

ISSN: 2616-1419



FUOYE Journal of Pure and Applied Sciences

Available online at www.fuoye.edu.ng

ESTIMATION OF Sn Activity In Ternary Lead-Free Sn-Ag-Au ALLOYS AND ITS SUBSYSTEMS

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Abstract

ARTICLEINFO

Received: 20 March 2020 Accepted: 13 June 2020

Keywords:

Activity; Prediction; Thermodynamic modeling; Ternary Sn-Ag-Au alloy

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The activities of components in the binary liquid Ag-Sn, Au-Sn and Ag-Au alloys were calculated using the molecular interaction volume model (MIVM) at 1250 K, 823 K and 800 K, respectively. The reasonable good agreement between the calculated and the corresponding experimental data for the constituents binary alloy form the basis of employing the MIVM to predict the activities of Sn in ternary liquid Sn-Ag-Au alloys at different temperatures of 973 K, 1073 K, 1173 K, 1273 K and 1373 K with a constant molar ratio of Ag:Au = 1:2, 2:2 and 2:1 across the three sections, respectively. The activity of tin in Sn-Ag-Au alloys was found to show a negative deviation from ideality across the entire concentration range. The calculated values of the tin activities compared quite well with the corresponding experimental values.

FJPAS Vol 5 (1)

1.0 Introduction

Thermodynamic properties of alloys can generally be obtained by experimental measurements. However, owing difficulties in performing experimental especially for multimeasurements component alloys due to technological complications; high cost of expenses and time consuming, it is not easy to obtain all thermodynamic data through experiments [1-4]. In particular, alloys containing highly reactive elements such as Ti. Zr and Hf are the most difficult to determine experimentally and therefore, it is necessary to estimate the missing values by theoretical models.

In order to meet the demands of thermodynamic modelling data. the thermodynamics of complex alloys have attracted great deal of attentions of researchers over the past decades [5-12]. processing Various methods of performing thermodynamic data and thermodynamic multicalculations component systems have been used. Some of the methods used include calculations of diagrams (CALPHAD), Chou's general solution model, neutron diffraction, x-ray scattering, small-angle electrical conductivity, calorimetric and electromotive force (EMF) measurements and other theoretical evaluations [5, 11, 13].

In this work, a relatively simple model known as the molecular interaction volume model (MIVM) proposed by Tao [3, 14] is employed to predict the thermodynamic activities of ternary lead-free solder Sn-Ag-Au alloys in order to alleviate the gap between theory and experiment especially in condition of severe shortage of thermodynamic data regarding the subsystem of the alloys. One of the major advantages of the MIVM over regular association model is that the model has capacity to predict

thermodynamic properties in a multicomponent liquid alloy system because the formulation does not require the assumptions regarding the presence and composition of associates or clusters and uses fewer fitting parameters [14-15]. Conventionally, Leadtin alloys have been used in microelectronics industry to attach integrated assembly to a printed circuit board (PCB) for decades [16]. The demands for lead-tin alloys in the industrial market for medium and small portable electronics devices have made interconnecting densities and packaging of technologies gained relevance [17]. Pb-Sn solder has the ability to lower the surface tension of pure tin and facilitate the formation of intermetallic compound for excellent soldering reliability [18-19].

However, since Lead is toxic in nature and its use has been known to cause serious health challenges to human and the environment due to the huge amount of printed circuits and electronic devices to be recycled from municipal waste dumps [20-22]. Lead hampers the development of nervous system and affects mental acuity in children. Over exposure to Lead can lead to anemia and persistent rise in blood pressure in middle and older aged people [23]. In order to mitigate the effect of Pb on the environment and human beings, European Union launched initiatives known as Restriction of Hazardous Substances (RoHS) directive in 2006 that constrained the use of Pb and other toxic metals (such as Hg and Cd) in electronics. Many other countries have placed a ban on the use of Lead in the electronic industry [12, 22]. Therefore, attentions have been focused on developing alternative lead-free solder alloys having physical, chemical and technological properties comparable to or better than the Pb-Sn alloys in use.

