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Thermodynamic, structural, surface and transport properties of Zn-Cd liquid alloy at 800 K

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Abstract

The mixing thermodynamic and structural properties of Zn-Cd liquid at 800K has been studied using Flory's model. To explain the mixing properties of binary liquid alloys, size factor (ϕ) and ordering energy (ω) are taken into account. Thermodynamic properties like free energy of mixing (G_M), activity (a), Heat of mixing (H_M) and entropy of mixing (S_M) and the microscopic properties like concentration fluctuation in the long wave length limit (Scc(0)) and chemical short range order parameter (α_1) have been calculated. Surface property has also been studied with the help of Buttler's model. The viscosity of the melt has been computed from Kaptay equation and BBK models. Both the viscosity and surface tension of the alloy increase with addition of zinc- component.

Keywords: Thermodynamic properties; Structural properties; Surface properties; Transport properties

1. Introduction

Zn-Cd alloy is a simple eutectic system which has been widely studied due to its low melting point and regular lamellar structure. Since cadmium is highly reactive element it has limited applications in its pure state. But the alloys of cadmium have several useful properties for industrial applications. When cadmium is alloyed with zinc, the properties of hardness, wear resistance, mechanical strength and fatigue strength are significantly improved. Cadmium and cadmium based alloys is the subject of both theoretical and experimental research studies [1-6].

Zinc–Cadmium alloys are used as solders at medium temperature which provide excellent corrosion resistance joints on most metals. Devices with such superior strength joints can work in high vibration

and high stress applications in electronics, lighting and electrical products. Moreover, use of cadmium in electroplating as in cyanide bath poses problems of environmental concern because cadmium is toxic. Zn-Cd alloys can be used in dry electroplating and can be a safer potential substitute for cadmium.

In metallurgical science, the study of mixing properties of liquid alloys is important because a good knowledge of their mixing properties in the liquid state is necessary for preparation of desired materials. Surface properties are required to understand the surface related phenomena such as corrosion, wetting characteristics of solders and kinetics of phase transformation. Transport properties such as viscosity and diffusivity of metals in the liquid state are required for many metallurgical processes and heterogeneous chemical reactions. Many researchers [7-12] have been working with several models to explain the mixing properties of binary liquid alloys. The alloying behavior of binary liquid alloys can be studied theoretically by computing thermodynamic, structural, transport and surface properties of alloys in the liquid state. The mixing behavior of binary liquid has been explained by several theoreticians of the basis of several models [13-19]. In this work we have used Flory's model [19, 20] to explain the thermodynamic and structural behavior of Zn-Cd liquid alloy at 800 K. In Flory's model the interaction energy parameter is considered as temperature dependent and is determined by fitting experimental values of thermodynamic functions at different concentrations.

2. Formalism

2.1 Thermodynamic function

2.1.1 Free energy of mixing

Flory's model [19] is found to be the best applicable for the determination of thermodynamic and microscopic properties of those alloys which has greater size mismatch. Homogenous solution of binary liquid alloy A-B consists of c_A (≡ c) mole of A and c_B {≡ c(1-c)} mole of B respectively, where c_A and c_B are the mole fractions of A (≡ Zn) and B (≡ Sn) in the binary liquid solution of A and B. Thus free energy of mixing of those alloys whose constituent atoms differ widely in sizes can be expressed as [19]

$$G_M = G(ideal) + G(size) + c(1-c)G(\omega) \quad (1)$$

where,

$$G(id) = [clnc + (1-c)\ln(1-c)]RT \quad (2)$$

G (size) and G (ω) are contributions due to the size effect and the interchange energy (ω) respectively. From Flory's model [19], we have

$$G(size) = RT[cln(1-\beta) - \ln(1-\beta c)] \quad (3)$$

$$G(\omega) = c(1-c)\omega/(1-\beta c) \quad (4)$$

$$\beta = 1 - 1/\Phi \quad (5)$$

with $\Phi = \vartheta_B/\vartheta_A$

where ϑ_A and ϑ_B are atomic volumes of the pure species A and B respectively [20]

