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Theoretical investigation on ordering nature of Cd-Bi alloys in the molten state

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Abstract

Experimental determination of thermo-physical mixing properties of binary liquid alloys is a long and expensive task that becomes more complicated for some system which may be chemically active or radioactive or even may contain scarce components. Theoretical method, on the other hand reduces the time and efforts required, and are of great importance in predicting the properties. Now we have focused on theoretical model to study of the alloying behaviour of Cd-Bi alloys in the molten state at 773K. We have used simple statistical model to report the ordering nature of Cd-Bi liquid alloys through the study of surface properties, transport properties and various thermodynamic and microscopic functions. The knowledge of surface phenomena like surface segregation and surface tension is essential for the processing of materials and productions in the metallurgical industry. At the microscopic level, transport properties such as viscosity and diffusion coefficient help to understand about the mixing behaviour of the alloys forming molten metals. Thermodynamic properties provide information on the interaction, stability and bonding strength among the constituent atoms in the alloys. The microscopic properties are useful in obtaining the microscopic information on structure of molten alloys. Our theoretical analysis gives the negative energy parameter, which is found to be temperature dependent. Negative deviation from Raoultian behaviour is observed in the computed surface tension, thermodynamic and structural parameters of the alloys. But in case of viscosity isotherm positive deviation from ideality is observed. The computed results are in good agreement with experimental data. The analysis concluded that the alloy is of weakly interacting and hetero-coordinating system.

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Keywords: Simple statistical model; Negative deviation; Hetero-coordination; Surface properties.

1. Introduction

It is very difficult to conduct diffraction experiment on cadmium metal because of its highly reactive nature. Therefore theoretical investigation giving mixing properties of cadmium alloys are highly desirable. Cadmium alloys have been widely used in fabricating solid state electronic components [1]. Cadmium-bismuth is one of the eutectic metal alloys, which has potential applications in bearing assemblies, ballasts, casting, step soldering, radiation soldering etc. Small content of cadmium lowers the melting point of cadmium-bismuth alloys, improves thermal and electrical conductivities and increases mechanical properties such as hardness, wear resistance, tensile and fatigue strength [1]. For the material preparation of metal alloys, the understandings of mixing properties of liquid alloys are important. Several theoretical models [2-8] have long been proposed to understand the thermodynamic, structural, transport and surface properties of binary liquid alloys. In present work, we study the concentration dependent mixing properties of liquid Cd-Bi at 773K by the simple statistical model [9-11]. The energetic of mixing as well as the positive/negative deviation from Raoultian behavior for the various bulk and surface properties have been discussed.

It is observed that system Cd-Bi is characterized by negative interaction energy, indicating the formation of two structures, as shown by their phase diagrams [12]. This characteristic behavior is likely to be a reflection of the interplay of the energetic and structural re-adjustment of the constituent elemental atoms.

The paper is arranged as follows: We present theoretical formalism in section 2, which is followed by results and discussions in section 3 and finally, conclusion of the paper is presented in section 4.

2. Theoretical formalism

2.1. Thermodynamic properties

In the framework of simple statistical model for regular solution the expression for the excess free energy of mixing [10, 11] is

$$G_M^{XS} = RT \int_0^x \ln \sigma^z dx = RT \{x \ln \gamma_A + (1-x) \ln \gamma_B\} \quad (1)$$

where $\sigma = (\beta + 2x - 1) \exp(-\omega / z k_B T) / 2x \quad (1a)$

$$\gamma_A = [(\beta - 1 + 2x) / x(1 + \beta)]^{z/2} \quad (1b)$$

$$\gamma_B = [(\beta + 1 - 2x) / (1-x)(1 + \beta)]^{z/2} \quad (1c)$$

$$\beta = \{1 + 4x(1-x) [\exp(2\omega / z k_B T) - 1]\}^{1/2} \quad (1d)$$

where R is universal molar constant; T, absolute temperature; x, concentration of the component; ω , interchange energy; z, coordination number and k_B , Boltzmann constant. The ideal free energy of mixing ($\omega = 0$) is

$$G_M^{id} = RT \{x \ln x + (1-x) \ln(1-x)\} \quad (2)$$

The expression for free energy of mixing [13], one obtains from the standard thermodynamic relation as

$$G_M = G_M^{id} + G_M^{XS} = RT \{x \ln x + (1-x) \ln(1-x)\} + RT \{x \ln \gamma_A + (1-x) \ln \gamma_B\} \quad (3)$$

