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Pb-Free Solders: Part II. Application of ADAMIS Database in Modeling of Sn-Ag-Cu Alloys with Bi Additions

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The ADAMIS database was used for calculation of the surface tension of the quaternary Sn-Ag-Cu-Bi liquid alloys by Butler's model. The resultant data were compared with those from the maximum bubble pressure measurements from Part I. The same thermodynamic database was next applied for the calculation of various phase equilibria. It was established that the Bi addition to the ternary Sn-Ag-Cu alloys (Sn-2.6Ag-0.46Cu and Sn-3.13Ag-0.74Cu in at.%; Sn-2.56Ag-0.26 Cu and Sn-2.86Ag-0.40Cu in mass%) causes lowering of the melting temperature and the surface tension to make the tested alloys closer to traditional Sn-Pb solders. The simulation of the solidification by Scheil's model showed that the alloys with the higher Bi concentration are characterized by the lifting-off failure due to the segregation of Bi at the solder/substrate boundary. Thus, in modeling of new Pb-free solders, a compromise among various properties should be taken into consideration.

Keywords phase equilibria, simulation of solidification, surface tension, Sn-Ag-Cu-Bi

1. Introduction

In the previous work,[1] the results were reported for the surface tension, density, and meniscographic investigations of the quaternary Sn-2.76Ag-0.46Cu and Sn-3.13Ag-0.74Cu (in at.%; Sn-2.56Ag-0.26 Cu and Sn-2.86Ag-0.40Cu in mass%) alloys together with the binary Sn-Ag eutectic and two Sn-Ag-Cu near eutectic alloys. It was established that the addition of Bi to the ternary Sn-Ag-Cu alloys remarkably decreases the surface tension and the wettability parameters (interfacial tension, wetting time, wetting force, contact angle) to the value comparable with the Pb-Sn solders. In this work, the data of the surface tension modeled by Butler's method[2] previously used for Sn-Ag,[3] Sn-Ag-Cu[4] are discussed. Also, the calculated isothermal and the vertical sections of the phase diagram, equilibrium, and nonequilibrium solidifications by Scheil's model using the Thermo-Calc software and the thermodynamic data from the ADAMIS database,[5] compiled at Tohoku University, Japan, is presented in this study.

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2. Modeling of the Surface Tension

Use of Butler's method for calculation of the surface tension was discussed in the authors earlier work on the surface tension of the binary[3,6-10] and ternary[4,8,11,12] liquid alloys. In the case of the quaternary Sn-Ag-Cu-Bi alloys, the following relation was used:

$$\begin{aligned}\sigma &= \sigma_{Ag} + \frac{RT}{A_{Ag}} \ln \frac{(1 - X^S_{Sn})}{(1 - X^B_{Sn})} + \frac{1}{A_{Ag}} \overline{G}^{E,S}_{Ag} - \frac{1}{A_{Ag}} \overline{G}^{E,B}_{Ag} \\ &= \sigma_{Sn} + \frac{RT}{A_{Sn}} \ln \frac{X^S_{Sn}}{X^B_{Sn}} + \frac{1}{A_{Sn}} \overline{G}^{E,S}_{Sn} - \frac{1}{A_{Sn}} \overline{G}^{E,B}_{Sn} \\ &= \sigma_{Cu} + \frac{RT}{A_{Cu}} \ln \frac{X^S_{Cu}}{X^B_{Cu}} + \frac{1}{A_{Cu}} \overline{G}^{E,S}_{Cu} - \frac{1}{A_{Cu}} \overline{G}^{E,B}_{Cu} \\ &= \sigma_{Bi} + \frac{RT}{A_{Bi}} \ln \frac{X^S_{Bi}}{X^B_{Bi}} + \frac{1}{A_{Bi}} \overline{G}^{E,S}_{Bi} - \frac{1}{A_{Bi}} \overline{G}^{E,B}_{Bi}\end{aligned} \quad \text{(Eq 1)}$$

where R is the gas constant in J mol^{-1} K^{-1}; T is the temperature in Kelvin; σ is the surface tension of the alloy; σ_i is the surface tension in mN m^{-1}; A_i is the molar surface area in a monolayer in square meters; X^S_i, X^B_i and $G^{E,B}_i$, $G^{E,S}_i$ are the mole fractions and the excess Gibbs free energy of component i (i = Ag, Bi, Cu or Sn) in the surface and bulk phase, respectively.

Molar surface area is calculated from the relation:

$$A_i = 1.091 V^{2/3} N^{1/3} \quad \text{(Eq 2)}$$

where V is the molar volume of component i, N is Avogadro's number, and 1.901 is set for liquid metals, assuming a close-packed structure.

