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ABSTRACT

The simple statistical model or simple theory of mixing has been used to study the structural behavior of cadmium based alloys at their molten state at a temperature of 800 K by computing thermodynamic functions and structural functions. The thermodynamic functions include free energy of mixing (G_M), activity (a), the heat of mixing (H_M), and the entropy of mixing (S_M). The structural functions include concentration fluctuation in the long-wavelength limit ($S_{CC}(0)$) and Warren-Cowley short-range order parameter (α_1). Interchange energy or interaction energy or ordering energy (ω) was calculated for the respective alloys system and found to be positive and temperature-dependent. Based on interchange energy (ω) and coordination number (Z), theoretical values of all the functions are calculated by applying the grand partition function. All the computed values for the mentioned functions are in good agreement with experimental values. For the cadmium based alloys, viz., Cd-Zn & Cd-In, both show the segregating in nature at temperature 800 K for the concentration of range 0.1 to 0.9, however, Cd-Zn is more segregating than Cd-In.

Keywords: Simple statistical model, Structural properties, Thermodynamical properties, Interchange energy.

1. INTRODUCTION

This world has been decorated with suitable materials which is the gift of present science. Furthermore, the demand of modern society is new and new reliable materials which could be fulfilled by the metallic mixture in addition to polymers. The metallic mixture may be formed by combing a metal to other metal or non-metals at their molten state which is termed also as an alloy. The determination of the thermodynamic, electrical, magnetic, surface, and structural properties of mixing of materials is one of the most intense research fields in present metallurgical science. Scientists and engineers frequently require the properties of different kinds of materials to design the structure involved in the construction. The mixing property of the binary liquid alloys results in producing new materials required for high-temperature applications.

Cadmium is the soft, bluish-white metal that is chemically related to zinc (demonstrates oxidation state +2 in most of its compounds) and mercury (lower melting point than the transition metals). It forms complex compounds although similar to zinc in many respects [1]. Cadmium is vastly used in batteries, coating, plating, alloys, etc. in various industries. Cadmium coated components, likewise, provide outstanding corrosion resistance along with low friction coefficient, low electrical resistivity, good galvanic comparability, good plating coverage, ability to coat a wide variety of surfaces, and good braze-ability and solder-ability. For these reasons, cadmium coated products are preferred for a wide range of critical and safety-related applications in the aerospace, electrical, defense, mining, nuclear, and offshore industries.

Despite cadmium has only limited use as a pure metal it forms many binary and more complex alloys that have useful properties for many commercial applications. Cadmium-containing alloys are generally important for their wide variety of applications as they possess improved

mechanical, thermal, and electrical properties such as tensile and fatigue strength, hardness, draw-ability, heat resistance, and electric conductivity, etc. Cadmium-Zinc alloys are used as solders at a medium temperature which provides excellent corrosion resistance joints on most metals. Eutectic cadmium-indium alloy can be used for soldering whisker wires, ribbons, stem leads, or other electrical connections to the emitter and collector electrodes without melting the In electrodes [2]. The alloying behavior of binary alloys can be studied theoretically by computing thermodynamic, structural, transport, and surface properties of alloys in the liquid state. For a long time, many researchers have been working with several models to explain the mixing properties of binary liquid alloys.

In the present work, we have used a simple statistical model [3] to study the thermodynamical properties such as free energy of mixing (G_M), activity (a), the heat of mixing (H_M) and entropy of mixing (S_M), and hence to study the structural properties such as concentration fluctuation in the long-wavelength limit $(S_{CC}(0))$ and Warren-Cowley short-range order parameter (α_1) of cadmium based alloys (Cd-Zn & Cd-In) alloys at molten state at 800 K.

2. THEORETICAL FORMALISM

2.1 Thermodynamic properties

The grand partition function for simple binary liquid alloys consisting of N_A (= Nx) and N_B (= N(1-x)) can be generalized as follows [3, 16]

$$\Xi = \sum_E q_A^{N_A}(T) q_B^{N_B}(T) \exp\left(\frac{\mu_A N_A + \mu_B N_B - E}{k_B T}\right) \ldots \ldots (1)$$

Where, the $q_i^{N_i}(T)$ (i =A, B) denote the partition functions associated with the inner and translational degrees of freedom of atoms i and μ_i are the chemical potentials. N_A and N_B are the numbers of A and B atoms in the alloy with the configurational energy E.

