

Thermodynamic Description of the Ternary Sb-Sn-Zn System

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The ternary Sb-Sn-Zn system is important for two reasons: the first one is that antimony-tin-zinc alloys are promising lead-free solders, the second one is, that zinc antimonides show thermoelectric properties. Based on available literature information, the Sb-Sn-Zn system was thermodynamically described using the Calphad approach. A good agreement between calculation and experimental information was found.

Key words: Lead-free solders, thermodynamic modeling, Calphad method, zinc, tin, antimony

INTRODUCTION

The ternary Sb-Sn-Zn alloys are important for the electronic industry as potential lead-free solders, especially for step soldering, which is a process that occurs at several steps because more than one soldering process is needed for some electronic products.^{1–3} Moreover, the Sn-Sb alloys are considered as a replacement for high-melting lead alloys (Sn-95 wt.%Pb).⁴ On the other hand, the Sn-Zn alloys have good mechanical properties and were also proposed as replacements for lead solder.⁵ Besides that, binary zinc antimonides ZnSb and Zn₄Sb₃ show thermoelectric properties, thus their modification by addition of low-melting Sn can improve the thermoelectric Sb-Zn frameworks.⁶ Since Zn is widely known as a metal with high vapor pressure that leads to easy evaporation, the experimental work on the Sb-Sn-Zn system is not easy and requires special treatment.⁷ In this case application of the Calphad⁸ method seems to be very useful due to the possibility of a phase diagram and thermodynamic properties calculation beyond available experimental data.

LITERATURE REVIEW

The ternary Sb-Sn-Zn system includes 12 phases: BCT_A5, HCP_ZN, RHOMBOHEDRAL_A7, SB2SN3, SB2ZN3_ETA, SB2ZN3_ZETA, SB3ZN4_GAMMA, SB3ZN4_GAMMAP, SBSN, SBZN, SB2SNZN, and

Liquid. The binary systems were described well in the literature. The Sb-Sn system was thermodynamically described by Jonsson and Argen,⁹ by the scientific community under COST 531 European action¹⁰ and COST MP-630 action¹¹ as well as by Chen et al.¹² The Sn-Zn system was optimized by Malakhov et al.,¹³ by Lee,^{14,15} under COST 507 action,¹⁶ by Ohtani et al.,¹⁷ by Moelans et al.,¹⁸ and by Yang et al.¹⁹ The system Sb-Zn was optimized by Zabdyr,²⁰ by Liu et al.,²¹ by Li et al.,²² and by Gierlotka.²³ In this work the Gibbs energies proposed by Chen et al.,¹² Yang et al.,¹⁹ and Gierlotka²³ were applied. Calculated binary phase diagrams Sb-Sn, Sn-Zn, and Sb-Zn are shown in Figs. 1, 2, and 3, respectively. Experimental information about the ternary Sb-Sn-Zn system is very limited. Lin et al.²⁴ determined an isothermal section at 423 K, Zobac et al.⁷ investigated isothermal sections at three temperatures: 473 K, 523 K, and 623 K. In general, the triangulations proposed by Lin et al.²⁴ and Zobac et al.⁷ agree well with each other except for a Sb-rich corner where data given by Zobac et al.⁷ is inconsistent. The inconsistency is shown in Fig. 4, which reveals two tie-triangles determined from experiment. A tie-triangle established from samples B4, B8, B11 shows equilibrium between SB2SNZN, SBSN, and SBZN phases. In contrast, the tie-triangle that was created from sample B2 exhibits equilibrium between SB2SNZN, SBZN, and Rhombohedral_A7. It is clear that intercepted tie-triangles can not exist, thus, one of the results must be wrong. To keep agreement with other experimental information, it was decided that tie-triangle SB2SNZN, SBZN, and Rhombohedral_A7 was incorrect. Besides information

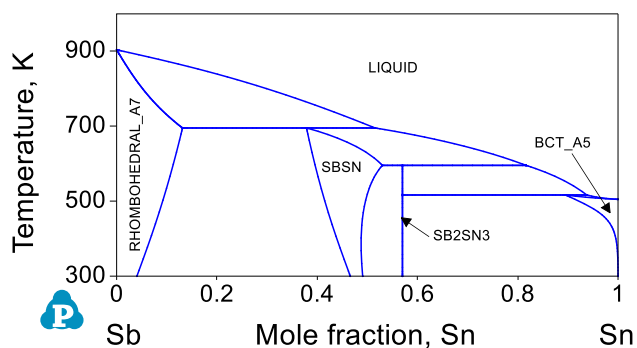


Fig. 1. Calculated binary Sb-Sn system.

