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Review

Enthalpies of mixing of Au–Pb and Ag–Au–Pb liquid alloys at 973 K

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Abstract

The enthalpies of mixing of the gold–lead and silver–gold–lead alloys were determined at 973 K by using a high temperature Calvet calorimeter ($400\,\text{K} < T < 1400\,\text{K}$). The mixing enthalpies of Au–Pb liquid are negative over the entire composition range with a minimum $\Delta_{\text{mix}}H_{\text{m}} = -1074\,\text{J}\,\text{mol}^{-1}$ at $X_{\text{Au}} = 0.40$. The enthalpies of formation of ternary liquid alloys have been carried out along the following sections: $X_{\text{Ag}}/X_{\text{Pb}} = 1/4$, $X_{\text{Au}}/X_{\text{Pb}} = 1/4$ and 2/3 at 973 K. Experimental values of $\Delta_{\text{mix}}H_{\text{m}} = f(X_{\text{Ag}}, X_{\text{Au}})$ are compared with those predicted from the Kohler, Muggianu and Toop relations. An assessment including the excess ternary parameter is presented.

Keywords: Calorimetry; Enthalpy of mixing; Alloys; Gold–lead and silver–gold–lead system

Contents

1. Introduction

Very few works were devoted to the Ag–Au–Pb ternary system. The first thermodynamic studies were undertaken by Hager and Zambrano [1] which determined the liquidus temperatures for seven alloys and a 1200 K liquidus isotherm derived from electromotive force data.

In our previous paper [2], we proposed the equilibrium phase diagram of this ternary system.

The present thermodynamic investigation is relative to the determination of the enthalpy of mixing of the Au–Pb binary and Ag–Au–Pb ternary systems at 973 K.

2. Bibliographic survey

Any estimate of the enthalpy of mixing function for a ternary system requires knowledge of the same function for the three limiting binary systems. Therefore, we reviewed the published

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reports on the enthalpies of mixing for the Ag–Au, Ag–Pb and Au–Pb alloys.

2.1. *Ag–Au system*

Gold and silver, which have very similar crystal structures and many close physico-chemical properties can be mixed in any amount. In both the liquid and the solid states, this system has been the subject of many thermodynamic investigations. The enthalpies of mixing obtained by various authors are in good agreement. These data were assessed by number of authors and the assessment of Hassam et al. [3] that which is generally retained in the thermodynamic databases [4].

2.2. *Ag–Pb system*

By calorimetry, numerous determinations were carried out in the Ag–Pb alloys by Kawakami [5] at 1323 K, Von Samson-Himmelstjerna [6] at 1000 K, Kleppa [7] at 723 K, Ehrlich [8] at 1248 K, Itagaki and Yazawa [9] at 1243 K, Kozuka et al. [10] at 1273 K and Castanet et al. [11] at 1280 K. All these results show a good agreement except for the higher values of Kawakami [5]. Other results were published; they derive from indirect measurements by e.m.f. and vapour pressure methods [12–15]. The enthalpy of mixing of the liquid Ag–Pb alloys were assessed by Lukas co-workers [16], Karakaya and Thompson [17], Lee et al. [18] and more recently by Lukas [19,20].

2.3. *Au–Pb system*

Several determinations of the molar enthalpies of mixing in liquid Au–Pb system have been performed.

The enthalpies obtained by Kleppa [7] by liquid Pb solution calorimetry at 623 and 723 K in the composition range $0.019 < X_{Au} < 0.327$ are very close to those of Béja [21] at 823 K.

Kleppa [22] deduced also the enthalpies of formation from e.m.f. measurements at 873 K but the values are in disagreement with his own previous calorimetric results [7]. The enthalpy data calculated from e.m.f. by Hager and Walker [23] at 1200 K and Kameda et al. [24] at 1223 K, respectively, show also a large discrepancy.

Okamoto and Massalski [25] have performed a first assessment of the Au–Pb diagram using the thermodynamic data reported by the previous authors but, because these data appeared to be contradictory, the thermodynamic calculations were made using two models according to the hypothesis that the $\Delta_{\mathrm{mix}} H_{\mathrm{m}}^{\circ}$ is dependent of the temperature or not. However, both models reproduced phase diagram equally well, so, no definite conclusion could be obtained.

