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Calculation of the thermodynamic properties of the Ga–Sb–Tl liquid alloys

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Abstract: The results of the calculation of the thermodynamic properties for liquid Ga–Sb–Tl alloys at the temperature 1073 K are presented in this paper. Initially, the most appropriate thermodynamic model for the investigated system was selected. Based on a comparison of the values calculated by different geometric models (Kohler, Muggianu, Toop, Hillert, Chou) with the existing experimental based data, asymmetric models of calculation were determined to give the best results. The asymmetric nature of the investigated ternary system was additionally confirmed by the Chou similarity coefficient concept. For these reasons, further complete thermodynamic calculations were performed according to the Hillert model in five sections of the ternary Ga–Sb–Tl system from each corner with the mole ratio of other two components being 9:1; 7:3; 5:5; 3:7 and 1:9. The obtained results include integral excess Gibbs energy dependences on composition for all the investigated sections. The calculated activity values at 1073 K for all components are given in the form of isoactivity diagrams. Comparison between the calculated and experimentally obtained gallium activities shows good agreement.

Keywords: thermodynamics of alloys, Ga–Sb–Tl system, geometric models, integral excess Gibbs energy, activities.

INTRODUCTION

The Ga–Sb binary system is a subsystem of various complex electronic materials. For this reason, many thermodynamic studies have been carried out on alloy systems containing the GaSb semiconducting compound. The present study was performed in order to create a set of thermodynamic data for liquid Ga–Sb–Tl ternary alloys which may be of use for further assessment of this system and to investigate the mutual agreement among the results obtained by some of the most frequently used methods for the prediction of thermodynamic properties of ternary systems based on the known data for the constitutive binaries.

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Literature thermodynamic data for the constitutive binary systems are numerous. Direct calorimetric measurements of the Ga–Sb binary system have been performed by Yazawa *et al.*,¹ Predel and Stein² and Gambino and Bros.³ Activities of the liquid Ga–Sb alloys have been measured by electrochemical techniques by Gerasimenko *et al.*,⁴ Danilin and Yatsenko,⁵ Anderson *et al.*⁶ and Katayama *et al.*,⁷ and vapor pressure measurements by Hsi-Hsiung *et al.*⁸ and Bergman *et al.*.⁹

Danilin and Yatsenko¹⁰ used the electromotive-force (EMF) measurements on fused salts to obtain the activity of Tl in liquid Ga–Tl alloys in the temperature range 695–1024 K over the whole composition range. Predel and Stein¹¹ measured the enthalpy of mixing of liquid Ga–Tl alloys using high-temperature calorimetry, and determined the entropy of mixing based on the results of the solubility equilibria evaluation. Katayama *et al.*¹² performed EMF measurements with zirconia as solid electrolyte over the entire composition range of liquid Ga–Tl alloys in the temperature range 973–1273 K to obtain the activity of gallium.

Thermodynamic properties of the liquid Sb–Tl alloys were investigated by Kameda.¹³ Based on the results of EMF measurements in the temperature range 953–1230 K, negative deviations from Raoult's law were obtained for both components over the whole concentration ranges.

Recently Katayama *et al.*¹⁴ performed EMF measurements with zirconia as solid electrolyte in the Ga–Sb–Tl system and derived gallium activity in three cross sections from the gallium corner (x_{Sb} : x_{Tl} =3:1;1:1;1:3) in the temperature range 973–1273 K.

As a contribution to a more complete knowledge of the thermodynamic behavior of the Ga–Sb–Tl system, the results of the calculation of thermodynamic properties are presented in this paper. The results using the Hillert, Toop, Kohler, Muggianu and Chou models were compared to available experimental data and mutually to each other. A so-obtained set of thermodynamic data for liquid Ga–Sb–Tl alloys may be useful for further assessment of this system.

RESULTS AND DISCUSSION

There are several traditional models from which thermodynamic properties of a ternary system can be extrapolated using the three constitutive binary systems as basis. These models are classified, according to Hillert,¹⁵ into two categories: symmetrical (Kohler,¹⁶ Muggianu¹⁷) and asymmetrical (Toop,¹⁸ Hillert¹⁵).