Here

$$\vartheta_{Zn} = \vartheta_M [1+\alpha_p (T-T_M)] \text{ and } \vartheta_{Cd} = \vartheta_M [1+\alpha_p (T-T_M)] \quad (6)$$

where,

ϑ_M = atomic volume at melting point

T_M = melting temperature and

α_p = volume coefficient at constant temperature

Then the expression for free energy of mixing is given by

$$G_M = RT\left[c\ln c + (1-c)\ln(1-c) + c\ln(1-\beta) - \ln(1-\beta c)\right] + \frac{\omega c(1-c)}{(1-\beta c)} \quad (7)$$

2.1.2 Chemical activity (a)

The activity (a_A) of the element A in the binary liquid alloy is given as

$$RT ln a_A = G_M + (1-c)\frac{\partial G_M}{\partial c} \quad (8)$$

From equation (1) and (8), we get

$$ln a_A = ln\left[c(1-\beta)\eta(c)\right] + \beta(1-c)\eta(c) + (1-c)^2\eta^2(c)\omega/RT \quad (9)$$

where, $\eta(c) = 1/(1-\beta c)$

2.1.3 Entropy of mixing (S_M)

The temperature derivative of G_M provides an expression for integral entropy of mixing

$$S_M = -RG(id) - RG(size) - \frac{c(1-c)\eta(c)\partial\omega}{\partial T} + RTc(1-c)\eta(c) \times \left[\beta/(1-\beta) - c\eta(c)\omega/RT\right]\partial\beta/\partial T \quad (10)$$

where,

$\partial\beta/\partial T = (\alpha^B - \alpha^A).\vartheta_A/\vartheta_B$ and $\eta(c) = 1/(1-\beta c)$

where, α^A and α^B are the coefficients of thermal expansion of pure species A and B respectively.

2.1.4 Heat of mixing (H_M)

The heat of mixing can be obtained from equation (1) and (10) from standard thermodynamic relation,

$$\frac{H_M}{RT} = \frac{S_M}{R} + \frac{G_M}{RT} \quad (11)$$

$$\frac{H_M}{RT} = c\ln c + (1-c)\ln(1-c) + c\ln(1-\beta) - \ln(1-\beta c) + \frac{c(1-c)}{(1-\beta c)}.\frac{\omega}{RT} - \alpha(c) - \Phi(c) - \frac{1}{R}\frac{c(1-c)}{(1-\beta c)}.\frac{\partial\omega}{\partial T} + \frac{Tc(1-c)}{(1-\beta c)}.\left[\frac{\beta}{(1-\beta)} - \frac{c}{(1-\beta c)}\frac{\omega}{RT}\right].\frac{\partial\beta}{\partial T} \quad (12)$$

where,

$\alpha(c) = [clnc + (1-c)\ln(1-c)]$ and $\phi(c) = [cln(1-\beta) - \ln(1-\beta c)]$

2.2 Microscopic functions

2.2.1 Concentration fluctuation in the long wavelength limit ($S_{cc}(0)$)

To study the atomic order of binary liquid alloy it is important to understand the behavior of the long wavelength limit of the concentration- concentration structure factor ($S_{cc}(0)$) and is given as [10,17,18]

$$S_{CC}(0) = \frac{RT}{\left(\frac{\partial^2 G_M}{\partial c^2}\right)_{T,P,N}} \quad (13)$$

The expression for concentration fluctuation in the long wavelength limit is obtained using equation (1) and (13) and is expressed as

$$S_{cc}(0) = \frac{c_A c_B}{[1-\frac{c_A c_B}{(1-\beta c)^3}\{2(1-\beta)\frac{\omega}{RT}-\beta^2(1-\beta c)\}]} \quad (14)$$

The experimental values of concentration fluctuation in the long wavelength limit $S_{cc}(0)$ derived from experimental data of the activities of the constituent species of the binary liquid alloys from the relation

$$Scc(0) = (1-c)a_A\left(\frac{\partial a_A}{\partial c}\right)^{-1}_{T,P,N} = ca_B\left(\frac{\partial a_B}{\partial c}\right)^{-1}_{T,P,N}$$

where a_A and a_B are the activities of the component of A and B respectively.

