

# Thermodynamic and phase diagram investigation of Pb–BiIn section in Pb–Bi–In ternary system

Dragana Živković\*, Dragan Manasijević, Živan Živković

*Department of Metallurgy, Technical Faculty, University of Belgrade, Vojske Jugoslavije 12, 19210 Bor, Serbia and Montenegro*

Received 10 November 2003; received in revised form 26 December 2003; accepted 22 January 2004

Available online 17 March 2004

## Abstract

The results of thermodynamic study of Pb–BiIn section of Pb–Bi–In system done experimentally by Oelsen calorimetry and calculated by general solution model, as well as measurements performed by DTA and optic microscopy in order to investigate phase diagram of the given section, are presented in this paper. Thermodynamic properties, such as integral molar enthalpies of mixing, activities, activity coefficients, partial and integral excess Gibbs energies at 600 and 650 K were determined and comparison between obtained experimental and calculated values were done. Also, based on cooling curves, DTA recordings and optic microscopy results phase diagram was constructed and compared with literature data.

© 2004 Elsevier B.V. All rights reserved.

*Keywords:* Thermodynamics; Pb–BiIn alloys; Calorimetry; General solution model; DTA

## 1. Introduction

Alloys in Pb–Bi–In ternary system are typical low melting alloys, which indicates their practical use as solder materials [1,2]. Having in mind that nowadays lead-free solder materials are used as their substitute, lead-based ternary alloys with low eutectics have been considered for different other applications. So, multicomponent and multiphase nanoscale precipitates consisting of nanodispersed Pb–Bi–In particles in aluminium matrix have been investigated recently [3,4], as well as possible application of such Pb–Bi-based alloys as a liquid metal cooling systems in the development of a design of natural-safety nuclear reactors and subcritical reactors controlled by a proton [5].

Considering phase diagram investigations of the Pb–Bi–In ternary system, there are a lot of references in literature [6–11], with the most recent assessment given by Boa and Ansara [12]. The liquidus projection of the Pb–Bi–In ternary system and a characteristic reaction scheme (taken from Ref. [6]) are given in Fig. 1.

Thermodynamic data are available in the literature for the binaries Pb–Bi [13,14], Bi–In [13,15] and In–Pb [13,16–18],

while thermodynamic properties of the Pb–Bi–In ternary system have been investigated by a few researchers: Vecher et al. [19] measured enthalpies of mixing of liquid alloys along five sections from the lead corner using DTA; Gregorczyk and Jurzyk [20] used EMF measurements to determine indium activities in the temperature range 673–873 K; Zheng and Kozuka [21] also used EMF measurements with solid electrolyte cell to obtain molar enthalpies of mixing and indium activities in the range 923–1123 K; while Boa and Ansara [12] did a thermodynamic assessment of the Pb–Bi–In system and obtained thermodynamic parameters for various phases using an optimization procedure.

As a contribution to a better knowledge of the investigated Pb–Bi–In system, since there is still a lack of complete experimental thermodynamic data, the results of an investigation of a Pb–BiIn section obtained experimentally by Oelsen calorimetry [22–24] and calculated by a general solution model [25], are presented in this paper. Also, a vertical section Pb–BiIn was investigated by means of DTA results, cooling curves and optic microscopy.

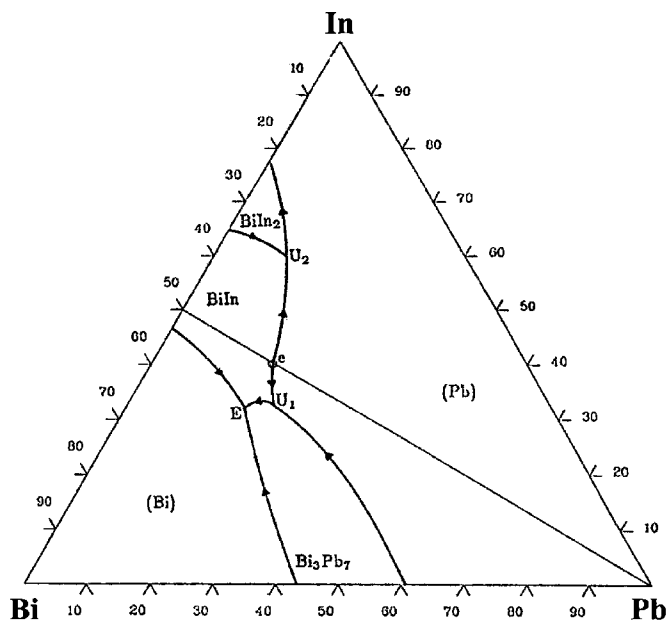
## 2. Experimental

Oelsen calorimetry was used for the thermodynamic analysis of Pb–BiIn section in the ternary system Pb–Bi–In. Detailed descriptions of this experimental technique are

\* Corresponding author. Tel.: +381-30-424-547;

fax: +381-30-424-547.