For the equiatomic composition ($x = \frac{1}{2}$), relation (1) reduces to

$$\frac{G_M^{XS}}{RT} = \ln 2^{z/2} [1 + \exp(-\omega / z k_B T)]^{-z/2} \quad (4)$$

The heat of mixing (H_M) and entropy of mixing (S_M) are obtained from the relations

$$H_M = G_M - T \left(\frac{\partial G_M}{\partial T} \right)_{x,P,N} \quad (5)$$

$$S_M = (H_M - G_M) / T \quad (6)$$

The activity of binary liquid alloys, f_i ($i=A$ or B), can be related to the free energy of mixing by the relation

$$RT \ln f_i = \left(\frac{\partial G_M}{\partial N_i} \right)_{T,P,N} = G_M + (1-x_i) \left(\frac{\partial G_M}{\partial x_i} \right)_{T,P,N} \quad (7)$$

Then from Eq.(8) we have [13],

$$\ln f_A = \ln x + (1-x)^2 \frac{\omega}{k_B T} \quad (8a)$$

$$\ln f_B = \ln(1-x) + x^2 \frac{\omega}{k_B T} \quad (8b)$$

2.2. Structural properties

Concentration fluctuations in the long-wavelength limit ($S_{cc}(0)$) is an essential structural function which has been widely used to study the nature of atomic order in binary liquid alloys [14,15]. $S_{cc}(0)$ is thermodynamically related to free energy of mixing (G_M) [14]. It is given as

$$S_{cc}(0) = RT \left(\frac{\partial^2 G_M}{\partial x^2} \right)^{-1}_{T,P,N} \quad (9)$$

Equations (3) and (9) gives the theoretical value of $S_{cc}(0)$:

$$S_{cc}(0) = x(1-x)[1-x(1-x)2\omega/k_B T]^{-1} \quad (10)$$

The ideal value of concentration-concentration fluctuations, when the ordering energy is zero, is usually computed from:

$$S_{cc}^{id}(0) = x(1-x) \quad (11)$$

There are difficulties in diffraction experiment, therefore theoretical determination of $S_{cc}(0)$ is of great importance when nature of interactions in the melt has to be analyzed. The mixing behavior of liquid alloys can be deduced from the deviation of $S_{cc}(0)$ from $S_{cc}^{id}(0)$. At a given composition if $S_{cc}(0) < S_{cc}^{id}(0)$, ordering in liquid alloy is expected while $S_{cc}(0) > S_{cc}^{id}(0)$ gives the indication of tendency of segregation [14].

The Warren-Cowley [16-17] short-range order parameter (α_1) is useful parameter to quantify the degree of chemical order in the alloy melt. It provides insight into the local arrangement of the atoms in the molten alloys. Although it is difficult to obtain the experimental values of α_1 , theoretical values of this parameter are easily obtain via conditional probability [A/B] which defines the probability of finding an A-atom as a nearest neighbor of a given B-atom. It can be evaluated theoretically [16-17]:

$$\alpha_1 = (S-1) / \{S(z-1)+1\}, \quad S = S_{cc}(0) / S_{cc}^{id}(0) \quad (12)$$

where z is the coordination number, which is taken as 10 for our purposes. For the equiatomic composition, the chemical short range order parameter is found to be $-1 \leq \alpha_1 \leq 1$. Negative values of α_1 indicate ordering in the melt, which is complete if $\alpha_1 = -1$. On the other hand, positive values of α_1 indicate segregation, leading to complete only if $\alpha_1 = 1$. But $\alpha_1 = 0$, corresponds to random distribution of the atoms in the mixture.