2.2.2 Short range order parameter (α_1)

The Warren-Cowley short range order parameter α_1 can be estimated from the knowledge of $S_{cc}(0)$ as

$$\alpha_1 = \frac{s-1}{s(Z-1)+1} \quad (15)$$

where

Z= coordination number and $S = \frac{S_{cc}(0)}{S^{id}_{cc}(0)}$

2.3 Surface tension

Buttler assumed the existence of surface monolayer at the surface of a liquid as a separate phase that is in thermodynamic equilibrium with the bulk phase and derived an equation, known as Buttler equation [7, 21, 22]. Buttler equation for the surface tension, σ of a binary A-B solution at temperature T reads as [7, 21, 22]

$$\Gamma = \Gamma_1 + \frac{1}{A_1}\left(G_1^{E,s} - G_1^{E,b}\right) + \frac{RT}{A_1}\left[\ln(1-X_2^s) - \ln\left(1-X_2^b\right)\right] = \Gamma_2 + \frac{1}{A_2}\left(G_2^{E,s} - G_2^{E,b}\right) + \frac{RT}{A_2}\left[\ln(X_2^s) - \ln\left(X_2^b\right)\right] \quad (16)$$

where, Γ_1 and Γ_2 are the surface tension of the pure component 1 and 2 respectively. $G_i^{E,s}$ and $G_i^{E,b}$ (i = 1,2) are partial excess free energy of component i in the surface and the bulk respectively. The molar surface area of the component i can be computed by using the relation

$$A_i = K.N_A^{1/3}.V_i^{2/3} \quad (17)$$

where, K (= 1.091) is geometrical factor for the liquid alloy [23, 24]

N_A is Avogadro's number and V_i is the molar volume of the component I.

For binary mixture $X_1^b + X_2^b = X_1^s + X_2^s = 1$, where X_i^s *and* X_i^b are mole fractions of component i in the surface and bulk respectively. R is universal gas constant and T stands for absolute temperature.

2.4 Viscosity

To study the atomic transport behavior in Zn-Cd alloys, we have computed its viscosity at 800 K by using Kaptay equation and BBK model [24, 25].

2.4.1 Kaptay equation

Kaptay [24] equation for the viscosity of the binary liquid alloys at temperature T is given as

$$\eta = \frac{hN_A}{\sum_K C_K \Omega_K + \Omega^E} \exp\left[\frac{\sum_K C_K G_K^* - \theta.H_M}{RT}\right] \quad (18)$$

where, h = Plank's constant, N_A is Avogadro's number, R is the ideal gas constant, $\Omega^E$is the excess molar volume upon alloy formation, H_M is enthalpy of mixing of the alloy, C_K (=A,B) represents concentration,Ω_K represents the molar volume and G_K^* is the Gibb's energy of activation of the viscous flow in pure component K and θ is a constant whose value is taken to be 0.155±0.015 [25].

$G_K^*$of component K can be calculated from the equation [25]

$$G_K^* = RT \ln\left(\frac{\eta_K \Omega_K}{hN_A}\right) \quad (19)$$

where, η_K is the viscosity of pure component K; is the planks constant. η_K at temperature T can be evaluated by

$$\eta_K = \eta_{OK} \exp\left[\frac{E_n}{RT}\right] \quad (20)$$

where, η_{OK} is constant (in unit of viscosity) and En is the energy of activation of viscous flow for pure metal (in unit of energy per mole).