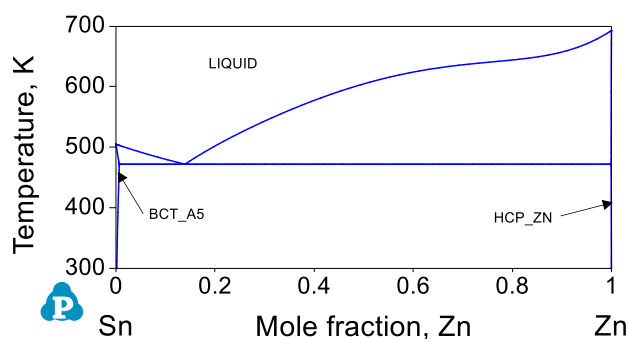


Fig. 2. Calculated binary Sn-Zn system.

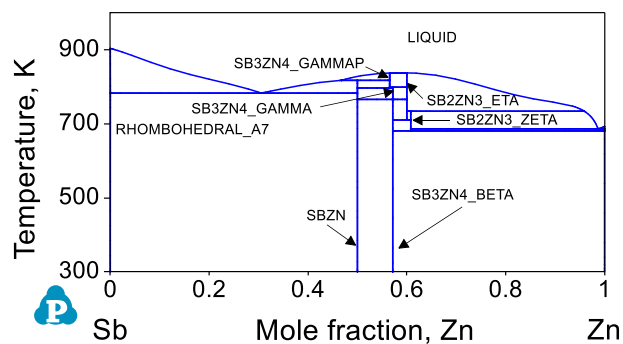
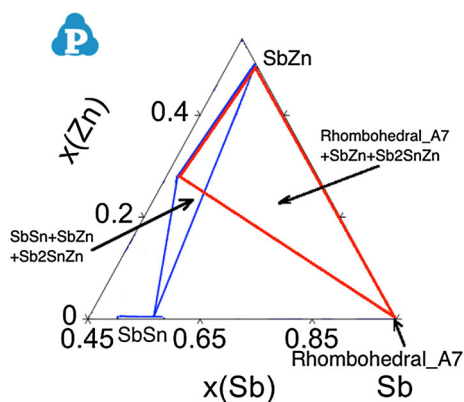


Fig. 3. Calculated binary Sb-Zn system.


 Fig. 4. Sb-rich corner of Sb-Sn-Zn system at 523 K with tie-triangles determine by Zöbacz et al.⁷

about isothermal sections, the Sb_2SnZn intermetallic compound was investigated by Tenga et al.⁶ who focused on crystal structure and determined peritectic decomposition at about 633 K. Moreover, Ko and Gierlotka²⁵ investigated the isothermal section at 523 K as well as the primary solidification phases in the Sb-Sn-Zn system. Thermodynamic properties of the liquid phase were described by Gancarz and Gasior²⁶ who determined activity of Zn in the liquid Sb-Sn-Zn system using the electromotive force (EMF) measurement technique. The activity was measured for five intersections: $\text{Sn}/\text{Sb} = 0.33$, $\text{Sn}/\text{Sb} = 1$, $\text{Sn}/\text{Sb} = 3$, $\text{Sn}/\text{Sb} = 4$, and $\text{Sn}/\text{Sb} = 9$ at temperature range 750–1023 K. Thermodynamic assessment of the ternary Sb-Sn-Zn system was not found in the literature.

THERMODYNAMIC MODEL

As it was mentioned before, the ternary Sb-Sn-Zn system includes 12 phases: BCT_A5, HCP_ZN, RHOMBOHEDRAL_A7, SB2SN3, SB2ZN3_ETA, SB2ZN3_ZETA, SB3ZN4_GAMMA, SB3ZN4_GAMMAP, SBSN, SBZN, SB2SNZN, and Liquid. The detailed description of their crystal structures is given in Table I.