Additionally, any promising potential substitute for Pb should not only satisfy environmental concerns, it must also meet some requirements as possession of low melting temperature; suitable wetting properties; excellent thermal conductivity; ability to produce good soldering joint that can withstand heat fatigue during service and be reliable over long-time usage [24]. It is important to note that thermodynamic properties of different liquid ternary alloys have already been determined using either the calorimetric or an EMF method. For instance, Zhongnan and coworkers [25] measured the thermodynamic activity of Sn in the ternary Au-Bi-Sn alloys using EMF method. The calorimetric method was used for measurement of the thermodynamic data of liquid ternary Sn-Ag-Au, Al-Sn-Zn, Ag-Sn-Zn alloys by [26-27]. While the EMF method has been used for the measurements of Cu-In-Zn, Bi-In-Zn, Sn-Ag-Au by [28-29] and [24]. Besides, enthalpies of formation in the liquid systems Ag-Au-Si, Ag-Au-Ge and Ag-Au-Sn were determined by [30] experimentally by the application of Hoch-Arpshofen model at 1373 K and 1550 K, respectively. Nonetheless, in all of these determinations the measurements were either made at only one or two temperatures of investigation. In the case of ternary Sn-Ag-Au alloys, the thermodynamic activity of Sn ternary Sn-Ag-Au system determined at the three cross-sections with constant molar ratio of 1:2, 1:1 and 2:1 by [24] at only 973 K using EMF method and this is the only experimental data available in literature for the system to the best of authors' knowledge.

$$G_{M}^{E} = x_{i} \ln \left(\frac{V_{mi}}{x_{i}V_{mi} + x_{j}V_{mj}B_{ji}} \right) + x_{j} \ln \left(\frac{V_{mj}}{x_{j}V_{mj} + x_{i}V_{mi}B_{ij}} \right)$$

$$-\frac{x_{i}x_{j}}{2}\left(\frac{Z_{i}B_{ji}\ln B_{ji}}{x_{i}+x_{j}B_{ji}}+\frac{Z_{j}B_{ij}\ln B_{ij}}{x_{j}+x_{i}B_{ij}}\right)$$

Therefore, the scope of the present work is to explore the thermodynamic behaviour of ternary Sn-Ag-Au alloys at different compositions with constant molar ratio of 1:2, 1:1 and 2:1 by predicting the Sn activity at different temperatures towards the quest of finding alternative lead-free solder alloys. Apart from high academic interest, Sn-Ag-Au system has been selected for theoretical study due to its suitable creeping properties and high tensile strength in eutectic form which makes it useful in flip-chip technology and optoelectronics [24, 31]. Also, Sn-Ag-Au alloys are considered for brazing alloys in jewelry [32]. Hence, a broad knowledge of the physical features of this system is important.

2.0 Basis of Molecular interaction volume model (MIVM)

The MIVM is a fluid-based model derived from statistical thermodynamics based on the physical perspective of molecular or atomic movement of liquid [12, 20-21]. The model takes into account the physical properties of the pure metals constituting the alloy, namely, the molar volume, V_m , and the first coordination number, Z, in the liquid state. These parameters are temperature dependent [33]. MIVM can predict the thermodynamic properties in a multi-component liquid alloy system using only the two infinite dilute coefficients, γ_i^{∞} and γ_i^{∞} [21]. activity According to MIVM, the molar excess Gibbs energy, G_{M}^{E} for binary liquid alloy i-j can be expressed as

(1)

where x_i and x_j are the molar fractions, V_{mi} and V_{mj} the molar volumes of component i and j, respectively. The pair-potential energy interaction parameter B_{ii} and B_{ij} are defined as:

$$B_{ij} = \exp\left[-\frac{\varepsilon_{ji} - \varepsilon_{ii}}{kT}\right] B_{ij} = \exp\left[-\frac{\varepsilon_{ij} - \varepsilon_{jj}}{kT}\right]$$
(2)

k is the Boltzmann constant; ε_{ii} , ε_{jj} and ε_{ji} are the i-i, j-j and i-j pair- potential energies, respectively, and $\varepsilon_{ij} = \varepsilon_{ji}$. T is the absolute temperature. Z_i in Eqn. (1) is the first coordination number of atom i defined as:

$$Z_{i} = \frac{4\sqrt{2\pi}}{3} \left(\frac{r_{mi}^{3} - r_{0i}^{3}}{r_{mi} - r_{0i}} \right) \rho_{i} r_{mi} \exp\left(\frac{\Delta H_{mi} (T_{mi} - T)}{Z_{c} R T T_{mi}} \right)$$
(3)