Taking into account the measurements of the enthalpy of the Au–Pb alloys of Michel and Castanet [26] at 695, 871 and 1123 K, Okamoto [27] attempted a new assessment but the results of the calculations are not good enough because the (Au) rich liquidus proposed by Michel and Castanet [26] is not in agreement with the experimental phase boundary data of Hager and Walker [23] and Evans [28]. A conclusion of this author was that further experimental studies are needed for reconciliation of the thermodynamic and phase diagram data.

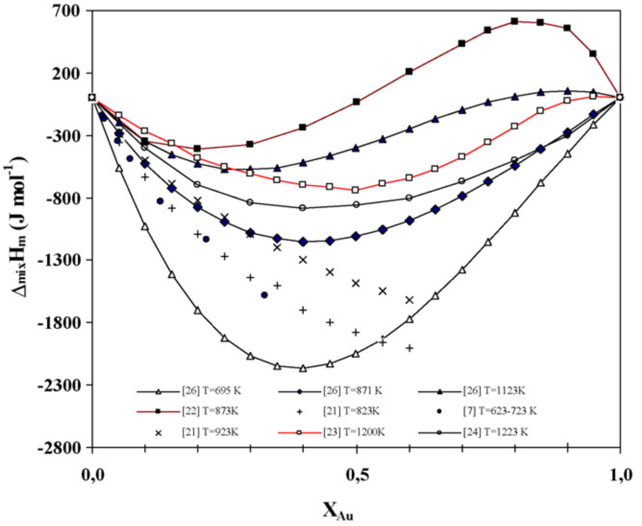

Fig. 1. Integral enthalpies of mixing of the Au–Pb binary system from literature.

In order to attempt to solve this conflict in the available experimental data and the disagreement between all the results as shown in Fig. 1, we propose a new calorimetric investigation at 973 K.

3. Experimental procedure

High purity metals Au (4 N) wire and Ag (4 N) (ingots) from Engelhard CLAL (Comptoir Lyon Alemand Louyot) and Pb (4 N) rods from Alfa Aesar, were employed in this study. The gold was rinsed with acetone p.a., and dried. The lead was scraped with a scalpel.

Measurements were performed by the direct drop method [29] using a high temperature Calvet calorimeter ($T < 1400$ K) with an experimental set up described elsewhere [30].

The alloying process took place in a graphite crucible (about 50 mm in height, 8 mm inner diameter and 9 mm outer diameter), located at the bottom of the calorimetric cell. The Au–Pb binary alloys were formed by multiple successive additions of a lot of pieces of gold, stabilized at room temperature, into the liquid lead at the experimental temperature. The ternary alloys were synthesized by additions of pieces of silver or gold in the liquid Au_xPb_{1-x} or Ag_xPb_{1-x} alloys. The integral enthalpy of mixing is obtained by summation of all the energy increments measured up to the considered concentration.

The calibrations were accomplished at the end of each experiment by dropping small samples of α-Al_2O_3 from N.I.S.T. [31] from the ambient temperature into the calorimeter.

The standard deviation of the calibration experiments is about 0.1 % and the uncertainty in the final value of the enthalpy of formation is estimated to be 5%.

Table 1
Experimental values of the integral enthalpy of mixing of the Au–Pb system at 973 K

X_{Au}	$\Delta_{\mathrm{mix}}H_{\mathrm{m}}$ (J mol^{-1})
0.0446	−240
0.0465	−181
0.0726	−383
0.0864	−464
0.0922	−345
0.1272	−631
0.1358	−531
0.1368	−643
0.1653	−729
0.1768	−665
0.1938	−789
0.2003	−847
0.2148	−782
0.2333	−876
0.2482	−885
0.2506	−854
0.2652	−952
0.2841	−920
0.2962	−1005
0.2966	−955
0.3161	−929
0.3226	−1019
0.3436	−985
0.3455	−969
0.3733	−985
0.3862	−1002
0.402	−1007
0.4248	−1007
0.4294	−1039
0.4416	−1040

4. Calorimetric results

4.1. Au–Pb system

The molar enthalpies of mixing of binary alloys are determined at 973 K over the composition range $0 < X_{\mathrm{Au}} < 0.46$. Results were referred to liquid undercooled gold and liquid lead with a heat of fusion for gold of 12.18 kJ mol^{-1} at 973 K [32].

