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# A NEW THERMODYNAMIC DESCRIPTION OF THE BINARY Sb – Zn SYSTEM

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***Abstract***

*The Sb – Zn alloys are important from technological point of view. Antimony – zinc alloys are not only part of lead-free solders but also thermoelectric materials. A good thermodynamic description of this system is essential for further investigations of multi-components materials that include Sb and Zn. A new assessment was proposed in this work based on recently published experimental reports. Good agreement between calculations and experiments was found. Moreover, two models were used for describing the liquid phase: substitutional solution and associated solution.*

*Keywords: CALPHAD; lead-free solder; thermoelectric materials;*

## 1. Introduction

The Sb – Zn alloys are in a big interest of material science community. Antimony and zinc are components very often used in lead-free soldering technology and the ternary Sb – Sn – Zn system was proposed as a lead-free solder material [1]. Moreover, the intermetallic compounds (IMCs) that can be found in the binary antimony – zinc system show thermoelectric properties. It is worth to mention, that Sb – Zn alloys were reported in the original Seebeck's work on thermoelectricity [2]. Furthermore, in 1997 Caillat et al. [3] reported very high figure of merit for the $Sb_3Zn_4$. It is obvious, that the knowledge of phase relationship as well as thermodynamic description will play a key-role in the future works on Me – Sb – Zn thermoelectric materials. Available descriptions of the Sb – Zn system show some conflicts about temperatures and kinds of phase transformations. Recently, Adjadj et al. [4, 5] published very detailed X-ray and differential scanning calorimetry (DSC) study on the antimony - zinc system. Based on their report a new thermodynamic description of this binary is proposed in this work.

## 2. Literature review

Thermodynamic properties of the liquid phase are described quite well in the literature. Enthalpy of mixing was measured in whole range of Zn concentrations by Rubin et al. [6] at 823 K and 973 K, Scheil and Lukas [7] at 843 K, Wittig and Gehring [8] at 913 K and Wittig et al. [9] at 923 K. Hultgren et al. [10] reported selected values of mixing enthalpy at 850 K. The experimental data agree each other except Rubin and Komarek's [6] work, which does not match temperature dependency shown by other data. Several authors determined activity of both components in a liquid phase. The electromotive force (EMF) measurement was used by Ptak and Zabdyr [11] for description of Zn activity in a temperature range 830 – 950 K. The same method was used by Seltz and De Witt [12] at 823 K, Gerling and Predel [13] at 860 – 1078 K, Kozuka et al. [14] at 823 – 923 K, Kleppa and Thalmayer [15] at 850 K and Rubin and Komarek [6] at 823 K. Jelinek and Wannow [16] applied vapor pressure measurement for Zn activity determination at 1058 K. Hultgren et al. [10] reported selected values of Sb an Zn activities at 850 K. Thermodynamic properties of the solid phases were determined by EMF method, by vapor pressure measurement and by calorimetric measurement. Eremenko and Lukashenko [17], Goncharuk and Lukashenko [18], Zabdyr [19] as well as Goryacheva and Geiderikch [20] carried out the EMF measurement. Vapor pressure method was applied by Nesmeyanov et al. [21] and Hirayama [22]. Shchukarev et al. [23] and Oelsen and Middel [24] used the calorimetric measurement for determination formation enthalpy of the intermetallic compounds in the Sb – Zn system.

The liquidus line of the binary Sb – Zn system was described by Monkmeyer [25], Zemczuzny [26], Curry [27], Gosselin [28] and Heycock and Neville [29] who used differential thermal analysis (DTA) technique. Liu et al. [30] reported a part of liquidus line determined by DSC measurement. Izard et al.

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*The work is dedicated to Professor Krzysztof Fitzner for his 70th birthday.*

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[31] described liquidus line and temperatures of phase transformations. They also discussed stability of intermediate phases with the aid of XRD experiment. Adjadj et al. [5] used the DSC technique for liquidus and phase transformation temperatures investigation. Takei [32] used resistometric as well as DTA measurement techniques to describe a liquidus line.