The Gibbs free energies of pure elements with respect to temperature ${}^0G_i(T) = G_i(T) - H_i^{\text{SER}}$ are represented by Eq. 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).²⁷ The reference states are RHOMBOHEDRAL_A7 (Sb), BCT_A5 (Sn), and HCP_ZN (Zn). The ${}^0G_i(T)$ expression may be 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 v.4.²⁷

Stoichiometric compounds: SB2SN3, SB2ZN3_ETA, SB2ZN3_ZETA, SB3ZN4_GAMMA, SB3ZN4_GAMMAP, SBZN, SB2SNZN.

The homogeneity range of those two compounds is negligible, thus the Gibbs energy is described as follows:

$$G_{\text{Sb}_n\text{Me}_m}^0 = a + bT + n \cdot \text{GHSESRB} + m \cdot \text{GHSESRME}, \quad (2)$$

where GHSESRME and GHSESRB are Gibbs energies of tin or zinc and antimony in BCT_A5, or HCP_ZN and RHOMBOHEDRAL_A7 structures, respectively, n, m are stoichiometric coefficients, a and b are adjustable coefficients, and T is absolute temperature.

Table I. Crystal structures of phases in the ternary Sb-Sn-Zn system⁶

Phase	Pearson symbol	Space group	Strukturbericht designation	Prototype
(Sb)	hR2	$R\bar{3}m$	A7	α As
(Sn)	tI2	$I4_1/amd$	B5	β Sn
(Zn)	hP2	$P6_3/mmc$	A3	Mg
SbZn	oP16	$Pbca$	B _e	CdSb
Sb ₃ Zn ₄ (β)	–	–	–	–
Sb ₃ Zn ₄ (γ)	–	–	–	–
Sb ₃ Zn ₄ (γ')	oP28	$Pmmn$	–	–
Sb ₂ Zn ₃ (ζ)	oI	–	–	–
Sb ₂ Zn ₃ (η)	oP32	$Pmmn$	–	–
SbSn	cF8	$Fm\bar{3}m$	–	α As
Sb ₂ Sn ₃	–	–	–	–
Sb ₂ SnZn	tI16	$I42d$	–	CuFeS ₂

Substitutional Solutions

Solid and liquid solution phases (Liquid, BCT_A5, HCP_ZN and RHOMBOHEDRAL_A7) were described by the substitutional solution model.²⁸

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

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

SBSN Phase

The two-sublattice model (Sb, Sn)₁:(Sb, Sn)₁ was applied in this optimization. The Gibbs energy of the SBSN is described as follows:

$$G_{\text{SbSn}}^0 = Y_{\text{Sb}}^I Y_{\text{Sb}}^{\text{II}} G_{\text{Sb:Sb}}^0 + Y_{\text{Sb}}^I Y_{\text{Sn}}^{\text{II}} G_{\text{Sb:Sn}}^0 + Y_{\text{Sn}}^I Y_{\text{Sb}}^{\text{II}} G_{\text{Sn:Sb}}^0 + Y_{\text{Sn}}^I Y_{\text{Sn}}^{\text{II}} G_{\text{Sn:Sn}}^0 + RT(Y_{\text{Sb}}^I \ln Y_{\text{Sb}}^I + Y_{\text{Sn}}^I \ln Y_{\text{Sn}}^I) + RT(Y_{\text{Sb}}^{\text{II}} \ln Y_{\text{Sb}}^{\text{II}} + Y_{\text{Sn}}^{\text{II}} \ln Y_{\text{Sn}}^{\text{II}}) + Y_{\text{Sb}}^I Y_{\text{Sn}}^{\text{II}} Y_{\text{Sb}}^{\text{II}} L_{\text{Sb,Sn:Sb}} + Y_{\text{Sb}}^I Y_{\text{Sn}}^{\text{II}} Y_{\text{Sn}}^{\text{II}} L_{\text{Sb,Sn:Sn}} + Y_{\text{Sb}}^{\text{II}} Y_{\text{Sb}}^{\text{II}} Y_{\text{Sn}}^{\text{II}} L_{\text{Sb,Sb:Sn}} + Y_{\text{Sb}}^{\text{II}} Y_{\text{Sb}}^{\text{II}} Y_{\text{Sn}}^{\text{I}} L_{\text{Sn,Sb:Sb}} \quad (4)$$

in which Y_i^N denotes the site fraction of element i on sublattice N , symbol “:” indicates separation of elements on the different sublattices, and “,” indicates separation of elements on the same sublattice.

