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Surface properties of the molten Al-Mg alloys

I.Koirala[1,2]*, I.S.Jha[3], B.P.Singh[1]

[1]University Department of Physics, T.M. Bhagalpur University, Bhagalpur, (INDIA)
[2]Central Department of Physics, Tribhuvan University, Kirtipur, (NEPAL)
[3]M.M.A.M. Campus, Biratnagar, Tribhuvan University, (NEPAL)
E-mail: ishwar_koirala@yahoo.com

Abstract : Simple statistical model has been used to report the thermodynamic, structural, transport and surface properties of liquid Al-Mg alloys at 1073K. In thermodynamic properties we have studied the free energy of mixing and the chemical activity of both components of the alloys. For the structural investigation, our study includes concentration fluctuation in long wave length limit and Warren-Cowley short range order parameter. Viscosity and diffusivity of the alloys have been studied to understand transport properties. The surface behavior has been analyzed by computing surface concentration and surface tension of the alloys. The structural behaviors of the alloys are found to be symmetric with respect to concentrations of the species. The computed results are in good agreement with experimental data. Positive deviation of viscosity isotherms from linear law is observed. Surface tension of the alloys is found to be smaller than ideal values throughout bulk concentration of Al. For all theoretical analysis, concentration independent energy parameter takes important role, which is found to be temperature dependent.

Keywords : Simple statistical model; Linear law; Hetero-coordination; Surface tension.

INTRODUCTION

Mg-based alloys as lightest structural metallic materials have been receiving considerable interest due to their good performances. Mg-based alloys with different additives such as Al, Ga, Pb, Hg and Tl have been widely used as anode materials due to their rapid activation, low density, high current capacity and low electrode potential[1]. The application of Al-Mg alloys has been extended in aerospace and automotive industries because of their light weight, high specific strength, high electrical and thermal conductivity, outstanding corrosion resistance, weldability and mechanical properties[2,3]. Thus knowledge of mixing properties of Al-Mg alloys is necessary for design and development of reliable materials. The liquidus line of Al-Mg alloys reveals that the constituent atoms form complexes at one or more stoichiometric composition[4]. For the material preparation of metal alloys, the understandings of mixing properties of liquid alloys are important. Several theoretical models[5-10] 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 Al-Mg at 1073K by the simple statistical model[11-13]. 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 Al-Mg investigated is characterized by negative interaction energy, indicating the formation of two structures, as shown by their phase diagrams[4]. This characteristic behavior is likely to be a reflection of the interplay of the energetic and structural re-adjustment of the constituent elemental atoms. However, this is discussed in term of metallurgical and chemical constructs, such as electro-negativity difference (=0.3) and size mismatch (=1.4) for Al-Mg alloy, which indicates the values that are characteristics for ordering alloy[14].

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

THEORETICAL FORMALISM

Thermodynamic properties

The grand partition function for simple binary liquid alloys AB which consists of N_A (=N x) number of A atoms and N_B (=N (1-x)) number of B atoms so that total number atoms N, becomes equal to N_A+N_B, may be expressed as[11]

$$\Xi = [\phi_A q_A(T)]^{N_A}\ [\phi_B q_B(T)]^{N_B} \exp(-\beta E), \beta = 1/k_B T \quad (1)$$

where $q_i^{N_i}$ are the partition functions of atoms (i =A or B) associated with inner and vibrational degree of freedom. q_i the same whether the atom i is located in the pure state or in alloy. ϕ_i (i = A or B) is given as

$$\phi_i = \exp(\mu_i / k_B T), \quad (2)$$

where μ_i, k_B and T refer to chemical potential of ith component, Boltzmann constant and absolute temperature, respectively. E is the configurational energy of the alloys. One can easily obtain a relation between excess free energy of mixing, G_M^{XS} and the ratio of activity coefficients, γ (= γ_A / γ_B) by using standard thermodynamic relations.