The basic analytical interpretations of these models are given by:

– Kohler model¹⁶

$$\Delta G^{\rm E} = (x_1 + x_2)^2 \Delta G_{12}^{\rm E} \left(\frac{x_1}{x_1 + x_2}; \frac{x_2}{x_1 + x_3} \right) + (x_2 + x_3)^2 \Delta G_{23}^{\rm E} \left(\frac{x_2}{x_2 + x_3}; \frac{x_3}{x_2 + x_3} \right) + (x_3 + x_1)^2 \Delta G_{31}^{\rm E} \left(\frac{x_3}{x_1 + x_3}; \frac{x_1}{x_1 + x_3} \right)$$
(1)

– Muggianu model¹⁷

$$\Delta G^{\rm E} = \frac{4x_1x_2}{(l+x_1-x_2)(l+x_2-x_1)} \Delta G^{\rm E}_{12} \left(\frac{l+x_1-x_2}{2}; \frac{l+x_2-x_1}{2}\right) + \frac{4x_2x_3}{(l+x_2-x_3)(l+x_3-x_2)} \Delta G^{\rm E}_{23} \left(\frac{l+x_2-x_3}{2}; \frac{l+x_3-x_2}{2}\right) + \frac{4x_3x_1}{(l+x_3-x_1)(l+x_1-x_3)} \Delta G^{\rm E}_{31} \left(\frac{l+x_3-x_1}{2}; \frac{l+x_1-x_3}{2}\right)$$
(2)

- Toop model¹⁸

$$\Delta G^{\rm E} = \frac{x_2}{1 - x_1} \Delta G_{12}^{\rm E}(x_1; 1 - x_1) + \frac{x_3}{1 - x_1} \Delta G_{13}^{\rm E}(x_1; 1 - x_1) + (x_2 + x_3)^2 \Delta G_{23}^{\rm E} \left(\frac{x_2}{x_2 + x_3}; \frac{x_3}{x_2 + x_3}\right)$$
(3)

- Hillert model¹⁵

$$\Delta G^{\rm E} = \frac{x_2}{1 - x_1} \Delta G^{\rm E}_{12}(x_1; 1 - x_1) + \frac{x_3}{1 - x_1} \Delta G^{\rm E}_{13}(x_1; 1 - x_1) + \frac{x_2 x_3}{v_{23} v_{32}} \Delta G^{\rm E}_{23} + (v_{23}; v_{32})$$
(4)

where $v_{ij} = (1 + x_i - x_j)/2$.

In all given equations, $\Delta G^{\rm E}$ and $\Delta G^{\rm E}_{ij}$ correspond to the integral molar excess Gibbs energy for ternary and binary systems, respectively, where x_1, x_2, x_3 correspond to the mole fraction of components in an investigated ternary system.

Graphical interpretation of these models is shown in Fig. 1.

The use of a symmetrical model when an asymmetrical model is more appropriate can often give rise to errors. Categorization of an investigated ternary system into one or the other of these two categories is in some cases, especially when adequate experimental data are lacking, an uncertain task. Also, a different choice of the arrangement of the three components to the three apexes of the triangle in the case of the application of an asymmetric model leads to a different result of the ternary Gibbs energy of mixing. For these reasons, Chou¹⁹ proposed a new model based on the "similarity coefficient concept", the advantage of which is that its application requires neither the predetermination of whether a system is symmetrical or not, nor the choice of the symmetric and asymmetric components in a particular ternary system. The correctness of this model has already been confirmed in some practical examples (Ga–Sb–Sn system²⁰; Ga–Sb–Bi system²¹; Ga–In–Te system²²). Therefore, besides the traditional models, this model is also applicable for the calculation of the thermodynamic properties of the Ga–Sb–Tl ternary system.

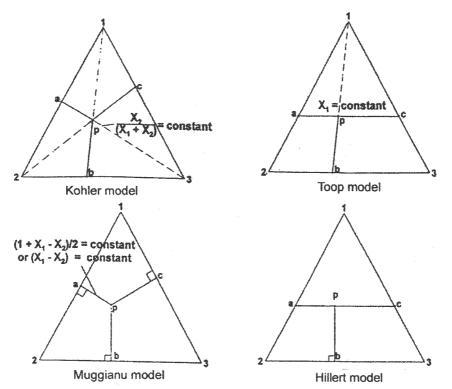


Fig.1. Selected compositions of the three constitutive binary systems for the calculation of the integral excess Gibbs energy of a ternary alloy according to the Kohler, Muggianu, Toop and Hillert model.