The first part of Eq. 4 describes a superposition of Gibbs energies of pure elements, the second part labels Gibbs energy of mechanical mixing, and the third one is the Gibbs energy of excess.

OPTIMIZATION PROCEDURE

The thermodynamic parameters for all phases in the system were optimized using ThermoCalc software²⁹ and Pandat software.³⁰ For this optimization,

thermodynamic data for the liquid phase and phase equilibrium data were used. To each piece of the selected information was given a certain weight based on experimental uncertainty. The optimization was carried out step by step in agreement with Schmid-Fetzer's et al.³¹ 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 II.

RESULT AND DISCUSSION

Calculated isothermal section of the ternary Sb-Sn-Zn system at 623 K superimposed with experimental data given by Zobac et al.⁷ is shown in Fig. 5. It can be easily seen, that calculation reproduces the experiment very well, except for the composition of the liquid phase, what can be explained by widely known difficulties of composition measurement in solidified liquids by an electro probe micro-analyzer (EPMA). The experimentally determined⁷ amount of Sb in the ternary intermetallic compound Sb₂SnZn varies between $x_{\text{Sb}} = 0.455$ and $x_{\text{Sb}} = 0.5154$. However, for sake of clarity, this work follows the composition of Sb₂SnZn determined by Ko and Gierlotka.²⁵ $x_{\text{Sb}} = 0.47$, $x_{\text{Sn}} = 0.25$ and $x_{\text{Zn}} = 0.28$. Figure 6 exhibits the calculated isothermal section at 523 K together with experimental data given by Zobac et al.⁷ As it was emphasized earlier, experimental data given by Ref. 7 at this temperature is inconsistent. The antimony-rich corner exhibits two kinds of tie-triangles that are shown in Fig. 4. Since, this is impossible, that tie-triangles intercept each other, then tie-triangle Sb₂SnZn-SBZN-RHOMBOHEDRAL_A7 was repudiated as a tie-triangle that does not agree with data given by Zobac et al.⁷ at 623 K and Lin et al.²⁴ at 423 K. The calculated isothermal section agrees well with experimental data. It should be mentioned here, that Zobac et al.⁷ reported several differential thermal analysis (DTA) data that was obtained

