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Bulk Properties of In-Bi Liquid Alloys 900 K

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Abstract:

The bulk properties i.e. thermodynamic as well as microscopic properties of In-Bi liquid alloys at 900 K as a function of In are computed on applying the four-parameter model [FPM]. The analytical expressions for the thermodynamic and microscopic functions of the binary molten alloys at a specified temperature are derived in the framework of FPM. The theoretical results for the excess free energy of mixing, free energy of mixing, activity, enthalpy of mixing, entropy of mixing and concentration fluctuations for In-Bi system at 900 K exhibit well agreement with the correlated experimental data. The concentration dependence of the shot-range order parameter has been theoretically predicted for In-Bi melts at 900 K. The results reveal that In-Bi liquid alloy at 900 K is a feebly interacting ordered alloy.

Keywords: Four-parameter model; excess free energy of mixing; activity; concentration fluctuations; short-range order parameter

1. Introduction

The In-Bi melts possess low melting points [1] and denotes chemical ordering. The Indium has a characteristics to lower the melting point of the alloy while the Bismuth enhances the wetting on the metallic surfaces and hence In-Bi alloy becomes useful to a lead-free solder alloy. It should be pointed out that In-Bi system is occasionally used as solders i.e. $In_{0.95}Bi_{0.05}$ is suitable for low temperature solders [2, 3]. However, In-Bi system has great impotence as base components for the advanced lead-free solders like In-Bi-Ag, In-Bi-Sb, In-Bi-Sn and In-Bi-Zn which have potentials

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applications in electronic industries [4]. The phase diagram [1] indicates that In-Bi system in its solid state has the existence of chemical complexes like In-Bi, In_5Bi_3 and In_2Bi. The composition dependence of the excess free energy of mixing, G_M^{XS}, free energy of mixing, G_M and entropy of mixing, exhibit the symmetric behavior of In-Bi melts at 900 K about C = 0.5 while the enthalpy of mixing, S_M shows the asymmetric nature of the alloys. The activity of In and Bi i.e. a_{In} and a_{Bi} illustrate the negative departures the ideal mixing behavior of the melts. This indicates the ordering nature of In-Bi melts at 90 K. This is also supported by the negative values of G_M^{XS} and H_M in the whole concentration range i.e. C_{In} = 0.1 to 0.9 of In-Bi molten alloys at 900 K. Therefore, the potential applications as well as the interesting thermodynamic properties of In-Bi melts have drawn the attentions of several researchers [2-6]. Earlier, Novakovic et al [4] have employed the complex formation model [7] to investigate the thermodynamic and surface properties of liquid In-Bi liquid alloys in the temperature region 553-873 K. Again, Koirala et al [6] have applied the simple statistical mechanical model [8] to study the thermophysical properties of In-Bi melts at 900 K. However, there is a scarcity of data related to In-Bi melts in the literature. Therefore, In-Bi melts still require a theoretical exploration.

In present work, the four-parameter model [9] has been applied to study the thermodynamic and structural properties of In-Bi melts at 900 K. In past, FPM model is successfully applied to analyze the thermophysical properties of the several alloys like Al-Cu, Al-Sn, Cu-Sn, Bi-Hg, Sn-Zn etc [10-14].

2. Formalism:

The thermodynamic and structural behavior of the binary alloys can be analyzed in the framework of FPM [9, 15] which is based on Maclaurin infinite series with suitable boundary conditions.

2.1 Thermodynamic and Microscopic Properties of Molten Binary System at a Specified Temperature

Consider a liquid binary mixture A-B having N_A and N_B atoms per mole of the elements A and B respectively such that $N_A = NC_A$ and $N_B = NC_B$; where, C_A and C_B indicate the concentrations of components A and B respectively of the binary alloy.

10.5281/zenodo.21819956

In the framework of FPM [9, 10], the expression for $\frac{G_M^{XS}}{RT}$ is given by

$$\frac{G_M^{XS}}{RT} = C_1C_2\left(a_1 + a_2C_2 + C_1C_2a_3 + C_1C_2^2a_4\right) \quad (1)$$

where, T= temperature (in K), R= molar gas constant, $C_A = C$ and $C_B = 1 - C$, $a_i (i \equiv 1,2,3,4)$ are the interaction parameters which depend on the temperature and does not depend upon the composition.