E-mail addresses: [dzivkovic@tf.bor.ac.yu](mailto:dzivkovic@tf.bor.ac.yu), [jmm@eunet.yu](mailto:jmm@eunet.yu) (D. Živković).



### Bi-In-Pb

e :	$L \rightleftharpoons \text{BiIn} + (\text{Pb})$	79°C	L =	39.7 at.% Bi , 39.7 at.% In
E :	$L \rightleftharpoons (\text{Bi}) + \text{BiIn} + \text{Bi}_3\text{Pb}_7(\text{HT})$	70°C	L =	48.2 at.% Bi , 31.0 at.% In
U <sub>1</sub> :	$L + (\text{Pb}) \rightleftharpoons \text{BiIn} + \text{Bi}_3\text{Pb}_7(\text{HT})$	73°C	L =	43.9 at.% Bi , 32.0 at.% In
U <sub>2</sub> :	$L + \text{BiIn} \rightleftharpoons \text{BiIn}_2 + (\text{Pb})$	74°C	L =	28.2 at.% Bi , 60.6 at.% In

Fig. 1. Liquidus projection of Pb–Bi–In ternary system and reaction scheme [6].

reported in [22–24]. Schematic representation of the used Oelsen water calorimeter is given in Fig. 2.

Such calorimeter consists of a Dewar flask with water, a stirrer and a thermometer. It also contains a housing from copper sheet into which the iron holder with the sample is added, after these have been heated in a separate oven to the initial temperature. The temperature of the sample is followed with NiCr–Ni thermocouple during cooling and simultaneously the temperature of the water is measured. The pair of values of a temperature measurement in the sample and in the water leads to a point on the heat content curve.

DTA curves were recorded on derivatograph MOM, Budapest (Hungary), at heating rate of  $10 \text{ K min}^{-1}$  in an air atmosphere, while  $\text{Al}_2\text{O}_3$  was used as a reference material during measurements.

Microstructure analysis of the samples was performed by optical microscopy, using a Reichert MeF2 microscope. Solution of (10%  $\text{HNO}_3$  + 2–3 drops of  $\text{H}_2\text{O}_2$ ) was applied for structure development.

The nine samples were chosen in the investigated section, along the line from the lead corner to the point in the binary Bi–In system, corresponding to the molar ratio of Bi:In = 1:1 (Fig. 1). According to the requirements of the method used, the total volume of all samples was constant  $0.2 \text{ cm}^3$ , while their composition and masses are given in Table 1.

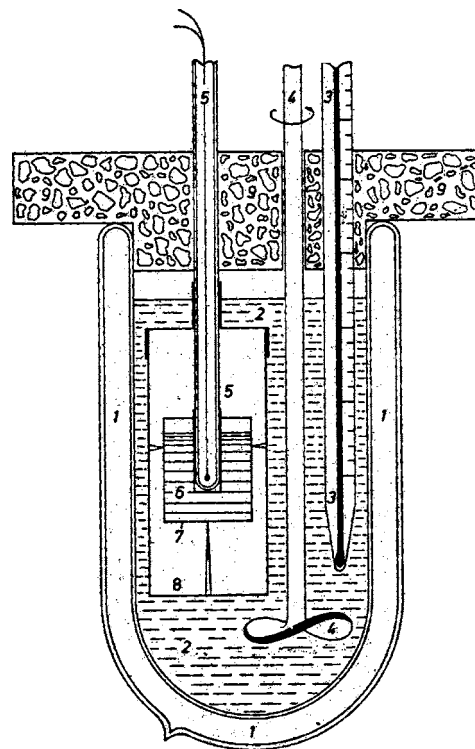


Fig. 2. Oelsen calorimeter. 1: Dewar flask; 2: water; 3: Beckman thermometer; 4: stirrer; 5: NiCr–Ni thermocouple; 6: sample; 7: sample holder; 8: copper sheet; 9: thermal insulation.

Table 1  
Composition and masses of the investigated samples

Alloy	$x_{\text{Pb}}$	$x_{\text{Bi}}$	$x_{\text{In}}$	Pb (mass%)	Bi (mass%)	In (mass%)	$m_{\text{tot}}$ (g)	$m_{\text{Pb}}$ (g)	$m_{\text{Bi}}$ (g)	$m_{\text{In}}$ (g)
e1	0.1	0.45	0.45	12.449	56.505	31.045	1.8010	0.2242	1.0177	0.5591
e2	0.18	0.41	0.41	21.932	50.385	27.683	1.8429	0.4042	0.9286	0.5102
e3	0.3	0.35	0.35	35.421	41.679	22.899	1.9061	0.6751	0.7944	0.4364
e4	0.5	0.25	0.25	56.137	28.309	15.554	2.0119	1.1294	0.5696	0.3129
e5	0.68	0.16	0.16	73.115	17.351	9.533	2.1079	1.5412	0.3657	0.2009
e6	0.8	0.1	0.1	83.658	10.547	5.795	2.1722	1.8172	0.2291	0.1259
e7	0.9	0.05	0.05	92.012	5.156	2.833	2.2259	2.0482	0.1148	0.0631
e8	0	0.5	0.5	0	64.539	35.460	1.7488	0	1.1287	0.6201

The water equivalent was determined by a standard method using dissolved  $\text{Na}_2\text{CO}_3$  and for the calorimeter used it was found to be  $3453 \text{ J K}^{-1}$ .