There are three intermediate phases in this binary system: SbZn, $Sb_3Zn_4$ and $Sb_2Zn_3$. According to recent examination by Adjadj et al. [5], phases $Sb_3Zn_4$ and $Sb_2Zn_3$ decompose by congruent melting at 839 K and 841 K, respectively. The phase SbZn decomposes by peritectic reaction Liquid + $Sb_3Zn_4$ (β) = SbZn at 779 K. Adjadj et al. [4] reported that $Sb_3Zn_4$ poses 3 forms: β, γ and γ'. The polymorphic transformation (βÛγ) occurs at 766 K and the order – disorder transformation (γÛγ') between 795 K and 802 K. The phase $Sb_2Zn_3$ exists in two modifications: low temperature ζ and high temperature η. The polymorphic transformation occurs at temperature range 437 – 460 K. The phase $Sb_2Zn_3$ breaks up, when cooling, towards 633 K in agreement with reaction 3 $Sb_2Zn_3$ (ζ) = 2 $Sb_3Zn_4$ (β) + (Zn). The intermediate phases were also investigated by Takei [32] and by Vuillard and Piton [33] who suggested existence of $Sb_5Zn_6$ phase. Adjadi et al. [4] and Mayer et al. [34], who used the XRD technique, did not confirm the $Sb_5Zn_6$ phase. Koneska and Mavrodiev [35] results agree with Vuillard and Piton [33] except existence of $Sb_5Zn_6$ phase. Stabilities of the intermediate phases were determined by Izard et al. [31]. Recently, Adjadj et al. [4] presented analysis of X-ray diffraction results obtained from examination of SbZn, $Sb_3Zn_4$ and $Sb_2Zn_3$ phases. Detailed information about solid phases in the binary Sb – Zn system is gathered in Table 1. The system was thermodynamically modeled by Zabdyr [36], Liu et al. [30] and Li et al. [37]. Zabdyr [36] proposed two different descriptions of the liquid phase: substitutional solution and associated liquid. The first description reproduced thermodynamic function well, the second one exhibits strange shape of mixing enthalpy at low (843 K) temperature. Besides that, in Zabdyr's [36] work the order-disorder transformation of $Sb_3Zn_4$ was omitted. Liu et al. [30] and Li et al. [37] used substitutional solution model for describing liquid phase; however their description is less complicated than proposed by Zabdyr [36]. The consequence of this description is awkward shape of calculated mixing enthalpy that exhibits additional extreme point for high concentration of antimony. The thermodynamic description of the Sb-Zn system was also included into the thermodynamic database COST MP0602 for materials for high-temperature lead-free soldering [38].

## 3. Thermodynamic models

This modeling of the binary Sb-Zn system includes 8 phases: Rhombohedral_A7 (Sb), HCP_ZN (Zn), $Sb_3Zn_4$(β), $Sb_3Zn_4$(γ), $Sb_3Zn_4$(γ'), $Sb_2Zn_3$(ζ), $Sb_2Zn_3$(η) and liquid. The description of the crystal structures is given in Table 1.

***Table 1.*** *Sb – Zn crystal structure data [48].*

| Phase | Pearson symbol | Space group | Strukturbericht designation | Prototype |
|---|---|---|---|---|
| (Sb) | hR2 | | A7 | αAs |
| SbZn | oP16 | *Pbca* | $B_e$ | CdSb |
| $Sb_3Zn_4$(β) | … | … | … | … |
| $Sb_3Zn_4$(γ) | … | … | … | … |
| $Sb_3Zn_4$(γ') | oP28 | *Pmmn* | … | … |
| $Sb_2Zn_3$(ζ) | oI | … | … | … |
| $Sb_2Zn_3$(η) | oP32 | *Pmmn* | … | … |
| (Zn) | hP2 | *$P6_3/mmc$* | A3 | Mg |