The activity coefficient, $\gamma_i (i = A, B)$ for i[th] component of the binary mixture is given by [16]

$$RT \ln \gamma_i = G_M^{XS} + (1 - C_i)\left(\frac{\partial G_M^{XS}}{\partial C_i}\right) \quad (2)$$

Equations (1) and (2) provide

$$ln\gamma_1 = C_B^2\left[a_1 - a_2 + 2C_Ba_2 + C_A(3C_B - 1)a_3 + 2C_AC_B(2C_B - 1)a_4\right] \quad (3)$$

$$ln\gamma_2 = C_A^2\left[a_1 + 2C_Ba_2 + C_B(2 - 3C_B)a_3 + C_B^2(3 - 4C_B)a_4\right] \quad (4)$$

For the binary mixture, the activity coefficient, γ_i is related with the activity, $a_i (i = A, B)$ as [16]

$$\Upsilon_i = \frac{a_i}{C_i} \quad (5)$$

The excess entropy for the i[th] component of the binary melt is expressed as [9]

$$\frac{S_i^{XS}}{R} = -\ln \gamma_i - T\left(\frac{\partial \ln \gamma_i}{\partial T}\right)_P \qquad (i \equiv A, B) \quad (6)$$

The equations (3), (4) and (6) yield

$$\frac{S_A^{XS}}{R} = -C_B^2\left[\left(a_1 + a_1'T\right) + (2C_B - 1)\left(a_2 + a_2'T\right) + (1 - C_B)(3C_B - 1)\left(a_3 + a_3'T\right) + 2C_B(1 - C_B)(2C_B - 1)\left(a_4 + a_4'T\right)\right] \quad (7a)$$

$$\frac{S_B^{XS}}{R} = -C_A^2\left[\left(a_1 + a_1'T\right) + 2C_B\left(a_2 + a_2'T\right) + C_B(2 - 3C_B)\left(a_3 + a_3'T\right) + C_B^2(3 - 4C_B)\left(a_4 + a_4'T\right)\right] \quad (7b)$$

where, a prime on a_1, a_2, a_3 and a_4 represents the temperature derivative of the interaction parameters i.e. $\frac{\partial a_i}{\partial T} (i \equiv 1,2,3,4)$.

The excess entropy of mixing, S_M^{XS} for a binary mixture is represented by [11]

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$$\frac{S_M^{XS}}{R}=C_A\frac{S_A^{XS}}{R}+C_B\frac{S_B^{XS}}{R} \tag{8}$$

Now, the entropy of mixing, S_M of the binary mixture is expressed as

$$\frac{S_M}{R}=\frac{S_M^{XS}}{R}+\frac{S_M^{id}}{R}$$

$$\frac{S_M}{R} = \frac{S_M^{XS}}{R} - \left(C_A \ln C_A + C_B \ln C_B\right) \tag{9}$$

The enthalpy of mixing, H_M for the binary system may be represented as [16]

$$H_M = G_M^{XS} - T\left[\frac{\partial G_M^{XS}}{\partial T}\right] \tag{10}$$

Equations (1) and (10) provide

$$\frac{H_M}{RT} = TC_A C_B\left[a_1' + C_B a_2' + C_A C_B a_3' + C_A C_B^2 a_4'\right] \tag{11}$$

The enthalpy of mixing, H_M may also be represented as [16]

$$\frac{H_M}{RT}=\frac{G_M^{XS}}{RT}+\frac{S_M^{XS}}{RT} \tag{12}$$

The microscopic function like $S_{cc}(0)$ [16, 17] is utilized to analyze the nature of atomic interactions in a binary system. For segregating system, $S_{cc}(0) > S_{cc}^{id}(0)$ which indicates like atoms (A-A or B-B) interactions exist in the binary solution. Similarly, for ordering system, $S_{cc}(0) < S_{cc}^{id}(0)$ which denotes unlike atoms (A-B) interactions in the binary mixture [16, 17].

The analytical expression for $S_{cc}(0)$ is expressed as [18]

$$S_{cc}(0)=RT\left[\frac{\partial G_M}{\partial c^2}\right]^{-1}_{T,P,N} \tag{13}$$

where, G_M is represented by

$$G_M = G_M^{XS} + G_M^{id}$$

$$G_M = G_M^{XS} + RT\left[C_A \ln C_A + C_A \ln C_A\right]$$

$$\frac{G_M}{RT}=\frac{G_M^{XS}}{RT}+\left[C_A \ln C_A + C_A \ln C_A\right]$$

10.5281/zenodo.21819956

$$\frac{G_M}{RT} = C_A C_B \left[a_1 + C_B a_2 + C_A C_B a_3 + C_A C_B^2 a_4 \right] + \left(C_A \ln C_A + C_B \ln C_B \right) \quad \text{(from equation(1))} \quad (14)$$