It was shown that in most investigated Pb-free sol-

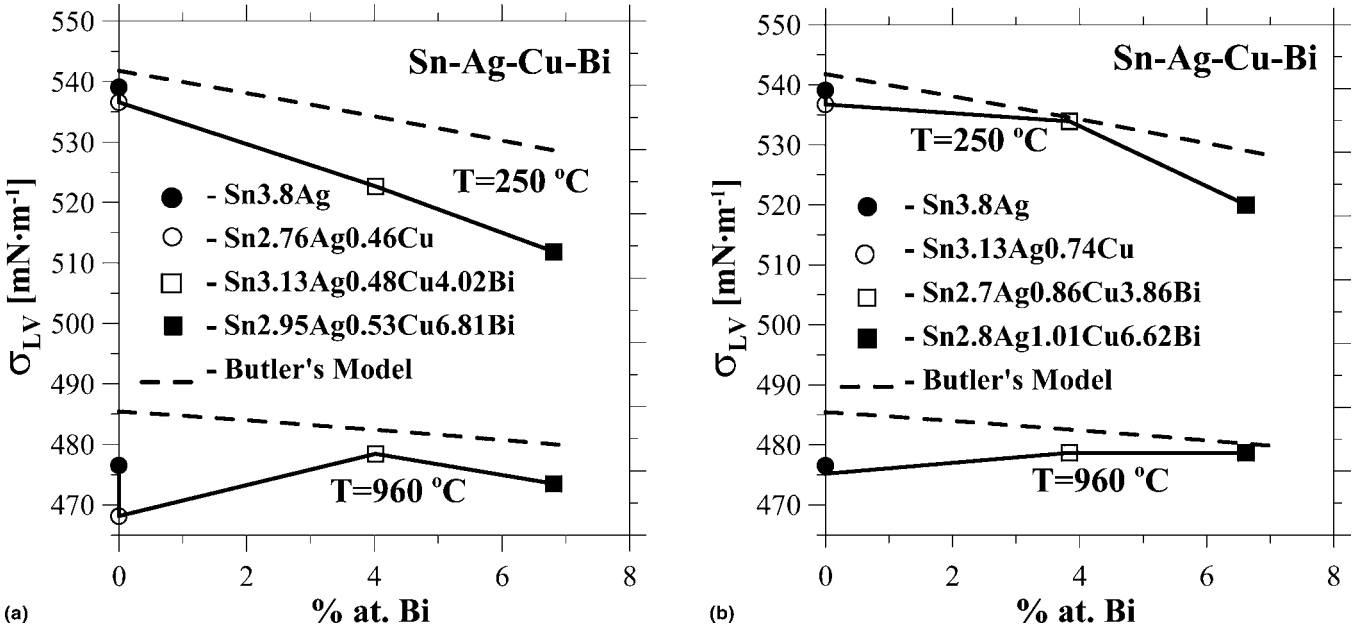

Fig. 1 Isotherms of the surface tension measured in protective atmosphere[1] at 250 and 960 °C compared with calculated data using Butler's method: (a) adding Bi to Sn-2.76Ag-0.47Cu alloy and (b) adding Bi to Sn-3.13Ag-0.74Cu alloy

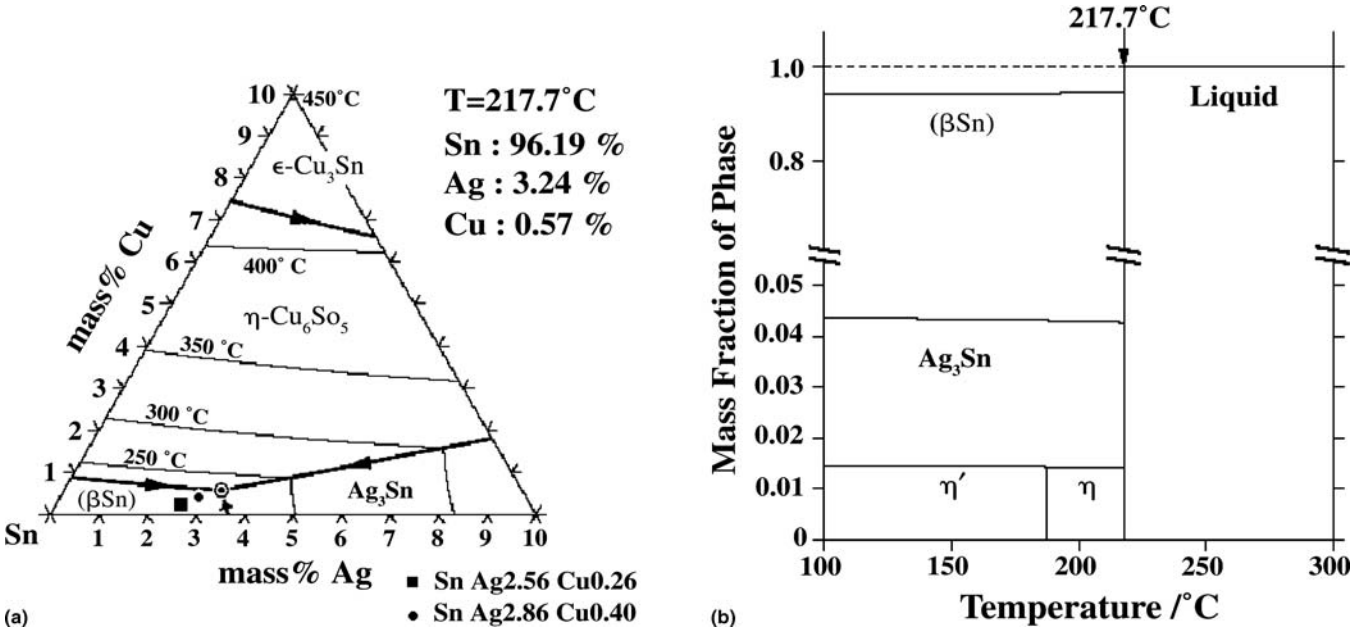

Fig. 2 (a) Projection of liquidus surface in Sn-based portion of the Sn-Ag-Cu system with two alloys discussed in this study Sn-2.56Ag-0.26 Cu and Sn-2.86Ag-0.40Cu in mass%. (b) Plot of mass fractions of phases versus temperature of Sn-Ag-Cu ternary eutectic alloy with two forms of Cu_6Sn_5, η' low temperature and η high temperature,[13] which exist in phase diagram calculations of the quaternary system Sn-Ag-Cu-Bi

ders,[3,4,6-12] Butler's method gives reasonable agreement with experimental results; however, the calculated temperature dependences of the surface tension were slightly curvilinear in comparison with the straight linear dependence of the experimental data. This problem was discussed in detail in the case of Ag-Bi[6] liquid alloys.

The excess Gibbs free energies of components were calculated from the following equation:

$$G_i^{ex} = G^{ex} + \left(\frac{dG^{ex}}{dX_i} - \sum X_j \frac{dG^{ex}}{dX_j} \right) \quad j \neq i \qquad \text{(Eq 3)}$$

using in the case of multicomponent alloys the binary formalism with ternary interactions of the form:

$$G^{ex} = \sum_{i=1}^{3}\sum_{j=2}^{4} X_i X_j W_{i,j} + \sum_{i=1}^{2}\sum_{j=i+1}^{3}\sum_{k=j+1}^{4} X_i X_j W_k W_{i,j,k} \qquad \text{(Eq 4)}$$

In Eq 3 and 4, X_i denotes mol fractions of components, $W_{i,j}$ and $W_{i,j,k}$ are interaction parameters of the binary and the ternary alloys, and G^{ex} and G_i^{ex} are the molar and partial excess Gibbs free energies of the components of the investigated alloys. In Eq 4, the binary Sn-Ag, Sn-Cu, Cu-Bi, Ag-Bi, Cu-Ag, and Sn-Bi and ternary Sn-Ag-Cu and Sn-Ag-Bi interaction parameters from ADAMIS database were used in calculations of the surface tension, and they are listed in the Appendix.

In Fig. 1(a) and (b), the calculated isotherms of the surface tension of the quaternary alloys are presented starting from ternary alloys Sn76Ag0.46Cu and Sn3.13Ag0.74Cu (in at.%). On vertical axes, the surface tension data from Ref 2 and 3 are given for ternary and binary data. The calculated surface tension at temperature 250 °C shows a decrease with the Bi additions similar to equivalent data. The experimental data at 960 °C show slightly increasing tendencies in comparison to those for binary and ternary Sn-Ag and Sn-Ag-Cu alloys (crossing of the temperature dependences of the surface tension results for the quaternary alloys with those of the binary Sn-Ag and ternary Sn-Ag-Cu as reported in Part I). The modeled surface tension values always show a decreasing tendency of the surface tension with Bi additions.