The Gibbs energies of pure elements with respect to temperature ${}^0G_i(T) = G_i(T) - H_i^{SER}$ are represented by Equation 1:

$${}^0G_i(T) = a + bT + cT\ln(T) + dT^2 + eT^{-1} + fT^3 + iT^4 + jT^7 + kT^{-9} \quad (1)$$

The ${}^0G_i(T)$ data are referred to the constant enthalpy value of the standard element reference $H_i^{SER}$ at 298.15 K and 1 bar as recommended by Scientific Group Thermodata Europe (SGTE) [39]. The reference states are Rhombohedral_A7 (Sb) and HCP_ZN (Zn). The ${}^0G_i(T)$ expression is given for several temperature ranges, where the coefficients *a, b, c, d, e, f, i, j, k* have different values. The ${}^0G_i(T)$ functions are taken from SGTE Unary (Pure elements) TDB v5.0 [39].

The solid Sb and Zn were treated as pure components because neither there is information about solubility of Sb in solid zinc nor about solubility of Zn in solid antimony. In this case, the Gibbs energies of Sb and Zn were directly taken from the SGTE Unary database [39].

Intermetallic compounds $Sb_3Zn_4$(β), $Sb_3Zn_4$(γ) and $Sb_3Zn_4$(γ'), $Sb_2Zn_3$(ζ), $Sb_2Zn_3$(η):

The IMCs were treated as the line compounds because the homogeneity range of them is negligible. In this case, the Gibbs energy is described as follows:

$$G^0_{Sb_iZn_j} = a + bT + i \cdot GHSERSB + j \cdot GHSERZN \quad (2)$$

where GHSERSB and GHSERZN are Gibbs energies of antimony and zinc in Rhombohedral_A7 and HCP_ZN structures, respectively.

### *3.1 Substitutional solution*

Liquid phase was described by the substitional solution model [40]:

$$G_m(T) = \sum_i x_i {}^0G_i(T) + RT\sum_i x_i \ln(x_i) + \sum_i \sum_{j>i} x_i x_j \left( \sum_v {}^vL_{ij}(x_i - x_j)^v \right) \qquad (3)$$

where the $\sum_i \sum_{j>i} x_i x_j \left( \sum_v {}^vL_{ij}(x_i - x_j)^v \right)$ part is the Redlich-Kister polynomial for excess Gibbs free energy.

### *3.2 Associated solution*

The liquid phase was also described by associated solution model [41]. The liquid associate was supposed to be $Sb_3Zn_4$. The Gibbs energy of one mole liquid is given by equation (4):

$$G_m(T) = \sum_i y_i {}^0G_i(T) + RT\left( \sum_i y_i \ln y_i \right) + \sum_i \sum_{j>i} \left( \sum_v {}^vL(y_i - y_j)^v \right)$$

where $y_i$ denotes a site fraction, *i, j* denote constituents of the liquid phase: Sb, Zn and liquid associate $Sb_3Zn_4$.

### *3.3 Optimization procedure*

The thermodynamic parameters for all phases in the system were optimized using ThermoCalc software [42]. For this optimization, thermodynamic data for the liquid phase, invariant reactions and phase equilibrium data were used. To each piece of the selected information was given a certain weight based on experiment accuracy. The optimization was carried out step by step in agreement with Schmid-Fetzer's et al. [43] guideline. First, the optimization of the liquid phase was performed, and then the solid phases were assessed. All parameters were finally evaluated together to provide the best description of the system. The calculated interaction parameters are shown in Table 2. For checking the results of optimization, the system was also calculated using Pandat [44] software. The last step of modeling was to change a description of the liquid model. During this step description of solid phases was kept unchanged. After optimization the results were checked again in Pandat [44] software.

## 4. Results and discussion

Figure 1 shows calculated phase diagram of the binary Sb – Zn system superimposed with experimental data given by Adjadj et al. [5], Liu et al. [31] and Takei [33]. The calculated liquidus line agrees well with experimental data.