Table II. Gibbs energies of phases in the ternary Sb-Sn-Zn system

Phase	Parameters
SbZn	${}^m G_{\text{Sb:Zn}}^{\text{SbZn}} = -24600.8631 + 11.4198 + \text{GHSERSB} + \text{GHSERZN}$ ${}^m G_{\text{Sb:Zn}}^{\text{SbZn}} = 10000 + \text{GHSERSN} + \text{GHSERZN}$ ${}^0 L_{\text{Sb,Sn:Zn}}^{\text{SbZn}} = -500$
Sb ₃ Zn ₄ (β)	${}^m G_{\text{Sb:Zn}}^{\text{Sb}_3\text{Zn}_4\beta} = -82174.639 + 32.8691 + 3\text{GHSERSB} + 4\text{GHSERZN}$
Sb ₃ Zn ₄ (γ)	${}^m G_{\text{Sb:Zn}}^{\text{Sb}_3\text{Zn}_4\gamma} = -81723.458 + 32.2801 + 3\text{GHSERSB} + 4\text{GHSERZN}$
Sb ₃ Zn ₄ (γ')	${}^m G_{\text{Sb:Zn}}^{\text{Sb}_3\text{Zn}_4\gamma'} = -361795.319 - 236.8462 + 30\text{GHSERSB} + 39\text{GHSERZN}$
Sb ₂ Zn ₃ (ζ)	${}^m G_{\text{Sb:Zn}}^{\text{Sb}_2\text{Zn}_3\zeta} = -397804.613 - 1.1087 + 20\text{GHSERSB} + 31\text{GHSERZN}$
Sb ₂ Zn ₃ (η)	${}^m G_{\text{Sb:Zn}}^{\text{Sb}_2\text{Zn}_3\eta} = -37109.540 - 3.6646 + 2\text{GHSERSB} + 3\text{GHSERZN}$
SbSn	${}^m G_{\text{Sb:Sn}}^{\text{SbSn}} = 3094.5934 + \text{GHSERSB}$ ${}^m G_{\text{Sb:Sn}}^{\text{SbSn}} = -5193.182 + 3.0434T + 0.5\text{GHSERSB} + 0.5\text{GHSERSN}$ ${}^m G_{\text{Sn:Sb}}^{\text{SbSn}} = -38885.575 + 1.9321T + 0.5\text{GHSERSB} + 0.5\text{GHSERSN}$ ${}^3 G_{\text{Sn:Sb}}^{\text{SbSn}} = 6940.7006 + \text{GHSERSN}$ ${}^m G_{\text{Sb:Zn}}^{\text{SbSn}} 1000 + 0.5\text{GHSERSN} + 0.5\text{GHSERZN}$ ${}^m G_{\text{Sb:Zn}}^{\text{SbSn}} 1000 + 0.5\text{GHSERSB} + 0.5\text{GHSERZN}$ ${}^m G_{\text{Zn:Sn}}^{\text{SbSn}} 1000 + 0.5\text{GHSERSN} + 0.5\text{GHSERZN}$ ${}^m G_{\text{Sb:Zn}}^{\text{SbSn}} 1000 + 0.5\text{GHSERSB} + 0.5\text{GHSERZN}$
Sb ₂ Sn ₃	${}^m G_{\text{Sb:Sn}}^{\text{Sb}_2\text{Sn}_3} = -5193.182 + 3.0434T + 0.43\text{GHSERSB} + 0.57\text{GHSERSN}$
Sb ₂ SnZn	${}^m G_{\text{Sb:Sn:Zn}}^{\text{Sb}_2\text{SnZn}} = -8428.2 + 2.8208T + 0.47\text{GHSERSB} + 0.25\text{GHSERSN} + 0.28\text{GHSERZ}$
Rhombohedral_A7	${}^0 L_{\text{Sb,Sn}}^{\text{RhombohedralA7}} = -2682.8508 + 8.0154T$
BCT_A5	${}^1 L_{\text{Sb,Sn}}^{\text{RhombohedralA7}} = -78.475$ ${}^0 L_{\text{Sb,Sn}}^{\text{BCTA5}} = -14643.227 + 6.66374T$ ${}^1 L_{\text{Sb,Sn}}^{\text{BCTA5}} = -37586.893 + 62.244T$ ${}^0 L_{\text{Sb,Sn}}^{\text{BCTA5}} = -14643.277 + 6.6374T$ ${}^1 L_{\text{Sb,Sn}}^{\text{BCTA5}} = -37586.893 + 62.244T$ ${}^0 L_{\text{Sn,Zn}}^{\text{BCTA5}} = 6772.1554 + 24.1579T$
HCP_ZN	${}^0 L_{\text{Sn,Zn}}^{\text{HCPZn}} = 4941.6236 + 50T$
Liquid	${}^0 L_{\text{Sb,Zn}}^{\text{Liquid}} = -47736.194 + 326.5303T - 42.2936T \ln T$ ${}^1 L_{\text{Sb,Zn}}^{\text{Liquid}} = -808.225 + 0.7409T + 0.3242T \ln T$ ${}^2 L_{\text{Sb,Zn}}^{\text{Liquid}} = 25540.912 - 17.6368T$ ${}^3 L_{\text{Sb,Zn}}^{\text{Liquid}} = -12308.192 + 6.1383T$ ${}^4 L_{\text{Sb,Zn}}^{\text{Liquid}} = -6050.661$ ${}^5 L_{\text{Sb,Zn}}^{\text{Liquid}} = -393.624$ ${}^0 L_{\text{SbSn}}^{\text{liquid}} = -5536.588 + 1.5399 * T$ ${}^1 L_{\text{SbSn}}^{\text{liquid}} = 177.9316$ ${}^2 L_{\text{SbSn}}^{\text{liquid}} = 883.186$ ${}^0 L_{\text{Sn,Zn}}^{\text{liquid}} = 21468.899 - 100.7913T + 12.0055T \ln(T)$ ${}^1 L_{\text{Sn,Zn}}^{\text{Liquid}} = -6355.4014 + 12.388401T - 1.12437T \ln(T)$ ${}^0 L_{\text{Sb,Sn,Zn}}^{\text{Liquid}} = 124315.16 - 118.169T$ ${}^1 L_{\text{Sb,Sn,Zn}}^{\text{Liquid}} = -336089.76 + 397.945T$ ${}^2 L_{\text{Sb,Sn,Zn}}^{\text{Liquid}} = 126925.125 - 124.503T$