All experiments were carried out in an air atmosphere, with metals of the analytical grade.

### 3. Results and discussion

Oelsen developed, through combination of simple classical thermal analysis with classical calorimetry at room temperature, a procedure with which the heat content curve of a material can be determined relatively rapidly over large temperature ranges in a single experiment.

The method is applied to determine the mixing enthalpies of liquid alloys. But, usually, further quantitative thermodynamics analysis, proposed by Oelsen [22,24], is applied. It includes graphical planimetry and indirect determination of activities for constitutive components.

The main theoretic fundamentals of Oelsen's approach are presented here for the eutectic system A–B. Concerning the alloy of the composition  $x_{\text{B}} = x$  at the temperature  $T$ , the integral Gibbs energy of mixing is given as [22]

$$\begin{aligned} \Delta G_{x,T}^{\text{M}} &= (1-x)G_{\text{A},T}^{\text{M}} + xG_{\text{B},T}^{\text{M}} \\ &= (1-x)RT \ln a_{\text{A},T} + xRT \ln a_{\text{B},T} \end{aligned} \quad (1)$$

or according to the Gibbs–Helmholtz equation

$$\Delta G_{x,T}^{\text{M}} = \Delta H_{x,T}^{\text{M}} - T \Delta S_{x,T}^{\text{M}} \quad (2)$$

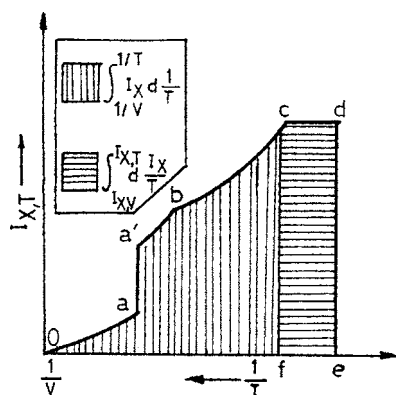


Fig. 3. Illustration of a graphical planimetry.

and the following expression is obtained:

$$\Delta H_{x,T}^{\text{M}} - T \Delta S_{x,T}^{\text{M}} = (1-x)RT \ln a_{\text{A},T} + xRT \ln a_{\text{B},T} \quad (3)$$

Further determination of the activities for the constitutive components is conceived on the knowledge of the enthalpy and entropy term. Under equilibrium conditions and constant pressure, the entropy term can be written as

$$dS_{x,T}^{\text{M}} = \frac{dH_{x,T}^{\text{M}}}{T} \quad (4)$$

and

$$S_{x,T}^{\text{M}} = S_{x,T=0}^{\text{M}} + \int_{H_{x,T=0}^{\text{M}}}^{H_{x,T}^{\text{M}}} \frac{dH_{x,T}^{\text{M}}}{T} \quad (5)$$

and having in mind that  $S_{x,T=0}^{\text{M}} = 0$ , it follows that

$$S_{x,T}^{\text{M}} = \int_{H_{x,T=0}^{\text{M}}}^{H_{x,T}^{\text{M}}} \frac{dH_{x,T}^{\text{M}}}{T} \quad (6)$$

Based on Eqs. (3) and (6), one could obtain

$$\Delta G_{x,T}^{\text{M}} = H_{x,T}^{\text{M}} - T \int_{H_{x,T=0}^{\text{M}}}^{H_{x,T}^{\text{M}}} \frac{dH_{x,T}^{\text{M}}}{T} \quad (7)$$

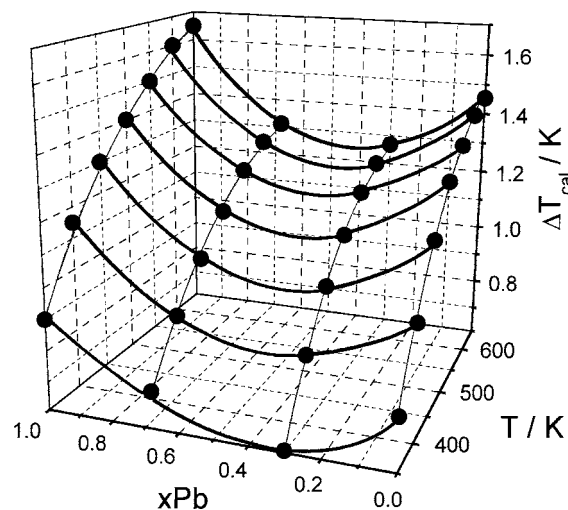


Fig. 4. Space enthalpy diagram for the investigated section.