2.3 Transport properties

The mixing behavior of binary liquid alloys can also be studied at the microscopic level in terms of transport properties such as viscosity, chemical diffusion coefficients etc. Viscosity (η) of liquid alloys is equally important in the understanding of the atomic level structure and interactions. As in the case of thermodynamic functions, the composition dependence of η of liquid alloys is also found to exhibit either a linear variation, or positive or negative deviations from the linear law. A simple formula for the investigation of viscosity of a binary solution [18] is

$$\eta = \eta_{id} \left[1 - x(1-x) \left(\frac{2\omega}{k_B T} \right) \right] \quad (13)$$

$$\text{with} \quad \eta_{id} = x \eta_A^0 + (1-x) \eta_B^0 \quad (14)$$

where η_i^0 is the viscosity of pure component i ($= A$ or B) and ω , as usual, is the interchange energy or order energy.

The mixing behavior of the alloys forming molten metal can also be studied at the microscopic level in terms of coefficient of diffusion. The mutual diffusion coefficient (D_M) of binary liquid alloys can be

expressed in terms of activity (f_i) and self- diffusion coefficient (D_{id}) of pure component with the help of Darken's equation [19],

$$D_M = D_{id} x_i \frac{d \ln f_i}{dx_i} \quad (15)$$

We obtain an expression for D_M in terms of $S_{cc}(0)$:

$$\frac{D_M}{D_{id}} = \frac{S_{cc}^{id}(0)}{S_{cc}(0)} \quad (16)$$

with
$$D_M = xD_B + (1-x)D_A \quad (17)$$

where D_A and D_B are the self-diffusion coefficients of pure components A and B respectively.

According to Eq. (17), $S_{cc}(0) \langle S_{cc}^{id} \rangle$ indicates typical compound forming alloys implying $D_M > D_{id}$. A peak on diffusivity curve, D_M / D_{id} verses x , suggests presence of maximum chemical order in the molten alloys system as well as the composition of the most probable associates formed in the liquid phase. The relationship between $S_{cc}(0)$ and the diffusivity expressed by the ratio of the mutual and self-diffusion coefficients, D_M / D_{id} , indicates the mixing behavior of the alloys, i.e. the tendency for compound formation ($D_M / D_{id} > 1$) or phase separation ($D_M / D_{id} < 1$) or ideal mixing (D_M / D_{id} approaches 1). Considering only the energetics of the nearest- neighbor bond, equation (14) becomes

$$D_M = D_{id} \left[1 - x(1-x) \left(\frac{2\omega}{k_B T} \right) \right] \quad (18)$$

2.4 Surface properties

Many properties of technological importance such as mechanical behavior, kinetics of phase transformation, catalytic activity of alloy catalyst and thin film cannot be explained without understanding surface phenomena i.e. surface segregation and surface tension. Surface segregation results from the interaction among the atoms and surface tension, is the enrichment of the surface by the atoms of the particular component element in a binary alloy [4, 5]. In the statistical formulation of Prasad et al. [4, 5] binary liquid alloy is considered to have a layered structure near the surface with thermodynamic equilibrium existing between the species at the surface and in the bulk. The surface properties of liquid alloys are influenced by their bulk thermodynamic properties. The surface grand partition function is related to the surface tension τ by the expression [5].

$$\Xi^s = \exp \left(\frac{-S\tau}{k_B T} \right) = \exp \left(\frac{-N^s \tau \xi}{k_B T} \right) \quad (19)$$

where S is the surface area and ξ is the mean area of the surface per atom and is defined as $\xi = S/N^s$, and N^s is the total number of atoms at the surface. k_B is the Boltzmann constant.

