Sn-Ni-Bi liquid phase thermodynamic properties

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Abstract: Experimental data of bismuth activity coefficients at 1773 K were obtained by isopiestic method and compared to calculated values. Thermodynamic properties of the Sn–Ni–Bi liquid phase were estimated by means of the general solution model and by the methods of Kohler. Description of the ternary liquid phase (Gibbs excess energy dependence on the temperature and the composition) was achieved by using available thermodynamic data of the constitutive binary systems (Ni–Bi, Sn–Bi, Sn–Ni). A comparison between calculated quantities and experimental data was conducted. The present assessment with thermodynamically optimized values of the system Sn–Ni–Bi (obtained by the CALPHAD approach) was in good agreement. The suggested appearance of a liquid phase miscibility gap at high temperatures is in agreement with the experimental bismuth activity data and with the assessed thermochemical functions.

Keywords: Thermodynamics • General solution model • Ternary interaction parameters • Bi–Ni–Sn system • Bismuth activity coefficients

1. Introduction

The joining of modules in the electronics industry is accomplished mostly by soldering technology. Thus the development of lead-free solder materials is a current challenge because of lead's toxicity. The replacement of Sn-Pb based solders is a very difficult task due to technological and materials’ properties challenges [1,2]. Therefore, fundamental research in regard to the development of the soldering processes, for example, the diffusion paths in the contact zone, is indispensable for modeling the phase transformations in solder/substrate systems.

Nickel is often used for metallization in electronic devices and in general is one of the metals for future solder alloys. The soldering is done in liquid phase, thus the knowledge of the alloy's thermochemical properties (e.g. wetting etc.) is noteworthy.

There is a common agreement that prospective solder materials may be based on Sn alloyed with Zn or/and Bi [3-5]. Oppositely, nickel metallization often appears as intrinsic part of the electronic devices. Thus, investigations on the Bi-Ni-Sn system have been reported [6-14]. These elements (Ni, Sn, Bi) and the respective binary phase diagrams were included in the thermodynamic database developed by the European concerted action COST 531, and reliable thermodynamic optimizations are available [15]. Nevertheless, the thermochemical properties of the ternary system (especially in liquid phase) are still quite uncertain in view of the possible existence of a miscibility gap [16]. The purpose of this work was to predict the liquid phase thermochemical properties in various ways and to compare them with new experimental data.

2. Computational Details

Hilert [17] has classified the geometric models as “symmetrical” and “asymmetrical”. More recently, discussions on the development and the applicability of the geometric models were published by Pelton et al. [18,19]. It was suggested [18] that when (in a ternary system) one of the components is strongly chemically different from the others, an asymmetric model is preferable. Also, preferences might be given to the combinations of methods e.g. the model of Kohler [20] or Toop [21] and the general solution model (GSM) developed by Chou [22,23]. However, there are no commonly accepted selection criteria for the models.

Assessments of the present work were conducted by means of the classic geometric models of Kohler [15] (belonging to the so called “symmetrical” models, [17]) and the general solution model [22,23]. The latter

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has already been proven, and details can be found in [24-27]. Nonetheless, a concise explanation of both models is depicted below. The molar excess Gibbs energy ($\Delta G^E$, J mol$^{-1}$) of the ternary liquid phase and the thermodynamic activity were selected as quantities whose values were calculated by various models and compared.

An assessment done by geometric models needs reliable values of the molar excess Gibbs energies for every binary system. These quantities were calculated by means of the Thermocalc software package, using optimized parameters for the binary Bi-Sn, Bi-Ni and Ni-Sn systems which have become recently available by the European concerted action COST 531 for development of lead-free solders thermodynamic database [15]. Meanwhile, new experimental data (liquid phase mixing enthalpy) were reported by Schmeterer et al. [28]. Thus the system Ni-Sn was optimized based on the new data [29].

In agreement with Redlich-Kister formalism [30], the composition dependence of the binary Gibbs molar excess energies ($\Delta G^E_{ij}$) is given by the following expression:

$$\Delta G^E_{ij} = X_i X_j (A^0_{ij} + A^1_{ij} X_i - X_j) + A^2_{ij} (X_i - X_j)^2 + \ldots + A^n_{ij} (X_i - X_j)^n$$  \hspace{1cm} (1)

Here, $A^0_{ij}, A^1_{ij}, A^2_{ij}$ are adjustable temperature dependent parameters belonging to the binary system "ij"; $X_i$ and $X_j$ indicate the mole fractions of the corresponding constituents.