2.4.2 Budai-Benko- Kaptay (BBK) model

The expression for estimation of the viscosity of a multi- component alloy from BBK model is given by [25]

$$\eta = A(T\sum_K c_K M_K)^{1/2}.(\sum_K c_K \Omega_K + \Omega^E)^{-\frac{2}{3}}.\exp[\left(\sum_K c_K T_{m,k} - \frac{H_M}{qR}\right).\frac{B}{T}] \quad (21)$$

where, A and B are fitting parameter equal to (1.80±0.39)x10^{-8} $(J^{-K}mol^{-1/3})^{1/2}$ and (2.34±0.20) respectively; c_K is the concentration , and M_K is the atomic mass of the given component K, q is the semi – empirical parameter equal to q≡25.4±2; Ω_K is the molar volume of the alloy, and $T_{m,k}$is the effective melting temperature of the component K given by

$$T_{m,k} = \frac{T}{B}\, ln\left(\frac{\eta_K \Omega_K^{\frac{2}{3}}}{AM_K^{\frac{1}{2}} T^{\frac{1}{2}}}\right) \quad (22)$$

where ,η_K is viscosity of the pure component K.

3. Result and discussion

3.1 Free energy of mixing

To compute the free energy of mixing G_M/RT of molten Zn-Cd as a function of concentration at 800 K, size factor (ϕ) and energy parameter are required. The interchange energy ω has been calculated from equation (7) with the help of experimental values of G_M [26] in the concentration range of c_{Zn}=0.1 to 0.9 by the method of successive approximation and the value of size factor ϕ is evaluated by equation (5) and (6) in alloying temperature.

In the present work the best fit value of ordering energy (ω) has been found to be 1.085RT. The size ratio (i.e. ϕ = V_{Cd}/V_{Zn}) where V stands for atomic volume for the constituent atom in Zn-Cd alloys at 800K is determined by using equation (5). The value of size ratio is found to be 1.43. Using these two input parameters the free energy of mixing G_M of Zn-Cd liquid alloys at 800 K has been computed using equation (7) for concentration range c_{Zn} =0.1 to 0.9. The experimental values of free energy of mixing are taken from the ref. [26].

The plot of computed and experimental values of free energy of mixing with respect to concentration is depicted in fig 3.1. The computed and experimental values of free energy of mixing are in good agreement in entire concentration range. The disagreement between the computed and experimental values is within 6%. The theoretical values is minimum around c_{Zn} = 0.4 (i.e. G_M/RT = -0.3919) while the experimental values is minimum around c_{Zn} = 0.5 (i.e. G_M/RT = -0.3927). The minimum value suggests that Zn-Cd alloy in liquid state at 800 K is weekly interacting system.

3.2 Activity

By putting the input parameters interchange energy (ω) and size factor (ϕ) in equation (8) and (9), we have computed the activity of Zn and Cd of Zn-Cd alloys in molten state at 800

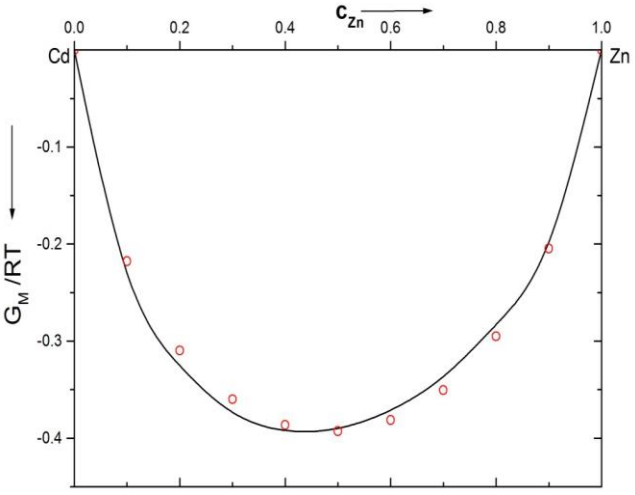

Fig. 3. 1: Free energy of mixing of Zn-Cd liquid alloys at 800 K. The solid line represents theoretical values and circle represents experimental values