A pair of equation for surface tension of the binary liquid alloys in terms of activity coefficient (γ_i) of the alloy components and interchange energy parameter (ω), at the given temperature T , proposed by Prasad et al., have been reduced in the simple form, using zeroth approximation as

$$\tau = \tau_A + \frac{k_B T}{\xi} \ln \frac{x^s}{x} + \frac{\omega}{\xi} \left[p(1-x^s)^2 + (q-1)(1-x)^2 \right] \quad (20a)$$

$$\tau = \tau_B + \frac{k_B T}{\xi} \ln(1-x^s) - \frac{k_B T}{\xi} \ln(1-x) + \{p(x^s)^2 + (q-1)x^2\} \frac{\omega}{\xi} \quad (20b)$$

where τ_A and τ_B are the surface tension values for the pure components A and B respectively; x and x^s are the bulk and surface concentrations of the alloy component; p and q are the surface coordination

fractions, which are defined as the fraction of the total number of nearest neighbors made by atom within its own layer and that in the adjoining layer. For p and q, the following relation is available [4-7].

$$P + 2q = 1$$

For closed packed structure, p = 0.5 while q = 0.25 (in view of the disordered structure and relaxation effect of the surface layer p and q should be treated as parameters [5]).

The pair of Eqs. 20(a) and 20(b) can be solved numerically to obtain x^s as a function of x. Obviously the surface concentration depends upon the surface tension of the ith component in the pure state (τ_i), surface area per atom (ξ), order energy (ω) and the coordination fractions (p and q). This approach is useful because it can be used to investigate the dependence of surface composition on order energy and surface coordination.

The mean atomic surface area ξ ($=A/N^s$) is given as

$$\xi = \sum c_i \xi_i \quad (i = A, B) \quad (21)$$

where the atomic area of hypothetical surface for each component is given as [5]:

$$\xi_i = 1.102 \left(\frac{\Omega_i}{N_0} \right)^{2/3} \quad (22)$$

where Ω_i is the molar volume of the species i and N_0 stands for Avogadro number.

On the basis of assumption of monatomic surface layer, Butler's approach [20] of surface tension τ of liquid solution can be expressed as

$$\tau = \frac{\mu_A^s - \mu_A^b}{\alpha_A} = \frac{\mu_B^s - \mu_B^b}{\alpha_B} = \dots = \frac{\mu_i^s - \mu_i^b}{\alpha_i} \quad (23)$$

where μ_i^s, μ_i^b and α_i represent respectively the chemical potential in the hypothetical surface and that in the bulk and molar surface area of pure component i (A or B).

Equation(23) yields the expressions for surface tension in terms of partial excess free energy of mixing in bulk ($G_i^{E,b}$) and at the surface ($G_i^{E,s}$), and concentration in the bulk (x) and at the surface (x^s) as

$$\tau = \tau_A + \frac{1}{\alpha_A} \left(G_A^{E,s} - G_A^{E,b} \right) + \frac{RT}{\alpha_A} \ln x^s - \frac{RT}{\alpha_A} \ln x \quad (24a)$$

$$\tau = \tau_B + \frac{1}{\alpha_B} \left(G_B^{E,s} - G_B^{E,b} \right) + \frac{RT}{\alpha_B} \ln(1 - x^s) - \frac{RT}{\alpha_B} \ln(1 - x) \quad (24b)$$

where τ_A and τ_B are surface tension of pure component A and B respectively.

The area of monatomic surface layer for the component i can be calculated by the relation [20]

$$\alpha_i = 1.091 N_0^{1/3} \Omega_i^{2/3} \quad (25)$$

where N_0 is Avogadro's number and Ω_i stands for molar volume of the component i, can be calculated from its molar mass and density.