The Gibbs molar excess energy of a ternary phase ($\Delta G^E_{123}$), consisting of the elements 1, 2 and 3, is given by the following expression:

$$\Delta G^E_{123} = X_1 X_2 \Delta G^E_{12} + X_2 X_3 \Delta G^E_{23} + X_3 X_1 \Delta G^E_{31} + \Delta G^E_{123}$$  \hspace{1cm} (2)

where $\Delta G^E_{12}$, $\Delta G^E_{23}$ and $\Delta G^E_{31}$ can be obtained as defined above (Eq. 1) and $\Delta G^E_{123}$ is the contribution of the ternary non-ideal mixing. In the simplest case of a regular ternary solution it may be assessed by means of Eq. 3:

$$\Delta G^E_{123} = X_1 X_2 X_3 f_{123}$$  \hspace{1cm} (3)

where $A_{123}$ is a ternary interaction parameter that might be temperature and concentration dependent.

The essential equations, associated with the a.m. geometrical models [18–21] are only shown below because a detailed presentation would reach beyond the scope of this work. Hence, in general a ternary system with constituents 1, 2 and 3 is considered where $X_1$, $X_2$ and $X_3$ are the mole fractions of the respective components. The molar excess Gibbs energies of the ternary liquid phase (at a specific composition, temperature and pressure), are estimated by means of Eq. 4 according to Kohler [20]:

$$\Delta G^E_{123} = \Delta G^E_{12}(1-x_3)^2 + \Delta G^E_{23}(1-x_1)^2 + \Delta G^E_{31}(1-x_2)^2$$  \hspace{1cm} (4)

Here $\Delta G^E_{123}$ is the molar Gibbs excess energy of the ternary liquid phase; $\Delta G^E_{ij}$ – the respective values for the two-component liquid phases, where the down indexes indicate the considered binary system.

The optimized adjustable parameters of a considered phase (i.e., liquid, in this case) were used to calculate the components' thermodynamic activities (respectively the activity coefficients) by the Thermocalc software package. $\Delta G^E_{123}$ is related to the activity coefficients by the following expression:

$$\Delta G^E_{123} = x_1 RT \ln \gamma_1 + x_2 RT \ln \gamma_2 + x_3 RT \ln \gamma_3$$  \hspace{1cm} (5)

where $x_1, x_2, x_3$ are the mole fractions of the respective component; $\gamma_1$, $\gamma_2$, $\gamma_3$, the activity coefficients, $R$, the gas constant and $T$, temperature, K.

Eqs. 6–8 shall be used for a brief introduction to the general solution model (GSM) of Chou [22,23]:

$$\Delta G^E_{123} = X_1 X_2 X_3 f_{123}$$  \hspace{1cm} (6)

Here $f_{123}$ is the ternary interaction coefficient, related to the Redlich-Kister ternary interaction parameters $A_{jk}$ (\(f_{123} = X_1 A^0_{123} + X_2 A^1_{123} + X_3 A^2_{123}\)). This parameter can be assessed as follows [22]:

$$f_{123} = (2\xi_{12} - 1)(A^2_{12} - (2\xi_{12} - 1)X_3 + 2(X_1 - X_2)) + \frac{A^1_{12}}{A^2_{12}} + (2\xi_{23} - 1)(A^2_{23} - (2\xi_{23} - 1)X_1 + 2(X_2 - X_3)) + \frac{A^1_{23}}{A^2_{23}} + (2\xi_{31} - 1)(A^2_{31} - (2\xi_{31} - 1)X_2 + 2(X_3 - X_1)) + A^1_{31}$$  \hspace{1cm} (7)

Here $\xi_{ij}$ are “similarity coefficients”, that are defined by the quantity $\eta_i$ called “deviation sum of squares”:

$$\xi_{ij} = \eta_i \sqrt{\eta_i + \eta_j}$$  \hspace{1cm} (8)

According to this model (GSM), the values of $\eta_i$ are explicit functions of $\Delta G^E_{ij}$, and are easily calculated when the adjustable coefficients of the three binary end-systems are known.

Basic thermodynamic information on the constitutive subsystems which were needed for the assessment,
was taken from [15]. Updated parameters were used [29] for the Ni-Sn liquid phase. The optimized parameters of each system (Table 1) are needed. They are used for the calculation (Eqs. 1-8) of the molar excess Gibbs energies of the binary end-systems liquid phases. The following sequence of components was adopted: Bi - 1, Ni - 2 and Sn - component 3.

3. Experimental Procedure

The use of vapor-pressure methods has lately been reviewed by Ipser [31]. The present work effectively applied such techniques [32-37]. The experiments were carried out by an equilibrium saturation method, i.e., a modified isothermal isopiestic technique using a set of graphite cells placed in an induction furnace (Fig. 1). The main part of the apparatus is an alundum crucible where graphite containers are embedded.

