removal of a surface oxide film in order to give the good wettability. This oxide removal reaction of flux is dissolution *i.e.*, corrosion of base metals and solders. The highly reactive flux can easily remove the oxide film, however, the residue causes problems due to the decrease in surface insulation resistance and migration resistance<sup>10-13)</sup> which has resulted in the lower reliability of assembled equipments.

In soldering assembly of electronics, a variety of metallic materials are used on printed wiring boards; especially, widely used metals as Cu, Ag and Au have relatively higher electrode potentials than solder constituents. Under the flux soldering condition, metals with different electrode potential can influence the wettability through the contact corrosion between solder and base metal which is based on the difference

in electrode potential. However, no study has been carried out to clarify the effect of the electrode potential between solder and base metal, which can form a local corrosion cell at the advancing wetting front based on the difference in potential. From this viewpoint, the lead-free solder contains noble element of silver or base element of zinc for substitution of lead. It is easily predicted that Sn-Ag has quite different electrode potential from that of Sn-Zn. The present study aimed to find the procedure to enhance wetting in lead-free soldering through the investigation on the influence of electrochemical reactions during the soldering process, especially we aimed to clarify the effect of electrode potential difference between solder and base metal on the wetting of lead-free solders.

## 2. Experimental Procedure

To investigate the electrochemical reaction between solder and base metal, the contact polarization measurement was conducted in soldering flux. The soldering flux used was RA type flux containing 0.2%Cl, 35% solid rosin and the rest of isopropyl alcohol as a dilution solvent. The experiment was performed at room temperature, however, the temperature was raised up to 353 K in some cases to confirm the effect of the temperature on the contact current. An oxygen free high conductivity copper plate was chosen as base metal. The nominal chemical compositions of lead-free solders used are listed in Table 1, several basic lead-free solders, lead added Sn-3.5Ag, Sn-Zn with addition of noble elements and low zinc were made to confirm the effect of electrode potential on wettability. The conventional Sn-37Pb solder was also used for comparison. The melting temperature ranges were determined by a differential scanning calorimetry analysis. The

Table 1 Nominal chemical compositions and melting temperature ranges of solders used.

Solders	Liquidus $T_L$ /K	Solidus $T_S$ /K
Sn-37Pb	456	456
Sn-8.8Zn	472	472
Sn-3.5Ag	494	494
Sn-3.5Ag-2Pb	493	453
Sn-3.5Ag-4Pb	490	453
Sn-3.5Ag-6Pb	488	453
Sn-3.5Ag-10Pb	483	453
Sn-8Zn-3Ag	488	472
Sn-5Zn-3Ag	492	472
Sn-3Zn-3Ag	491	491
Sn-3Zn	490	472
Sn-7.8Zn-5.4Au	501	472
Sn-8.2Zn-1Cu	482	471

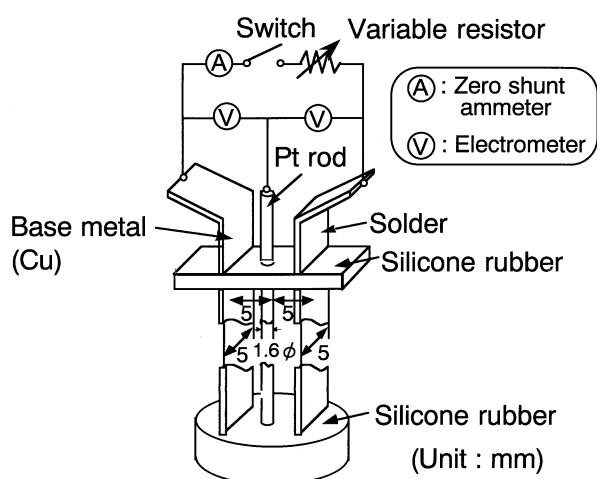


Fig. 1 Schematic illustration of apparatus for measurement of contact polarization curves between solder and copper base metal.

solders used were made of high purity metals. They were cast and cold rolled to 0.5 mm thickness and cut in to 5 mm wide and 80 mm in length. Prior to the test, the specimens were polished by emery paper of number 600.