3. Results and discussion

The energy parameter used for the calculation for Cd-Mg liquid alloys at 773K has been determined from equations (4) and (5) by using experimental value of G_M^{XS} and H_M for equiatomic composition [12]. The best fitting values of parameter are found as:

$$\frac{\omega}{k_B T} = -0.224 \quad \text{and} \quad \frac{1}{k_B} \frac{d\omega}{dT} = -0.07084 \quad (26)$$

The negative value of energy parameter $\omega/k_B T$ suggests that there is higher tendency for unlike atoms to pair in the alloy which implies a hetero-coordination system. However, the tendency of pairing is weak since energy parameter is small. We have observed that if energy parameters are supposed to be independent of temperature ($d\omega/dT = 0$), then S_M and H_M so obtained are in poor agreement with experimental data. This suggests the importance of temperature dependence of ordering energy, ω .

The free energy of mixing, heat of mixing and entropy of mixing for Cd-Mg liquid alloys at 773K have been computed from equations (3), (5) and (6). The plot of G_M/RT , H_M/RT and S_M/R versus x_{Cd} are depicted in Fig. 1. The calculated and experimental values of G_M/RT , H_M/RT and S_M/R are in good agreement throughout whole concentrations of cadmium. Therefore our choice of ω for the alloy is good because it is confirmed from the qualitative agreement between calculated and experimental values. The value of G_M/RT and H_M/RT are minimum but S_M/R is maximum at $x_{Cd} = 0.5$, which show that Cd-Bi liquid alloy is symmetric about equiatomic concentration. The negative small values of free energy of mixing throughout the entire compositions indicate that the Cd-Bi alloy at 773K in molten state is weakly interacting system. The activity is one of the important thermodynamic functions which are obtained directly from experiment. The deviation from ideal behaviour is incorporated into activity. We have used the same value of the energy parameter in Eq. (8a) and (8b) for the evaluation of chemical activities of the components of the alloy. There is well agreement between calculated and experimental values of the activities of the components Cd and Bi of the alloy (Fig. 2).

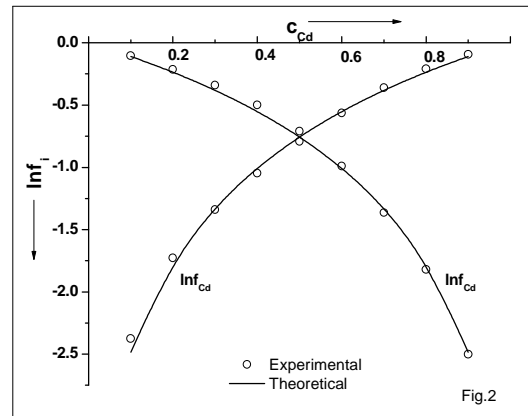
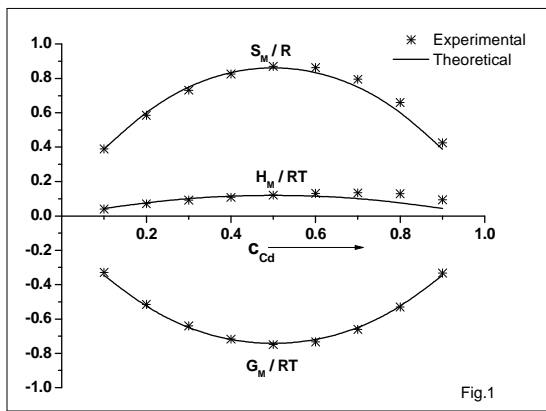


Figure 1: Free energy of mixing (G_M), heat of mixing (H_M) and entropy of mixing (S_M) Vs concentration of cadmium (x_{Cd}) in liquid Cd-Bi alloy at 773K.

Figure 2: Chemical activity (f_i) versus concentration of cadmium (x_{Cd}) in liquid Cd-Bi alloy at 773K.