In Eqn. (3), $\rho_i = \frac{N_i}{V_i} = \frac{0.6022}{V_{mi}}$ is the molecular

number density, ΔH_{mi} and T_{mi} are the melting enthalpy and the melting temperature, respectively. Where $Z_c = 12$ is a closed-packed coordination, R is the gas

$$\ln \gamma = \ln \left(\frac{V_{mi}}{x_i V_{mi} + x_j V_{mj} B_{ji}} \right) + x_j \left(\frac{V_{mj} B_{ji}}{x_i V_{mi} + x_j V_{mj} B_{ji}} - \frac{V_{mi} B_{ij}}{x_j V m_j + x_i V_{mi} B_{ij}} \right)$$

constant, r_{mi} and r_{0i} are the beginning and first peak values of radial distance distribution function of the liquid metal i near its melting point, respectively. For a binary mixture i-j, the activity coefficients of components i and j are expressed as:

$$-\frac{x_{j}^{2}}{2} \left[\frac{Z_{i}B_{ji}^{2} \ln B_{ji}}{(x_{i} + x_{j}B_{ji})^{2}} + \frac{Z_{j}B_{ij} \ln B_{ij}}{(x_{j} + x_{i}B_{ji})^{2}} \right]$$
(4)

$$\ln \gamma_{j} = \ln \left(\frac{V_{mj}}{x_{j}V_{mj} + x_{i}V_{mi}B_{ij}} \right) + x_{i} \left(\frac{V_{mj}B_{ji}}{x_{i}V_{mi} + x_{j}V_{mj}B_{ji}} - \frac{V_{mi}B_{ij}}{x_{j}V_{mj} + x_{i}V_{mi}B_{ij}} \right)$$

$$-\frac{x_i^2}{2} \left[\frac{Z_j B_{ij}^2 \ln B_{ij}}{(x_i + x_i B_{ii})^2} + \frac{Z_i B_{ji} \ln B_{ji}}{(x_i + x_j B_{ii})^2} \right]$$
 (5)

Now, allowing the investigated Ag–Sn–Au ternary alloy to be the 1–2–3 system, the activity coefficient of the component 1 of this system can be obtained using the expression given by [12]:

$$\begin{split} & \ln \gamma_{\mathrm{I}} \! = \! 1 \! + \! \ln \! \left(\frac{V_{\mathrm{ml}}}{x_{1}V_{\mathrm{ml}} + x_{2}V_{\mathrm{m2}}B_{2\mathrm{l}} + x_{3}V_{\mathrm{m3}}B_{3\mathrm{l}}} \right) \! - \! \frac{x_{1}V_{\mathrm{ml}}}{x_{1}V_{\mathrm{ml}} + x_{2}V_{\mathrm{m2}}B_{2\mathrm{l}} + x_{3}V_{\mathrm{m3}}B_{3\mathrm{l}}} \\ & - \frac{x_{2}V_{\mathrm{ml}}B_{12}}{x_{1}V_{\mathrm{ml}}B_{12} + x_{2}V_{\mathrm{m2}} + x_{3}V_{\mathrm{m3}}B_{32}} - \frac{x_{3}V_{\mathrm{ml}}B_{13}}{x_{1}V_{\mathrm{ml}}B_{13} + x_{2}V_{\mathrm{m2}}B_{23} + x_{3}V_{\mathrm{m3}}} \\ & - \frac{1}{2} \! \left(\frac{Z_{1}(x_{2}B_{2\mathrm{l}} + x_{3}B_{3\mathrm{l}})(x_{2}B_{2\mathrm{l}} \ln B_{2\mathrm{l}} + x_{3}B_{3\mathrm{l}} \ln B_{3\mathrm{l}})}{(x_{1} + x_{2}B_{2\mathrm{l}} + x_{3}B_{3\mathrm{l}})^{2}} \end{split}$$

$$+\frac{Z_{2}x_{2}B_{12}[(x_{2}+x_{3}B_{32})\ln B_{12}-x_{3}B_{32}\ln B_{32}]}{(x_{1}B_{12}+x_{2}+x_{3}B_{32})^{2}} + \frac{Z_{3}x_{3}B_{13}[(x_{2}B_{23}+x_{3})\ln B_{13}-x_{2}B_{23}\ln B_{23}]}{(x_{1}B_{13}+x_{2}B_{23}+x_{3})^{2}}$$
(6)