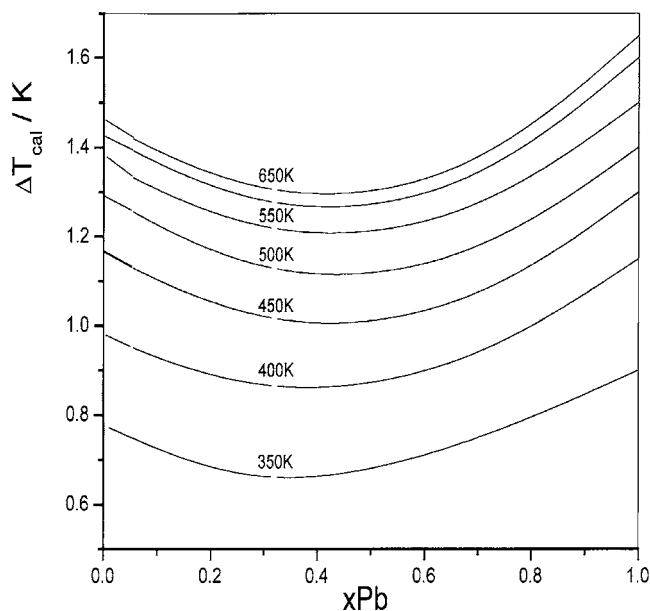


Fig. 5. The enthalpy isotherm diagram for the temperature interval 350–650 K.

For a better survey, Oelsen introduced the quantity  $I_{x,T}$  [22], which represents the value of the enthalpy, directly determined in a calorimeter:

$$I_{x,T} = H_{x,T}^M - H_{x,V}^M \quad (8)$$

where  $V$  is the room temperature of 298 K. Multiplying Eq. (7) with  $(-T)$  and using Eq. (8), it follows:

$$-\frac{G_{x,T}^M}{T} = \int_{I_{x,T=0}}^{I_{x,T}} \frac{dI_{x,T}^M}{T} \int_{1/V}^{1/T} I_{x,T} d\left(\frac{1}{T}\right) \quad (9)$$

which presents the important Oelsen integral. Solution of this integral is done using a graphical planimetry of the area below the cooling curve, as presented in Fig. 3.

Therefore, the basic equation of the Oelsen quantitative thermodynamic analysis [22] is

$$-\frac{G_i^M}{T} = \int_{1/T_0}^{1/T} H_{x,T} d\left(\frac{1}{T}\right) = -R \ln a_i \quad (10)$$

Table 2

Results of the Oelsen's quantitative thermodynamic analysis at 600 and 650 K (energies in  $\text{J mol}^{-1}$ )

Alloy	$x_{\text{Pb}}$	600 K				650 K			
		$a_{\text{Pb}}$	$\gamma_{\text{Pb}}$	$G_{\text{Pb}}^E$	$G_{\text{Pb}}^M$	$a_{\text{Pb}}$	$\gamma_{\text{Pb}}$	$G_{\text{Pb}}^E$	$G_{\text{Pb}}^M$
e1	0	0	–	–	0	0	–	–	0
e2	0.1	0.130	1.300	1309	–10177	0.129	1.290	1376	–11067
e3	0.18	0.211	1.173	798	–7761	0.227	1.261	1254	–8013
e4	0.3	0.351	1.170	783	–5223	0.356	1.187	925	–5581
e5	0.5	0.587	1.174	800	–2657	0.582	1.164	821	–2925
e6	0.68	0.754	1.109	515	–1409	0.731	1.075	391	–1693
e7	0.8	0.835	1.044	214	–900	0.829	1.036	192	–1013
e8	0.9	0.912	1.013	66	–460	0.91	1.011	60	–510

Table 3

Binary parameters and similarity coefficients at 650 K for systems Pb–Bi, Bi–In and In–Pb

System $ij$	System $ij$				
	$A_{ij}^0$	$A_{ij}^1$	$A_{ij}^2$	$A_{ij}^3$	$\xi_{ij}$
Pb–Bi	–5382.24	–663.407	74.84	0	0.946
Bi–In	–7409.010	1231.681	142.365	0	0.036
In–Pb	3145.45	207.2	0	0	0.605

where are:  $G_i^M$  the partial Gibbs energy of mixing for the component  $i$ ,  $T_0$  the starting temperature,  $T$  the final temperature,  $H_{x,T}$  the enthalpy value measured in the Oelsen calorimeter for the temperature change from  $T_0$  to  $T$ ,  $R$  the gas constant, and  $a_i$  the activity of the component  $i$ .