It can be seen, that data given by Liu et al. [31] and Takei [33] shows discrepancy at the composition range 0.6 – 0.8 mole fraction of Zinc; however, the calculated line is located between these 2 datasets. Zoomed part of the phase diagram for composition range from 0.45 to 0.65 mole fraction of zinc is shown in Figure 2 together with DSC data obtained by Adjadj et al. [5]. One can see, that all the phase transformation temperatures agree well with Adjadj et al. [5] data. Calculated order-disorder transformation occurs 2 degree lower than it was revealed by experiment.

***Table 2.*** *Calculated nteraction parameters.*

| Phase | Parameters |
|---|---|
| SbZn | ${}^mG$ = -21827.976+7.9927+GHSERSB+GHSERZN |
| $Sb_3Zn_4(\beta)$ | ${}^mG$ = -82174.639+32.8691+3GHSERSB+4GHSERZN |
| $Sb_3Zn_4(\gamma)$ | ${}^mG$ = -81723.458+32.2801+3GHSERSB+4GHSERZN |
| $Sb_3Zn_4(\gamma')$ | ${}^mG$ = -361795.319-236.8462+30GHSERSB+39GHSERZN |
| $Sb_2Zn_3(\zeta)$ | ${}^mG$ = -397804.613-1.1087+20GHSERSB+31GHSERZN |
| $Sb_2Zn_3(\eta)$ | ${}^mG$ = -37109.540-3.6646+2GHSERSB+3GHSERZN |
| Liquid substitional | ${}^0L$ = -47736.194+326.5303T-42.2936TlnT |
| | ${}^1L$ = -808.225+0.7409T+0.3242TlnT |
| | ${}^2L$ = 25540.912-17.6368T |
| | ${}^3L$ = -12308.192+6.1383T |
| | ${}^4L$ = -6050.661 |
| | ${}^5L$ = -393.624 |
| Liquid associated | ${}^LG^{Sb_3Zn_4}_{Sb:Zn}$ = -109299.271+494.7556T+17.8590TlnT +3GHSERSB+4GHSERZN |
| | ${}^0L_{Sb,Sb_3Zn_4}$ = -208903.328+2503.8572T-441.8588TlnT |
| | ${}^1L_{Sb,Sb_3Zn_4}$ = 216478.405-1917.5183T+289.6028TlnT |
| | ${}^2L_{Sb,Sb_3Zn_4}$ = -46646.4274-114.7774T |
| | ${}^0L_{Sb_3Zn_4,Zn}$ = -192800.132+166.0501T-85.5694TlnT |
| | ${}^1L_{Sb_3Zn_4,Zn}$ = -293498.096+3.3584T+36.0324TlnT |

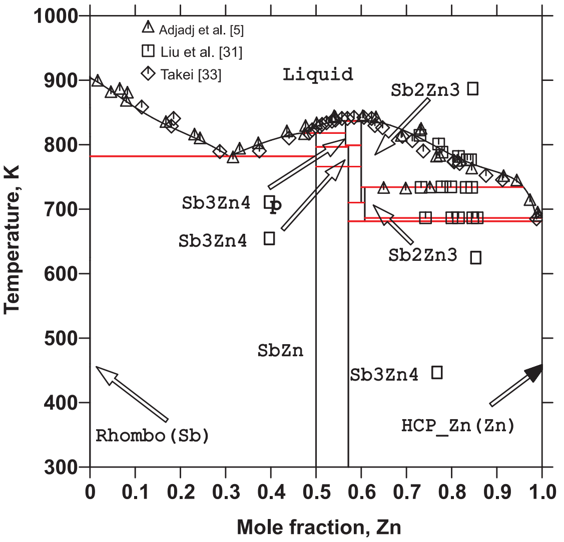

***Figure 1.*** *Calculated Sb – Zn phase diagram superimposed with experimental data given by Adjadj et al [5], Liu et al. [31] and Takei [33].*