3. Phase Equilibria and Simulation of Solidification of Ternary Sn-Ag-Cu and Quaternary Sn-Ag-Cu-Bi Alloys

Two eutectics, Ag-Sn and Ag-Sn-Cu, have been proposed as substitutes for traditional Pb-Sn solders. The first one is characterized by a melting temperature of 221 °C and the second by a melting temperature of 217-219 °C; both are much higher than the Pb-Sn eutectic melting temperature of 183 °C. (Surface tension data show similar shifts.) Therefore, the mentioned modification of lead-free alloys should result in wettabilities and melting temperatures closer to those for Pb-Sn.

The liquidus surface of the Sn-Ag-Cu phase diagram, published by Ohnuma,[13] is shown in Fig. 2(a). The calculated ternary eutectic melts at 217.7 °C and the eutectic reaction is as follows:

$$\text{Liquid} \rightarrow (\beta\text{Sn}) + \text{Ag}_3\text{Sn} + \eta\text{Cu}_6\text{Sn}_5$$

The composition of the components are equal to 3.24 mass% Ag and 0.57 mass% Cu; the mass fractions of the phases are presented in Fig. 2(b). The influence of the Bi addition at the melting temperature and the concentration of Ag and Cu in solidified alloy are shown in Fig. 3(a) and (b).

The influence of Bi additions on the eutectic ternary Sn-Ag-Cu alloy and on the change of silver and copper was also calculated and is shown in Fig. 3.

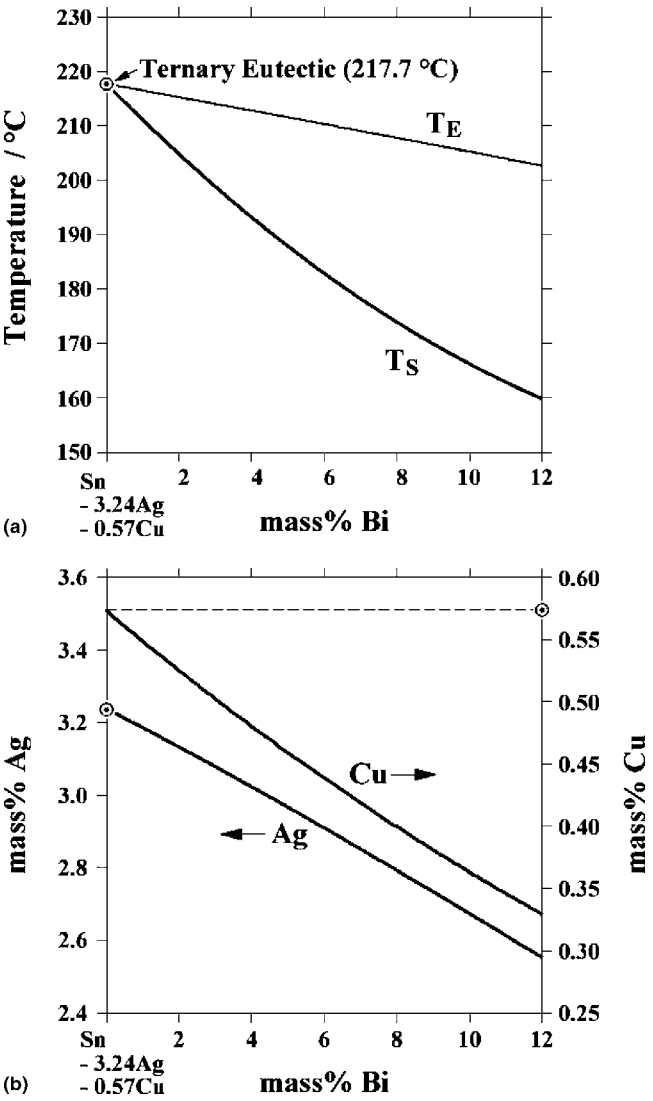

Fig. 3 Indicates (a) the influence of Bi additions on the ternary eutectic temperature and (b) the change of Ag and Cu content with Bi addition

To investigate the influence of Bi additions on melting temperatures using the ADAMIS database, various phase equilibria were calculated for the quaternary Sn-Ag-Cu-Bi system starting from Sn2.56Ag0.26 Cu and Sn2.86Ag0.40Cu in mass%, adding to each various amounts of Bi. Results of calculations starting from Sn2.56Ag0.26 Cu alloy are shown in Fig. 4 on the vertical sections.

In practical solidification processes, nonequilibrium rather than equilibrium solidification is often observed, and this can be easily simulated by the Scheil module of Thermo-Calc (Stockholm, Sweden). Although the Scheil model assumes that local equilibrium exists at the liquid/solid interphase, such calculations can still provide a prediction close to reality.[14] In Fig. 5 in both cases, under equilibrium and Scheil-model solidification conditions, solidification starts at 222 °C with the primary crystals of βSn, and is terminated when it reaches the ternary eutectic reac-