3.0 Results and Discussion

3.1. Activities of constituents in the liquid binary Ag-Sn, Au-Sn and Ag-Sn alloys

For predicting the activities of the components in the ternary liquid Sn-Ag-Au alloys, the relative parameters of the pure components are required, and are shown in Table 1 [33]. The experimental activities data for the constituents' binary liquid alloys of the ternary Sn-Ag-Au system were obtained from [34]. The values of B_{ij} and

 B_{ji} can be obtained by the Newton-Raphson methodology [21], and are listed in Table 2. Substituting the corresponding parameters B_{ij} , B_{ji} into Eqns. (4) and (5), the activity of binary liquid Ag-Au, Au-Sn and Sn-Zn at 800 K, 823 K and 1250 K can be predicted, as shown in Figs. 1, 2 and 3. It can be seen that the predicted values are in good agreement with the experimental data from [34], this shows that the calculations of activities of the binary constituents of the ternary system Sn-Ag-Au from the MIVM are reasonable.

Table 1: Parameters of the pure metals used in the model (Iida and Guthrie, 1988)

Metal	$\Delta H_{mi}(KJ/mo)$	$\sigma_i \times 10^{-8} cm$	$r_{cd} \times 10^{-8} cm$	$V_{mi} = V_m [1 + \alpha (T - Tm)]$
Sn	7.07	3.16	2.59	$17.00[1 + 0.87 \times 10^{-4}(T - 505)]$
Ag	11.09	2.82	2.34	$11.60 + 0.98 \times 10^{-4} (T - 1236)$
Au	12.76	2.80	2.34	$11.30[1+0.98\times10^{-4}(T-1336)]$

Table 2: Computed values of B_{ij} and B_{ji} for the constituents' binary liquid alloys

-j	T(K)	B_{ij}	B_{ji}
Ag-Au	800	1.3335	0.9796
Au-Sn	823	1.1538	1.9856
Ag-Sn	1250	0.2980	1.6709

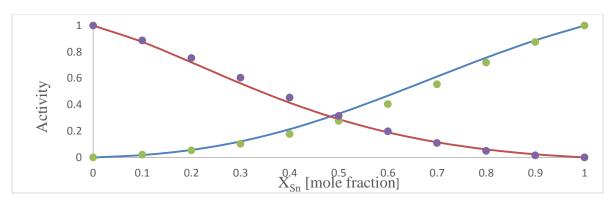
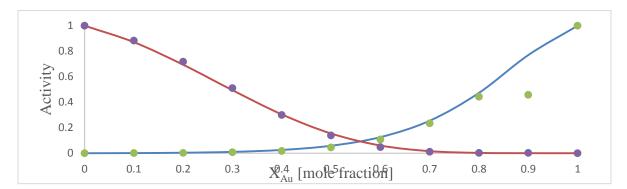


Figure 1: Comparison of the predicted activities (a_{Au} and a_{Ag}) of components of binary liquid Ag-Au alloys with experimental data at 800 K. The solid and dotted line represent predicted and experimental activity data, respectively. (—————the Raoult's law).



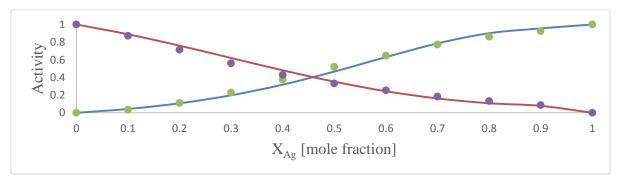


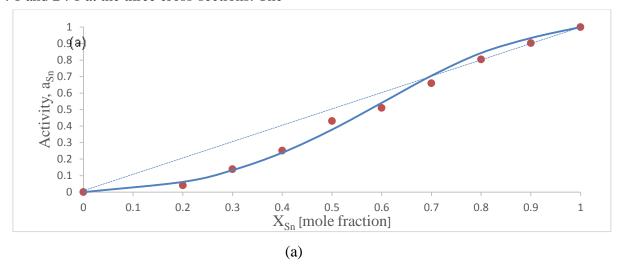
Figure 3: Comparison of the predicted activities (a_{Ag} and a_{Sn}) of components of binary liquid Ag-Sn alloys with experimental data at 1250 K. The solid line and dots represent predicted and experimental activity data, respectively. (------- the Raoult's law).