Based on the cooling curves obtained for investigated samples in Pb–BiIn section using Oelsen's calorimetry, the temperature changes of the calorimeter were determined for all samples in the investigated temperature range 350–650 K. That allowed to determine the dependence of the calorimeter temperature change on composition and temperature, shown in Fig. 4, and the construction of the enthalpy isotherm diagram for the investigated temperature interval 350–650 K, presented in Fig. 5. (Note: in presented diagrams 1 K correspond to determined water equivalent of  $3453 \text{ J K}^{-1}$ .)

Based on Eq. (1) and the results of graphic planimetry, the tangent was constructed for the calculation of  $-R \ln a_{\text{Pb}}$  at 600 and 650 K, which enabled the determination of lead activities, activity coefficients and partial molar quantities. The results are given in Table 2.

A positive deviation from Raoult's law was noticed in the whole concentration range at both investigated temperatures. Partial molar excess Gibbs energies for lead exhibit positive values in the whole concentration area at both temperatures.

Since Oelsen's calorimetry results are related only to lead, thermodynamic calculation according to Chou's general solution model [25] was applied in order to obtain the integral thermodynamic quantities in the investigated section Pb–BiIn of the ternary system Pb–Bi–In. This model whose successful application has been confirmed in recent

Table 4

Results of the thermodynamic calculation according to Chou's general solution model at 650 K (energies in  $\text{J mol}^{-1}$ )

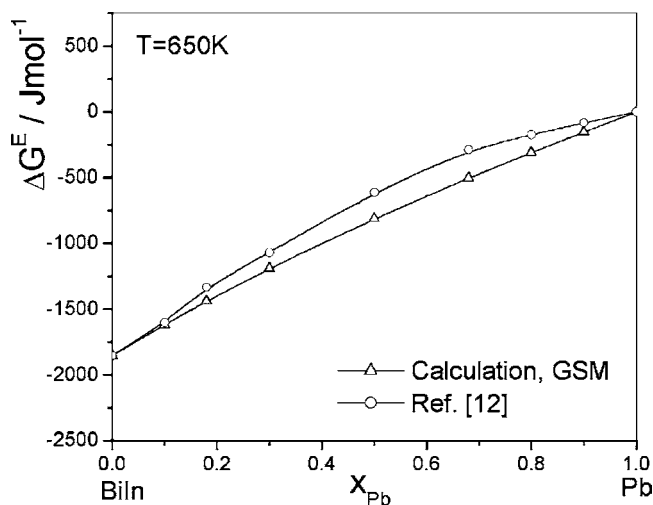
Alloy	$x_{\text{Pb}}$	$f$	$\Delta G^E$	$G_{\text{Pb}}^E$	$\gamma_{\text{Pb}}$	$a_{\text{Pb}}$
e1	0	-1227.99	-1852	-	-	0
e2	0.1	-1257.35	-1617	578	1.112885	0.111289
e3	0.18	-1286.71	-1440	429	1.08262	0.194872
e4	0.3	-1321.94	-1191	329	1.062771	0.318831
e5	0.5	-1374.79	-812	279	1.052983	0.526492
e6	0.68	-1433.52	-503	103	1.019242	0.693085
e7	0.8	-1468.75	-309	46	1.008548	0.806839
e8	0.9	-1492.24	-153	6	1.001111	0.901

literature [26–30] for thermodynamic calculation is based on the knowledge of thermodynamic data for constituent binary subsystems. The basic theoretical interpretations of this method [25] are given:

$$\begin{aligned} \Delta G^E = & x_1 x_2 (A_{12}^0 + A_{12}^1 (x_1 - x_2) + A_{12}^2 (x_1 - x_2)^2) \\ & + x_2 x_3 (A_{23}^0 + A_{23}^1 (x_2 - x_3) + A_{23}^2 (x_2 - x_3)^2) \\ & + x_3 x_1 (A_{31}^0 + A_{31}^1 (x_3 - x_1) + A_{31}^2 (x_3 - x_1)^2) \\ & + f x_1 x_2 x_3 \end{aligned} \quad (11)$$

where  $A_{ij}^0$ ,  $A_{ij}^1$ ,  $A_{ij}^2$  are the parameters for binary system “ $ij$ ” independent of composition, only depending on temperature, and  $x_1$ ,  $x_2$ ,  $x_3$  correspond to the mole fraction of the components in investigated ternary system, which have been used in the regular type equation [25]:

$$\begin{aligned} \Delta G_{ij}^E = & X_i X_j (A_{ij}^0 + A_{ij}^1 (X_i - X_j) + A_{ij}^2 (X_i - X_j)^2 \\ & + \dots + A_{ij}^n (X_i - X_j)^n) \end{aligned} \quad (12)$$