We have used Eq.(10) to compute the $S_{cc}(0)$ for Cd-Bi. It can also be obtained directly from the measured activity [12] data as

$$S_{cc}(0) = (1-x)f_{Cd} \left(\frac{\partial f_{Cd}}{\partial x} \right)^{-1} = x f_{Bi} \left(\frac{\partial f_{Bi}}{\partial (1-x)} \right)^{-1} \quad (27)$$

where f_{Cd} and f_{Bi} are the observed chemical activities of cadmium and bismuth respectively. The $S_{cc}(0)$, obtained from Eq. (27) are taken as experimental values. Figure 3 shows a plot of the calculated and experimental values of $S_{cc}(0)$ along with the ideal values. The calculated values of $S_{cc}(0)$ are in good agreement with the experimental values of $S_{cc}(0)$ except in the region $0.7 < x_{Bi} < 0.3$. In the region $0.3 < x_{Bi} < 0.7$, the computed values lie below the experimental values but in the whole range of concentrations of aluminium, computed values of $S_{cc}(0)$ lying below the corresponding ideal values. The result $S_{cc}(0) < S_{cc}^{id}(0)$, clearly described this system as compound forming.

Figure 4 shows the plots of α_1 against chemical composition of cadmium, obtained from Eq. (12) for Cd-Bi at 773K. It is observed that the plots are symmetrical about equiatomic composition and negative throughout the whole concentration range of cadmium. The negative values of α_1 (minimum at $x_{Cd} = 0.5$) throughout whole concentration range are the signatures of hetero-coordination system in the Cd-Bi liquid alloys at 773K.

The viscosity of the Cd-Bi liquid alloy has been computed numerically from Eq. (13). From the plot of η verses bulk concentration of x_{Cd} (Fig.5) in Cd-Bi liquid alloy, positive deviation from the linear law (Raoult's law) in viscosity isotherms $\eta(c)$ have been observed for the regular alloy.

The calculated values of $S_{cc}(0)$ are used in Eq. (16) to evaluate the ratio of the mutual and intrinsic-diffusion coefficients, D_M/D_{id} . For the consistency of the estimated order energy parameter, ω , we have also calculated D_M/D_{id} using Eq. (18). Fig.6 shows plot of D_M/D_{id} against the concentration of cadmium. In that plot the value of D_M/D_{id} is found greater than 1 in the entire range of concentration which is indicative for the compound formation in the mixture. A maximum value of $D_M/D_{id} = 1.112$ for $x_{Cd} = 0.5$, confirms a weak tendency for chemical ordering, as observed by the $S_{cc}(0)$ and CSRO parameter.

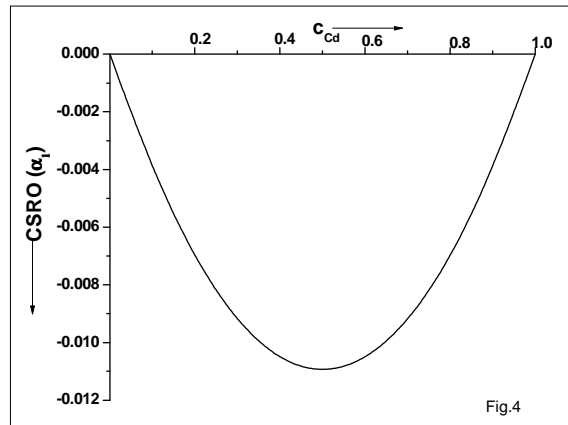
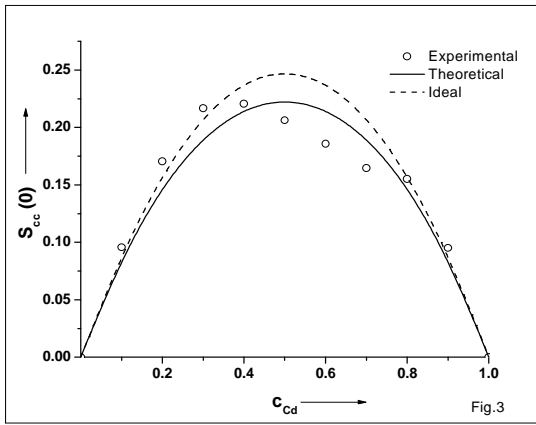


Figure 3: Concentration fluctuation at long wavelength limit ($S_{cc}(0)$) Vs concentration of cadmium (x_{Cd}) in liquid Cd-Bi alloy at 773K.