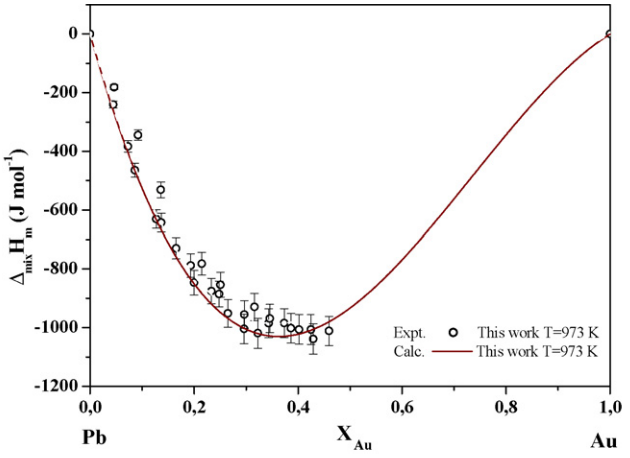

Fig. 2. Integral enthalpies of mixing of the liquid Au–Pb system, experimental results at 973 K. Our assessment at 973 K.

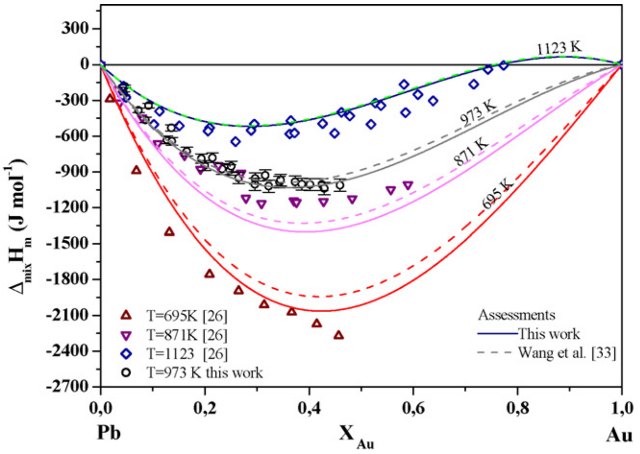

Fig. 3. Comparison between our results, Michel's data [26], our assessment and Wang's assessment [33].

The experimental results are recorded in Table 1 and are plotted in Fig. 2.

The molar integral enthalpies are negative over the entire investigated composition range. The minimum of the enthalpy vs. concentration is located at about $X_{\mathrm{Au}} = 0.40$ and $\Delta_{\mathrm{mix}}H^{\circ}_{\mathrm{m}} = -1074$ J mol^{-1}.

Fig. 3 represents the comparison between our results, our assessment, the Michel and Castanet's [26] experimental values at three temperatures and the calculated curves deduced from the recent Wang's optimization [33]. It can be noticed that Wang's optimization is in good agreement with our results.

4.2. Ag–Au–Pb system

The integral enthalpies of mixing of ternary alloys were determined at 973 K along the following sections:

$$\frac{X_{\mathrm{Ag}}}{X_{\mathrm{Pb}}} = \frac{1}{4} \quad \text{with} \quad 0 < X_{\mathrm{Au}} < 0.44$$

$$\frac{X_{\mathrm{Au}}}{X_{\mathrm{Pb}}} = \frac{1}{4} \quad \text{with} \quad 0 < X_{\mathrm{Ag}} < 0.42$$

$$\frac{X_{\mathrm{Au}}}{X_{\mathrm{Pb}}} = \frac{2}{3} \quad \text{with} \quad 0 < X_{\mathrm{Ag}} < 0.45$$

The results of the measurements are given in Table 2 and shown in Figs. 4–6.

5. Thermodynamic modelling

The molar Gibbs energy of the binary liquid mixture can be described as random mixtures of elements A and B by a sub regular-solution type model [34,35]

$$G_{\mathrm{A-B}} = x_{\mathrm{A}}G^{\circ}_{\mathrm{A}} + x_{\mathrm{B}}G^{\circ}_{\mathrm{B}} + RT(x_{\mathrm{A}}\ln x_{\mathrm{A}} + x_{\mathrm{B}}\ln x_{\mathrm{B}}) + x_{\mathrm{A}}x_{\mathrm{B}}\sum_{\nu=0}^{\nu} L^{\nu}_{\mathrm{A-B}}(x_{\mathrm{A}} - x_{\mathrm{B}})^{\nu} \quad (1)$$