Half of these cells contained the studied Bi-Ni-Sn alloy while the other half made reference to Bi-Cu alloys. During the experiment the system had to gain a state of equilibrium (concerning the chemical potential of bismuth), i.e., equal bismuth activities in all alloys. All of them were in a liquid state at the working temperature of 1773 K. As the bismuth vapor pressure ($p_{\text{Bi}}^\ast$) is much higher than that of the other elements (at T=1773 K: $p_{\text{Sn}}^\ast = 0.32$ Pa, $p_{\text{Ni}}^\ast = 4.7$ Pa, $p_{\text{Bi}}^\ast = 32800$ Pa [38]), pure bismuth vapor was the dominant constituent of the gaseous phase. And so, the equilibrium was achieved through bismuth redistribution through the gaseous phase, common for all samples.

Before starting the saturation experiments, Sn-Bi-Ni and Cu-Bi alloys of appropriate compositions were prepared by mixing and annealing weighed masses of metals (in quartz ampoules). The amounts of copper, tin and nickel did not change during the synthesis, but a certain amount of bismuth evaporated. Therefore, its contents were expected to be lower at the end of each experiment and the alloy compositions had to be verified. This was completed by weighing and by the glow discharge spectroscopic method which made use of the Glow Discharge Spectrometer Horiba Yobin Yown.

A preliminary test at 1773 K found that the time necessary to reach equilibrium was 2 hours with argon pressure maintained in the apparatus at 20 kPa. Such a high pressure was necessary to limit the evaporation process but to keep the bismuth vapour pressure sufficiently high without boiling. In these tests Bi-Cu alloys (purity of copper 99.9995 and purity of bismuth 99.999 mass per cent) of various compositions (Table 2) were used. In order to establish the required time to achieve equilibrium, alloys were saturated with bismuth vapour for 0.5, 1, 1.5, 2 and 2.5 hours. After the saturation, argon was introduced to the apparatus,

![Figure 1. Schematic diagram of the apparatus used for the activity measurements: 1 – thermocouple, 2 – graphite cover, 3 – graphite blocks with groves for alloys, 4 – vacuum chamber.](image)
and the bismuth content was determined in each alloy at room temperature. This was accomplished by weighing and glow discharge spectroscopic methods. The results are presented in Table 2. The equilibrium was considered attained when the bismuth mole fractions in all alloys became equal. It was determined that the required time to reach equilibrium in the test alloys was 1.5 hours. Nevertheless, for the equilibrium saturation measurements the Sn-Bi-Ni samples were saturated for two hours.

In a state of equilibrium, the bismuth activities \(a_{\text{Bi}}\) in the ternary liquid alloys since the bismuth activity coefficients \(\gamma_{\text{Bi}}\) in the reference Bi-Cu alloys (as well as their compositions) are known (Eq. 10):

\[
\begin{align*}
\gamma_{\text{Bi}} & = \frac{x_{\text{Bi}}^{\text{Bi-Cu}}}{x_{\text{Bi}}^{\text{Bi-Ni-Sn}}} \\
\gamma_{\text{Bi}} & = \frac{x_{\text{Bi}}^{\text{Bi-Cu}}}{x_{\text{Bi}}^{\text{Bi-Ni-Sn}}} \\
\end{align*}
\]

Here \(x_{\text{Bi}}^{\text{Bi-Cu}}\) and \(x_{\text{Bi}}^{\text{Bi-Ni-Sn}}\) denote the experimentally determined equilibrium mole fractions of Bi in Bi-Cu and Bi-Ni-Sn, respectively.

In order to obtain \(\gamma_{\text{Bi}}\), from the experimental data through equation (10) (Table 3) it is critical to know the exact compositions and the bismuth activity coefficients of the equilibrated alloys and \(\gamma_{\text{Bi}}\). This was made possible due to available thermodynamic optimization of the Bi-Cu system. The latter has been calculated using the optimised coefficients of Teppo et al. [39]. We would like to point out that the above-cited optimisation of the Cu-Bi system has been retained for the thermodynamic database of the European action for development of lead-free solders COST 531 [15].

MOOB grade (oxygen free) copper rolls (99.99 mass%) as well as nickel, tin and bismuth of purity 99.9 mass% have been used for all the synthesis experiments. The temperature has been measured by a \{Pt-(Pt+Rh)\} thermocouple and the pressure – by means of Balzers vacuum-meter APG-010.
The accuracy of the pressure and temperature measurements was ± 10 Pa and ± 5 K, respectively. The estimated accuracy of the measured bismuth activity coefficients was approximately 2%. The graphite container (outer part of the apparatus) is made of EK-412 type graphite produced by Ringsdorf (ash residue less than 0.1 mass %). No contact was allowed between the alloys and the graphite so that a carbon contamination of the samples would not occur.