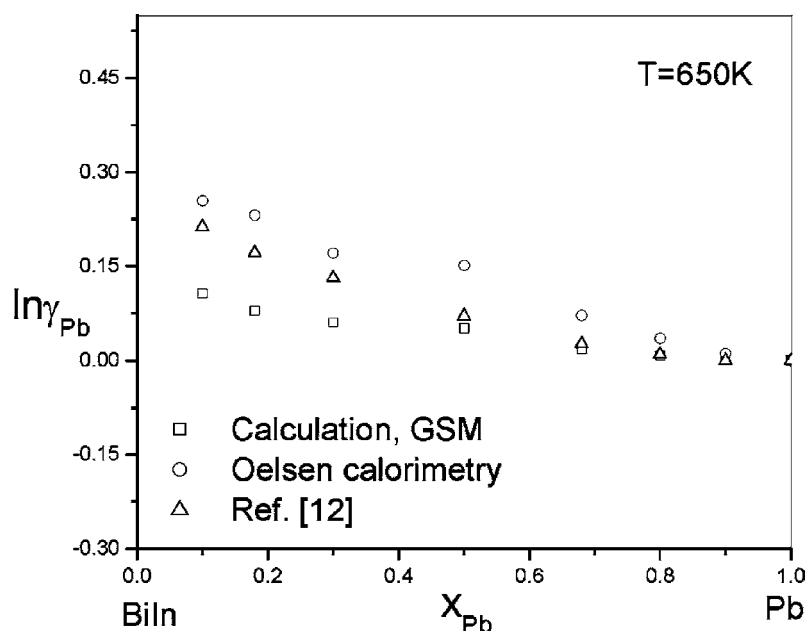
Fig. 6. Comparison of  $\Delta G^E$  on  $x_{\text{Pb}}$  at 650 K.

$X_i$  and  $X_j$  are the mole fractions of component “ $i$ ” and “ $j$ ” in the “ $ij$ ” binary system. In Eq. (11)  $f$  is the ternary interaction coefficient expressed by [25]

$$\begin{aligned} f = & (2\xi_{12} - 1) \{ A_{12}^2 ((2\xi_{12} - 1)x_3 + 2(x_1 - x_2)) + A_{12}^1 \} \\ & + (2\xi_{23} - 1) \{ A_{23}^2 ((2\xi_{23} - 1)x_1 + 2(x_2 - x_3)) + A_{23}^1 \} \\ & + (2\xi_{31} - 1) \{ A_{31}^2 ((2\xi_{31} - 1)x_2 + 2(x_3 - x_1)) + A_{31}^1 \}, \end{aligned} \quad (13)$$

where  $\xi_{ij}$  are the similarity coefficients defined by  $\eta_i$  called the deviation sum of squares [25]:

$$\xi_{ij} = \frac{\eta_i}{\eta_i + \eta_j} \quad (14)$$

Fig. 7. Comparison between experimentally obtained, predicted and literature data for  $\ln \gamma_{\text{Pb}}$  at 650 K.

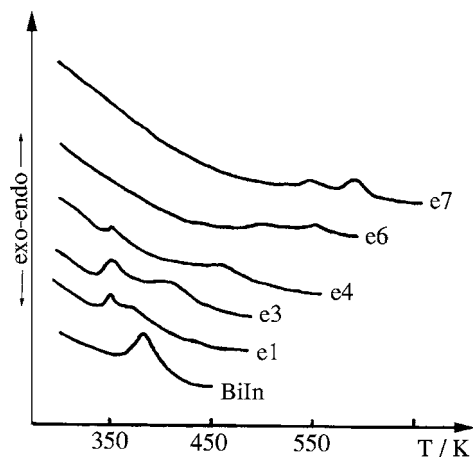


Fig. 8. DTA curves for samples with  $x_{\text{Pb}} = 0.1, 0.3, 0.5, 0.8$  and  $0.9$  obtained at the heating rate of  $10^\circ\text{C min}^{-1}$  in an air atmosphere.

where, according to Chou's model [25], deviation sum of squares are defined as:

$$\begin{aligned} \eta_{\text{I}} &= \int_{X_i=0}^{X_i=1} (\Delta G_{12}^{\text{E}} - \Delta G_{13}^{\text{E}})^2 dX_1, \\ \eta_{\text{II}} &= \int_{X_i=0}^{X_i=1} (\Delta G_{21}^{\text{E}} - \Delta G_{23}^{\text{E}})^2 dX_2, \\ \eta_{\text{III}} &= \int_{X_i=0}^{X_i=1} (\Delta G_{31}^{\text{E}} - \Delta G_{32}^{\text{E}})^2 dX_3 \end{aligned} \quad (15)$$

and where are

$$\begin{aligned} X_{1(12)} &= x_1 + x_3\xi_{12}, & X_{2(23)} &= x_2 + x_1\xi_{23}, \\ X_{3(31)} &= x_3 + x_2\xi_{31} \end{aligned} \quad (16)$$

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

Values for the integral molar excess Gibbs energies for the constituent binary systems Pb–Bi, Bi–In and In–Pb, available in the literature for temperature of 650 K [12], were used as the starting data for calculation. The binary parameters were calculated using Eq. (12) and presented in Table 3. Also, similarity coefficients for three constituent binary sys-

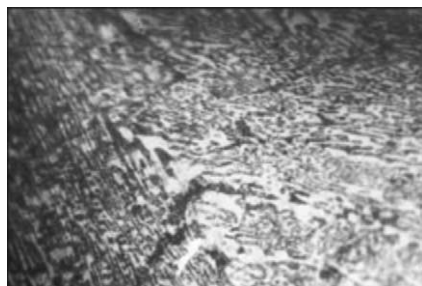


Fig. 9. Microphotograph for sample e2.