Figure 4: Chemical short range order parameter (α_1) Vs concentration of cadmium (x_{Cd}) in liquid Cd-Bi alloy at 773K.

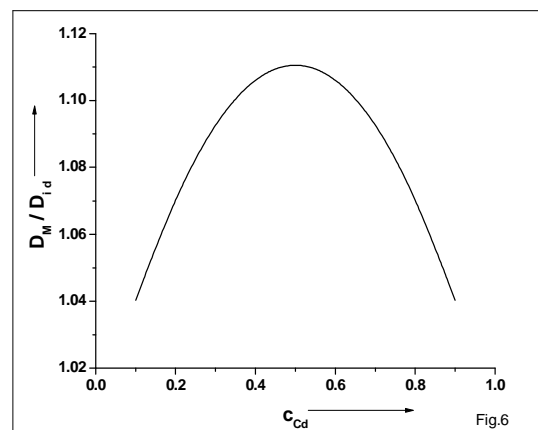
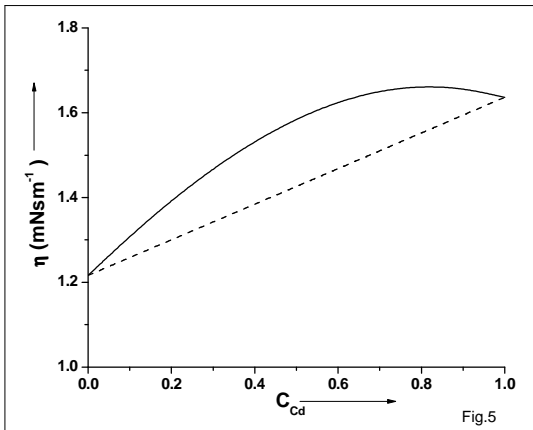


Figure 5: Viscosity (η) Vs concentration of cadmium (x_{Cd}) in liquid Cd-Bi alloy at 773K.

Figure 6: Ratio of mutual and intrinsic diffusion coefficients (D_M/D_{id}) Vs concentration of cadmium (x_{Cd}) in liquid Cd-Bi alloy at 773K.

The surface concentrations and surface tension of Cd-Bi have been computed numerically from the Eqs. 20(a) and 20(b). For this, we need experimental data of density and surface tension of the components at the working temperature. We have calculated the density and surface tension for the components Cd and Bi [21,22] at the working temperature $T=773\text{K}$ by using the following equations

$$\rho_{\text{Cd}}(T) = 8.70904 - 1.16 \times 10^{-3} T \quad (28a)$$

$$\rho_{\text{Bi}}(T) = 10.79152 - 1.33 \times 10^{-3} T \quad (28b)$$

$$\tau_{\text{Cd}}(T) = 0.585444 - 2.6 \times 10^{-5} T \quad (29a)$$

$$\tau_{\text{Bi}}(T) = 0.41608 - 7.0 \times 10^{-5} T \quad (29b)$$

The mean atomic surface area ξ has been calculated by using Eqs. (21) and (22). For calculating surface tension we used same estimated energy parameter, ω . At first, surface concentrations of cadmium in alloy Cd-Bi have been obtained as a function of bulk concentration by concurrently solving the Eqs. 20(a) and 20(b). Using the obtained values of surface concentrations we computed the surface tension of Cd-Bi liquid alloys at temperature 773K for whole concentration range.