4. Results and Discussion

Coefficients \( f_{123} \) calculations were done along five sections of the Bi-Ni-Sn system with molar Sn/Bi ratios of 1/3, 1/1, 3/1, 1/25 and 25/1 within an interval of 1000 – 2000 K, as well as (for every section) molar fractions of nickel equal to 0, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9 and 1. A matrix was created to derive the parameters \( A_{ijk} \). The results are shown in Table 4. These parameters will be useful for the future thermodynamic optimization of this ternary system.

The binary integral Gibbs excess energies should be initially obtained as previously mentioned. This is accomplished by the Thermocalc software package and by calculating the binary activity values (referred to the liquid phases of the pure elements) at specific points. The ternary integral molar Gibbs excess energies assessed by a.m. geometrical methods are graphically compared to those obtained by Thermocalc, using the optimized adjustable parameters of the end-systems only (Table 1). Sections with constant molar ratios between two of the components are plotted.

Results for a section with constant molar Bi/Sn equal to 1/3, at 1700 K are plotted in Fig. 2. It can be determined that the ternary molar integral Gibbs excess energies calculated with the binary parameters of the thermodynamic database COST 531 [15] (curve 1) deviate from those calculated by GSM (curve 2), by the updated parameters [29] (curve 3) and by the assessment using the Kohler method. The updated

### Table 3
Comparison at \( T = 1773 \) K between experimental Bi activity coefficients \( (\gamma_{Bi}^{Bi-Ni-Sn}) \) and activities \( (a_{Bi}^{Bi-Ni-Sn}) \) from one side, and calculated activity coefficients (using binary end-systems parameters only) of Bi-Ni-Sn by Seo et al. [11], by COST MP 0602 thermodynamic database [29] and by GSM (assessed in this work). The reference state for the activity values is the liquid phase.

| N | \( x_{Ni} \) | \( x_{Sn} \) | \( x_{Bi} \) | \( \gamma_{Bi}^{Bi-Ni-Sn} \) | \( a_{Bi}^{Bi-Ni-Sn} \) | \( \gamma_{Bi}^{[11]} \) | \( \gamma_{Bi}^{[29]} \) | \( \gamma_{Bi}^{[GSM]} \) |
|---|---|---|---|---|---|---|---|---|
| 1 | 0.0181 | 0.9265 | 0.0554 | 1.048 | 0.058 | 1.227 | 1.245 | 1.264 |
| 2 | 0.0474 | 0.9006 | 0.0520 | 1.117 | 0.058 | 1.231 | 1.308 | 1.327 |
| 3 | 0.0956 | 0.8591 | 0.0453 | 1.282 | 0.058 | 1.258 | 1.435 | 1.479 |
| 4 | 0.1442 | 0.6178 | 0.0380 | 1.529 | 0.058 | 1.342 | 1.605 | 1.684 |
| 5 | 0.1930 | 0.7731 | 0.0339 | 1.716 | 0.058 | 1.475 | 1.829 | 1.976 |
| 6 | 0.2427 | 0.7281 | 0.0292 | 1.860 | 0.054 | 1.712 | 2.158 | 2.363 |
| 7 | 0.2928 | 0.6837 | 0.0235 | 2.316 | 0.054 | 2.043 | 2.553 | 2.851 |
| 8 | 0.3430 | 0.6371 | 0.0199 | 2.733 | 0.054 | 2.462 | 3.065 | 3.417 |
| 9 | 0.3919 | 0.5917 | 0.0164 | 3.304 | 0.054 | 2.988 | 3.720 | 4.146 |
| 10 | 0.4444 | 0.5413 | 0.0143 | 3.809 | 0.054 | 3.706 | 4.476 | 5.035 |
| 11 | 0.4817 | 0.5043 | 0.0140 | 2.123 | 0.030 | 4.286 | 5.071 | 5.714 |
| 12 | 0.5406 | 0.4446 | 0.0148 | 2.007 | 0.030 | 5.405 | 6.081 | 6.824 |
| 13 | 0.5978 | 0.3852 | 0.0170 | 1.747 | 0.030 | 6.529 | 7.000 | 7.882 |
| 14 | 0.6457 | 0.3495 | 0.0048 | 6.241 | 0.030 | 7.917 | 8.333 | 9.375 |
| 15 | 0.6887 | 0.2968 | 0.0145 | 2.040 | 0.030 | 8.483 | 8.621 | 9.517 |
| 16 | 0.7442 | 0.2384 | 0.0174 | 2.319 | 0.040 | 9.138 | 9.023 | 9.885 |
| 17 | 0.7953 | 0.2002 | 0.0045 | 8.977 | 0.040 | 10.667 | 10.222 | 11.111 |
| 18 | 0.8336 | 0.1494 | 0.0170 | 2.375 | 0.040 | 9.471 | 9.235 | 9.824 |
| 19 | 0.8941 | 0.0963 | 0.0096 | 4.200 | 0.040 | 9.479 | 9.375 | 9.792 |
| 20 | 0.9352 | 0.0527 | 0.0121 | 3.337 | 0.040 | 8.347 | 8.512 | 8.760 |