The standard thermodynamic relation for free energy of mixing is [3, 6 – 15],

$$G_M^{XS} = G_M - G_M^{id} \ldots \ldots (2)$$

Where, G_M^{id} is the free energy of mixing for ideal solution. For ideal solution, interaction between A-A, B-B and A-B are identical ($\omega = 0$), then we get,

$$G_M^{id} = NK_B T[x \ln x + (1 - x)\ln(1 - x)] \ldots (3)$$

and,

$$G_M^{XS} = RT \int_0^x \ln \sigma^z dx = RT\{x \ln \gamma_A + (1 - x)\ln \gamma_B\} \ldots (4)$$

With

$$\sigma = \frac{(\beta + 2x - 1)\exp\left(\frac{-\omega}{ZK_B T}\right)}{2x} \ldots (5)$$

$$\gamma_A = \lfloor (\beta - 1 + 2x)/x\,(1 + \beta)\rfloor^{z/2},$$

$$\gamma_B = \lfloor (\beta + 1 - 2x)/(1 - x)\,(1 + \beta)\rfloor^{z/2} \ldots (6)$$

$$\beta = \{1 + 4x(1 - x)\left[\exp\left(\frac{2\omega}{zk_B T}\right) - 1\right]\}^{1/2} \ldots (7)$$

For the equiatomic composition (x = 1/2), relation (4) reduces to

$$G_M^{XS} = \ln 2^{z/2}\left[1 + \exp\left(\frac{-\omega}{zk_B T}\right)\right]^{-Z/2} \ldots (8)$$

To calculate $\omega/K_B T$ in terms of excess free energy of mixing,

$$\omega/K_B T = -[Z = 10]\ln\left[2\exp\left[\frac{-G_M^{XS}}{5RT}\right] - 1\right] \ldots (9)$$

Here Z = 10 because in the liquid and amorphous states, the atoms are randomly distributed in a nearly close-packed structure and the coordination number of the nearest neighbors is about 10 [4].

Using relations (2) to (8), the free energy of mixing [3] will be

$$\frac{G_M}{RT} = x(1 - x)\frac{\omega}{K_B T} + x \ln x + (1 - x)\ln(1 - x) \ldots (10)$$

To calculate $\omega/K_B T$ in terms of free energy of mixing,

$$\frac{\omega}{K_B T} = \frac{\left[\frac{G_M}{RT} - x \ln x - (1 - x)\ln(1 - x)\right]}{x(1 - x)} \ldots (11)$$

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

$$RT \ln a_A = G_M + (1 - x)\left(\frac{\partial G_M}{\partial x}\right)_{T,P,N} \ldots (12)$$

$$RT \ln a_B = G_M + x\left(\frac{\partial G_M}{\partial(1-x)}\right)_{T,P,N} \ldots (13)$$

From Equation (12) and (13) we get [3]:

$$a_A = x \exp\left(\frac{\omega}{K_B T}(1 - x)^2\right) \ldots (14)$$

$$a_B = (1 - x)\exp\left(\frac{\omega}{K_B T}(1 - x)^2\right) \ldots (15)$$

The heat of mixing (H_M) can be obtained from the standard thermodynamic expression as

$$H_M = G_M - T\left(\frac{\partial G_M}{\partial T}\right)_P$$

$$(16)$$

$$\frac{H_M}{RT} = x(1-x)\frac{\omega}{K_B T} - x(1-x)\frac{1}{K_B}\frac{\partial \omega}{\partial T} \dots\dots\dots (17)$$

And entropy of mixing (S_M) can be obtained from the standard thermodynamic expression as

$$S_M = -\left(\frac{\partial G_M}{\partial T}\right)_{P,x,N} \dots\dots\dots\dots\dots\dots (18)$$

$$\frac{S_M}{R} = -x(1-x)\frac{1}{K_B}\frac{\partial \omega}{\partial T} - x\ln x - (1-x)\ln(1-x) \; .. \; (19)$$

If $-1 < G_M/RT < 0$, weakly interacting systems, if $-2 < G_M/RT < -1$, moderate interacting systems, if $G_M/RT \leq -2$, strongly interacting systems [3, 6-15]. The magnitude of activity of more interacting systems is smaller in comparison to the activity of less interacting systems [11-14].