For the computation of surface concentration and surface tension of the Cd-Bi alloy on using Butler's approach, the partial excess free energies of mixing of pure components of the alloys have been taken from the ref. [12]. By concurrently solving the equations 24(a) and 24(b) surface concentrations of Cd in alloy Cd-Bi have been obtained as a function of bulk concentration and then using the obtained values of surface concentrations we computed the surface tension of Cd-Bi liquid alloys at temperature 773K for whole concentration range.

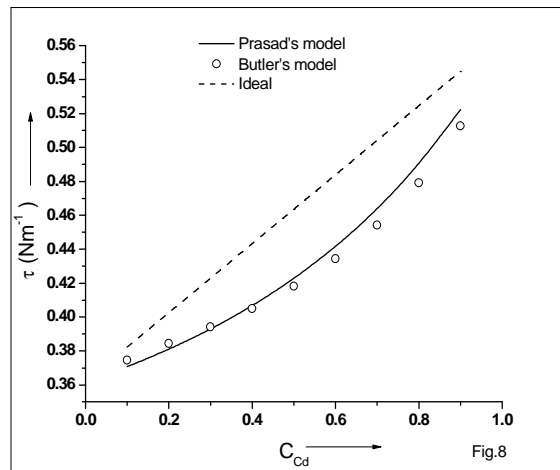
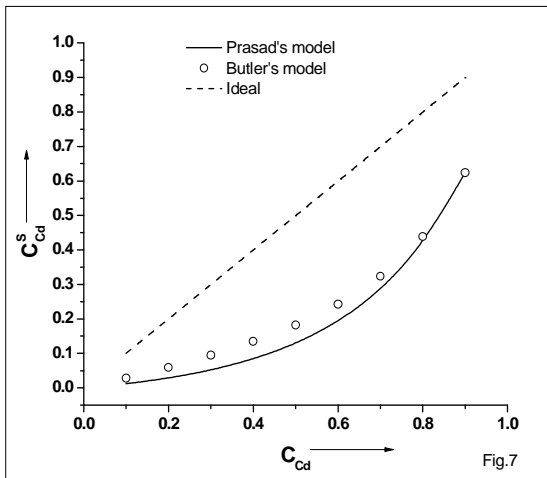


Figure 7: Surface concentration of cadmium (x_{Cd}^{S}) Vs bulk concentration of cadmium (x_{Cd}) in liquid Cd-Bi alloy at 773K.

Figure 8: Surface tension (τ) Vs bulk concentration of cadmium (x_{Cd}) in liquid Cd-Bi alloy at 773K.

The study of surface concentration shows the usual pattern for surface concentration increasing with increasing bulk compositions (Fig. (7)). This indicates that Bi-atoms (having lower value of surface tension relative to Cd-atoms in Cd-Bi) segregate at the surface of Cd-Bi liquid alloys in preference to Cd-atoms throughout the entire compositions. The computed values of surface tension for molten Cd-Bi alloys at 773K from two approaches, mentioned above are depicted in Fig. (8). Due to lack of surface tension experimental data we could not compare our computed surface tension with the experimental data, nonetheless, we observed that the isothermal plots of surface tension of the system exhibits negative

deviation from the ideal values ($\tau = \tau_A x + \tau_B (1-x)$) as it is the case with most binary liquid mixtures [21]. It is noticed that in two approaches computed values of τ and x_{Cd}^S are found to be smaller than the ideal values throughout bulk concentration of cadmium.

4. Conclusion

Following conclusions are drawn from the theoretical investigation on Cd-Bi liquid alloys at 773K:

- Order energy is small, negative and found to temperature dependent.
- The alloy is of a weakly interacting nature.
- It is of ordering system.
- Negative departure of thermodynamic and structural parameters from ideality is observed.
- Viscosity isotherm of the alloy is found to deviate positively from ideality.
- Consistency is occurred in the values of surface tension obtained from two approaches, negative departure of surface tension from ideality is observed.
- The surface of liquid Cd-Bi alloys is enormously rich with bigger Bi atoms.

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