The basic equation of the Chou model is given as follows (in detail see Ref.19):

$$\Delta G^{\rm E} = x_1 x_2 \left(A^0_{12} + A^1_{12} \left(x_1 - x_2 \right) + A^2_{12} \left(x_1 - x_2 \right)^2 \right) + x_2 x_3 \left(A^0_{23} + A^1_{23} \right) \left(x_2 - x_3 \right) + A^2_{23} \left(x_2 - x_3 \right)^2 \right) + x_3 x_1 \left(A^0_{31} + A^1_{31} \left(x_3 - x_1 \right) \right) + A^2_{31} \left(x_3 - x_1 \right)^2 \right) + f x_1 x_2 x_3$$
(5)

where $\Delta G^{\rm E}$ is the integral excess Gibbs energy for a ternary system, x_1, x_2, x_3 are the mole fractions of a ternary alloy, A^0_{ij} , A^1_{ij} , A^2_{ij} are parameters for the binary system "*ij*" which can be temperature dependent.

The function f is the ternary interaction coefficient expressed by:

$$f = (2x_{12} - 1) \{A^{2}_{12} ((2\xi_{12} - 1)x_{3} + 2(x_{1} - x_{2})) + A^{1}_{12}\} + (2\xi_{23} - 1) \\ \{A^{2}_{23} ((2\xi_{23} - 1)x_{1} + 2(x_{2} - x_{3})) + A^{1}_{23}\} + (2\xi_{31} - 1) \\ \{A^{2}_{31} ((2\xi_{31} - 1)x_{2} + 2(x_{3} - x_{1})) + A^{1}_{31}\}$$
(6)

where ξ_{ij} are the similarity coefficients defined by v_i called the deviation sum of squares:

$$\xi_{ij} = \eta_i / (\eta_i + \eta_j) \tag{7}$$

where:

$$\eta_{\rm I} = \int_{0}^{1} (\Delta G_{12}^{\rm E} - \Delta G_{13}^{\rm E})^2 \, \mathrm{d}X_1$$

$$\eta_{\rm II} = \int_{0}^{1} (\Delta G_{21}^{\rm E} - \Delta G_{23}^{\rm E})^2 \, \mathrm{d}X_2$$

$$\eta_{\rm III} = \int_{0}^{1} (\Delta G_{31}^{\rm E} - \Delta G_{32}^{\rm E})^2 \, \mathrm{d}X_3$$
(8)

and

$$X_{1(12)} = x_1 + x_3\xi_{12}$$

$$X_{2(23)} = x_2 + x_1\xi_{23}$$

$$X_{3(31)} = x_3 + x_2\xi_{31}$$
(9)

In order to investigate which among the chosen geometric models give the most accurate results, experimentally based data¹⁴ of the excess Gibbs energy at 1073 K for the sections from the gallium corner with mole ratio of x_{Sb} : x_{Tl} equal to 3:1, 1:1 and 1:3 were compared with the results calculated according to the following calculation models – Kohler, Muggianu, Toop, Hillert and Chou.

The basic data for the model calculations were the values of the parameters of the Redlich-Kister equation given as $\Delta G^{\rm E} = X_i X_j \left(\sum_{\nu=0}^n (X_i - X_j)^{\nu} L^{(\nu)}_{ij}({\rm T})\right)$ at 1073 K for the constitutive binary systems taken from Ref. 7 for the Sb–Ga system, Ref. 12 for the Ga–Tl system and Ref. 13 for the Tl–Sb system given in Table I.

TABLE I. The Redlich–Kister coefficients for the Sb–Ga⁷; Ga–Tl¹² and Tl–Sb¹³ binary systems at 1073 K

Sb–Ga		Ga–Tl		Tl–Sb	
$L^{(0)}_{\text{SbGa}}$	-8341.95	$L^{(0)}_{\text{GaTl}}$	14210.5	$L^{(0)}$ _{TISb}	-11227.3
$L^{(1)}_{\text{SbGa}}$	1810.725	$L^{(1)}_{GaTl}$	1150	$L^{(1)}$ _{TISb}	-5197.6
$L^{(2)}_{\text{SbGa}}$	363.025	$L^{(2)}_{\text{GaTl}}$	1740.5	$L^{(2)}$ _{TISb}	146.725

The binary regular-solution parameters of the binary systems Sb–Ga, Ga–Tl and Tl–Sb are equal to the Redlich–Kister coefficients shown in the Table I, while related similarity coefficients were calculated according to Eq.(7) and are shown in the Table II.

The values of ΔG^{E} was done using data from Tables I and II and the results for the three sections with mole ratio of Sb:Tl equal to 3:1, 1:1 and 1:3 at 1073 K, are shown in Fig.2, together with literature data¹⁴ for comparison.