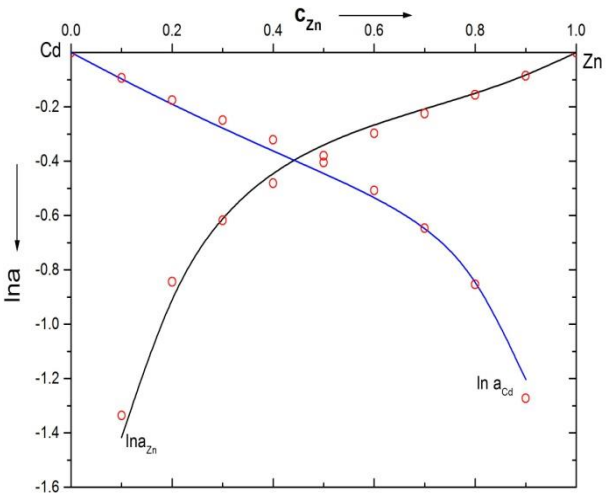

Fig.3. 2: Activities a_{Zn} and a_{Cd} of Zn-Cd liquid alloys at 800 K. The solid line represents theoretical values and circle represents experimental values.

The computed values of activity of both the components of Zn-Cd alloys at 800 K are in good agreement with the experimental values for whole range of concentration with some discrepancies. For zinc the maximum departure from experimental value [26] of lna_{Zn} from experimental value is 9.4% around c_{Zn} = 0.4 as shown in figure 3.2.

3.3 Heat of mixing

The heat of mixing of Zn-Cd liquid alloy at 800 K has been determined by using equation (10). Both of the computed and experimental values of heat of mixing are positive in whole range of concentrations. The computed and experimental values of heat of mixing are in reasonable agreement in all compositions.

There is small negative deviation of computed values from experimental values in the region C_{Zn}>0.4 whereas small positive deviation has been observed in the region C_{Zn}<0.4 at C_{Zn}=0.4. The computed and experimental values are almost same. The graphical comparison between computed and experimental values [26] of heat of mixing with respect to concentration is displayed in fig 3.3.

3.4 Entropy of mixing

Following equation (11) the entropy of mixing of Zn-Cd alloy in molten state at 800 K has computed. For the computation of entropy of mixing the basic input parameters are interchange energy (ω), size factor (ϕ) and temperature derivative of ordering energy (dω/dT). For the consistency we have used the same values of these parameters as used in the calculation of free energy of mixing, activity and heat of mixing.

The computed and experimental values are found to be in excellent agreement. The disagreement between computed and experimental values is within 1%. The computed entropy of mixing is minimum (i.e.S_M/R = 0.7050) at C_{Zn} =0.5. The computed values of entropy of mixing together with experimental values are plotted as a function of concentration as shown in fig 3.4.

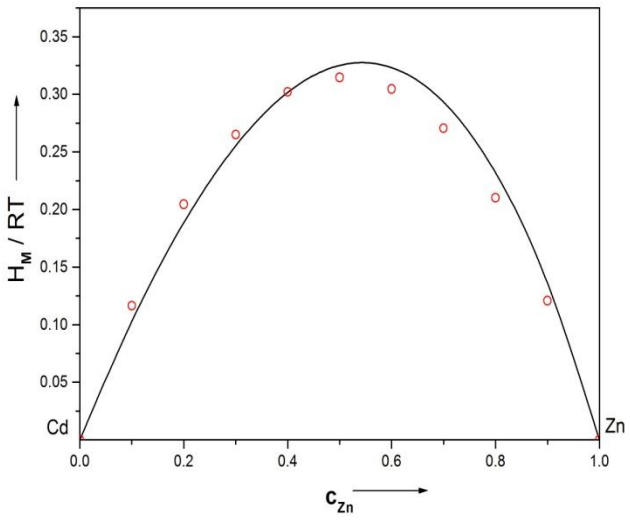

Fig. 3. 3*:* Heat of mixing of Zn-Cd liquid alloys at 800 K. The solid line represents theoretical values and circle represents experimental values.