Equations (13) and (14) provide

$$S_{cc}(0) = \left[-2a_1 + 2(3C_A - 2)a_2 + 2(1 - 6C_A + 6C_A^2)a_3 + 2(1 - 9C_A + 18C_A^2 - 10C_A^3)a_4 + \frac{1}{C_A C_B} \right]^{-1} \quad (15)$$

The expression for the concentration fluctuations, $S_{cc}(0)$ in terms of activity may be represented by [, 16]

$$S_{cc}(0) = C_B a_A \left[\frac{\partial a_A}{\partial C_A} \right]^{-1}_{T,N,P} = C_A a_B \left[\frac{\partial a_B}{\partial C_B} \right]^{-1}_{T,N,P} \quad (16)$$

The calculated values of $S_{cc}(0)$ on using equation (16) are usually assumed to be the experimental data of $S_{cc}(0)$ [16, 18].

The ideal value of $S_{cc}(0)$ for the binary system is given by

$$S_{CC}^{id}(0) = C_A C_B \quad (17)$$

The microscopic function like shot-range order parameter (SRO), α_1 [16, 18, 19] provides the knowledge of the atomic arrangement in a binary mixture. The negative value of α_1 stands for the ordering behavior of the mixture whereas the positive value of α_1 refers to the segregating character of the mixture and $\alpha_1 = 0$ denotes the random distribution of atoms in the mixture. The SRO, α_1 in terms of $S_{cc}(0)$ is represented as [16]

$$\alpha_1 = \frac{S - 1}{S(Z - 1) + 1} \quad \text{with,} \quad S = \frac{S_{cc}(0)}{S_{cc}^{id}(0)} \quad (18)$$

where, Z = coordination number (usually taken to be 10).

3. Result and Discussions

The concentration dependency of the thermodynamic and microscopic properties of In-Bi system at 900 K are investigated on employing the analytical expressions deduced in the framework of FPM.

3.1 Results for G_M^{XS}, G_M, a_i, H_M and S_M of In-Bi Melts at 900 K

The analytical expression (1) indicates that the computation of G_M^{XS} for In-Bi molten system at 900 K requires the interaction parameters i.e. a_1, a_2, a_3 and a_4. In present case, the interaction

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parameters are estimated by the least square method on using the experimental data of G_M^{XS} for In-Bi melts at 900 K[1] in equation (1). The estimated values of a_1, a_2, a_3 and a_4 are found to be

$$a_1 = -1.0990 \qquad a_2 = 0.4604$$

$$a_3 = -0.5973 \qquad a_4 = -0.0728$$

It is to remarkable that the above mentioned values of a_1, a_2, a_3 and a_4 are utilized for the computations of the thermodynamic and microscopic properties of In-Bi melts at 900 K in order to maintain the consistency. The equations (1) and (14) are applied to determine the theoretical values of $\frac{G_M^{XS}}{RT}$ and $\frac{G_M}{RT}$ as a function of concentration respectively for In-Bi melts at 900 K. An excellent harmony is observed between the theoretical and corresponding experimental data [1] as shown in Fig. 1. Obviously, both $\frac{G_M^{XS}}{RT}$ and $\frac{G_M}{RT}$ are negative at each composition of In in the region, C_{In} = 0.1 to 0.9. The theoretical and experimental values of $\frac{G_M^{XS}}{RT}$ are minimum i.e. – 0.2568 (Theory) and - 0.2569 (Experiment) both at C_{In} = 0.5. Again, the minimum values of $\frac{G_M}{RT}$ are - 0.9500 (Theory) and – 0.9505 (Experiment) both at C_{In} = 0.5. Thus, the observed symmetries in G_M^{XS} and G_M for In-Bi melts at 900 K are well reproduced. The minimum value of $\frac{G_M}{RT}$ signifies that In-Bi melt at 900 K is a weakly interacting ordered alloy [18].