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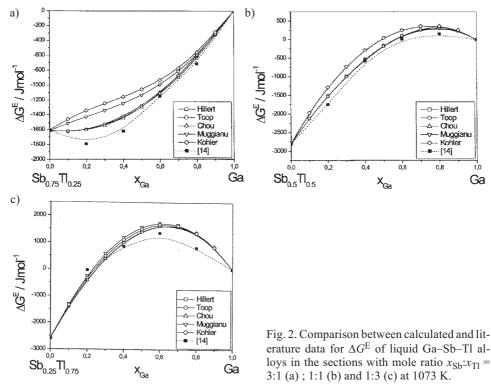
TABLE. II. Binary solution parameters and similarity coefficients calculated by the Chou model for the Ga–Sb–Tl liquid alloys at 1073 K

$A^{(0)}$ _{SbGa}	$A^{(1)}_{\rm SbGa}$	$A^{(2)}_{\rm SbGa}$	$\eta_{ m I}$	$\xi_{ m SbGa}$
-8341.95	1810.725	363.025	517419.8	0.029114
$A^{(0)}_{\rm GaTl}$	$A^{(1)}_{\rm GaTl}$	$A^{(2)}_{\rm GaTl}$	$\eta_{ m II}$	$\xi_{ m GaTl}$
14210.5	1150	1740.5	17254719	0.437869
$A^{(0)}_{\text{TISb}}$	$A^{(1)}_{\text{TISb}}$	$A^{(2)}_{\text{TISb}}$	$\eta_{ m III}$	ξ _{TISb}
-11227.3	-5197.6	146.725	22151446	0.977175

Further, root mean square deviation analysis was applied on the ΔG^{E} data obtained for the mentioned three sections in order to determine accurately which of the applied models was the most adequate one for liquid Ga–Sb–Tl alloys:

$$S_{\rm t} = 1/N [\Sigma (\Delta G^{\rm E}_{\rm exp} - \Delta G^{\rm E}_{\rm calc})^2]^{1/2}$$

where: S_t – root mean square deviation, N – the number of counting points, ΔG^E_{exp} – literature results¹⁴ for ΔG^E and ΔG^E_{calc} – calculated values for ΔG^E . The results of such an analysis are presented in Table III, which indicates that: a) the investigated system Ga–Sb–Tl should be regarded as an asymmetric system and b) the Hillert model is the most adequate model for its thermodynamic description.



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TABLE III. The root mean square deviation from the experimental data for each calculation model

Section	Hillert	Тоор	Chou	Muggianu	Kohler
$x_{\rm Sb}: x_{\rm Tl} = 3:1$	70	76	70	139	174
$x_{\rm Sb}: x_{\rm Tl} = 1:1$	72	74	78	156	158
$x_{\rm Sb}:x_{\rm Tl} = 1:3$	179	182	184	198	185

The Chou similarity coefficient concept¹⁹ was used as an additional tool in the process of selecting an appropriate method for thermodynamic prediction. This concept, depending on similarity coefficients values for the three binaries in the ternary system considered, indicates whether an asymmetrical or a symmetrical model is to be used in a specific case. In the case of the Ga–Sb–Tl system, the calculated similarity coefficients for the three constitutive binaries are presented in Table II and the obtained relations between the ternary and binary composition according to Eq. (9) are shown in Fig. 3 (for a temperature of 1073 K).

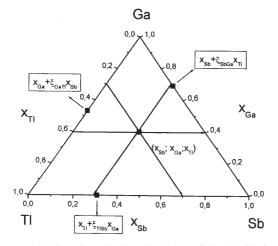


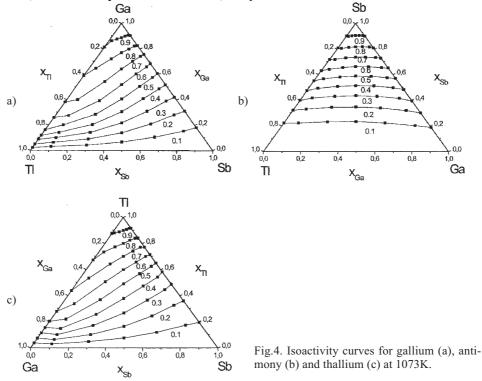
Fig. 3. Selected binary compositions for the three binaries in the ternary system Ga–Sb–Tl according to the Chou model at 1073 K.