Table 2
Experimental values of the integral enthalpy of mixing of the Ag–Au–Pb ternary system along the sections $X_{\mathrm{Ag}}/X_{\mathrm{Pb}} = 1/4$, $X_{\mathrm{Au}}/X_{\mathrm{Pb}} = 1/4$ and $X_{\mathrm{Au}}/X_{\mathrm{Pb}} = 2/3$ at 973 K

$T = 973$ K					
$X_{\mathrm{Ag}}/X_{\mathrm{Pb}} = 1/4$		$X_{\mathrm{Au}}/X_{\mathrm{Pb}} = 1/4$		$X_{\mathrm{Au}}/X_{\mathrm{Pb}} = 2/3$	
X_{Au}	$\Delta_{\mathrm{mix}}H_{\mathrm{m}}$ (J mol^{-1})	X_{Ag}	$\Delta_{\mathrm{mix}}H_{\mathrm{m}}$ (J mol^{-1})	X_{Ag}	$\Delta_{\mathrm{mix}}H_{\mathrm{m}}$ (J mol^{-1})
0.0000	2322	0.0000	−886	0.0000	−1074
0.0550	1741	0.0450	−435	0.0408	−880
0.1153	1275	0.0864	−43	0.0925	−627
0.1711	834	0.1284	306	0.1469	−411
0.2224	527	0.1755	697	0.1957	−240
0.2688	266	0.2212	990	0.2420	−110
0.3110	8	0.2637	1191	0.2836	44
0.3507	−173	0.3037	1364	0.3255	153
0.3830	−311	0.3454	1493	0.3648	182
0.4199	−477	0.3851	1615	0.4089	205
0.4436	−520	0.4241	1674	0.4491	315

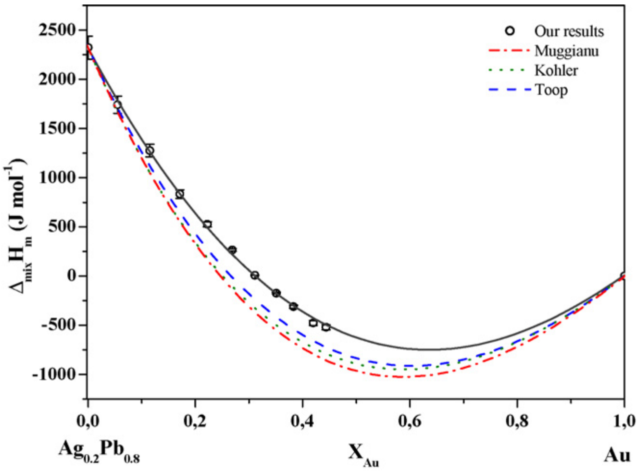

Fig. 4. Integral enthalpies of mixing of the Ag–Au–Pb ternary system along the section $X_{\mathrm{Ag}}/X_{\mathrm{Pb}} = 1/4$ at 973 K. Comparison of the experimental values with results obtained using the Kohler, Muggianu and Toop relations.

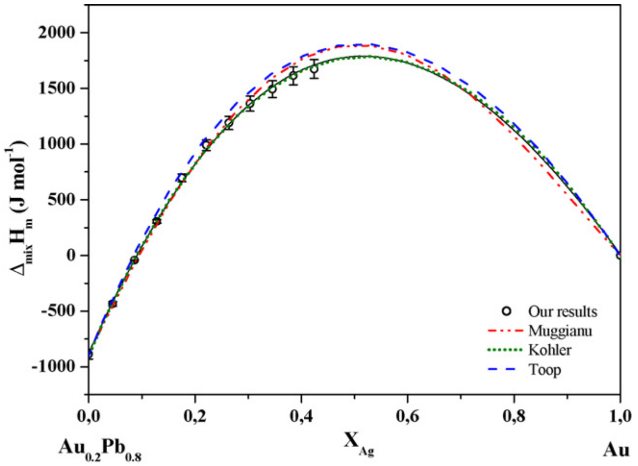

Fig. 5. Integral enthalpies of mixing of the Ag–Au–Pb ternary system along the section $X_{\mathrm{Au}}/X_{\mathrm{Pb}} = 1/4$ at 973 K. Comparison of the experimental values with results obtained using the Kohler, Muggianu and Toop relations.