Figure 1 shows the schematic of contact polarization test. The platinum rod was used as a reference electrode to measure the electrode potential. Copper and solder materials were vertically placed 5 mm apart from the platinum rod with 1.6 mm in diameter. A variable resistor and a zero shunt ammeter were set in series between copper and solder material. After immersion in soldering flux in depth of 30 mm from the bottom of immersed specimen, the electrode potentials of copper and solder were measured against platinum electrode by opening the switch of a series circuit. The electrode potential was measured by using the electrometers with internal impedance of  $10^{11} \Omega$ . The contact corrosion circuit was made by switching the circuit on under the maximum resistance of the variable resistor in series. The contact polarization curves were made by plotting the each electrode potential of copper and solder against the contact corrosion current measured by

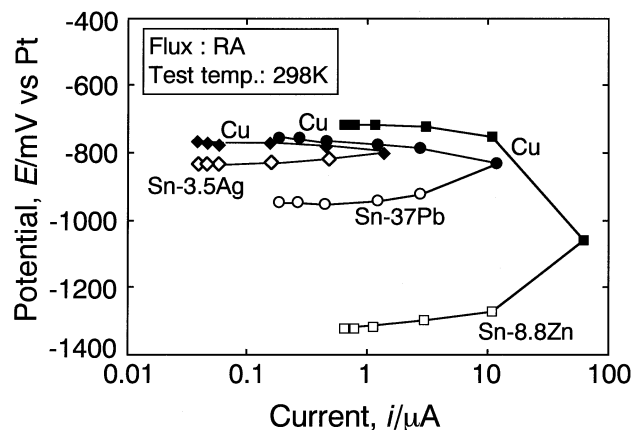


Fig. 2 Contact polarization curves between copper and three eutectic solders in soldering flux.

reducing the resistance of the variable resistor.

To evaluate the wettability, the spread test similar to the method specified in JIS Z 3197 was conducted. A molten solder bath was used to heat the spread test specimen. The base metal for the spread test was octagonal oxygen free high conductivity copper sheet with periphery length of 20 mm and thickness of 0.4 mm. The surface of copper was cleaned in 5% HCl solution, sufficiently cleaned in tap water, and dried before the test. The predetermined weight of solder corresponding to a volume of  $70 \text{ mm}^3$  was placed at the center of the spread test specimen. The set volume of flux,  $0.1 \text{ cm}^3$ , was supplied on the specimen and preheated at 373 K for 60 s to evaporate the solvent. The specimen was horizontally placed on the molten bath kept at 523 K by using a wetting balance tester, having up and down mechanism. The specimen was heated from the back side touching on the surface of molten solder bath. After having contacted the molten bath for 60 s, the solder placed on the spread specimen was molten, the spread specimen was pulled up. The temperature of the spread test specimen became the same as the molten solder bath after melting of the solder placed on copper. The contact angle and the spread area were measured for evaluation of wettability.

### 3. Results

#### 3.1 Comparison of lead-free solders

As a basic test, the electrode potential of copper and solder was measured in the soldering flux. Copper showed higher potential than solders, indicating that copper will act as a cathode when in contact with solder. Figure 2 shows the contact polarization curves between copper and three eutectic solders. The contact current is the anodic dissolution current of solder generated by the contact with copper cathode, and the current flows from the copper cathode to the solder anode. At the final stage of the contact polarization test, the resistance of a variable resistor was reduced to zero. The situation is known as short circuited, where the electrode potential of copper and solder becomes almost the same. The short current at zero resistance is defined as contact current or current at short. The potential of Sn-3.5Ag is quite similar to that of copper, in this case the contact current is small, about  $1 \mu\text{A}$ , *i.e.*,  $0.3 \mu\text{A}/\text{cm}^2$

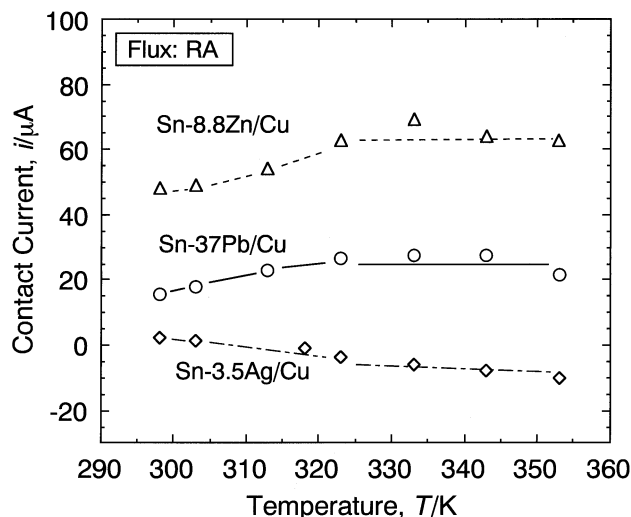


Fig. 3 Changes in contact current in RA flux during heating of short circuit specimens up to 353 K.