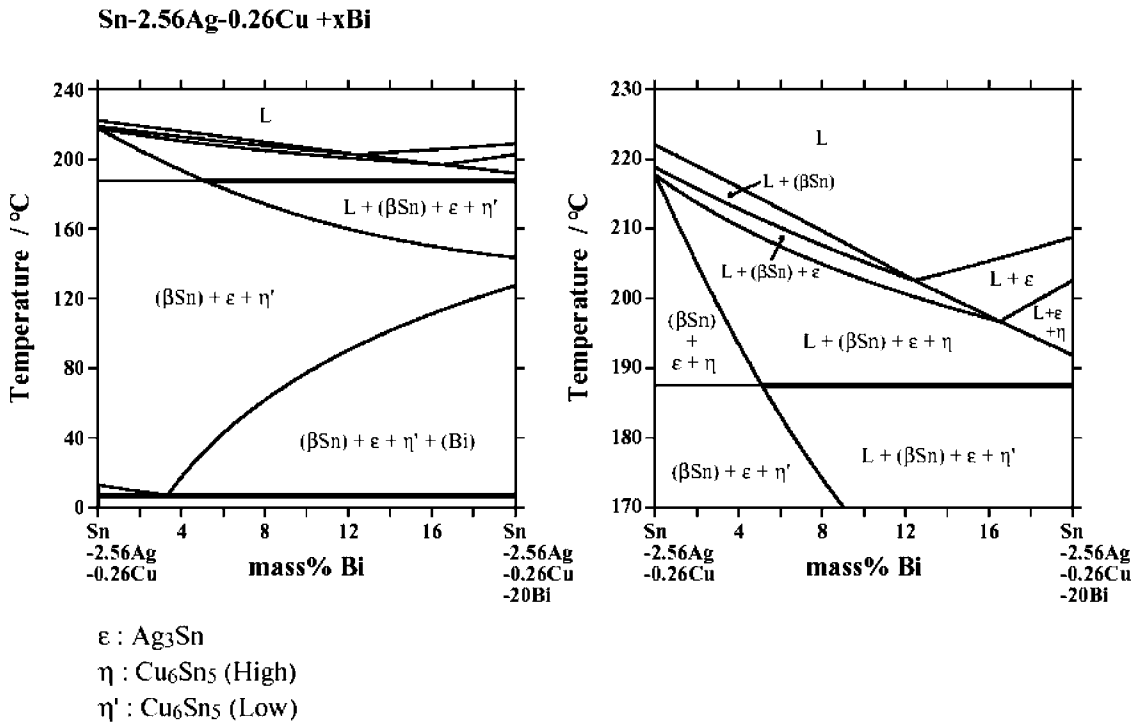

Fig. 4 Vertical section of the Sn-Ag-Cu-Bi phase diagram for Sn-rich portion starting from Sn-2.56Ag-0.26Cu alloy in mass %; the right side shows the magnified left part at higher temperatures

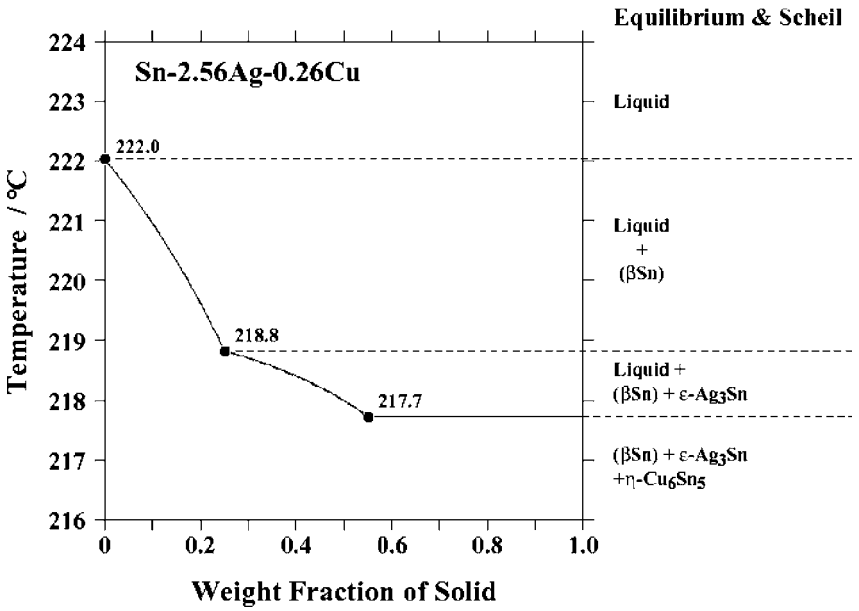

Fig. 5 Calculated mass fraction of the solid phase versus temperature variation of Sn-2.56Ag-0.26Cu alloy under equilibrium and Scheil-model solidification conditions

tion at 217.7 °C. The difference between the two cases seems to be quite small. It is due to the fact that the primary crystal βSn has no solubility of Ag and Cu. The secondary one Ag_3Sn has no solubility of Cu either (the same applies to Fig. 9).

The solidification of Sn2.77Ag0.26Cu6.91Bi and Sn2.56Ag0.27Cu11.45Bi alloys shown in Fig. 6 and 7 is different from that shown in Fig. 5, and the existence of respective phases corresponds to the vertical section in Fig. 6. The solidification starts with the primary crystals of ηCu_6Sn_5; it proceeds substantially, however, with the growth of βSn at the beginning. After commencement of the crystallization of Ag_3Sn phase, the liquid phase would disappear at 179 °C under equilibrium solidification conditions

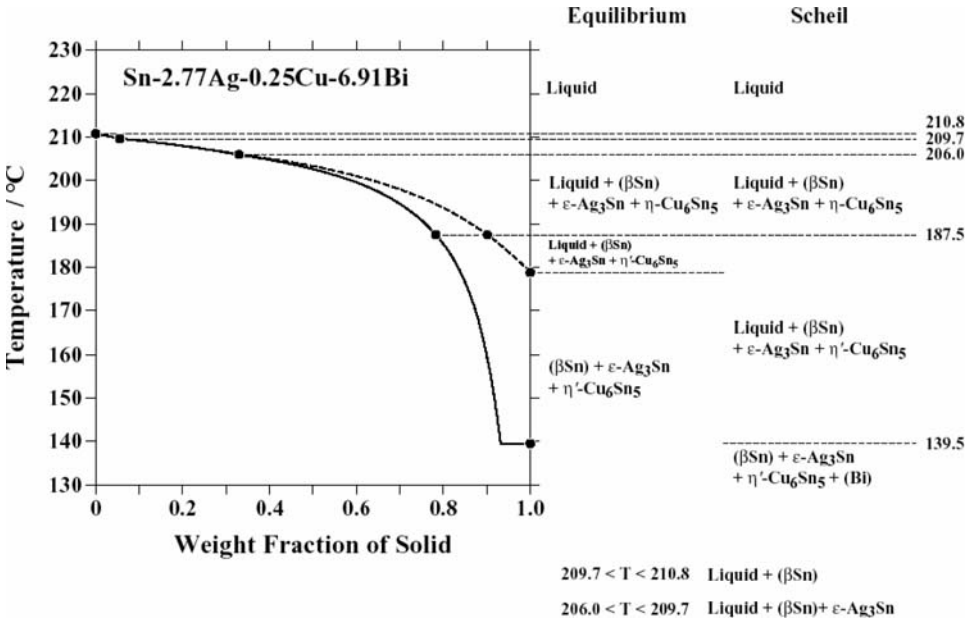

Fig. 6 Calculated mass fraction of the solid phase versus temperature variation of Sn-2.56Ag-0.26Cu alloy under equilibrium and Scheil-model solidification conditions with 6.91 mass% Bi additions