$$G_M^{XS} = RT\int_0^x \ln\gamma\, dx = RT[x\ln\gamma_A + (1-x)\ln\gamma_B] \quad (3)$$

with $\gamma_A = [(\beta-1+2x)/x(1+\beta)]^{z/2}$ (3a)

$$\gamma_B = [(\beta+1-2x)/(1-x)(1+\beta)]^{z/2} \quad (3b)$$

$$\beta = \{1+4x(1-x)[\exp(2\omega/zk_B T)-1]\}^{1/2} \quad (3c)$$

Where R is universal molar constant; x,concentration of the component; ω,interchange or ordering energy and z, coordination number. For the equiatomic composition ($x=\frac{1}{2}$), relation (3) reduces to

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

The standard thermodynamic relation for free energy of mixing (G_M) is

$$G_M = RT[x\ln x + (1-x)\ln(1-x)] + G_M^{xs} \quad (5)$$

Using the relations (3) and (5), one obtains an expression for the free energy of mixing[15] as

$$\begin{aligned} G_M &= RT[x\ln x + (1-x)\ln(1-x) + x\ln\gamma_A + (1-x)\ln\gamma_B] \\ &= RT[x\ln x + (1-x)\ln(1-x) + x(1-x)\frac{\omega}{k_B T}] \end{aligned} \quad (6)$$

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

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

Then we have[15] from Eq.(7),

$$\ln a_A = \ln x + (1-x)^2 \frac{\omega}{k_B T} \quad (7a)$$

$$\ln a_B = \ln(1-x) + x^2 \frac{\omega}{k_B T} \quad (7b)$$

Structural properties

The theoretical determination of concentration-concentration fluctuations in the long-wavelength limit ($S_{cc}(0)$) is of great importance when nature of interactions in the melt has to be analyzed. $S_{cc}(0)$ is an essential structural function which has been widely used to study the nature of atomic order in binary liquid alloys[16,17]. It is thermodynamically related to free energy of mixing (G_M)[16,17] through the equation.

$$S_{cc}(0) = RT\left(\frac{\partial^2 G_M}{\partial x^2}\right)^{-1}_{T,P,N} \tag{8}$$

Equations (6) and (8) gives the theoretical value of $S_{cc}(0)$:

$$S_{cc}(0) = x(1-x)[1+z(1-\beta)/2\beta]^{-1} \tag{9}$$

For the ideal mixing, when the ordering energy is zero, concentration-concentration fluctuations are usually computed from:

$$S^{id}_{cc}(0) = x(1-x) \tag{10}$$

The Warren-Cowley[18,19] short-range order parameter (α_1) is another important structural function which is useful to quantify the degree of chemical order in the alloy at 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[18,19]:

$$\alpha_1 = (S-1)/\{S(z-1)+1\}, \quad S = S_{cc}(0)/S^{id}_{cc}(0) \tag{11}$$

where z is the coordination number, which is taken as 10 for our purposes.

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 isotherm (η) of liquid alloys is an important transport parameter for 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[20] is

$$\eta = \eta_{id}\left[1 - x(1-x)\left(\frac{2\omega}{k_B T}\right)\right] \tag{12}$$

with $\eta_{id} = x\eta^0_A + (1-x)\eta^0_B$ **(13)**

where η^0_i is the viscosity of pure component i (= A or B) and ω as usual, 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 (a_i) and self- diffusion coefficient (D_{id}) of pure component with the help of Darken's equation[21]

$$D_M = D_{id} x_i \frac{d \ln a_i}{dx_i} \tag{14}$$

We obtain an expression for in terms of $S_{cc}(0)$[14]:

$$\frac{D_M}{D_{id}} = \frac{S^{id}_{cc}(0)}{S_{cc}(0)} \tag{15}$$

With $D_M = xD_B + (1-x)D_A$ **(15a)**

where D_A and D_B are the self-diffusion coefficients of pure components A and B respectively. According to Eq. (14), is typical compound forming alloys implying D_M Ã D_{id}. A peak on diffusivity curve, D_M / D_{id} verses x_{Al}, 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[14]. 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

$$D_M = D_{id}\left[1 - x(1-x)\left(\frac{2\omega}{k_B T}\right)\right] \tag{16}$$