for one side of the specimen. The potential difference of Sn–Pb eutectic from copper stands the middle between Sn–Ag and Sn–Zn. The relation between contact current and potential difference corresponded well; the larger difference gave larger contact current, which means that Sn–Zn solder will dissolve preferentially when contacting with the copper base metal. Sn–3.5Ag solder has close potential to copper, therefore, the contact current is extremely small.

To investigate the effect of temperature on the contact current, the glass tube containing flux and short circuited specimens were heated in a mantle heater up to 353 K. Figure 3 shows the changes in contact current during heating stage. The contact current seems to gradually increase during the heating of Sn–Zn and Sn–Pb up to about 330 K, however, it slightly decreased during the heating of Sn–3.5Ag. The contact current became negative in Sn–3.5Ag, which means that Sn–3.5Ag solder became a cathode against copper, *i.e.*, copper became an anode. However, the contact current flow between Sn–3.5Ag and copper is small, less than 10  $\mu\text{A}$ . The contact current of higher temperatures corresponded well to those at room temperature. Accordingly, the electrochemistry obtained at room temperature could be expanded to the soldering temperature.

Figure 4 indicates the appearance of spread test specimen. The contact angle and spread area are also shown in the figure. As clearly shown, Sn–Pb solder exhibited a wide spread area, whereas both the lead-free solders showed small spreading, especially Sn–Zn solder showed the minimum spreading with the maximum contact angle within the experiment. The results shown in Fig. 4 could be expected by the previous reports, mentioning the poor wettability of Sn–Zn solders.<sup>14–16</sup> Lead-free solders contain elements that form more stable oxides than that of Sn–Pb solder.<sup>7</sup> The free energy of oxide formation for the elements in lead-free solders stands the following order from the most stable element to unstable one,<sup>7</sup> Zn, Sn, Cu, Ag. Zinc and tin form stable oxides, therefore, it is necessary that both tin oxide and zinc oxide are removed by contacting copper especially in Sn–Zn.

Table 2 shows the results of atomic absorption analysis of the flux after heating a glass tube containing 0.9 g of solder

Table 2 Analysis of elements in RA flux dissolved from solders after heating at 523 K for 60 s.

Materials	Dissolved tin, $C_{\text{Sn}}$ (ppm)	Dissolved element, $C_X$ (ppm)
Pure tin	81	—
Sn–3.5Ag	83	Ag: 0
Sn–37Pb	168	Pb: 11
Sn–8.8Zn	0	Zn: 2670

and 2.5 g of flux. A glass tube was immersed in a solder bath at 523 K and maintained for 60 s. It is confirmed that the noble element of silver was not dissolved into the flux in Sn–Ag solder. In Sn–Pb solder, the dissolution rate of tin is more than one order larger than that of lead. The reason may attribute to the following substitute reaction between the dissolved lead and tin.<sup>17)</sup>



In Sn–8.8Zn, only the dissolution of zinc was observed which corresponds to the low electrode potential of zinc. In this alloy no dissolution of tin was observed which means that the surface oxide of tin was not removed, this suggests that the extreme preferential dissolution of zinc deteriorates the wettability. The results indicate that the adjustment of the electrode potential difference between base metal and solder is the key to control the wettability. To enhance the wettability of Sn–Ag, it will be effective to lower the electrode potential of Sn–Ag solder by addition of base elements. On the other hand, to improve the wettability of Sn–Zn, the addition of noble elements will be effective. Raising the electrode potential of Sn–Zn solder may reduce the preferential dissolution of zinc.