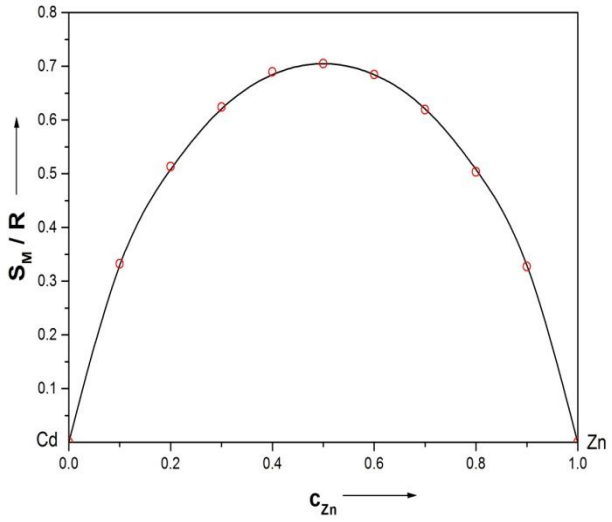

Fig. 3. 4: Entropy of mixing of Zn-Cd liquid alloys at 800 K. The solid line represents theoretical values and circle represents experimental values.

3.5 Concentration – Concentration structure factor in long wavelength limit $S_{cc}(0)$

The theoretical value of $S_{cc}(0)$ of Zn-Cd liquid alloys at 800 K computed from equation (12) using the same energy parameter ω and size factor ϕ for the full range of concentration (i.e. C_{Zn}= 0.1 to 0.9) as used for the computation of free energy of mixing, activity, heat of mixing and entropy of mixing.

Both the computed and experimental values of $S_{cc}(0)>S_{cc}^{id}(0)$. The plot of computed and experimental values of $S_{cc}(0)$ along with the ideal values with respect to concentration are also depicted in figure 3.5. The computed values of $S_{cc}(0)$ are in good agreement with the experimental values in entire range. The disagreement between the computed and experimental values is within 12%. The theoretical value is maximum at C_{Zn}= 0.6 (=0.6407), but the experimental value is maximum at C_{Zn} = 0.5 (=0.5522). The $S_{cc}(0)$ is used to understand the nature of atomic order in binary alloys. Any deviation of $S_{cc}(0)$ from ideal value is of interest in reflecting the extent of interactions in the mixture. At the given composition if $S_{cc}(0)<S_{cc}^{id}(0)$, ordering of liquid alloy is expected and if $S_{cc}(0)>S_{cc}^{id}(0)$, there is tendency of segregation or phase separation. In present case $S_{cc}(0)>S_{cc}^{id}(0)$ which indicates that Zn-Cd liquid alloys is segregating in nature.

3.6 Chemical short range order parameter (α_1)

The chemical short range parameter has been computed by using equation (13) as a function of concentration of Zn-Cd liquid alloys at 800 K for full range of concentrations. In our present work we have computed the value of short range order parameters for different values of coordination numbers (i.e. Z= 7,8,10).

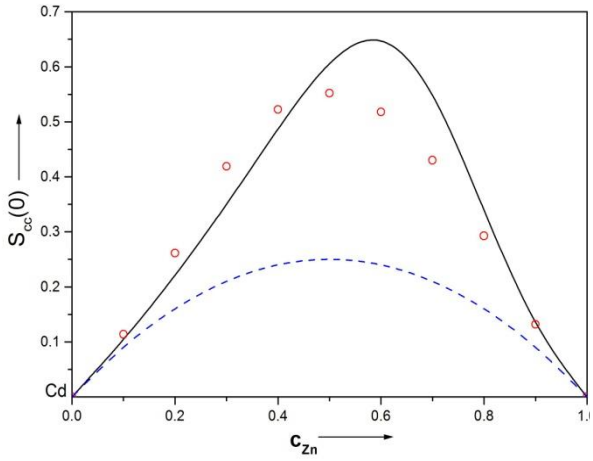

Fig. 3. 5: Concentration fluctuation of Zn-Cd liquid alloys at 800 K. The solid line represents theoretical values, dotted line represents ideal values and circle represents experimental values.