Surface properties

The knowledge of surface phenomena i.e. surface segregation and surface tension of the alloys can help to explain the technological important various properties such as mechanical behavior, kinetics of phase transformation, catalytic activity of alloy catalyst and thin film etc. Surface segregation is the enrichment of the surface by the atoms of the particular component element in a binary alloy which results from the interaction among the atoms and surface tension[7,8]. 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[7, 8]. 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[7, 8]

$$\Xi^S = \exp\left(\frac{-S\tau}{k_B T}\right) = \exp\left(\frac{-N^S \tau \xi}{k_B T}\right) \qquad \textbf{(17)}$$

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 modified in the simple form, using zeroth approximation as

$$\tau = \tau_A + \frac{k_B T}{\xi}\ln x^s - \frac{k_B T}{\xi}\ln x + \{p(1-x^s)^2 + (q-1)(1-x)^2\}\frac{\omega}{\xi} \qquad \textbf{(17a)}$$

$$\tau = \tau_B + \frac{k_B T}{\xi}\ln(1-x^s) - \frac{k_B T}{\xi}\ln(1-x) + \{p(x^s)^2 + (q-1)x^2\}\frac{\omega}{\xi} \qquad \textbf{(17b)}$$

where and 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[7-10].

$$\mathbf{P + 2q = 1} \qquad \textbf{(18)}$$

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[8]).

The pair of Eqs.17(a) and 17(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

$$\xi = \sum c_i \xi_i \qquad (i = A, B) \qquad \textbf{(19)}$$

Where the atomic area of hypothetical surface for each component is given as[7]:

$$\xi_i = 1.102\left(\Omega_i / N_0\right)^{2/3} \qquad \textbf{(20)}$$

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

In order to compare the behavior of concentration- concentration fluctuations and Warren-Cowley short rang order parameter in the bulk and at the surface of the alloys, we need an expression for determination of concentration- concentration fluctuations and warren-Cowley short rang order parameter at the surface. On the basis of earlier works of Singh et al. in ref[5] for the bulk phase, an expression for concentration- concentration fluctuations at the surface obtained via solving the surface grand partition function is given by[5,11]

$$S^S_{cc}(0) = x^S(1-x^S)[1 + z^S(1-\beta^S)/2\beta^S]^{-1} \qquad \textbf{(21)}$$

with $\boldsymbol{\beta^s = \{1 + 4x^s(1-x^s)\left[\exp(2\omega/z^s k_B T) - 1\right]\}^{1/2}}$ **(22)**

Here z^s is the coordination number of surface atoms which is obtained from $z^s = (p+q)\,z$ and z is the coordination number in bulk. In the case of ideal mixing (i.e. ω = 0), Eq.(21) reduces to

$$S^S_{cc}(0, \text{ideal}) = x^S(1-x^S) \qquad \textbf{(23)}$$

The expression for the surface Warren-Cowley short range order parameter, deduced from Eq.(11) is given by

$$\alpha_1^S = (S^S - 1)/\{S^S\,(z^S - 1) + 1\}, \qquad S^S = S^S_{cc}(0)/x^S(1-x^S) \qquad \textbf{(24)}$$

RESULT AND DISCUSSION

The energy parameter used for the calculation for Al-Mg liquid alloys at 1073K has been determined from equation (4) by using experimental value of G_M^{xs} for equiatomic composition[4].The best fitting value of parameter is found as

$$\frac{\omega}{k_B T} = -1.12 \qquad \textbf{(25)}$$

The negative value of energy parameter ($\frac{\omega}{k_B T}$) 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.