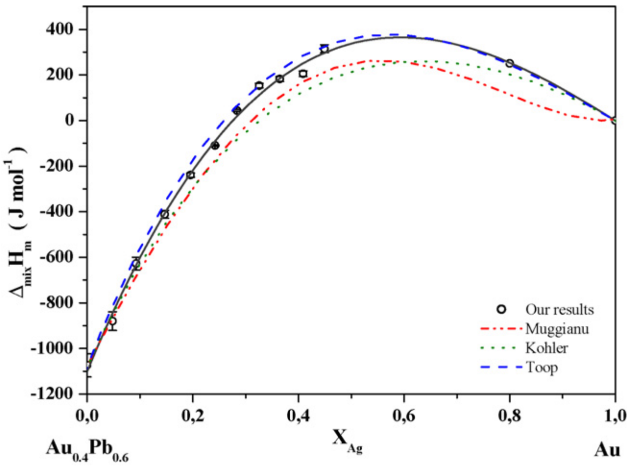

Fig. 6. Integral enthalpies of mixing of the Ag–Au–Pb ternary system along the section $X_{\mathrm{Au}}/X_{\mathrm{Pb}} = 2/3$ at 973 K. Comparison of the experimental values with results obtained using the Kohler, Muggianu and Toop relations.

where x_{i} denotes the mole fractions, ν an integer and G_i° the standard Gibbs energy of the pure elements i relative to the standard element reference (SER) at 29,815 K according to the SGTE description. The first two terms represent the mechanical mixture of the elements, the third the entropy of ideal mixture and the fourth, the excess molar Gibbs energy. This excess Gibbs energy, $G^{\mathrm{ex}}_{\mathrm{A-B}}$, is represented by Redlich–Kister polynomials which are characterized by a set of interaction parameters L^{ν}_{i-j} depending on temperature [35]:

$$L^{\nu}_{i-j}(T) = a^{\nu}_{i-j} + b^{\nu}_{i-j}T + c^{\nu}_{i-j}T\ \ln T \tag{2}$$

These binary parameters are extended in enthalpy contribution

$$H^{\mathrm{mix}}_{\mathrm{A-B}} = x_{\mathrm{A}}x_{\mathrm{B}}\sum_{\nu=0}^{\nu} W^{\nu}_{\mathrm{A-B}}(x_{\mathrm{A}} - x_{\mathrm{B}})^{\nu} \tag{3}$$

where W^{ν}_{i-j} is the enthalpic part to the interaction parameter depending on temperature as

$$W^{\nu}_{i-j}(T) = a^{\nu}_{i-j} - Tc^{\nu}_{i-j}T \tag{4}$$

The binary interaction parameters $L^{\nu}_{\mathrm{Ag-Au}}$ and $L^{\nu}_{\mathrm{Ag-Pb}}$, respectively, assessed by Hassam et al. [3] and Lukas [19,20] (Table 3) are retained in this work. These interaction parameters depend linearly with temperature. Thus, the enthalpy of mixing of Ag–Au and Ag–Pb liquid alloy is considered independent of temperature. For the Au–Pb binary liquid alloys the previous experimental work at different temperature (Figs. 1 and 3) show that the enthalpy of mixing must be considered as temperature dependent.

Considering our calorimetric data at 973 K and those of Michel and Castanet [26] at, respectively, 695, 871 and 1123 K we carried out an assessment of enthalpy of mixing of Au–Pb liquid alloys. In the assessment, we give higher weigh to our data at 973 K than those of Michel and Castanet at 871 K. Then we determined enthalpic part of the interaction parameter described by the temperature dependent parameter $W^{0}_{\mathrm{Au-Pb}}$ and the temperature independent parameter $W^{1}_{\mathrm{Au-Pb}}$. These parameters are given in Table 3.