### 3.2 Effect of addition of lead to Sn–3.5Ag

The potential measurement on lead added Sn–3.5Ag solder revealed that the addition of lead to Sn–3.5Ag lowered the electrode potential, therefore, the difference in potential from copper is increased by addition of lead. The contact polarization measurement was conducted on lead added Sn–3.5Ag solders. The increased contact current was observed in lead containing solders. Figure 5 summarizes the results of the contact polarization tests showing the effect of lead addition to Sn–3.5Ag on the potential difference from copper cathode and contact current at short. The potential difference is defined as follows; (potential of copper vs. Pt)–(potential of solder vs. Pt). Of course all solders behave as anodes when contacting with copper, the short circuit current is the anodic dissolution current of solders. The two plots behave similarly; the addition of 2%Pb drastically increased both potential difference and short circuit current. Further addition of lead gradually increased both values.

Figure 6 shows the effect of lead addition on the spreadability of Sn–Ag solders on a copper base metal. The spread area was gradually increased with the increase of lead addition. Corresponding to the increment of spread area, contact angle also gradually decreased with increasing lead. The plots in Figs. 5 and 6 clearly show that the contact current flow be-

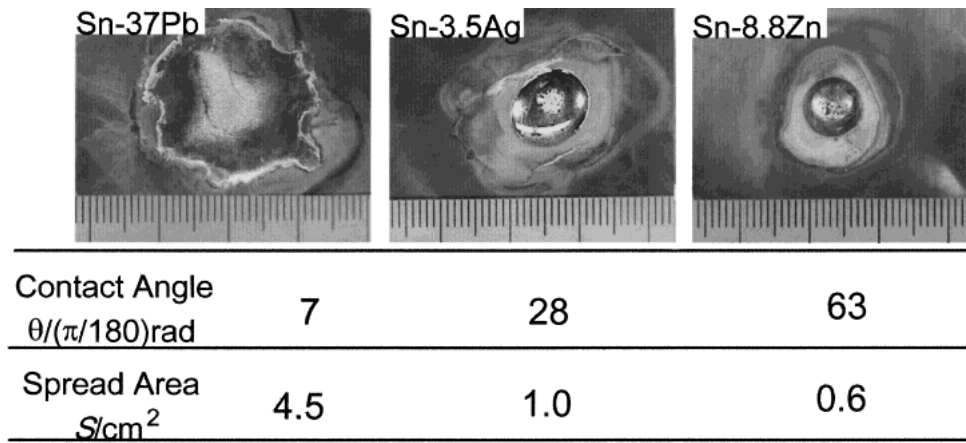


Fig. 4 Appearance of spread test specimen, the contact angle is indicated in the figure.

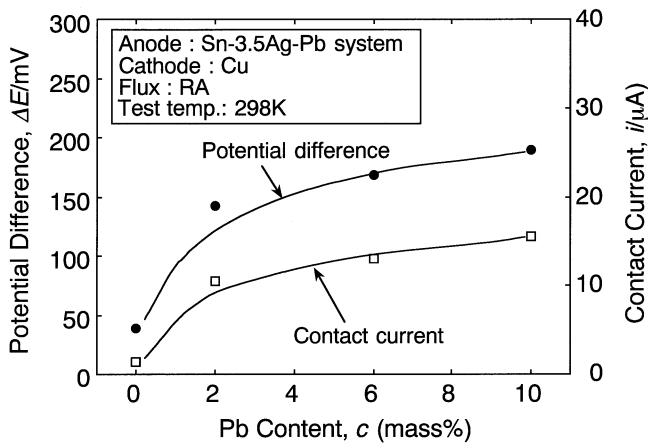


Fig. 5 Effect of lead addition to Sn-3.5Ag solder on potential difference from copper and contact current obtained by contact polarization measurement in RA flux.