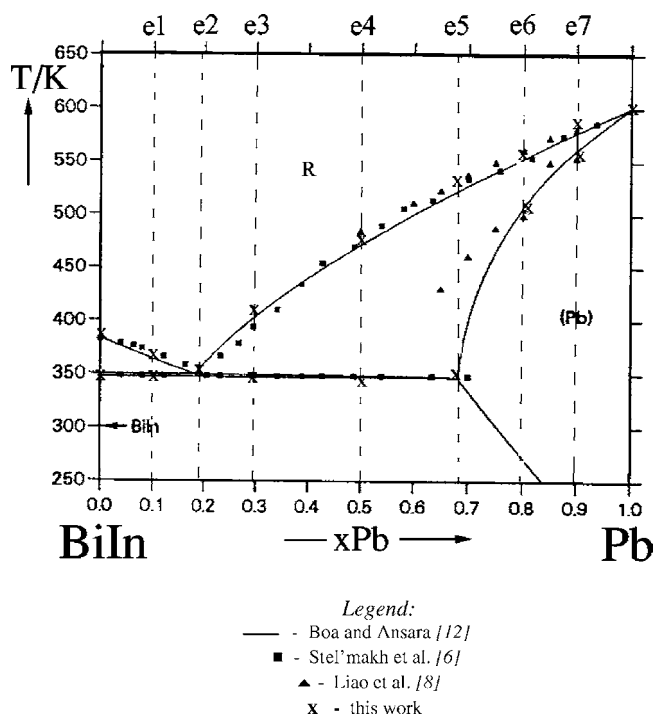


Fig. 10. Phase diagram of investigated Pb–BiIn quasibinary section—comparative review.

tems, calculated according to Chou's analytical procedure [25], are presented in the same table.

Based on these data and according to Eq. (11), thermodynamic calculations were performed for the selected alloys (Table 1) in section Pb–BiIn of the Pb–Bi–In system, and the values of integral molar excess Gibbs energies and ternary interaction coefficients were determined. In order to compare the experimental results with the calculations, partial thermodynamic quantities for lead were derived according to the following equation:

$$G_{\text{Pb}}^{\text{E}} = G^{\text{E}} + (1 - x_{\text{Pb}}) \left( \frac{\partial G^{\text{E}}}{\partial x_{\text{Pb}}} \right)$$

The results of the thermodynamic calculation, as well as the derived values for lead thermodynamic quantities, are shown in Table 4, while the dependence of integral molar Gibbs excess energies on composition is given in Fig. 6 compared to calculated data in Ref. [12].

As can be seen, negative values for the integral molar excess Gibbs energies were obtained in the whole concentration range of the Pb–BiIn section, while positive values for partial molar Gibbs excess energies for lead were derived. Slight positive deviation from Raoult's law is noticed, similarly to experimentally obtained results by Olsen calorimetry.

Furthermore, the comparison between experimental, calculated and literature data [12] for  $\ln \gamma_{\text{Pb}}$  at investigated temperature of 650 K is presented in Fig. 7. As can be seen, there is a good agreement between compared data in the range of  $x_{\text{Pb}} = 0.5-1$ , while for lower molar contents of lead, slight



disagreement occurs. For the characterization of investigated alloys, DTA results and optic microscopy were used.

Results obtained by DTA for alloys with  $x_{\text{Pb}} = 0.1, 0.3, 0.5, 0.8$  and  $0.9$  are shown in Fig. 8, while microphotograph of the eutectic (sample e2) is presented in Fig. 9.

Based on the presented results of characterization of alloys, the phase diagram of Pb–BiIn section was constructed and the occurrence of eutectic reaction: liquid  $\rightarrow$  (Pb) + BiIn, was confirmed at 349 K and  $x_{\text{Pb}} = 0.19$ . The comparison with different literature data, given in Fig. 10, shows relatively good mutual agreement.

#### 4. Conclusion

Comparative thermodynamic analysis of Pb–BiIn section in the Pb–Bi–In ternary system was done based on the experimental calorimetric investigations according to Oelsen's method and thermodynamic calculation by Chou's general solution model. Values for the activities, activity coefficients, partial molar quantities for lead at 600 and 650 K, and integral molar quantities at a temperature of 650 K were obtained and their comparison shows reasonable agreement. Characterization of the investigated alloys was done by DTA and optic microscopy.