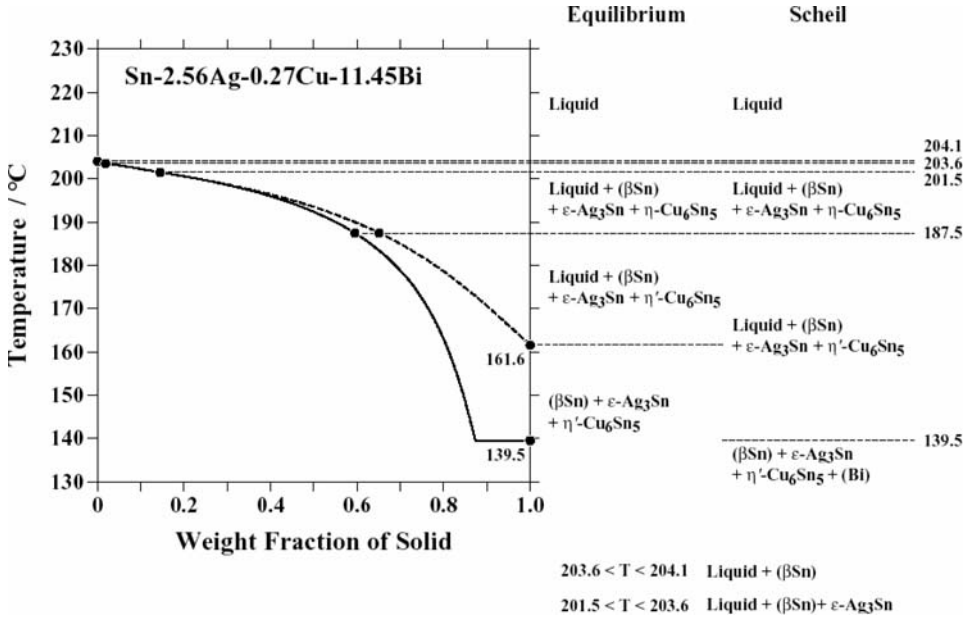

Fig. 7 Calculated mass fraction of the solid phase versus temperature variation of Sn-2.56Ag-0.26Cu alloy under equilibrium and Scheil-model solidification conditions with 11.45 mass% Bi additions

at 6.91 mass% Bi and at 161.6 °C at 11.45 mass % Bi. According to the Scheil model, Bi concentrates in the liquid phase during solidification (Fig. 6 and 7), which causes an extensive fall of the terminating temperature of solidification. The temperature 139.5 °C corresponds to the eutectic reaction of the Sn-Bi binary system. The behavior of such a residual liquid phase at low temperatures is considered to cause the previously mentioned lifting-off failure at the interface between the solder material and Cu substrates.

Similar calculations were undertaken for the second ternary alloy Sn2.86Ag0.40Cu (mass%) and are presented in Fig. 8-11. Because of small differences in the compositions of both starting ternary alloys Sn2.56Ag0.26Cu and Sn2.86Ag0.40Cu and Bi additions (instead of 6.91 and 11.45 mass%; 6.65 and 11.17 mass%), in Fig. 8-11 only slight differences can be observed in the temperatures at the beginning of solidification of the ternary and quaternary alloys and in the temperatures at which the liquid phase disappears in the equilibrium solidification conditions.

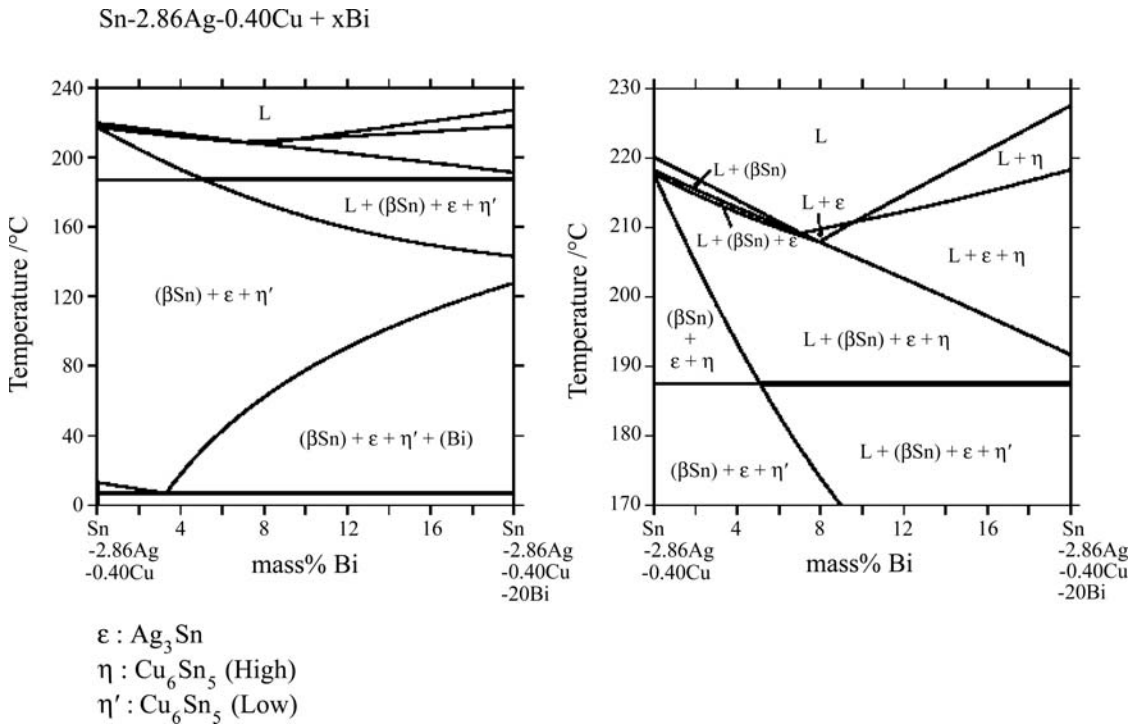

Fig. 8 Vertical section of the Sn-Ag-Cu-Bi phase diagram at Sn-rich portion starting from Sn-2.86Ag-0.40 Cu alloy; the right side shows the magnified left part at higher temperatures

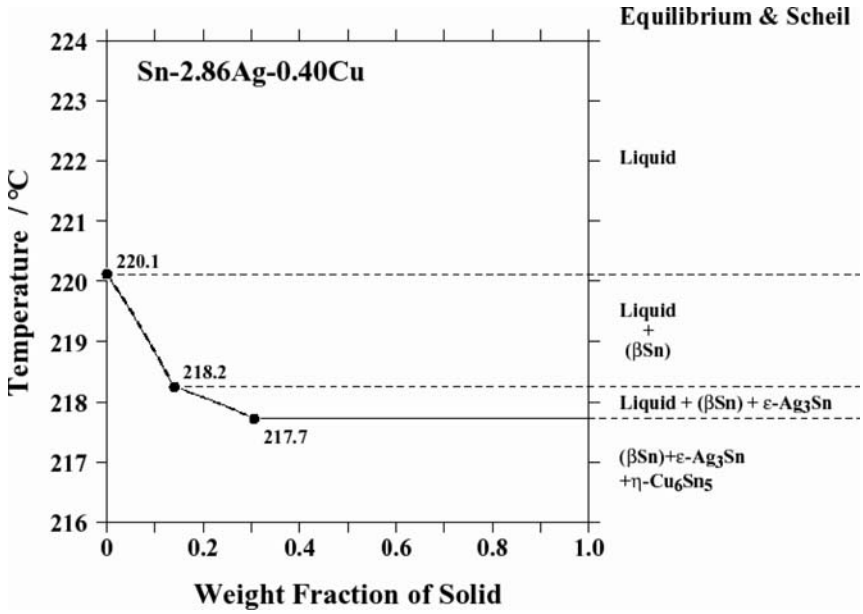

Fig. 9 Calculated mass fraction of the solid phase versus temperature variation of Sn-2.86 Ag-0.40Cu alloy under equilibrium and Scheil-model solidification conditions

There is always the same extensive fall of terminating temperature of solidification at 139.5 °C. In Fig. 6-7, Fig. 10-11, and Fig. 12 under the equilibrium solidification Bi is distributed into βSn and liquid phases enabling equilibrium condition. When the temperature reaches the solidus boundary, the solidification terminates. Therefore, the terminating temperature decreases with increasing Bi content.