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 [16, 17]. $S_{CC}(0)$ is thermodynamically related to free energy of mixing (G_M) [16]. It is given as

$$S_{cc}(0) = NK_B T \left(\frac{\partial^2 G_M}{\partial x^2}\right)^{-1}_{N,P,T} \dots\dots\dots\dots (20)$$

Equations (10) and (20) gives the theoretical value of $S_{CC}(0)$:

$$S_{cc}(0) = x(1-x)\left[1 - \frac{x(1-x)2\omega}{k_B T}\right]^{-1} \dots\dots\dots (21)$$

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

$$S_{cc}^{id}(0) = x(1-x) \dots\dots\dots\dots\dots\dots\dots (22)$$

There are difficulties in the diffraction experiment, therefore theoretical determination of $S_{CC}(0)$ is of great importance when the 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 $S_{cc}^{id}(0)$. Theoreticians have demonstrated that any deviation of $S_{CC}(0)$ from ideal value $S_{cc}^{id}(0)$ is of great importance to visualize the degree of ordering in the alloys [3]. They have demonstrated that $S_{cc}(0) > S_{cc}^{id}(0)$,

corresponds to segregation (preference of like atoms pairing as nearest neighbors or homo-coordination or phase separation) and $S_{cc}(0) < S_{cc}^{id}(0)$, refers to ordered alloys (preference for unlike atoms pairing as nearest neighbors or hetero-coordination).

The Warren-Cowley [26, 27] short-range order parameter (α_1) is a 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 the nearest neighbor of a given B-atom. It can be evaluated theoretically [26, 27]:

$$\alpha_1 = \frac{(S-1)}{\{S(z-1)+1\}} \dots\dots\dots\dots\dots\dots\dots\dots(23)$$

Where,

$$s = \frac{S_{CC}(0)}{S_{CC}^{id}(0)} = \frac{S_{CC}(0)}{x(1-x)} \dots\dots\dots\dots\dots\dots(24)$$

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.

3. RESULTS AND DISCUSSION

3.1. Thermodynamic properties

The energy parameter $\frac{\omega}{K_B T}$ and temperature derivative of energy parameter $\frac{1}{K_B}\frac{\partial \omega}{\partial T}$ used for the calculation for Cd-based alloys at 800 K are estimated as,

Table 1: Values of $\frac{\omega}{K_B T}$ and $\frac{1}{K_B}\frac{\partial \omega}{\partial T}$ for Cd-based alloys at 800 K

Cd-based alloys	$\frac{\omega}{K_B T}$	$\frac{1}{K_B}\frac{\partial \omega}{\partial T}$
Cd-Zn	1.2	- 0.058
Cd-In	0.6	- 0.262

Where values of ω/K_BT have been determined from equations (9) and (11) by using experimental value of excess free energy of mixing and free energy of mixing and values of $1/K_B \ \partial\omega/\partial T$ have been determined with successive approximation from equation (17) by using the experimental values [5] of H_M.

The positive value of energy parameter ω/K_BT suggests that there is a higher tendency for like atoms to pair in the alloy which implies a homo-coordination system. However, the tendency of pairing is weak since the energy parameter is small. We have observed that if energy parameters are
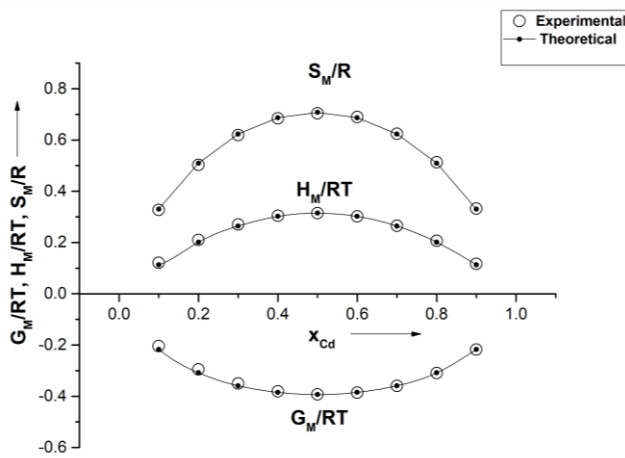
supposed to be independent of temperature ($\partial\omega/\partial T = 0$), then S_M and H_M so obtained are in poor agreement with experimental data [5]. This suggests the importance of temperature dependence of ordering energy, ω.