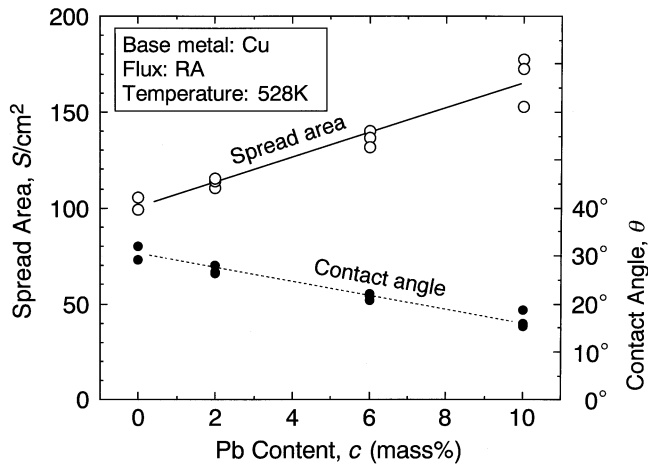


Fig. 6 Effect of lead content in Sn-3.5Ag on spread area and contact angle obtained by spread test at 528 K.

tween base metal cathode and solder anode has significant influence on the wetting in flux soldering process.

### 3.3 Effect of composition on wettability of Sn-Zn

To improve the wettability on Sn-Zn, the raise of electrode potential of solder will be effective from the electrochemical view point. To raise the potential, the addition of noble

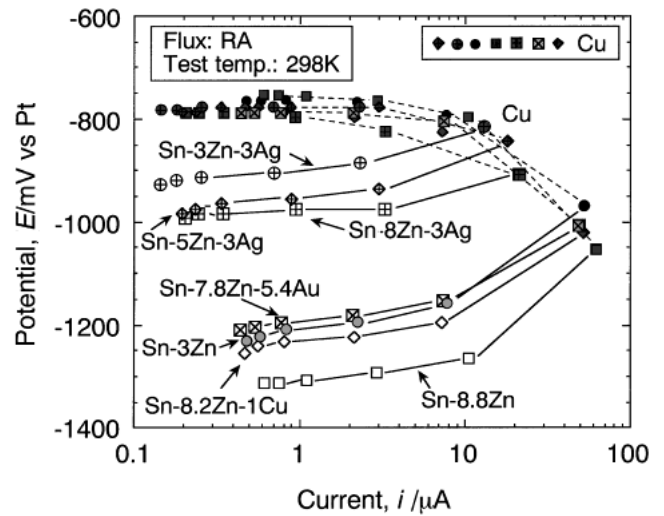


Fig. 7 Contact polarization curves between copper and Sn-Zn system solders with some elements and different zinc content.

elements such as Au, Ag, and Cu, and the decrease of zinc content will be acceptable from wettability view point. To confirm these factors, the contact polarization measurements were conducted on several Sn-Zn solders. The results are shown in Fig. 7. It is clear that the additions of Ag, Au and Cu are effective to raise the electrode potential of Sn-9Zn. According to the rise of the electrode potential of solders, the decrease in contact current was observed. Among these elements, the addition of silver extremely decreased both electrode potential and contact current. The decrease of zinc was also found to be effective to raise the electrode potential and the reduction of contact current in both Sn-Zn and Sn-Zn-Ag. Among these solder alloys, Sn-3Zn-3Ag showed the highest electrode potential.

Figure 8 indicates the relation between the potential difference and the contact current for Sn-Zn system, Sn-Ag-Pb system and Sn-37Pb solders. The increase in potential difference seems to increase the contact current linearly irrespective to the solder alloy composition system. The potential difference is a measure for estimating the contact current between solder and copper.

Figure 9 shows the results of spread test. The obtained contact angle is plotted according to the zinc content of solder.

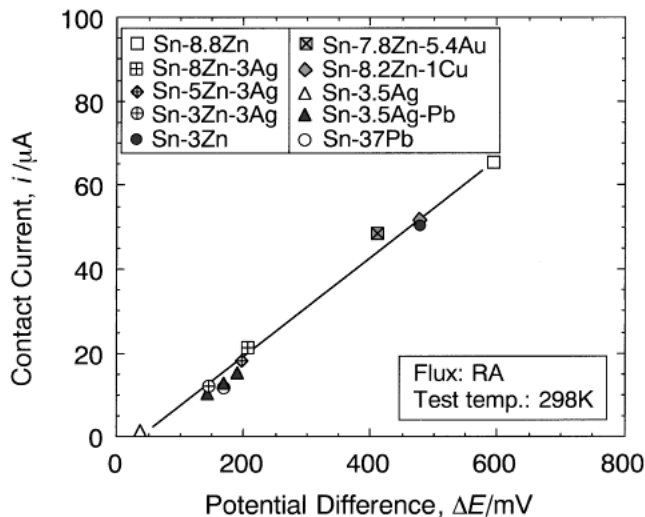


Fig. 8 Plots of potential difference between copper and solder against contact current.