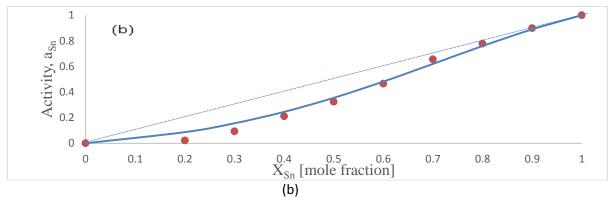
ISSN: 2616-1419

3.2 Prediction of activity of Sn in ternary Sn-Ag-Au Systems at 973 K

On the basis of the reasonable good agreement between predicted and the experimental values of activities of the constituents binary liquid alloys of the ternary liquid Sn-Ag-Au systems, we proceeded to employ the MIVM to predict the activity of Sn in ternary Sn-Ag-Au alloys at temperature of 973 K and compared our results with the experimental data obtained by [29] at the same temperature using the same constant molar ratio of Ag: Au = 2:1, 1:1 and 2:1 at the three cross-sections. The

results are presented in Fig. 4(a, b and c). From the Figures, it is clearly evident that the activity of Sn in ternary Sn-Ag-Au systems exhibits negative deviation from Raoult's law with a slight tendency towards a transformation from negative deviation to a positive deviation at the concentration, where $x_{Sn} > 0.9$ especially at molar ratio Ag : Au = 1 : 1. In addition, It is observed from Figures 4(a, b and c) that the predicted activities values using the MIVM gave a reasonable representation of the experimental data across the molar ratios at 973 K.





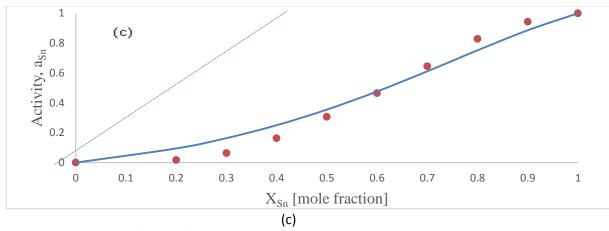


Figure 4a-c Comparison of the predicted Sn activity in ternary Sn-Ag-Au systems calculated using Eqn. (5) at 973 K with the experimental data (Hindler $et\ al.$, 2010). The dots and the solid line denote the experimental and calculated activity values, respectively. The dash line is the Raoult law. (a) For Ag:Au= 2:1 cross-section, (b) For Ag:Sn = 1:1 cross-section, and (c) For Au:Sn 1:2 cross-section.

Following the agreement obtained between the predicted activity of Sn in ternary Sn-Ag-Au at 973 K with the experimental data of [24] as shown in Fig. 4, the activities of Sn in the ternary liquid Sn-Ag-Au alloys at different temperatures of 1073 K, 1173 K, 1273 K and 1373 K for which there is currently no thermodynamic activity data in literatures were predicted. This was done by substituting the corresponding values of B_{ij} ,

$$RT \ln a_i(c) = -zFE(c) \tag{7}$$

where F is the Faraday constant, z = 3, is the valence of the ion in the liquid or solid

 B_{ji} and other parameters in Table 2 into Eqn. (6). The predictions were made in the ternary systems at each of the three different cross-sections with constant molar ratios of the other two components Ag: Au remaining as 2:1,1:1 and 1:2, respectively. The results are presented in Table 3 through Table 6, respectively, along with the measured activity of Sn computed using the relation in [12, 24, 35]:

electrolyte, and E is the measured EMF of the cell whose values were taken from [24].

ISSN: 2616-1419

Table 3: Predicted and experimental values of Sn activity in ternary Sn-Ag-Au alloys at 1073 K at the three sections