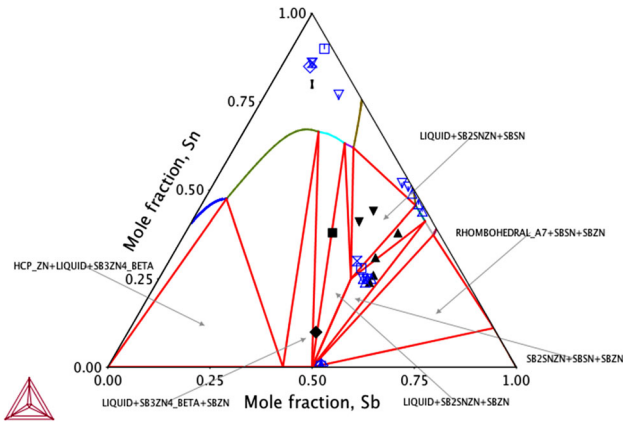


Fig. 5. Calculated isothermal section of Sb-Sn-Zn system at 623 K superimposed with data given by Zobac et al.⁷ Open symbols represent equilibrium composition, closed symbols represent nominal compositions of samples.

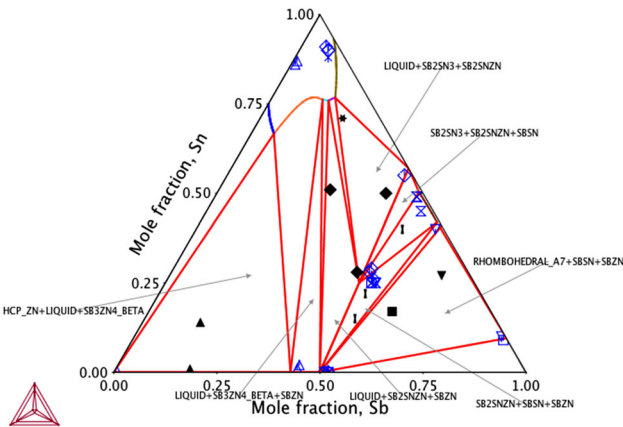


Fig. 6. Calculated isothermal section of Sb-Sn-Zn system at 523 K superimposed with data given by Zobac et al.⁷ Open symbols represent equilibrium composition, closed symbols represent nominal compositions of samples.

from their samples equilibrated at 623 K. Unfortunately, the compositions do not lay at a straight line and the authors⁷ do not provide information about primary solidification phases, thus this data can not be simply used during the optimization process. The calculated isothermal section at 423 K is shown in Fig. 7. The calculation agrees with triangulation of Gibbs triangle at 423 given by Lin et al.²⁴ Unluckily, Lin et al.²⁴ did not provide information about nominal compositions of the samples and the compositions of phases in equilibrium, thus direct comparison of calculations with their results was impossible. Figure 8 displays a calculated liquidus projection together with experimental information given by Ko and Gierlotka.²³ As it can be seen from this figure, there are eleven primary solidification regions: BCT_A5, SB2SN3, SBSN, RHOMBOHEDRAL_A7, SBZN, SB3ZN4_GAMMAP, SB3ZN4_GAMMA, SB3ZN4_BETA, SB2ZN3_ETA, SB2ZN3_ZETA, HCP_ZN,

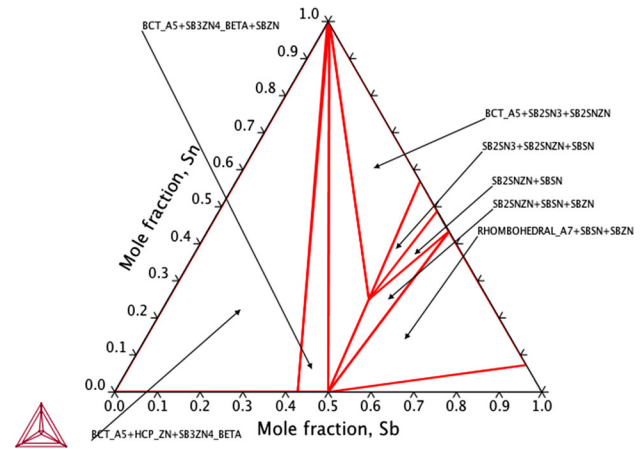


Fig. 7. Calculated isothermal section of Sb-Sn-Zn system at 423 K.