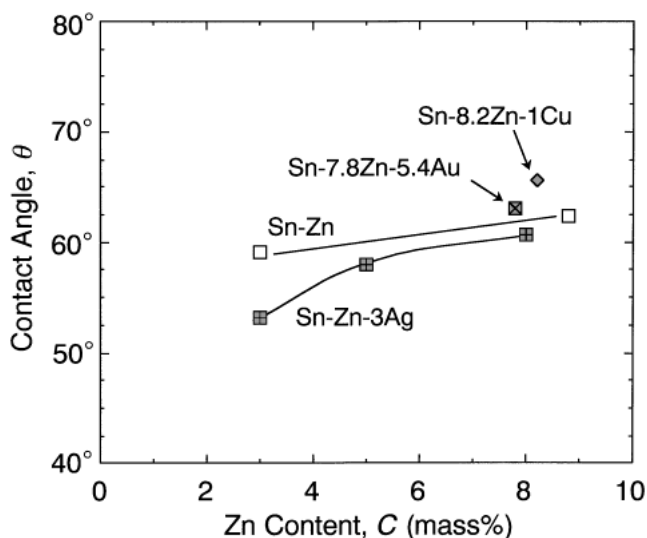


Fig. 9 Results of spread test on Sn-Zn system solders with additional elements and different zinc content.

Both the decrease in zinc content and the addition of silver slightly reduced the contact angle showing they slightly enhanced wettability. The addition of Au and Cu has no improvement on wettability even they reduced the contact current by reducing the electrode potential difference between solder and copper as shown in Fig. 8. The reason that the addition of these elements gives little effect on wettability is not clear.

#### 4. Discussions

Several factors have been known to influence the wettability.<sup>18)</sup> They are surface tension, interfacial tension between flux and molten solder in case of soldering, interfacial tension between solder and base metal, intermetallic compound formation between solder and base metal,<sup>19)</sup> stability of oxide, activity of flux and *etc.* In the present study, the effect of difference of electrode potential between solder and base metal was studied. The difference of potential could affect the wettability by changing the contact corrosion current flow-

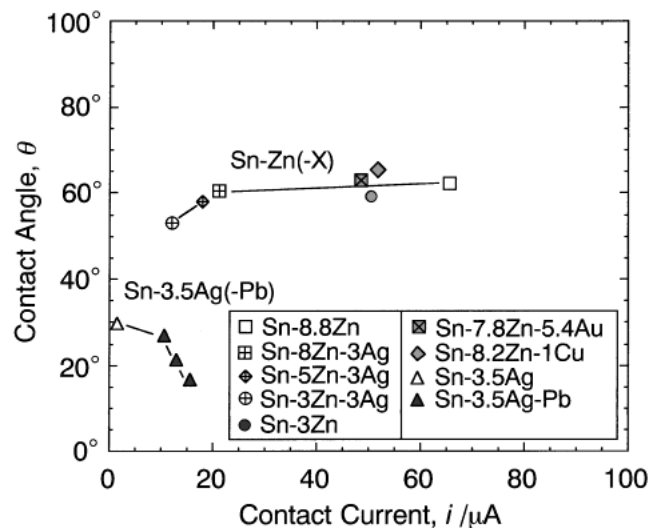


Fig. 10 Relations between contact current and contact angle for Sn-3.5Ag(-Pb) and Sn-Zn(-X) lead-free solders.

ing between them. In Sn-Zn solder, the potential difference between copper and solder is extremely high due to the low electrode potential of zinc. Accordingly, the dissolution of tin could not be sufficiently occurred, Table 2, the reaction is important to remove the surface oxide film on solder. The preferential dissolution of zinc is responsible for the poor wetting, therefore, to raise the electrode potential of Sn-Zn alloy may be effective to suppress the preferential dissolution of zinc, which may enhance the dissolution of tin resulted in the improved wetting. However, the addition of noble elements of Au and Cu was found to have little effect on wettability even they lowered the potential difference and the contact current between copper and solder. The addition of Ag only slightly lowered the contact angle, of course it raised the electrode potential of solder. To improve the wettability of Sn-Zn system solder, both the decrease in zinc content and addition of Ag were effective, which was in accordance with the decrease in the contact current between copper and solder. The decrease of zinc content in Sn-Zn-2Ag-0.8Cu solder has been found to improve the wettability.<sup>14)</sup>