On the other hand, in the Scheil simulation of solidification, no diffusion of Bi in βSn phase is assumed and then an extraordinary amount of Bi is concentrated into the liquid phase. Finally, the composition of the liquid enriches in Bi with terminated temperature of all alloys 139.5 °C.

The problem of mutual interactions in quaternary alloys Sn-Ag-Cu-Bi has been discussed by Hwang,[15] suggesting

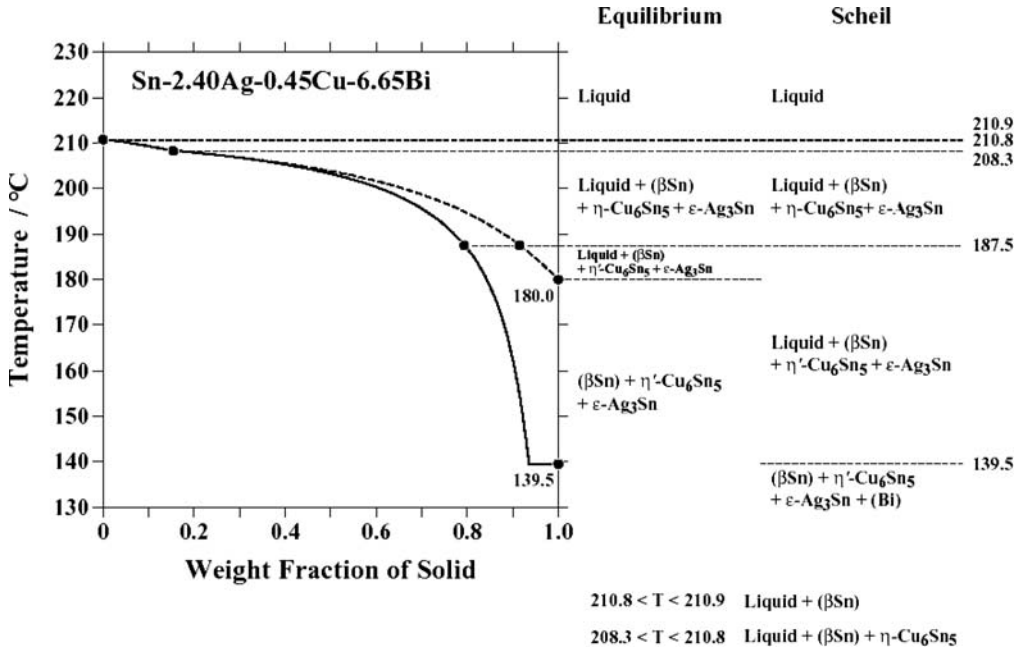

Fig. 10 Calculated mass fraction of the solid phase versus temperature variation of Sn2.86Ag0.40Cu alloy under equilibrium and Scheil-model solidification conditions with 6.65 mass% Bi additions

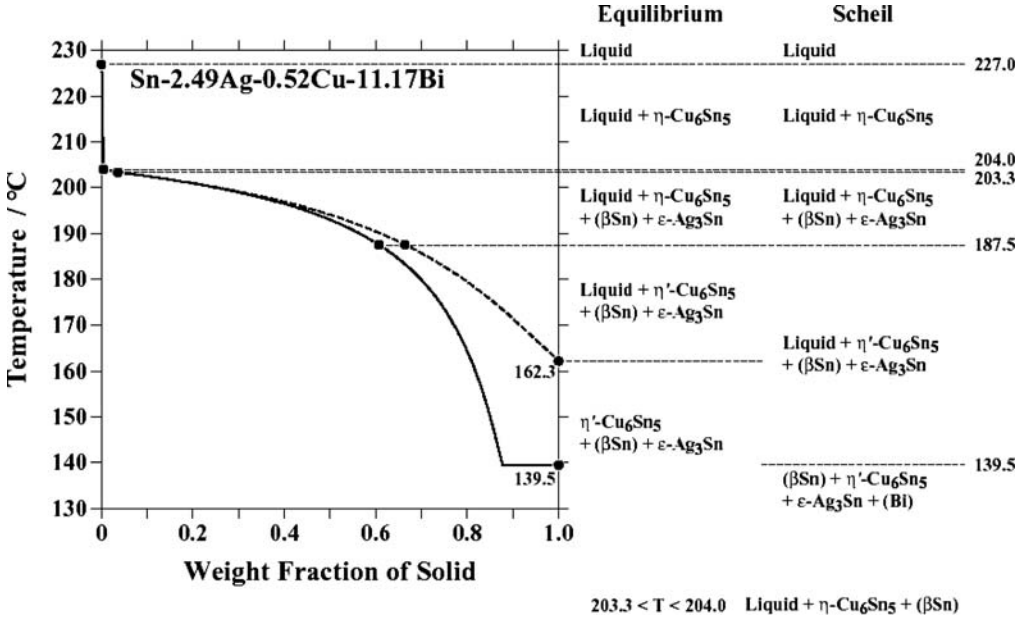

Fig. 11 Calculated mass fraction of the solid phase versus temperature variation of Sn2.86Ag0.40Cu alloy under equilibrium and Scheil-model solidification conditions with 11.17 mass% Bi additions

tin-based alloys for use as Pb-free solders. Interactions between Ag and Sn form a Ag_3Sn intermetallic compound, and Cu reacts with Sn to form a Cu_6Sn_5 intermetallic compound. For Sn-Bi interactions, Bi atoms are expected to enter into lattice sites as substitutional atoms up to 1 mass%; beyond that percentage, Bi atoms precipitate out as an independent second phase. This was documented in this study in the simulation of solidification by Scheil's method.