Table 3
Binary and ternary interaction parameters

Parameters	Reference	ν	J mol^{-1}
Binary interaction			
$L^{\nu}_{\text{Ag–Au}}$	[3]	0	$-16402 + 1.14T$
$L^{\nu}_{\text{Ag–Pb}}$	[19,20]	0	$14665.80 - 7.92409T$
		1	$-1350.24–0.11456T$
		2	$-2670.89 + 0.69284T$
$L^{\nu}_{\text{Au–Pb}}$	[33]	0	$-17577.988 + 105.97303T$ $-14.43519T \ln T$
		1	$2673.2826 - 5.379358T$
$W^{\nu}_{\text{Au–Pb}}$	This work	0	$-18843.253 + 15.531T$
		1	2640.126
Ternary interaction			
$L^{\text{Ag}}_{\text{Ag–Au–Pb}}$	This work		-3112
$L^{\text{Au}}_{\text{Ag–Au–Pb}}$			17762
$L^{\text{Pb}}_{\text{Ag–Au–Pb}}$			-6908

Thereafter we compare our assessment to the experimental data on the one hand and to the Wang et al.'s assessment [33] on the other hand. The comparison is shown in Figs. 2 and 3. Both assessments lead to almost equivalent description of the enthalpy of mixing of the liquid mixture within the error margin. The enthalpy of mixing at 671, 973 and 1123 K is suitably described by the present assessment and that of Wang et al. [33] whereas the enthalpy of mixing at 871 K is in worst agreement.

The binary parameters are used to describe the ternary excess molar Gibbs energy $G^{\text{ex}}_{\text{A–B–C}}$, as follows [36]:

$$\begin{aligned} G^{\text{ex}}_{\text{A–B–C}} = {} & x_{\text{A}}x_{\text{B}}\sum_{\nu=0}^{\nu} L^{\nu}_{\text{A–B}}(x_{\text{A}}-x_{\text{B}})^{\nu} + x_{\text{A}}x_{\text{C}}\sum_{\nu=0}^{\nu} L^{\nu}_{\text{A–C}}(x_{\text{A}}-x_{\text{C}})^{\nu} \\ & + x_{\text{B}}x_{\text{C}}\sum_{\nu=0}^{\nu} L^{\nu}_{\text{B–C}}(x_{\text{B}}-x_{\text{C}})^{\nu} \\ & + x_{\text{A}}x_{\text{B}}x_{\text{C}}[x_{\text{A}}L^{\text{A–B–C}}_{\text{A}} + x_{\text{B}}L^{\text{A–B–C}}_{\text{B}} + x_{\text{C}}L^{\text{A–B–C}}_{\text{C}}] \end{aligned} \tag{5}$$

The last term includes the ternary interaction parameter $L^{\text{A–B–C}}$ which is assumed temperature independent. In this work, it will be also assumed. The ternary enthalpy is written as

$$\begin{aligned} H^{\text{mix}}_{\text{A–B–C}} = {} & x_{\text{A}}x_{\text{B}}\sum_{\nu=0}^{\nu} W^{\nu}_{\text{A–B}}(x_{\text{A}}-x_{\text{B}})^{\nu} + x_{\text{A}}x_{\text{C}}\sum_{\nu=0}^{\nu} W^{\nu}_{\text{A–C}} \\ & \times (x_{\text{A}}-x_{\text{C}})^{\nu} + x_{\text{B}}x_{\text{C}}\sum_{\nu=0}^{\nu} W^{\nu}_{\text{B–C}}(x_{\text{B}}-x_{\text{C}})^{\nu} \\ & + x_{\text{A}}x_{\text{B}}x_{\text{C}}[L^{\text{A–B–C}}_{\text{A}}x_{\text{A}} + L^{\text{A–B–C}}_{\text{B}}x_{\text{B}} + L^{\text{A–B–C}}_{\text{C}}x_{\text{C}}] \end{aligned} \tag{6}$$

To compare our experimental results of enthalpy of mixing with the various predictive extensions from the binary to ternary systems of Kohler [37]:

$$\begin{aligned} H^{\text{mix}}_{\text{A–B–C}} = {} & (x_{\text{A}}+x_{\text{B}})^2 H^{\text{mix}}_{\text{A–B}}\left(\frac{x_{\text{A}}}{x_{\text{A}}+x_{\text{B}}}; \frac{x_{\text{B}}}{x_{\text{A}}+x_{\text{B}}}\right) \\ & + (x_{\text{A}}+x_{\text{C}})^2 H^{\text{mix}}_{\text{A–C}}\left(\frac{x_{\text{A}}}{x_{\text{A}}+x_{\text{C}}}; \frac{x_{\text{C}}}{x_{\text{A}}+x_{\text{C}}}\right) \\ & + (x_{\text{B}}+x_{\text{C}})^2 H^{\text{mix}}_{\text{B–C}}\left(\frac{x_{\text{B}}}{x_{\text{C}}+x_{\text{B}}}; \frac{x_{\text{C}}}{x_{\text{C}}+x_{\text{B}}}\right) \end{aligned} \tag{7}$$