Ag: Au = 2:1			MIVM	EXPT.*	a _{Sn,EXPT} – a _{Sn,MIVM}	
XSn	XAg	XAu	a_{Sn}	a_{Sn}	' '	
0.2	0.5333	0.2667	0.0158	0.0093	0.0065	
0.3	0.4667	0.2333	0.0384	0.0538	0.0154	
0.4	0.4000	0.2000	0.0806	0.1198	0.0392	
0.5	0.3333	0.1667	0.1529	0.2774	0.1245	
0.6	0.2667	0.1333	0.2669	0.3551	0.0882	
0.7	0.2000	0.1000	0.4293	0.5214	0.0921	
0.8	0.1333	0.0667	0.6330	0.7172	0.0842	
0.9	0.0667	0.0333	0.8447	0.8569	0.0122	
Ag:Au :	= 1:1					
0.2	0.4000	0.4000	0.0292	0.0045	0.0247	
0.3	0.3500	0.3500	0.0638	0.0311	0.0327	
0.4	0.3000	0.3000	0.1207	0.0976	0.0231	
0.5	0.2500	0.2500	0.2075	0.1830	0.0245	
0.6	0.2000	0.2000	0.3303	0.3128	0.0175	
0.7	0.1500	0.1500	0.4898	0.5366	0.0468	
0.8	0.1000	0.1000	0.6753	0.6839	0.0086	
0.9	0.0500	0.0500	0.8600	0.8533	0.0067	
Ag:Au :	= 1:2					
0.2	0.2667	0.5334	0.0509	0.0035	0.0474	
0.3	0.2333	0.4667	0.1012	0.0183	0.0829	
0.4	0.2000	0.4000	0.1745	0.0689	0.1056	
0.5	0.1667	0.3334	0.2745	0.1748	0.0997	
0.6	0.1333	0.2666	0.4023	0.3257	0.0766	
0.7	0.1000	0.2000	0.5539	0.5220	0.0319	
0.8	0.0667	0.1334	0.7183	0.7744	0.0561	
0.9	0.0333	0.0666	0.8760	0.9113	0.0353	
*Hindler et al. (2010)						

^{*}Hindler *et al.*, (2010)

Table 4: Predicted and experimental values of Sn activity in ternary Sn-Ag-Au alloys at 1173 K at the three sections

Ag:Au	1 = 2:1		MIVM	EXPT*	$a_{Sn,EXPT} - a_{Sn,MIVM}$
XSn	XAg	XAu	a_{Sn}	a_{Sn}	
0.2	0.5333	0.2667	0.0276	0.0077	0.0199
0.3	0.4667	0.2333	0.0581	0.0431	0.0150
0.4	0.4000	0.2000	0.1075	0.0946	0.0129
0.5	0.3333	0.1667	0.1842	0.2420	0.0578
0.6	0.2667	0.1333	0.2969	0.3129	0.0160
0.7	0.2000	0.1000	0.4517	0.4803	0.0286
0.8	0.1333	0.0667	0.6439	0.6936	0.0497
0.9	0.0667	0.0333	0.8468	0.8414	0.0054
Ag:Au	= 1:1				
0.2	0.4000	0.4000	0.0231	0.0037	0.0194
0.3	0.3500	0.3500	0.0508	0.0237	0.0271
0.4	0.3000	0.3000	0.0975	0.0801	0.0174
0.5	0.2500	0.2500	0.1723	0.1551	0.0172
0.6	0.2000	0.2000	0.2846	0.2772	0.0074
0.7	0.1500	0.1500	0.4410	0.5077	0.0667
0.8	0.1000	0.1000	0.6371	0.6593	0.0222
0.9	0.0500	0.0500	0.8445	0.8407	0.0038
Ag:Au :	= 1:2				
0.2	0.2667	0.5334	0.0206	0.0031	0.0175
0.3	0.2333	0.4667	0.0467	0.0143	0.0324
0.4	0.2000	0.4000	0.0921	0.0558	0.0363
0.5	0.1667	0.3334	0.1663	0.1521	0.0142
0.6	0.1333	0.2666	0.2794	0.2974	0.0180
0.7	0.1000	0.2000	0.4383	0.4897	0.0514
0.8	0.0667	0.1334	0.6379	0.7567	0.1188
0.9	0.0333	0.0666	0.8474	0.8993	0.0519
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^{*}Hindler et al., (2010)

ISSN: 2616-1419

Table 5: Predicted and experimental values of Sn activity in ternary Sn-Ag-Au alloys at $1273~\mathrm{K}$ at the three sections