The tendency to precipitate Bi from liquid alloys as a separate phase is connected with the fillet-lifting phenomena at the interface between the solder material and the Cu usually used as a substrate. This problem in Sn-Ag-Bi alloys was discussed by Takao et al.,[16] indicating that a peculiar composition dependence exists on the fillet-lifting. With fillet-lifting, compositions are 3-20 mass% Bi. Without fillet-lifting, the compositions are 0-1 mass% Bi, 30 mass%

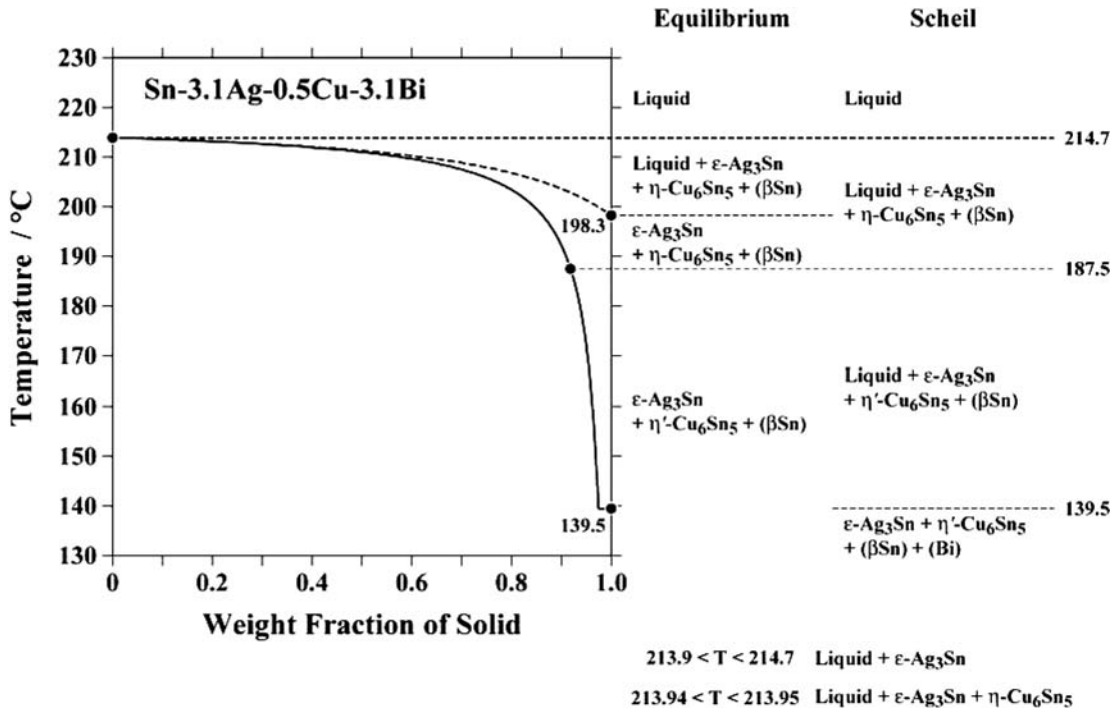

Fig. 12 Calculated mass fraction of the solid phase versus temperature variation of Sn3.1Ag0.5Cu3.1Bi alloy under equilibrium and Scheil-model solidification conditions

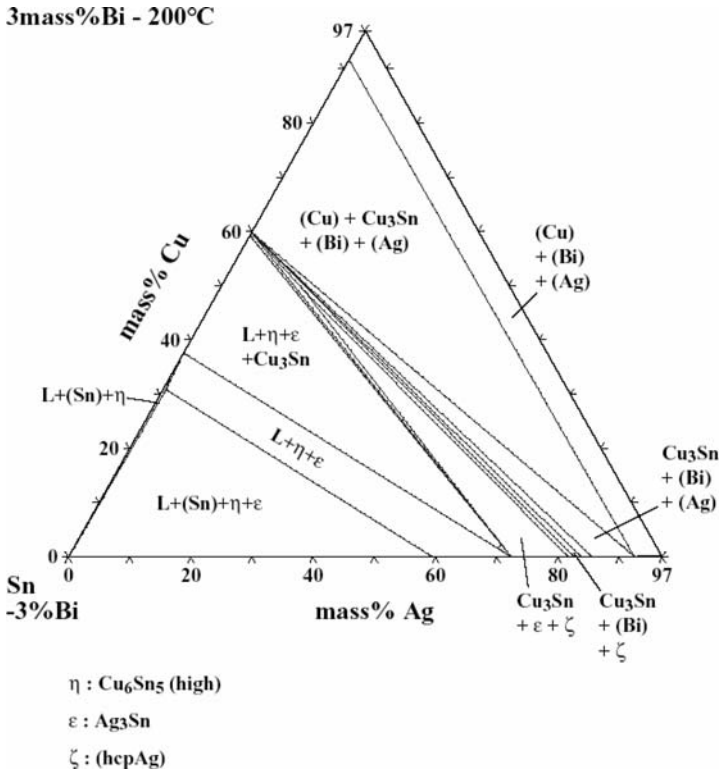

Fig. 13 Isothermal section of quaternary tin-based alloys at 3 mass% Bi at 200 °C

Bi, and above. Takao et al.[16] also suggested that there is a slight correlation between the fillet-lifting and the formation of a Bi concentration at the solder/Cu interface, which may occur as a consequence of the Sn consumption by the intermetallic compound formation with Cu dissolving into the molten solder. Dissolving Cu in Sn with formation of