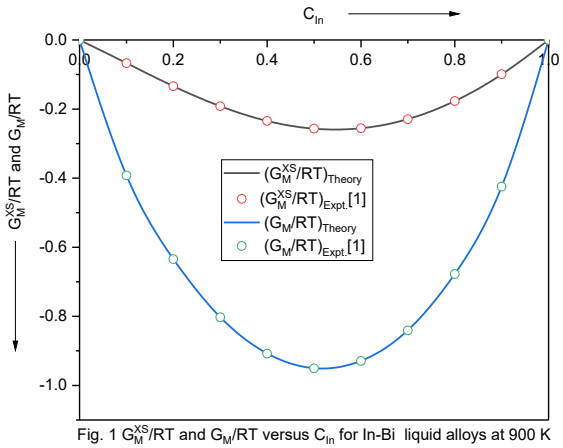

Fig. 1 G_M^{XS}/RT and G_M/RT versus C_{In} for In-Bi liquid alloys at 900 K

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International Journal of Innovative Research in Science and Engineering
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The theoretical values of the activity coefficients i.e γ_{In} and γ_{Bi} of In and Bi respectively, are determined from equations (4) and (5) respectively as a function of concentration of In for In-Bi melts at 900 K. The theoretical values of γ_{In} and γ_{Bi} are employed to compute the values of a_{In} and a_{Bi} respectively for In-Bi melts at 900 K from equation (6). The theoretical data and corresponding experimental data for a_{In} and a_{Bi} [1] exhibit excellent agreement as illustrated in Fig.2. In whole composition region i.e. C_{In} = 0.1 to 0.9, both a_{In} and a_{Bi} show the negative deviations from the ideal mixing behavior for In-Bi melts at 900 K. This indicates the ordering character of In-Bi melts at 900 K. Therefore, the excellent agreement between the theory and experiment for a_{In} and a_{Bi} of In-Bi melts at 900 K validates the values of the model parameters, a_1, a_2, a_3 and a_4.

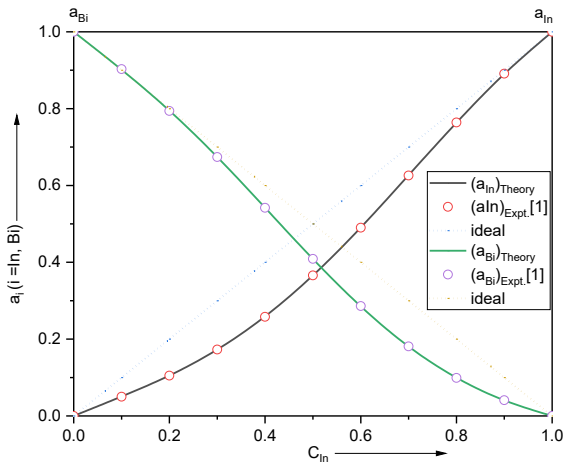

Fig.2 a_i (i= In,Bi) versus C_{In} for In-Bi liquid alloys at 900 K

In order to determine the values of S_M and H_M as function of concentration from equations (9) and (11) respectively, the temperature derivatives i.e. $a_1^{'}, a_2^{'}, a_3^{'}$ and $a_4^{'}$ of a_1, a_2, a_3 and a_4 respectively are required. For this, the least square method is utilized on employing the experimental data of H_M for In-Bi melts at 900 K [1] in equation (11). The estimated values of $a_1^{'}, a_2^{'}, a_3^{'}$ and $a_4^{'}$ are

$a_1^{'}$ = 0.0007 K^{-1} $\quad$ $a_2^{'}$ = 0.0003 K^{-1}

$a_3^{'}$ = 0.0027 K^{-1} $\quad$ $a_4^{'}$ = - 0.0036 K^{-1}

The estimated values of $a_1^{'}, a_2^{'}, a_3^{'}$ and $a_4^{'}$ are used to compute S_{In}^{XS} and S_{Bi}^{XS} as a function of concentration of In from equations (7a) and (7b) respectively for In-Bi melts at 900 K. Again, equation (8) is utilized to compute S_M^{XS} as a function of In for In-Bi melts at 900 K. Further,

10.5281/zenodo.21819956

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Volume 12, Issue 07, 2026
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equation (9) is used to determine the theoretical values of $\frac{S_M}{R}$ relative to the composition of In for In-Bi melts at 900 K. The theoretical results and experimental results [1] for $\frac{S_M}{R}$ are in well agreement as depicted in Fig.3. Both the theoretical and experimental values [1] are maximum at $C_{In} = 0.5$ i.e. $\frac{S_M}{R} =$ 0.7050 (Theory) and 0.7059 (Experiment), Thus, symmetry in S_M is observed for In-Bi liquid alloys at 900 K.