The free energy of mixing for Al-Mg liquid alloys at 1073K has been computed from equation (6). The plot of $\frac{G_M}{RT}$ versus x_{Al} are depicted in Figure 1. The calculated and experimental values of $\frac{G_M}{RT}$ are in good agreement throughout whole concentrations of aluminium. 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 $\frac{G_M}{RT}$ (= -0.97332) is minimum at x_{Al} =0.5, which show that Al-Mg liquid alloy is symmetric about equiatomic concentration. The negative small values of free energy of mixing throughout the entire compositions indicate that the Al-Mg alloy at 1073K in molten state is weakly interacting system. The activity is one of the important thermodynamic functions which is obtained directly from experiment. The deviation from ideal behavior is incorporated into activity. We have used the same value of the energy parameter in Eq. (7a) and (7b) 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 Al and Mg of the alloy (Figure 1).

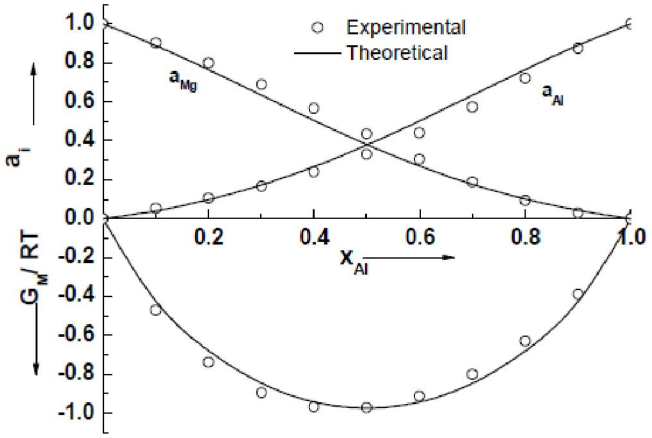

Figure 1 : Free energy of mixing (G_M) and Chemical activity (a_i) Vs concentration of aluminium($x_{Al)}$ in liquid Al-Mg alloy at 1073K

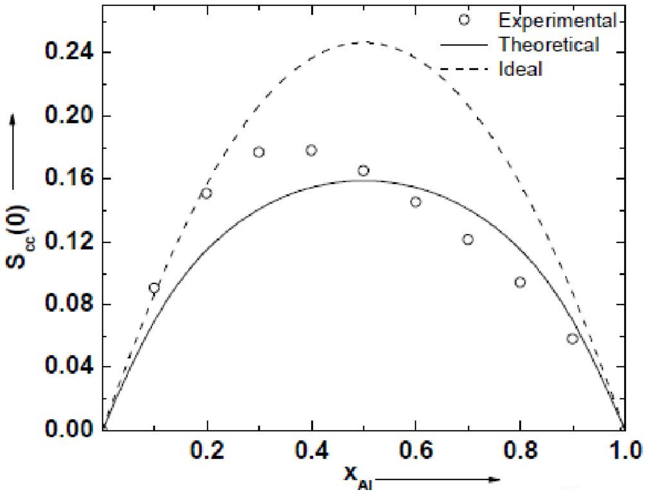

Figure 2 : Concentration- concentration fluctuation at long wavelength limit (S_{cc}(0)) Vs concentration of aluminium($x_{Al)}$ in liquid Al-Mg alloy at 1073K

The mixing behavior of liquid alloys can be deduced from the deviation of concentration- concentration fluctuations at long wavelength limit (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.

We have used Eq.(9) to compute the S_{cc}(0) for Al-Mg. It can also be obtained directly from the measured activity[4] data as

$$S_{cc}(0) = (1-x)a_{Al}\left(\frac{\partial a_{Al}}{\partial x}\right)^{-1} = xa_{Mg}\left(\frac{\partial a_{Mg}}{\partial(1-x)}\right)^{-1} \qquad (26)$$

where a_{Al} and a_{Mg} are the observed chemical activities of aluminium and magnesium respectively. The S_{cc}(0), obtained from Eq.(26) are taken as experimental values. Figure 2 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) in the region 0.5 x_{Mg} 0.1. In the region 0.4 x_{Mg} 0.9, 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, clearly described this system as compound forming.