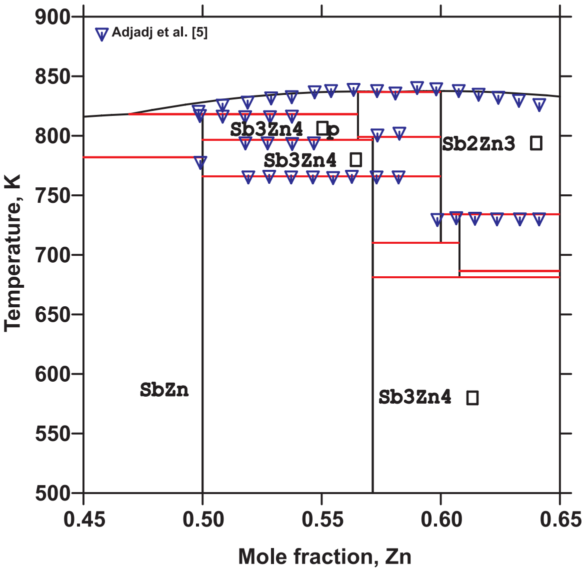

***Figure 2.*** *Zoomed part of calculated Sb – Zn phase diagram superimposed with experimental data given by Adjadj et al [5].*

Table 3 shows detailed comparison of calculated invariant reactions with experimental data. The Gibbs energies of formation of IMCs obtained from EMF experiment were taken into account for this optimization. Generally speaking, calculated Gibbs energies of formation show quite good agreement with experimental data, except results for $Sb_3Zn_4(\beta)$ phase.

The comparison between calculated energies of formation and literature data is given in Table 4. Figure 3 shows calculated mixing enthalpy of liquid phase at 843 K and 913 K superimposed with data given by Scheil and Lukas [7] and Witting and Gehring [8]. The calculated functions reproduce experimental information well; only two points reported by Scheil and Lukas [7] exhibit significant deviation from calculation. In the same picture it is shown enthalpy of mixing calculated from previous optimization given by Li et al. [38]. It can be easy found out, that previously proposed description produces extra maximum of the function near composition 0.1 mole fraction of Zn.

The experimental information does not suggest this shape of a function and the additional extreme seems to be an artifact connected with proposed description. The same additional maximum on mixing enthalpy of liquid can be found in Liu's et al. [31] proposition. Figure 4 shows calculated activity of Sb and Zn at 823 K superimposed with experimental data reported by Rubin and Komarek [6] and Hultgren [10]. The calculation shows very good agreement with experimental data.

The literature reveals, that atoms in the liquid phase tend to have short-range ordering (SRO) and

***Table 3.*** *Temperatures of invariant reactions in the Sb – Zn system.*

| Reaction | Temperature [K] | Reference |
|---|---|---|
| Liquid = $Sb_2Zn_3\beta$+$Sb_3Zn_4\gamma p$ | 836.78 | This work |
| | 835.65 | 44 [37] |
| | 839 | 3 [32] |
| | 836 | [39] |
| | 836.5 | [5] |
| Liquid+ $Sb_3Zn_4\gamma p$ = SbZn | 818.18 | This work |
| | 814.95 | [37] |
| | 819 | 24 [31] |
| | 820 | [32] |
| | 817 | [39] |
| | 817 | [5] |
| $Sb_3Zn_4\gamma p$+$Sb_2Zn_3\eta$ = $Sb_3Zn_4\gamma$ | 799 | This work |
| | 805 | [32] |
| | 802 | [39] |
| | 803 | [5] |
| $Sb_3Zn_4\gamma p$+SbZn = $Sb_3Zn_4\gamma$ | 796.6 | This work |
| | 798 | [32] |
| | 801.5 | [39] |
| | 795 | [5] |
| Liquid = Rhombohedral_A7(Sb)+SbZn | 782 | This work |
| | 779 | [37] |
| | 780 | [31] |
| | 782 | [32] |
| | 774 | [39] |
| SbZn + $Sb_3Zn_4\gamma$ = $Sb_3Sn_4\beta$ | 765.97 | This work |
| | 764.55 | [31] |
| | 766 | [5] |
| $Sb_3Zn_4\gamma$ + $Sb_2Zn_3\eta$ = $Sb_3Sn_4\beta$ | 765.97 | This work |
| | 766 | [5] |
| $Sb_2Zn_3\eta$ + Liquid = $Sb_2Zn_3\zeta$ | 734.12 | This work |
| | 728.15 | [31] |
| | 731 | [32] |
| | 733.2 | [39] |
| | 733 | [5] |
| $Sb_2Zn_3\eta$ = $Sb_3Sn_4\beta$ + $Sb_2Zn_3\zeta$ | 710 | This work |
| | 713.15 | [31] |
| | 714 | [32] |
| | 715 | [39] |
| | 710 | [5] |
| Liquid = $Sb_2Zn_3\eta$ + HCP_ZN(Zn) | 686.5 | This work |
| | 684.5 | [37] |
| | 682.1 | [31] |
| | 686 | [39] |
| | 687 | [39] |
| | 687 | [5] |
| $Sb_2Zn_3\eta$ = $Sb_3Sn_4\beta$ + HCP_ZN(Zn) | 681.2 | This work |
| | 678.25 | [37] |
| | 681.15 | [31] |
| | 677 | [39] |
| | 678 | [39] |
| | 680 | [5] |