Ag:Au	= 2:1		MIVM	EXPT*	$a_{Sn,EXPT} - a_{Sn,MIVM}$
XSn	XAg	XAu	a_{Sn}	XSn	
0.2	0.5333	0.2667	0.0111	0.0063	0.0048
0.3	0.4667	0.2333	0.0328	0.0345	0.0017
0.4	0.4000	0.2000	0.0770	0.0747	0.0023
0.5	0.3333	0.1667	0.1548	0.2111	0.0563
0.6	0.2667	0.1333	0.2758	0.2757	0.0001
0.7	0.2000	0.1000	0.4430	0.4424	0.0006
0.8	0.1333	0.0667	0.6453	0.6708	0.0255
0.9	0.0667	0.0333	0.8498	0.8263	0.0235
Ag:Au	1 = 1:1				
0.2	0.4000	0.4000	0.0191	0.0031	0.0160
0.3	0.3500	0.3500	0.0475	0.0181	0.0294
0.4	0.3000	0.3000	0.0984	0.0658	0.0326
0.5	0.2500	0.2500	0.1808	0.1315	0.0493
0.6	0.2000	0.2000	0.3022	0.2457	0.0565
0.7	0.1500	0.1500	0.4647	0.4803	0.0156
0.8	0.1000	0.1000	0.6583	0.6356	0.0227
0.9	0.0500	0.0500	0.8538	0.8282	0.0256
	1 = 1:2				
0.2	0.2667	0.5334	0.0318	0.0026	0.0292
0.3	0.2333	0.4667	0.0682	0.0112	0.0570
0.4	0.2000	0.4000	0.1264	0.0452	0.0812
0.5	0.1667	0.3334	0.2133	0.1323	0.0810
0.6	0.1333	0.2666	0.3346	0.2716	0.0630
0.7	0.1000	0.2000	0.4914	0.4594	0.0320
0.8	0.0667	0.1334	0.6749	0.7395	0.0646
0.9	0.0333	0.0666	0.8597	0.8874	0.0277

*Hindler *et al.*, (2010)

ISSN: 2616-1419

Table 6: Predicted and experimental values of Sn activity in ternary Sn-Ag-Au alloys at 1373 K at the three sections

Ag: Au = 2:1			MIVM	EXPT*	a _{Sn,EXPT} – a _{Sn,MIVM}
XSn	XAg	x_{Au}	a_{Sn}	a_{Sn}	
0.2	0.5333	0.2667	0.0180	0.0052	0.0128
0.3	0.4667	0.2333	0.0428	0.0276	0.0152
0.4	0.4000	0.2000	0.0874	0.0589	0.0285
0.5	0.3333	0.1667	0.1611	0.1841	0.0230
0.6	0.2667	0.1333	0.2743	0.2429	0.0314
0.7	0.2000	0.1000	0.4335	0.4075	0.0260
0.8	0.1333	0.0667	0.6332	0.6487	0.0155
0.9	0.0667	0.0333	0.8436	0.8114	0.0322
Ag:Aı	ı = 1:1				
0.2	0.4000	0.4000	0.0151	0.0026	0.0125
0.3	0.3500	0.3500	0.0376	0.0133	0.0243
0.4	0.3000	0.3000	0.0795	0.0541	0.0254
0.5	0.2500	0.2500	0.1510	0.1115	0.0395
0.6	0.2000	0.2000	0.2631	0.2178	0.0453
0.7	0.1500	0.1500	0.4233	0.4543	0.0310
0.8	0.1000	0.1000	0.6264	0.6127	0.0137
0.9	0.0500	0.0500	0.8412	0.8160	0.0252
Ag:Aı	ı = 1:2				
0.2	0.2667	0.5334	0.0143	0.0023	0.0120
0.3	0.2333	0.4667	0.0359	0.0087	0.0272
0.4	0.2000	0.4000	0.0768	0.0366	0.0402
0.5	0.1667	0.3334	0.1474	0.1151	0.0323
0.6	0.1333	0.2666	0.2592	0.2480	0.0112
0.7	0.1000	0.2000	0.4201	0.4309	0.0108
0.8	0.0667	0.1334	0.6250	0.7226	0.0976
0.9	0.0333	0.0666	0.8416	0.8757	0.0341

^{*}Hindler et al., (2010)

4.0 Conclusion

The predicted results of activities of components of the constitutive binary liquid Ag-Au, Ag-Sn and Au-Sn alloys of the ternary Sn-Ag-Au system are in good agreement with experimental data. Then the predicted results of activities of Sn in ternary Sn-Ag-Au at 973 K using the MIVM are also in fairly reasonable agreement with experimental data of [24] across the constant molar ratio of 2 : 1, 1 : 1, and 1 : 2,

respectively. In addition, the MIVM was used to predict the activity of the first component, Sn, in the ternary Sn-Ag-Au system at different temperatures ranging from 1073 K to 1373 K, which has not been done before to the best of authors' knowledge. The thermodynamic properties showed mostly negative deviation from ideality, indicating strong mutual mixing tendencies in the investigation system. Due to the fact that experimental determination at

high temperatures is a rather difficult task, substitute for thermodynamic data of this multi-component Sn-based system.

the predicted results can be a good

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