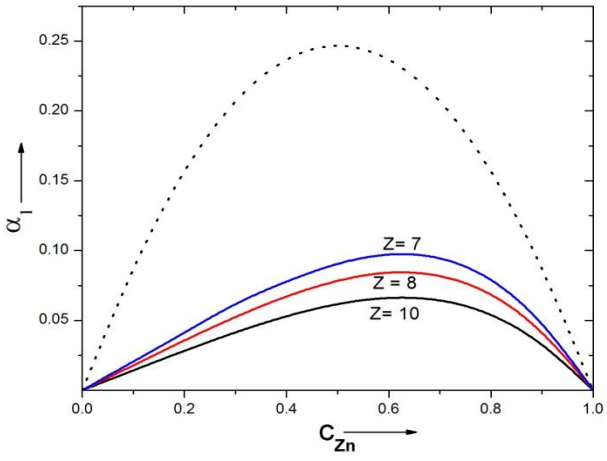

Fig.3 . 6: Chemical short range parameter (α_1) of Zn-Cd liquid alloys at 800 K for different co-ordination number Z= 7, 8 and 10. Dotted line represents ideal $S_{cc}(0)$.

The values of short range order parameter have been found positive in all concentration ranges. The value of short range order parameter has been found maximum at C_{Zn} = 0.6 in all three coordination numbers. The plot of computed values of Zn-Cd liquid alloy at 800 K is depicted in fig3.6.

The knowledge of short range order parameter (α_1) provides an immediate insight into the nature of the local arrangement of atoms in the mixture. The minimum possible value of α_1 is -1 and it implies complete ordering of unlike atoms pairing at nearest atoms. On the other hand the maximum value of α_1 is +1 which implies complete segregation leading to the phase separation and α_1=0 corresponding to the random distribution of atoms. Figure 3.6 shows that α_1 is positive in the entire concentration range (i.e. C_{Zn}= 0.1 to 0.9) showing that α_1 in Zn-Cd is segregating system of like atoms pairing (i.e. Zn-Zn and Cd-Cd) as nearest neighbours.

3.7 Surface tension

The surface tension of the liquid alloys can be computed using equation (14). The ratio of partial excess Gibbs energy in the bulk and that in the surface can be expressed as

$$\beta = \frac{G_i^{E,s}}{G_i^{E,b}}$$

where $G_i^{E,s}$ and $G_i^{E,b}$ are the partial excess free energy in the surface and that in the bulk. The values of excess mixing of the pure componenst are taken from ref. [26]. The value of parameter β has been taken as 0.83 as suggested by different researchers to compute surface tension of liquid alloys [27-29]. We have taken the surface tension of Zn-Cd and temperature coefficients for pure Zn and Cd components from the ref. [30]. The surface tension of the pure component at the temperature of study have been computed by the equation

$$\Gamma(T) = \Gamma_m + \frac{\partial \Gamma}{\partial T}(T - T_m)$$

T=800 K; Tm= melting temperature (T_m= 692.5 K for Zn, and 594 K for Cd); $\frac{\partial \Gamma}{\partial T}$ (= -0.17mNm^{-1}K^{-1} for Zn, and -0.26 mNm^{-1}K^{-1} for cadmium) is the temperature coefficient of surface tension.