The free energy of mixing, heat of mixing and entropy of mixing for Cd-based liquid alloys at 800 K have been computed from Equation (10), Equation (17) and Equation (19) respectively. The plot of $\frac{G_M}{RT}, \frac{H_M}{RT}$ and $\frac{S_M}{R}$ against x_{Cd} for Cd-Zn and Cd-In alloys system are depicted in Figure (1.1) and Figure (1.2) respectively.
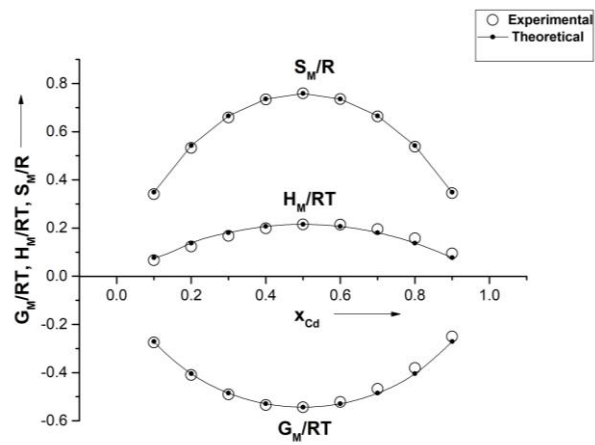

Fig 1.1: $\frac{G_M}{RT}, \frac{H_M}{RT}$ and $\frac{S_M}{R}$ versus x_{Cd} for Cd-Zn alloy at 800 K

Fig 1.2: $\frac{G_M}{RT}, \frac{H_M}{RT}$ and $\frac{S_M}{R}$ versus x_{Cd} for Cd-In alloy at 800 K

The calculated and experimental values [5] of $\frac{G_M}{RT}, \frac{H_M}{RT}$ and $\frac{S_M}{R}$ are in good agreement at all compositions of Cd. The qualitative agreement between the calculated and experimental values is a confirmation that our choice of ω for the alloy is correct. The value of $\frac{G_M}{RT}$ is minimum but $\frac{H_M}{RT}$ and $\frac{S_M}{R}$ are maximum at $x_{Cd} = 0.5$, which shows that both Cd-based liquid alloys are symmetric about equiatomic concentration. The negative small values of free energy of mixing throughout the entire compositions indicate that both Cd-based alloys at 800 K in the molten state are weakly interacting systems. The most negative value of ω/K_BT in alloy Cd-In is -0.5431 which is nearer to -1, and in alloys, Cd-Zn is -0.3931 which is nearer to 0 at concentration 0.5. Since the system that has the more negative value of ω/K_BT tends to have a strongly interacting

system, it indicates that Cd-Zn is a more weakly interacting system than Cd-In.