#### References

- [1] W. Hofmann, *Lead and Lead Alloys*, McGraw Hill, New York, 1970.
- [2] See at [http://www.indium.com/products/alloy/alloy\\_tablep1.html](http://www.indium.com/products/alloy/alloy_tablep1.html) and <http://www.frymetals.com/pdf/uploads/tinleadcadmiumindiumbismuthalloys.pdf>.
- [3] E. Johnson, A. Johansen, V.S. Touboltsev, U. Dahmen, *International Symposium on Metastable, Mechanically Alloyed and Nanocrystalline Materials (ISMANAM-2000)*, Oxford, UK, Session 8, Immiscible Systems, 11-08-01I, July 2000.
- [4] V. Bhattacharya, K. Chattopadhyay, *International Symposium on Metastable, Mechanically Alloyed and Nanocrystalline Materials (ISMANAM-2000)*, Oxford, UK, Session 8, Immiscible Systems, 11-08-04T, July 2000.
- [5] See at <http://www.tech-db.ru>.
- [6] S.I. Stel'makh, V.A. Zimmergaki, I.A. Sheka, *Ukr. Khim. Zh.* 40 (1974) 471.
- [7] S.I. Stel'makh, V.A. Zimmergaki, I.A. Sheka, *Ukr. Khim. Zh.* 40 (1974) 762.
- [8] K.C. Liao, D.L. Johnson, R.C. Nelson, *Mater. Res. Bull.* 10 (1975) 1225.
- [9] V.D. Bushmanov, V.G. Il'ves, S.P. Yatsenko, *Izv. Akad. Nauk. SSSR, Metally* 1 (1988) 216.
- [10] A. Prince, *Mater. Res. Bull.* 11 (1976) 1105.
- [11] V.G. Il'ves, V.V. Filippov, S.P. Yatsenko, *Izv. Akad. Nauk. SSSR, Metally* 5 (1992) 166.
- [12] D. Boa, I. Ansara, *Thermochim. Acta* 314 (1998) 79.
- [13] R. Hultgren, P.D. Desai, D.T. Hawkins, M. Gleiser, K. Kelley, *The Selected Values of Thermodynamic Properties of Binary Alloys*, ASM, Metals Park, OH, 1973.
- [14] S.W. Yoon, H.M. Lee, *Calphad* 22 (1998) 167.
- [15] Y. Cui, S. Ishihara, Y.J. Liu, I. Ohnuma, R. Kainuma, H. Ohtani, K. Ishida, *Mater. Trans.* 43 (2002) 1879.
- [16] J.P. Nabot, I. Ansara, *Bull. Alloy Phase Diagr.* 8 (3) (1987) 246.
- [17] A. Bolcavage, C.G. Rao, S.L. Chon, Y.A. Chang, in: P. Nash, B. Sundman (Eds.), *Proceedings of the Symposium on Application of Thermodynamics in Synthesis and Processing of Materials*, TMS, Warrendale, PA, 1995, p. 171.
- [18] D. Minic, D. Zivkovic, Z. Zivkovic, *Thermochim. Acta* 372 (2001) 85.
- [19] A.A. Večer, L.D. Zal'tsman, L.A. Mechkovskii, A.S. Skoropanov, *Russ. J. Phys. Chem.* 49 (1975) 1297.
- [20] Z. Gregorczyk, E. Jurzyk, *J. Chem. Thermodyn.* 16 (1984) 551.
- [21] M. Zheng, Z. Kozuka, *Trans. Jpn. Inst. Met.* 28 (1987) 925.
- [22] W. Oelsen, P. Zuhlke, *Arch. Eisenhüttenwess.* 27 (12) (1956) 743; W. Oelsen, F. Johannsen, A. Podgornik, *Zeitsch. Erzbergbau Metallhüttenwus.* 9 (10) (1956) 1; W. Oelsen, E. Schürmann, H.J. Weigt, O. Oelsen, *Arch. Eisenhüttenwess.* 27 (12) (1956) 487.
- [23] B. Predel, I. Arpshofen, M.J. Pool, *Thermochim. Acta* 22 (1978) 211.
- [24] A. Kostov, D. Zivkovic, Z. Zivkovic, *Thermochim. Acta* 338 (1999) 35–45.
- [25] K.C. Chou, *Calphad* 19 (3) (1995) 315.
- [26] D. Zivkovic, Z. Zivkovic, Y.H. Liu, *J. Alloys Comp.* 265 (1998) 176.
- [27] D. Zivkovic, Z. Zivkovic, J. Sestak, *Calphad* 23 (1) (1999) 113.
- [28] I. Katayama, K. Yamazaki, Y. Hattori, T. Iida, *Mater. Trans. JIM* 41 (2) (2000) 251.
- [29] D. Zivkovic, Z. Zivkovic, L. Stuparevic, *J. Therm. Anal. Calor.* 65 (2001) 805.
- [30] I. Katayama, K. Yamazaki, M. Nakano, T. Iida, *Scand. J. Metall.* 32 (2003) 1.