### Table 4
Ternary interaction parameters, \( A_{ijk} \), for the Bi–Ni–Sn liquid phase obtained by GSM (from the \( f_{123} \) coefficients). T – temperature.

| System, ijk | \( A_{100}^{i}(T) \) J mol\(^{-1}\) | \( A_{010}^{i}(T) \) J mol\(^{-1}\) | \( A_{001}^{i}(T) \) J mol\(^{-1}\) |
|---|---|---|---|
| Bi-Ni-Sn | +16494.00-5.62*T | +15686-4.273*T | +15961.64-4.818*T |

The accuracy of the pressure and temperature measurements was ± 10 Pa and ± 5 K, respectively. The estimated accuracy of the measured bismuth activity coefficients was approximately 2%. The graphite container (outer part of the apparatus) is made of EK-412 type graphite produced by Ringsdorf (ash residue less than 0.1 mass %). No contact was allowed between the alloys and the graphite so that a carbon contamination of the samples would not occur.

Results for a section with constant molar Bi/Sn equal to 1/3, at 1700 K are plotted in Fig. 2. It can be determined that the ternary molar integral Gibbs excess energies calculated with the binary parameters of the thermodynamic database COST 531 [15] (curves 1, 2) deviate from those calculated by GSM (curve 3) and by the assessment using the Kohler method. The updated
Ni-Sn liquid phase parameters [29] were used to calculate the $\Delta G^{E}_{r23}$ values, represented by curves 2 – 4. Small positive values of the ternary molar integral Gibbs excess energy were observed close to the Sn-Bi region. The shapes of the curves at sections with constant molar Bi/Sn equal to 1 were similar.

Fig. 3 represents ternary integral molar Gibbs excess energies assessed along a molar Bi/Sn ratio equal to 3/1, at 1700 K. In this case, the $\Delta G^{E}_{r23}$ values were positive in the Bi-rich region (up to around 15 at. % Ni) and in the Ni-rich melts. The trend toward positive deviations from the Raoult’s law rises with the increase
in the Bi-content and the decrease in temperature (see Fig. 4, T=1500 K, Bi/Sn ratio equal to 25/1).

Comparison at T=1773 K, between experimental bismuth activity coefficients ($\gamma_{\text{Bi}}^{\text{Bi-Ni-Sn}}$) and activities values ($a_{\text{Bi}}^{\text{Bi-Ni-Sn}}$) and calculated ones (using binary end-systems parameters only) by Seo et al. [11], by COST MP 0602 thermodynamic database [29] and by GSM (assessed in this work) is shown in Table 3. It is evident that the experimental bismuth activity coefficients have values between 1 and 11, thus indicating positive deviations from the Roult’s law of the ternary liquid phase. Consequently, a liquid phase miscibility gap is expected in the Bi-Ni-Sn system. These suggestions are backed up by the calculations of isothermal sections done by the software packages either Thermocalc (Fig. 5a) either Pandat (Fig. 5b).

5. Conclusions

Bismuth activity coefficients of the ternary liquid phase were measured at 1773 K indicating positive deviations from the Roult’s law of the ternary liquid phase. Some thermodynamic properties (molar Gibbs excess energies and Bi activity coefficients) of the Bi–Ni–Sn liquid phase were predicted using the general solution model developed by Chou. The respective ternary interaction parameters were assessed.

The values of the ternary liquid phase molar Gibbs excess energies assessed by the general solution model have been compared to the ones calculated by the CALPHAD method using binary optimized parameters. These were in agreement indicating that the interaction parameters for the ternary system would have weak values.

The calculated parameters indicate the existence of a liquid phase miscibility gap. The experimental Bi activity coefficients of the liquid phase also gives evidence of strong positive deviations from the ideal solutions behaviour.

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