Now, equation (11) is employed to obtain the theoretical values of H_M for In-Bi liquid alloys at 900 K. The theoretical values of H_M is compared with correlated experimental data [1] for In-Bi melts at 900 K as shown in Fig. 3. An excellent harmony is seen between theory and experiment. Again, the theoretical and experimental values of H_M are found to be minimum at $C_{In} = 0.6$ i.e. $\frac{H_M}{RT} = -$ 0.2456 (Theory) and – 0.2455 (Experiment). Thus, the symmetry in H_M for In-Bi melts at 900 K is successfully explained.

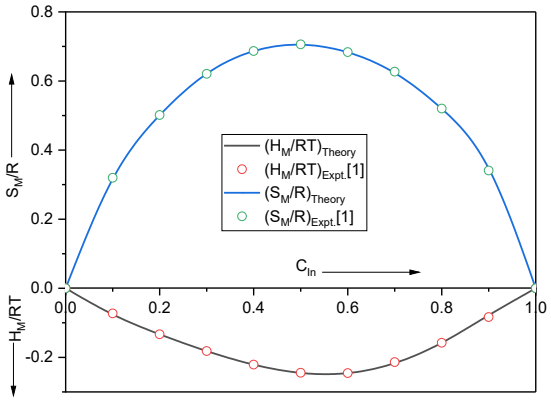

Fig. 3 H_M/RT and S_M/R versus C_{In} for In-Bi liquid alloys at 900 K

3.2 Results for $S_{cc}(0)$ and α_1 of In-Bi Melts at 900 K

The equation (15) is employed to compute the composition dependency of $S_{cc}(0)$ in terms of composition of In for In-Bi melts at 900 K. The theoretical values and experimental values [1] of $S_{cc}(0)$ are shown in Fig. 4. An excellent agreement is observed between theory and experiment with maximum values i.e. 0.161 (Theory) and 0.161 (Experiment) both at $C_{In} = 0.4$. Thus, $S_{cc}(0)$ shows

10.5281/zenodo.21819956

asymmetric nature of In-Bi melts at 900 K with respect to the concentration. The values of $S_{cc}(0)$ are found to be less than $S_{cc}^{id}(0)$ in the whole concentration range, C_{In} = 0.1 to 0.9 which indicates that In-Bi melt at 900 K is an ordered alloy.

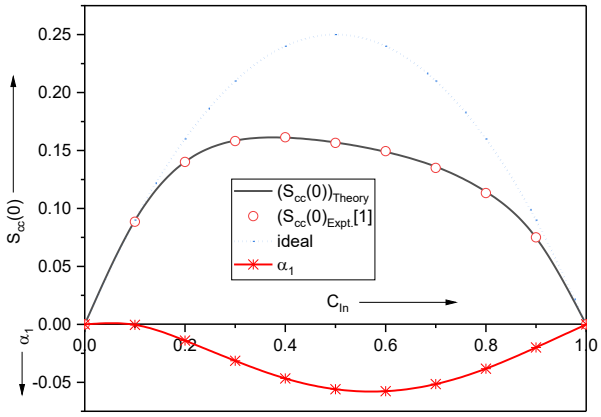

Fig. 4 $S_{cc}(0)$ and α_1 versus C_{In} for In-Bi liquid alloys at 900 K

The theoretical values of short-range order parameter, α_1 determined from equation (18) as a function of concentration of In are also included in Fig.4 for Bi-In melts at 900 K. The $\alpha_1 - C$ isotherm denotes the negative value of α_1 at each composition with minimum value of - 0.0577 at C_{In} = 0.6 which indicates that In-Bi melt is an ordered system.

4. Conclusions

The four-parameter model is utilized to deduce the analytical expressions for the thermodynamic functions like G_M^{XS}, G_M, a_i, H_M and S_M as well as the microscopic functions namely $S_{cc}(0)$ and α_1 of the binary liquid alloys at a given temperature. The model parameters a_1, a_2, a_3 and a_4 are estimated on employing the experimental data of G_M^{XS} for In-Bi melts at 900 K. The estimated values of a_1, a_2, a_3 and a_4 are used to compute the thermodynamic and microscopic properties of In-Bi melts at 900 K in terms of the composition of In. The theoretical results for G_M^{XS}, G_M, a_i, H_M, S_M and $S_{cc}(0)$ agree well with the correlated experimental results for In-Bi melts at 900 K. The concentration dependence of G_M^{XS}, a_i, H_M, $S_{cc}(0)$ and α_1 indicate that In-Bi melt at 900 is an ordered system. Further, the model parameters a_1, a_2, a_3 and a_4 are temperature dependent.

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