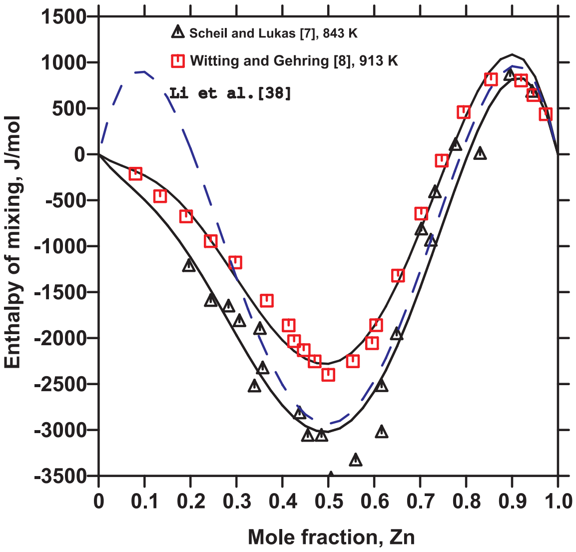

***Figure 3.*** *Calculated enthalpy of mixing at 843 K and 913 K superimposed with experimental data given by Scheil and Lukas [7] and Witting and Gehring [8]. Dashed line represents calculated enthalpy at 843 K based on Li et al. [38] proposition.*

***Table 4.*** *Gibbs energies of intermetallic compounds in the binary Sb – Zn system.*

| Phase | Gibbs energy | T[K] | Reference state | | References |
|---|---|---|---|---|---|
| | | | Sb | Zn | |
| SbZn | -9236 | 420 | Rhombohedral | HCP_ZN | This work |
| SbZn | -9700 | 420 | Rhombohedral | HCP_ZN | [20] |
| SbZn | -8204 | 804 | Liquid | Liquid | This work |
| SbZn | -8115 | 804 | Liquid | Liquid | [12] |
| $Sb_3Zn_4$( γ’) | -8018 | 826 | Liquid | Liquid | This work |
| $Sb_3Zn_4$( γ’) | -8032 | 826 | Liquid | Liquid | [12] |
| $Sb_3Zn_4$( γ) | -8189 | 806 | Liquid | Liquid | This work |
| $Sb_3Zn_4$( γ) | -7903 | 806 | Liquid | Liquid | [12] |
| $Sb_3Zn_4$( γ) | -7332 | 800 | Rhombohedral | Liquid | This work |
| $Sb_3Zn_4$( γ) | -7126 | 800 | Rhombohedral | Liquid | [19] |
| $Sb_3Zn_4$(β) | -9767 | 420 | Rhombohedral | HCP_ZN | This work |
| $Sb_3Zn_4$(β) | -8800 | 420 | Rhombohedral | HCP_ZN | [20] |
| $Sb_3Zn_4$(β) | -8922 | 600 | Rhombohedral | HCP_ZN | This work |
| $Sb_3Zn_4$(β) | -6990 | 600 | Rhombohedral | HCP_ZN | [19] |
| $Sb_2Zn_3$(ζ) | -7123 | 800 | Rhombohedral | Liquid | This work |
| $Sb_2Zn_3$(ζ) | -6916 | 800 | Rhombohedral | Liquid | [19] |