Analyzing the calculated values of the similarity coefficients for the Ga–Sb–Tl system (Table II) it may be seen that one similarity coefficient (ξ_{SbGa}) is close to zero, while one is close to unity (ξ_{TlSb}). The selected binary compositions according to the Chou model in this case are very close to the selected binary compositions according to the asymmetric Hillert and Toop models (Fig. 3). It could be concluded that this particular situation reduces the Chou model to an asymmetric model very similar to the Toop or Hillert model when antimony is chosen as component "1" (Eqs. (3) and (4)).

Taking into account Table III and Fig. 3, it is obvious that in the case of the Ga–Sb–Tl system, the results of asymmetrical models are more precise than the symmetric methods, while the results of the Chou model results are positioned very close to the results of the asymmetric models for this particular ternary system. Hence, the graphical representation given in Fig. 3 confirms asymmetric na-

ture of the investigated system and finally indicates the application of the Hillert model as the most adequate in the thermodynamic investigation of the Ga–Sb–Tl liquid alloys. For this reason, all further detailed thermodynamic analysis was performed using the Hillert model with antimony as component "1" and gallium and thallium as components "2" and "3", respectively.

Further thermodynamic calculations of the investigated ternary system Ga–Sb–Tl were preformed according to Eq. (4) in five sections from any corner with mole ratios of other two components equal to 9:1; 7:3; 1:1; 3:7 and 1:9 at a temperature 1073 K. The values of the integral molar Gibbs excess energies for the chosen sections, calculated by the Hillert model, are presented in Table IV.



Using the calculated values for the integral excess Gibbs energies as the starting data and the known relation between integral and partial excess Gibbs energy given by the following expression: $G^{E}_{i} = RT \ln \left(\frac{a_{i}}{x_{i}}\right) = \Delta G^{E} + (1 - x_{i}) \frac{d(\Delta G^{E})}{dx_{i}}$, the

activities of all three components in the investigated sections were determined. The isoactivity curves for gallium, antimony and thallium at 1073 K are graphically presented in Fig. 4.

The calculated isoactivity curves for gallium and antimony are very similar to those shown in our previous paper about the Ga–Sb–Pb liquid alloys.²³ With in-

 $\Delta G^{\rm E}$ / J mol⁻¹ $x_{\text{Ga}}:x_{\text{Tl}} = 9:1$ $x_{\text{Ga}}:x_{\text{Tl}} = 7:3$ $x_{Ga}: x_{Tl} = 1:1$ $x_{\text{Ga}}:x_{\text{Tl}}=3:7$ $x_{\text{Ga}}:x_{\text{Tl}} = 1:9$ $x_{\rm Sb}$ 0 1422 3139 3610 2944 1251 0.1 205 1503 1793 1159 -323 0.2 - 697 253 411 - 163 - 1416 0.3 - 1315 - 576 -1070-2089-645-24030.4 -1678- 1226 -1212- 1612 0.5 -1812- 1524 - 1538 - 1839 -24150.6 - 1745 - 1575 - 1596 -1802-2185 0.7 - 1503 - 1413 - 1429 - 1549 -17700.8 - 1112 -1071-1078- 1130 -12280.9 - 599 - 586 - 585 - 595 -617b) Gallium corner ΔG^{E} / J mol⁻¹ $x_{\rm Sb}: x_{\rm Tl} = 7:3$ $x_{\rm Sb}: x_{\rm Tl} = 9:1$ $x_{Sb}:x_{Tl} = 1:1$ $x_{\rm Sb}: x_{\rm Tl} = 3:7$ $x_{\rm Sb}:x_{\rm Tl} = 1:9$ x_{Ga} 0 - 632 - 1917 -2803-2790- 1380 -1031- 95 0.1-1786-2125-16850.2 - 1360 - 1639 -1518- 741 949 - 1606 - 1475 - 985 37 1763 0.3 0.4 - 1755 - 1297 - 534 640 2335 -17980.5 -1104- 173 1060 2656 0.6 -1720-89893 1285 2712 0.7 - 1512 -681255 1308 2493 0.8 - 1160 -452305 1116 1985 0.9 - 654 -213237 701 1178 c) Thallium corner $\Delta G^{\rm E}$ / J mol^-1 $x_{Ga}:x_{Sb} = 9:1$ $x_{Ga}:x_{Sb} = 7:3$ $x_{Ga}: x_{Sb} = 1:1$ $x_{Ga}:x_{Sb}=3:7$ $x_{\text{Ga}}:x_{\text{Sb}} = 1:9$ x_{Tl} 0 -610- 1589 -2075- 1893 - 871 - 1038 - 1577 - 968 404 0.1 - 1558 0.2 -1488-1578- 1194 -2441363 0.3 - 1911 - 1620 -904296 2042 0.4 - 2259 - 1652 - 690 666 2455 0.5 -2482- 1646 - 533 880 2615 0.6 -2530-1570-414950 2534 0.7 -2354- 1395 -316889 2225 1700 0.8 - 1904 - 1090 -220710 0.9 -1132-626-108424 972