We have used the same values of the energy parameters in Equation (14) and Equation (15) for the evaluation of chemical activities of the components of the alloy. There is good agreement between calculated and experimental values of the activities of the components of both cadmium based alloys which are depicted in Figure (2.1) and Figure (2.2). It has been found that the magnitude of activity of more interacting systems is smaller in comparison to the activity of less interacting systems. From the point of view of activity at the same equi-atomic concentration, Cd-In has less value (i.e., 0.581) than that of Cd-Zn (i.e., 0.675). So, Cd-Zn is a more weakly interacting alloys system than Cd-In. This conclusion agrees with the conclusion obtained based on the free energy of mixing.
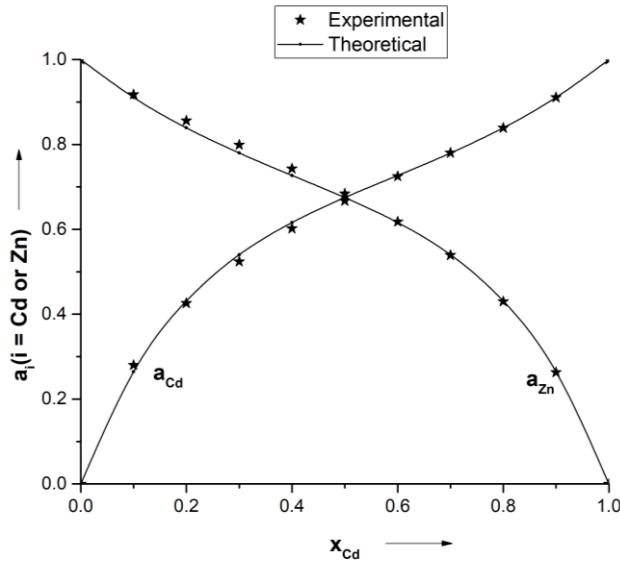
Fig 2.1: a_{Cd} and a_{Zn} versus x_{Cd} for Cd-Zn alloys at 800 K
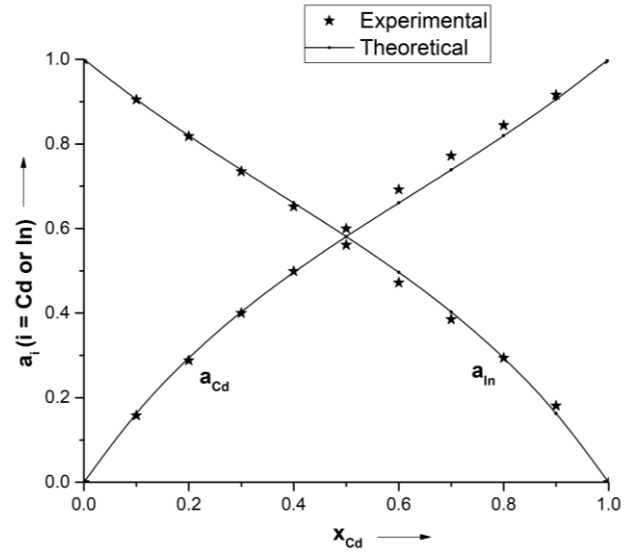

Fig 2.2: a_{Cd} and a_{In} versus x_{Cd} for Cd-In alloys at 800 K

3.2. Structural properties

We have used Equation (21) and Equation (23) to compute $S_{CC}(0)$ and α_1 respectively for both Cd-based alloys. The plot of theoretical, experimental, and ideal values of $S_{CC}(0)$ and computed values of α_1 for Cd-based alloys are shown in Figure (3.1) and Figure (3.2) respectively.
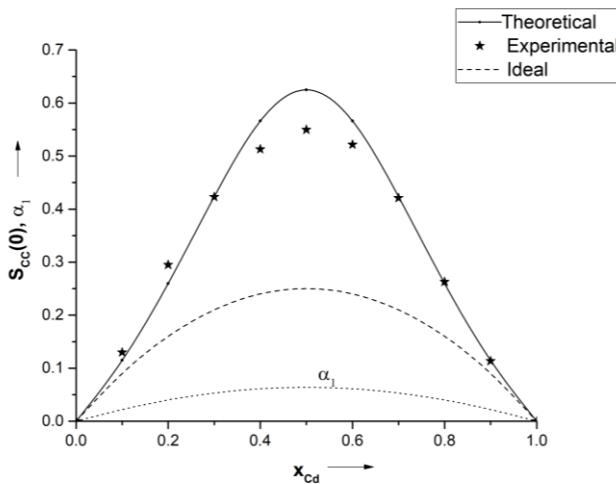