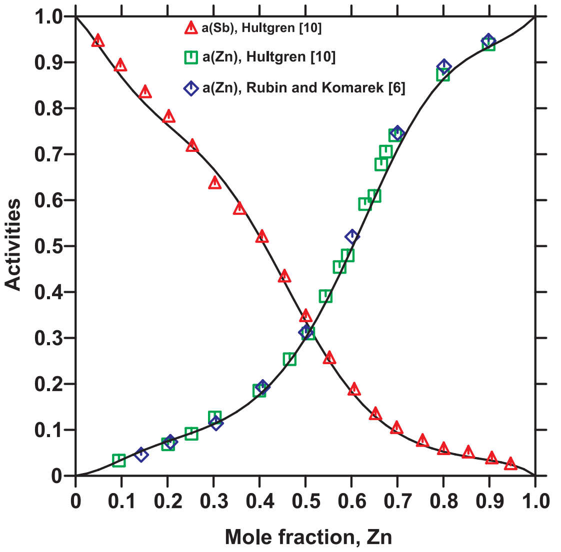

***Figure 4.*** *Calculated activities of Sb and Zn at 823 K superimposed with experimental data given by Hultgren [10] and Rubin and Komarek [6].*

form liquid associate $Sb_3Zn_4$. The existence of the associate was confirmed by Asanovich et al. [45] who used resistivity measurement, Sinha and Miller [46] by viscosity measurement and Matuyama [47] who determined density of a liquid phase. Considering presence of the SRO in a liquid phase, the associate solution model [40] was also applied for an assessment of the liquid phase in this work. During optimization of the liquid, Gibbs energies of solid phases were taken from the previous step (optimization with substational solution model for liquid phase) and kept unchanged. As it can be seen from Table 2, number of parameters is the same as in the case of substitional solution. The results of calculation are shown in Figures 5 – 7. Figure 5 shows calculated Sb – Zn phase diagram superimposed with experimental data. The liquidus line is reproduced well. Also temperatures of phase transformation agree with experimental data given by Adjadj et al.[5]. Figure 6 shows calculated mixing enthalpy of liquid phase at 843 K and 913 K superimposed with data given by Scheil and Lukas [7] and Witting and Gehring [8]. It can be seen, that calculated function agree with experimental data well; however, for the Zn-rich compositions, the calculation exhibits slightly less exothermic values than experimental points at temperature 843K. Comparing calculated enthalpy of mixing to Zabdyr’s [37] resultsit can be seen that in [37] calculated enthalpy of mixing at 843K poses an additional extreme for compositions close to pure antimony. This artifact is identical with artifacts obtained by Liu et al. [31] and Li et al. [38] for substitional solution model. Calculated activity of antimony and zinc at 823 K together with experimental data is shown in Figure 7. The

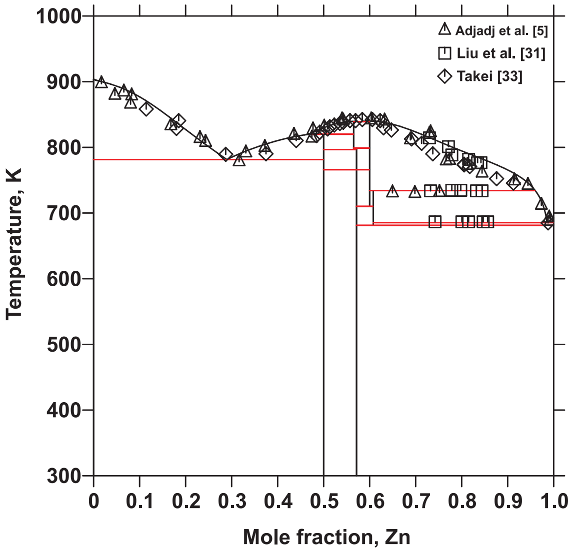

***Figure 5.*** *Calculated phase diagram of the Sb – Zn system superimposed with experimental data given by Adjadj et al. [5], Liu et al. [31] and Takei [33]. Associated solution model applied for a liquid phase description.*