Figure 3 shows the plots of Warren-Cowely chemical short range order parameter (α_1) against chemical composition of aluminium, obtained from

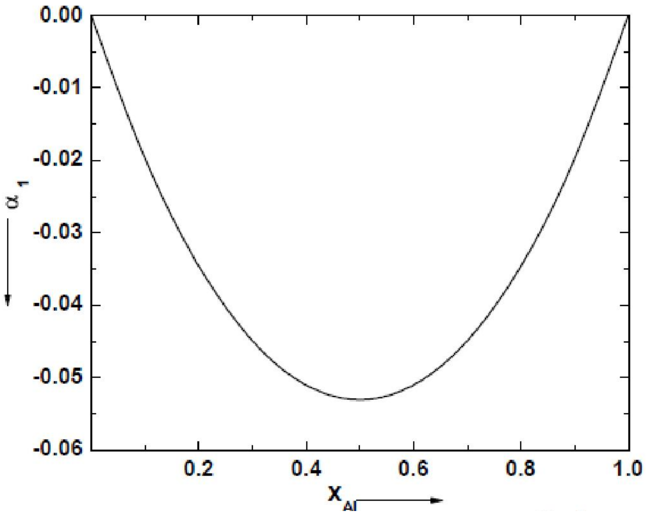

Figure 3 : Warren-Cowely short range order parameter (α_1) Vs concentration of aluminium($x_{Al)}$ in liquid Al-Mg alloy at 1073K

Eq.(11) for Al-Mg at 1073K. For the equiatomic composition, α_1 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 $\alpha_1$indicate segregation, leading to complete only if $\alpha_1 = 1$. But $\alpha_1 = 0$, corresponds to random distribution of the atoms in the mixture. It is observed that the plots are symmetrical about equiatomic composition and negative throughout the whole concentration range of aluminium. The negative values of α_1 (minimum at $x_{Al} = 0.5$) throughout whole concentration range are the signatures of hetero-coordination system in the Al-Mg liquid alloys at 1073K.

The viscosity of the Al-Mg liquid alloy has been computed numerically from Eq. (12). From the plot of η verses bulk concentration of x_{Al} (Figure 4) in Al-Mg liquid alloy, positive deviation from the linear law (Raoult's law) in viscosity isotherms η(c) have been observed for the regular alloy.

The calculated values of $S_{cc}(0)$ are used in Eq. (15) to evaluate the ratio of the mutual and intrinsic-diffusion coefficients, $\frac{D_M}{D_{id}}$. For the consistency of the estimated order energy parameter, ω, we have also calculated using Eq.(15). Figure 5 shows plotof $\frac{D_M}{D_{id}}$ against the concentration of aluminium. In that plot the value of $\frac{D_M}{D_{id}}$ is found greater than 1 in the entire range ofconcentration which is indicative for the compound formation in the mixture. A maximum value of $\frac{D_M}{D_{id}} = 1.56$ for $x_{Al} = 0.5$, concentration which is indicative for the compound formation in the mixture. A maximum value of $\frac{D_M}{D_{id}} = 1.56$ for $x_{Al} = 0.5$, confirms a weak tendency for chemical ordering, as observed by the $S_{cc}(0)$ and CSRO parameter.