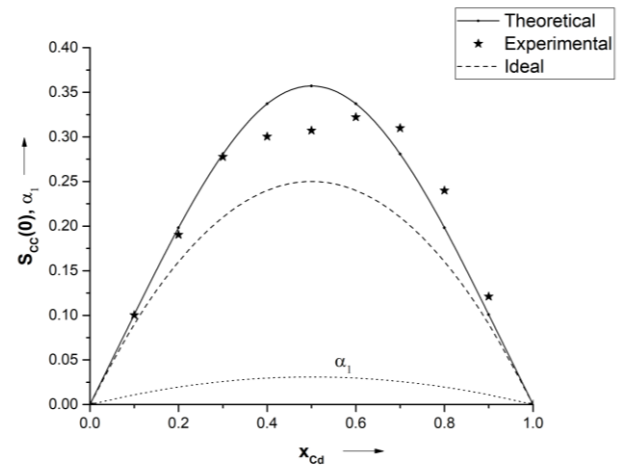**Fig 3.1:** $S_{CC}(0)$ and α_1 versus x_{Cd} for Cd-Zn alloy at 800 K

Fig 3.2: $S_{CC}(0)$ and α_1 versus x_{Cd} for Cd-In alloy at 800 K

The computed values of $S_{CC}(0)$ are in good agreement with the experimental values. Figure (3.1) and Figure (3.2) show that value of $S_{CC}(0)$ for both Cd-based alloys is greater than the ideal value at every concentration (i. e., $S_{cc}(0) > S_{cc}^{id}(0)$). The figures also show that α_1 is positive but less than 1 throughout the concentration of cadmium (i. e., $1 > \alpha_1 > 0$) and is found to be maximum at $x_{Cd} = 0.5$. This result suggests that both Cd-based alloys are

the homo-coordinating system. Between the alloys considered, Cd-Zn is closest to the ideal behavior whereas Cd-In exhibit the largest deviation which could be seen while the computed $S_{CC}(0)$ and $S_{cc}^{id}(0)$ values of both alloys are plotted against the concentration of cadmium in the same graph as shown in Figure (4.1) below. And also between the alloys considered, Cd-Zn has a greater peak of α_1 than Cd-In which could be seen while the computed

values of α_1 of both alloys are plotted against the concentration of cadmium in the same graph as shown in Figure (4.2) below. Concerning
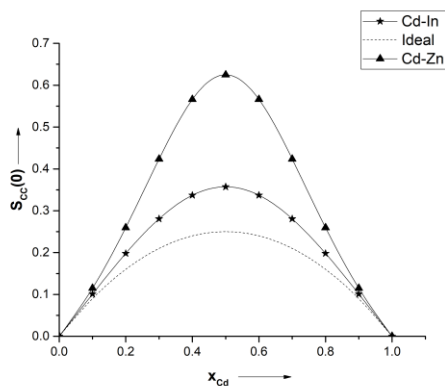
magnitude of deviation of $S_{CC}(0)$ from $S_{CC}^{id}(0)$ and the height of the peak of α_1, it has resulted that Cd-In is more segregation in nature than Cd-In.
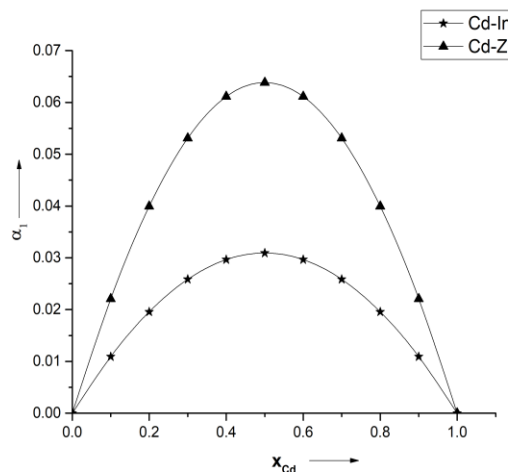

Fig 4.1: $S_{CC}(0)$ versus x_{Cd} for Cd-In and Cd-Zn alloys at 800 K

Fig 4.2: α_1 versus x_{Cd} for Cd-In and Cd-Zn alloys at 800 K

4. CONCLUSIONS

The analysis of the present research work reveals that:

- Ordering energy (ω) is positive and temperature-dependent. The value of ω for Cd-Zn is greater than that of Cd-In.
- Thermodynamic functions and structural functions are strongly dependent on ordering energy (ω).
- Alloys are weakly interacting in nature. Cd-Zn is more weakly interacting in nature than Cd-In.
- Alloys are segregating in nature. Cd-Zn is more segregating in nature than Cd-In.

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