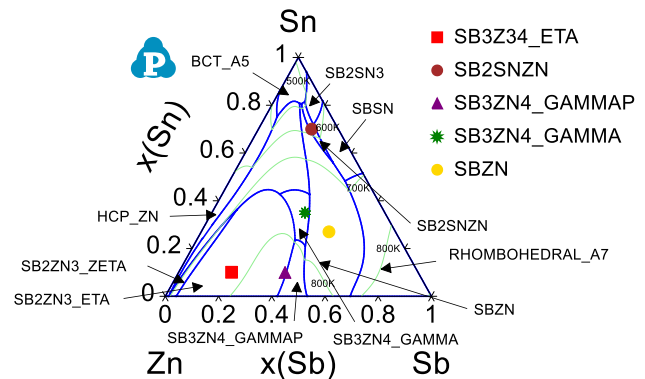


Fig. 8. Calculated liquidus projection together with experimental information given by Ko and Gierlotka.²³

and SB2SNZN. The ternary intermetallic Sb2SnZn solidification region is quite narrow; however, calculation agrees with the experimentally determined composition given by Ref. 25. Moreover, other calculated primary solidification regions also agree with experimental determination except the SB3ZN4_GAMMAP that is located on the boundary between the experimentally determined phase and SB2ZN3_ETA. According to the calculation, ternary intermetallic compound SB2SNZN decomposes at 649 K, which is slightly above the temperature provided by Tenga et al.⁶ It was impossible to obtain the same decomposition temperature as Ref. 6 and triangulations of isothermal section at 423 as Ref. 19. It should be emphasized here, that, if the heat capacity of an intermetallic compound is known, then its Gibbs energy can be described more accurately. Unfortunately, in the case of SB2SNZN, the heat capacity is unknown. Calculated activity of Zn in liquid at 923 K for an intersection $x_{Sn}/x_{Sb} = 0.33$ is shown in Fig. 9 together with data given by Gancarz and Gasior.²⁶ The calculated activity follows experimental data pretty well. Similarly, calculated activity at 923 K for intersection $x_{Sn}/x_{Sb} = 3$



Fig. 9. Calculated activity of Zn at 923 K for intersection $x_{\text{Sn}}/x_{\text{Sb}} = 0.33$ superimposed with data given by Gancarz and Gasior.²⁶

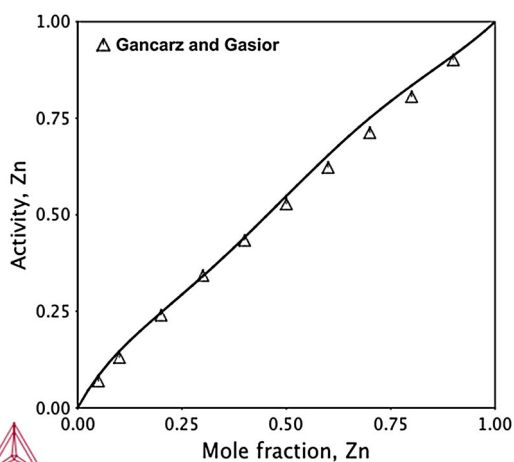


Fig. 10. Calculated activity of Zn at 923 K for intersection $x_{\text{Sn}}/x_{\text{Sb}} = 3$ superimposed with data given by Gancarz and Gasior.²⁶

that is shown in Fig. 10 agrees well with experimental determination.²⁶ Summarizing, the proposed thermodynamic description of the Sb-Sn-Zn system reproduces all available experimental data quite well, thus the proposed set of Gibbs energies can be used for further expansion to the quaternary Me-Sb-Sn-Zn system.

SUMMARY

The ternary Sb-Sn-Zn system was described by the Calphad approach using available experimental data. The isothermal sections, liquidus projection, and thermodynamic properties were calculated and compared with literature data. A good agreement between reported in literature information and simulation was found.

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