of Muggianu et al. [38]:

$$\begin{aligned} H^{\text{mix}}_{\text{A–B–C}} = {} & \frac{4x_{\text{A}}x_{\text{B}}}{(1+x_{\text{A}}-x_{\text{B}})(1+x_{\text{B}}-x_{\text{A}})} \\ & \times H^{\text{mix}}_{\text{A–B}}\left(\frac{(1+x_{\text{A}}-x_{\text{B}})}{2}; \frac{(1+x_{\text{B}}-x_{\text{A}})}{2}\right) \\ & + \frac{4x_{\text{A}}x_{\text{C}}}{(1+x_{\text{A}}-x_{\text{C}})(1+x_{\text{C}}-x_{\text{A}})} \\ & \times H^{\text{mix}}_{\text{A–B}}\left(\frac{(1+x_{\text{A}}-x_{\text{C}})}{2}; \frac{(1+x_{\text{C}}-x_{\text{A}})}{2}\right) \\ & + \frac{4x_{\text{B}}x_{\text{C}}}{(1+x_{\text{B}}-x_{\text{C}})(1+x_{\text{C}}-x_{\text{B}})} \\ & \times H^{\text{mix}}_{\text{A–B}}\left(\frac{(1+x_{\text{B}}-x_{\text{C}})}{2}; \frac{(1+x_{\text{C}}-x_{\text{B}})}{2}\right) \end{aligned} \tag{8}$$

and Toop [39]:

$$\begin{aligned} H^{\text{mix}}_{\text{A–B–C}} = {} & \frac{x_{\text{B}}}{1-x_{\text{A}}} H^{\text{mix}}_{\text{A–B}}(x_{\text{A}}; 1-x_{\text{A}}) \\ & + \frac{x_{\text{C}}}{1-x_{\text{A}}} H^{\text{mix}}_{\text{A–C}}(x_{\text{A}}; 1-x_{\text{A}}) \\ & + (x_{\text{B}}+x_{\text{C}})^2 H^{\text{mix}}_{\text{A–B}}\left(\frac{x_{\text{B}}}{x_{\text{B}}+x_{\text{C}}}; \frac{x_{\text{C}}}{x_{\text{B}}+x_{\text{C}}}\right) \end{aligned} \tag{9}$$

The Kolher and Muggianu ponderations are symmetric, based on random behaviour of the liquid mixture, whereas the Toop's ponderation is characterized by an asymmetry of the binary system that exhibits the most ideal behaviour and that receives a different weight with respect to the other two.

Our measurement of enthalpy of mixing of the ternary liquid alloy Ag–Au–Pb for the three sections $X_{\text{Ag}}/X_{\text{Pb}} = 1/4$, $X_{\text{Au}}/X_{\text{Pb}} = 1/4$ and $X_{\text{Au}}/X_{\text{Pb}} = 2/3$ are compared to the predicted. In the calculation we considered the assessment of Hassam et al. [3] for the Ag–Au system, of Lukas [19,20] for the Ag–Pb system and our assessment for the Au–Pb system. For the Toop's extrapolation the Ag–Au binary system exhibits the most ideal behaviour compared to the Ag–Pb and the Au–Pb binary systems.

As it can be noted in Figs. 4–6, the extrapolated ternary enthalpies are in good agreement with the experimental measurements. Nevertheless the Toop's relation lead to a better prediction of enthalpy of mixing than the two other. In order to obtain a better representation of the ternary enthalpy of mixing, the ternary interaction parameters were also considered. These parameters, listed in Table 3, were obtained by fitting our experimental results on Eq. (6).

6. Conclusion

By using a high temperature Calvet microcalorimeter, molar enthalpies of mixing of the liquid Au–Pb and Ag–Au–Pb alloys have been determined at 973 K.

An assessment of enthalpy of mixing of Au–Pb and Ag–Au–Pb alloys has been carried out. A comparison between the experimental and calculated results from the extrapolations methods gives a suitable representation of the enthalpy of mixing in the liquid Ag–Au–Pb ternary system. A ternary interaction term has been added to the Muggianu relation. These results will be taken into account for the assessment of the equilibrium phase diagram using the CALPHAD method.

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