The addition of In has been found to be effective to improve the wettability of Sn-Zn.<sup>15)</sup> The electrode potential of In is higher than that of Zn,<sup>20)</sup> and it gives little effect on surface tension of tin,<sup>21,22)</sup> therefore, the improvement of wettability of Sn-Zn by addition of In may attribute to the change of electrode potential of Sn-Zn-In solder.

In Sn-Ag solder, the addition of lead increased the potential difference that increased the contact current correspondingly. The addition of lead to Sn-Ag accordingly enhanced wetting.

Figure 10 summarizes the relation between the contact current and the contact angle for lead free solders. In Sn-Ag system, the increase in the contact current enhanced the wettability corresponding to the reduction of contact angle. The effect enhanced the contact corrosion of Sn-Ag-Pb solders when contacted with copper. The chemical analysis of flux after heating to the soldering temperature revealed that the dissolved tin content was increased with increasing the amount of added lead, Table 3. The table also shows that the disso-

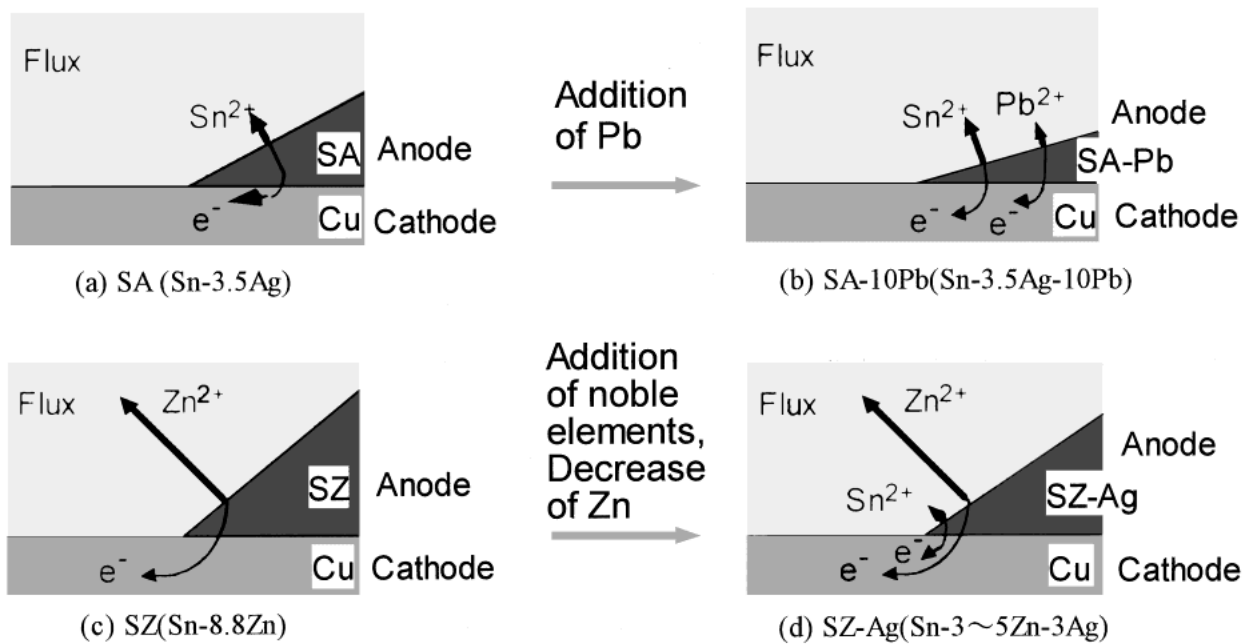


Fig. 11 Schematic illustration of the effect of electrochemical local corrosion cell between copper and solders in soldering flux RA on contact angle at the advancing wetting front.

Table 3 Dissolution of tin into soldering flux RA after heating at 523 K for 120 s with and without contacting to Cu plate.