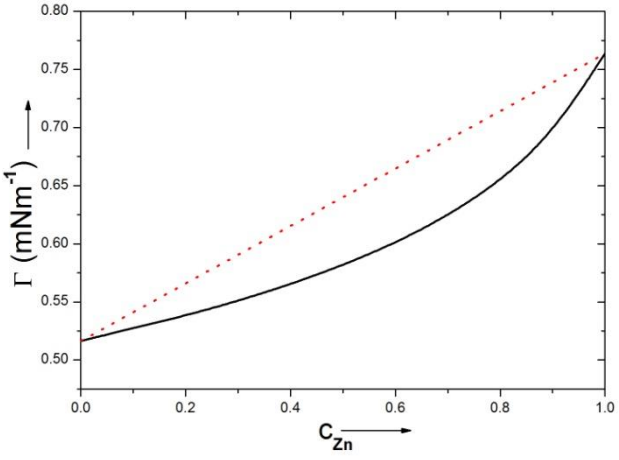

Fig 3. 7: Surface tension of Zn-Cd liquid alloy at 800 K. Solid line is calculated surface tension and dotted line is ideal surface tension.

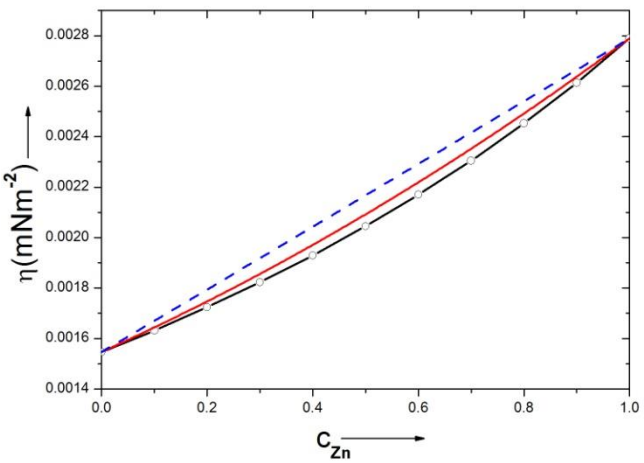

Fig 3. 8 : Viscosity (η) of Zn-Cd liquid alloy at 800K versus concentration of Zn. Dotted line represents ideal viscosity, solid line represents BBK and solid with circle represents Kaptay.

The graph shows that the computed surface tension for Zn-Cd system at 800 K is less than ideal value (= $C_1\Gamma_1 + C_2\Gamma_2$) at all the concentration of Zn as shown in figure 3.7 i.e. there is negative departure of surface tension from ideality. It is found that surface tension of Zn in Zn-Cd alloy is increased on increasing the bulk concentration of Zn.

3.8 Viscosity

To compute viscosity of Zn-Cd alloy at 800 K, the viscosities of the pure components Zn and Cd at 800 K are required. The viscosity of pure component can be obtained with the help of constants η_{ok} and E for the metals [31]. We have used Kaptay equation (15) and BBK model (16) to evaluate viscosity of the alloy and compared the results as shown in figure 3.8.

To calculate the viscosity by Kaptay equation enthalpy of mixing (H_M), the Gibbs free energy of activation of various viscous flow of the pure components (G^*) and excess molar volume of the alloy (Ω^E) are required. The enthalpy of mixing is taken from Flory's model calculations. The value of G* for each component was calculated from equation (17) with knowing the viscosity and molar volumes of pure components. Due to lack of experimental data we have taken the volume of Ω^E to be zero in our calculations for simplicity.

Viscosity from BBK model requires heat of mixing (H_M), excess molar volume of the component (Ω^E) and effective melting temperature of the component ($T_{m,k}$). Heat of mixing is calculated from Flory's model calculation and Ω^E is taken as zero in our calculation for simplicity.

The viscosities of the alloy calculated from different methods are compared along with ideal values ($=c_A\eta_A + c_B\eta_B$). Both Kaptay equation and BBK model show small negative deviation from ideal value.

4. Conclusion

Flory's model successfully explains the thermodynamic, microscopic, surface and transport properties of Zn-Cd binary liquid alloy at 800 K. The ordering energy is temperature dependent. The Zn-Cd liquid alloy is segregating in nature. The ordering energy and size factor play an important role to explain the alloying behavior of the liquid Zn-Cd alloy.

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