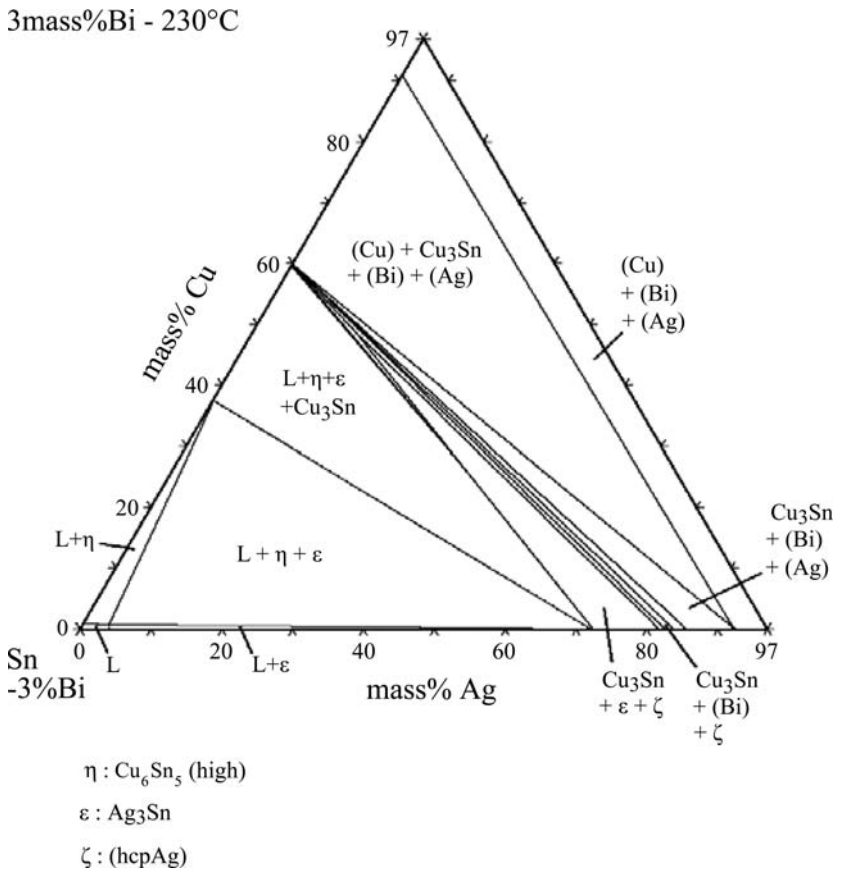

Fig. 14 Isothermal section of quaternary tin-based alloys at 3 mass% Bi at 230 °C

Cu_6Sn_5 may extend the suggestion of Takao et al.[16] from Sn-Ag-Bi to include the quaternary alloys discussed here, in Part 2. It was proved by simulation of solidification using Scheil's method that fillet-lifting tendency can be observed in Sn-Ag-Cu-Bi alloys with various Bi additions, contained in 3-20 mass% Bi (Fig. 6-7 and 10-11).

Because of the observed lifting-off failure tendency in Fig. 6-7 and 10-11, Hwang[15]made suggestions that indicated a quaternary Sn-Ag-Cu-Bi alloy with the composition 93.3Sn3.1Ag0.5Cu3.1Bi offered the best balance of the melting temperature, strength, plasticity, and fatigue life. This choice is in agreement with U.S. Patent No. 5 520 752. Simulation of solidification of this alloy, as presented in Fig. 12, still shows the tendency for lifting-off failure. The only difference in simulation of solidification of various quaternary alloys Sn-Ag-Cu-Bi in the Sn-rich part in equilibrium conditions is the starting temperature of solidification and the temperature at which the liquid phase disappears. In a Scheil's simulation of quaternary alloys, the terminating temperature is always 139.5 °C.

For industrial applications, the temperature of soldering should be 20-30 °C higher than the melting temperature of the solder. For the suggested composition Sn3.1Ag0.5Cu3.1Bi, isothermal calculations at 200 and 230 °C were made for 3 mass% Bi (Fig. 13 and 14). In agreement with the previous presentation on vertical sections (Fig. 4 and 8), the range of the liquid phase at 230 °C is visible in Fig. 14.

4. Discussion

In the current Part II and in the earlier Part I, various aspects of the influence of Bi additions to the near-ternary eutectic Sn-Ag-Cu alloys were studied. Beneficial influence of Bi additions on wettability and decreasing of melting temperature seems evident, but its suitable concentration is a compromise of various factors, especially in view of the tendency for lifting-off failure discussed in this work.

Butler's thermodynamic modeling is a useful approximation of the surface tension and offers preliminary information on the influence of the modifiers on the decrease or increase of wettability. Therefore, in all tested candidates for the substitution of the traditional Sn-Pb solders,[17] the authors have used this method, and it was also included to the SURDAT database[18] of the surface tension, density, and the molar volume of lead-free solder alloys.

The experimentally measured surface tension in the protective atmosphere can be treated only as the basic information on the wettability. For practical application, the meniscographic studies are most important because they supply information about the wetting force, wetting time, and contact angle [calculated combining the wetting force and the solder/flux interfacial tension measured in air by the Miyazaki method[19] (Ref 4)]. The contact angle and the solder/flux interfacial tension are very important because they allow the best flux and composition of the solder to be

found. In this respect, the conclusions are in agreement with those of Lopez et al.,[20] and our results of quinary alloys Sn-Ag-Cu-Bi-Sb.[21]

Because of the role and chemistry of the flux, the surface tension and the interfacial tension measured in air are lower than those measured in a protective atmosphere, but the tendency toward the change is very similar.

Applying the ADAMIS thermodynamic database enabled the modeling of the surface tension by Butler's model and the calculation of the various phase equilibria and the nonequilibrium solidification of the quaternary Sn-Ag-Cu-Bi alloys by Scheil's method. Analysis of the results showed that the advantageous influence of Bi on the melting temperature and the wettability properties of the Sn-Ag-Cu near eutectic alloys had to be strongly limited because the fillet-lifting failure observed at the solder/substrate interface (Bi precipitation) and the solder composition proposed by Hwang[15] (Sn3.1Ag0.5Cu3.1Bi in mass%) have shown the same tendency.

From the present study and the work of Hwang[15] and Takao et al.,[16] the conclusion can be drawn that the optimal concentration of Bi in the quaternary Sn-Ag-Cu-Bi solders should be located in the range of 1-3.1 mass%.

Appendix

Table 1 Binary and ternary interaction parameters in Eq 4

Ag-Bi	$W_{Ag,Bi} = [4589.8 + 23.73047T - 3.93814T\ln(T)] + (-5716.6 - 0.91452T)(X_{Ag} - X_{Bi}) + (-2630.2 + 0.88522T)(X_{Ag} - X_{Bi})^2$
Ag-Cu	$W_{Ag,Cu} = (17{,}534.6 - 4.45479T) + (2251.3 - 2.6733T)(X_{Ag} - X_{Cu}) + 492.7(X_{Ag} - X_{Cu})^2$
Ag-Sn	$W_{Ag,Sn} = (-4902.5 - 4.30532T) + (-16{,}474 + 3.12507T)(X_{Ag} - X_{Sn}) - 7298.6(X_{Ag} - X_{Sn})^2$
Bi-Cu	$W_{Bi,Cu} = (24{,}105.9 - 9.93287T) + (-2584.5 + 0.00906T)(X_{Bi} - X_{Cu})$
Bi-Sn	$W_{Bi,Sn} = -32 - 0.235T$
Cu-Sn	$W_{Cu,Sn} = (-9002.8 - 5.8381T) + (-20{,}100.4 + 3.6366T)(X_{Cu} - X_{Sn}) - 10{,}528.4(X_{Cu} - X_{Sn})^2$
Ag-Bi-Sn	$W_{Ag,Bi,Sn} = (11{,}000 + 4T)X_{Ag} + (20{,}000 - 38.95T)X_{Bi}$
Ag-Cu-Sn	$W_{Ag,Cu,Sn} = -83{,}524.551X_{Ag} - 99{,}690.536X_{Cu} + 29{,}237.648X_{Sn}$

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