Solder	Dissolved tin, $C_{Sn}$ (ppm)	
	Solder only	Contact with copper
Sn-3.5Ag	118	276
Sn-3.5Ag-2Pb	104	279
Sn-3.5Ag-4Pb	—	288
Sn-3.5Ag-6Pb	—	338
Sn-3.5Ag-10Pb	78	347

—: Not tested

lution of tin was accelerated by contact with copper. In this case, solder and copper plate were placed together in a glass vessel containing flux.

In Sn-Zn system, the contact angle remained almost constant even the contact current was reduced from about  $60\mu\text{A}$  to about  $20\mu\text{A}$ . Further decrease in contact current slightly reduced contact angle indicating that the wettability was slightly improved by addition of silver and reduction of zinc. Of course the contact current is not the only measure for evaluating the wettability, therefore, the contact angle is quite different in both systems at the same contact current.

The work has revealed that the potential difference between solder and copper base metal could influence the wettability through the contact polarization between them. The potential of tin based lead-free solder should be lower than the copper base metal, because the tin oxide with more stable oxide formation free energy than copper should be removed sufficiently to achieve good wetting. To possess the adequate wettability, the solders should be anodic by contacting with base

metal. The solder alloy should have a lower potential than the base metal and also should not contain elements having an extremely low electrode potential such as zinc. The excess preferential dissolution of zinc inhibited the dissolution of tin, which derives the poor wetting.

Figure 11 shows the schematic illustration of the effect of electrode potential on the electrochemical local corrosion cell between solder and base metal during flux soldering. In the combination with solder and base metal having similar electrode potentials such as Sn-3.5Ag and copper, the dissolution of tin is not accelerated by the contact with copper base metal, whereas in the solder with extremely low electrode potential, Sn-Zn for example, the excess selective dissolution of zinc hinders the dissolution of tin that resulted in the poor wetting. The solder alloy with an adequate electrode potential lower than copper is taken accelerated dissolution of tin by contact with copper that enhances the wettability through the increased contact current between copper and solder; Sn-3.5Ag-Pb and Sn-37Pb correspond to this case. The accelerated dissolution of solder components including tin at the wetting front by contacting the copper base metal plays an important role for solder spreading. This concept will be effectively used for the development of new lead-free solder with excellent wettability. It is well known that gold plating on the base metal is effective to improve wetting. From the point of view of electrode difference, the gold plating on base metal is effective to raise both the electrode potential of base metal and contact current with Sn-Ag solder. It is easily imaginable that the wettability on gold plated base metal was improved by this electrochemical mechanism.

Of course the gold plating inhibits the oxidation of under layer and the addition of lead may reduce the surface tension, therefore, the further investigation will be necessary using

fluxless process for example. The removal of surface oxide by the enhanced corrosion, *i.e.* dissolution, of tin is the key to secure the excellent wettability in lead-free soldering. The additional elements give many effects such as surface tension and intermetallic formation between solder and base metal interface, therefore, further investigation is necessary by using a fluxless process that can separate the electrochemical reaction.

## 5. Conclusion

The effect of electrode potential difference between solder and copper base metal on the wettability was investigated. The work pointed out that the contact current between solder and copper has significant influence on wettability. The adjustment of electrode potential and base metal is important to secure the excellent wettability in flux soldering. The results obtained are summarized as follows.

(1) The copper base metal has noble potential than Sn–Pb, Sn–Ag and Sn–Zn eutectic solders. These solders become anodes in soldering flux by contact with copper. The difference in electrode potential between copper and these solders increased in the following order; Sn–3.5Ag < Sn–37Pb < Sn–8.8Zn; the order corresponds well to the anodic corrosion current of solder when contacted with copper.

(2) The contact anodic current basically improves the wetting of solder by enhancing the removal of oxide film on solder surface by anodic dissolution of solder, however, the extreme preferential dissolution of the specific elements such as zinc deteriorates the wettability of Sn–Zn.

(3) The addition of lead up to 10% to Sn–3.5Ag lowered the electrode potential and increased the contact corrosion current between copper base metal and solder. The effectiveness of the lead addition on the wettability was confirmed by the spread test on copper.

(4) The addition of silver to Sn–Zn raised the electrode potential of solder that reduced the contact current between copper base metal and solder. The suppression of contact current by addition of silver slightly improved the wettability.

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