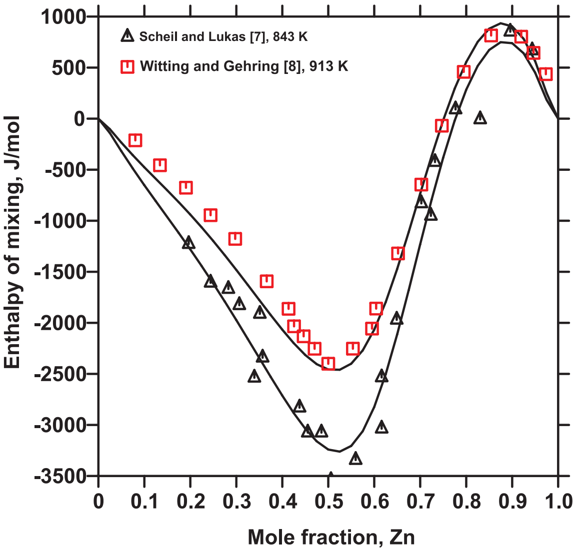

***Figure 6.*** *Calculated liquid enthalpy of mixing at 843 K and 913 K superimposed with experimental data given by Scheil and Lukas [7] and Witting and Gehring [8]. Associated solution model applied for a liquid phase description.*

calculated function shows good agreement with experimental data. From the results discussed above one can see that both models (substitutional solution and associated solution) described liquid phase well. It can be also found out that even the associated solution model is more suitable from physical point of view, the number of parameters is not smaller than in a case of substitional solution model application. The asymmetrical “S” shapes of liquid mixing enthalpy and activity functions need quite complicated mathematical description for fitting all the data. The associate solution model did not make description easier, what is in agreement with previous comparison of application both models done by Zabdyr [37]. Both models reproduced thermodynamic properties and liquidus line well; however, the associated solution model could be preferred as more realistic than substitional solution.

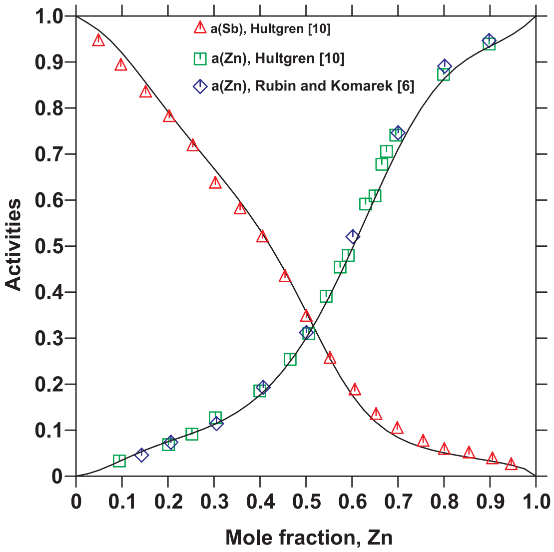

***Figure 7.*** *Calculated activities of Sb and Zn compared with experimental data given by Hultgren [10] and Rubin and Komarek [6]. Associated solution model applied for a liquid phase description.*

## 5. Summary

The binary Sb – Zn system was remodeled according to recent phase equilibria data given by Adjadj et al. [5]. Substitional and associated solution models were used for description of the liquid phase. Both of them reproduce experimental data well; however, the associated solution model could be preferred as more realistic. Modeled liquidus and solidus lines as well as solid phases equilibrium are in good agreement with experimental data.

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