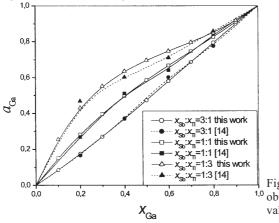
TABLE IV. Integral molar excess Gibbs energies, $\Delta G^{\rm E}$ (in J/mol), for the chosen sections in the Ga–Sb–Tl system at 1073 K according to the Hillert model

a) Antimony corner

creasing content of antimony in Ga–Sb–Tl alloys, both the gallium and thallium activity decrease strongly. On the other hand, replacement between gallium and thallium has only a small effect on the antimony activity.

The calculated activities of gallium from this work are graphically compared with the experimentally obtained gallium activities in Fig. 5.

The results presented in Fig. 5 show that there is good agreement between experimentally obtained and calculated activities of gallium in the $x_{Sb}:x_{Tl} = 3:1$ and 1:1 sections while for the section with a mole fractions ratio $x_{Sb}:x_{Tl} = 1:3$, the agreement is not so good.



4 Fig. 5. Comparison of the experimentally ^{1,0} obtained gallium activity and the calculated values in the Ga–Sb–Tl system at 1073 K.

The activity of gallium shows slight negative deviations from the Raoult law in the section with $x_{Sb}:x_{Tl} = 3:1$, moderately positive deviations in $x_{Sb}:x_{Tl} = 1:1$ section and large positive deviations from the ideal thermodynamic behavior in the section with $x_{Sb}:x_{Tl} = 1:3$.

CONCLUSION

The thermodynamic properties of the liquid Ga–Sb–Tl system were investigated using different models for thermodynamic predictions.

Comparison of the predicted integral excess Gibbs energies with the experimentally based results indicated the Hillert model to be the most accurate model for the thermodynamic description of the Ga–Sb–Tl system. The asymmetric nature of the investigated ternary system was additionally confirmed by the Chou similarity coefficient concept. Further thermodynamic calculation using the Hillert model was done in five sections of the ternary Ga–Sb–Tl system from each corner with mole ratios of other two components equal to 9:1; 7:3; 5:5; 3:7 and 1:9. The obtained results include integral excess Gibbs energy dependences *vs*. composition for all the investigated cross sections and isoactivity diagrams for all components of the Ga–Sb–Tl system at 1073 K. Comparison between the calculated and experimentally obtained gallium activities shows good agreement in the section with $x_{Sb}:x_{Tl} = 3:1$ and 1:1 and slight disagreement in the section with $x_{Sb}:x_{Tl} = 1:3$.

The results presented in this paper are a contribution to a more complete thermodynamic description of the Ga–Sb–Tl system and may be useful for the further thermodynamic assessment of this system.

ИЗВОД

ПРОРАЧУН ТЕРМОДИНАМИЧКИХ ОСОБИНА ТЕЧНИХ Ga-Sb-Tl ЛЕГУРА

ДРАГАН МАНАСИЈЕВИЋ 1 , ДРАГАНА ЖИВКОВИЋ 1 , IWAO КАТАУАМА 2 и ЖИВАН ЖИВКОВИЋ 1

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У овом раду је изложен прорачун термодинамичких величина у течним Ga–Sb–Tl легурама на 1073 К. На основу поређења прорачунатих интегралних ексцесних Гибсових енергија по различитим геометријским моделима термодинамичког предвиђања (Kohler, Muggianu, Toop, Hillert, Chou) са резултатима базираним на експерименталним подацима утврђено је да асиметрични модели дају тачније резултате. Асиметричност испитиваног система је додатно потврђена Chou-овим концептом коефицијената сличности. Из ових разлога потпуни прорачун термодинамичких карактеристика у тернарном Ga–Sb–Tl систему је извршен Hillert-овим моделом. Резултати обухватају зависности интегралних ексцесних Гибсових енергија од састава за петнаест испитиваних пресека као и дијаграме изоактивних линија за све три компоненте испитиваног система на 1073 К. Добијени резултати показују добро слагање са постојећим експерименталним резултатима.

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