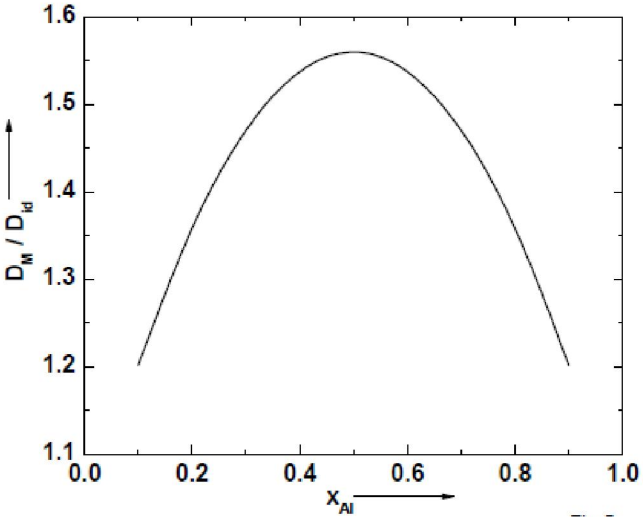

Figure 4 : Viscosity (η) Vs concentration of aluminium($x_{Al)}$ in liquid Al-Mg alloy at 1073K

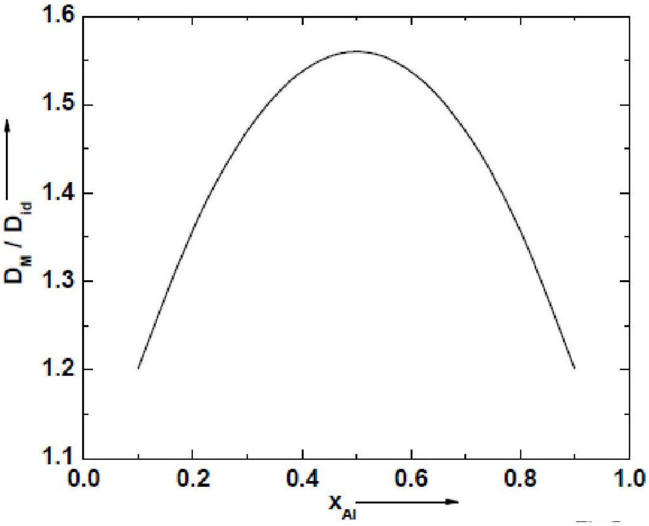

Figure 5 : Ratio of mutual and intrinsic diffusion coefficients (D_M /D_{id}) Vs concentration of aluminium($x_{Al)}$ in liquid Al-Mg alloy at 1073K

The surface concentrations and surface tension of Al-Mg have been computed numerically from the Eqs.17(a) and 17(b). For this, we need experimental data of density and surface tension of the components at the working temperature. The density and surface tension for the components Al and Mg[22,23] at the working temperature T =1073K have been calculated by using the following equations

$$\rho_{Al}(T) = 2.64625 - 2.8\times10^{-4}T \tag{27a}$$

$$\rho_{Mg}(T) = 1.8346 - 2.65\times10^{-4}T \tag{27b}$$

$$\tau_{Al}(T) = 1.24155 - 3.5\times10^{-4}T \tag{28a}$$

$$\tau_{Mg}(T) = 0.8824 - 3.5\times10^{-4}T \tag{28b}$$

The mean atomic surface area ξ has been calculated by using Eqs.(19) and (20). For calculating surface tension we used same estimated energy parameter, ω. At first, surface concentrations of aluminium in alloy Al-Mg have been obtained as a function of bulk concentration by concurrently solving the equations 17(a) and 17(b). Using the obtained values of surface concentrations we computed the surface tension of Al-Mg liquid alloys at temperature 1073K for whole concentration range.

The study of surface concentration as shown in Figure 6 shows the usual pattern for surface concentration increasing with increasing bulk compositions. This indicates that Mg-atoms (having lower value of surface tension relative to Al-atoms in Al-Mg) segregate at the surface of Al-Mg liquid alloys in preference to Al-atoms throughout the entire compositions. The computed values of surface tension for molten Al-Mg alloys at 1073K from modified prasad's model, mentioned above are depicted in Figure 7. 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[22].

It is noticed that computed values of τ and x^S_{Al} are found to be smaller than the ideal values throughout bulk concentration of aluminium. The absence of irregularities in the form of minimum, maximum or inflection point in the isothermal plots of the surface tension of the alloys Al-Mg is a signature of the absence of stoichiometric intermetallic compound

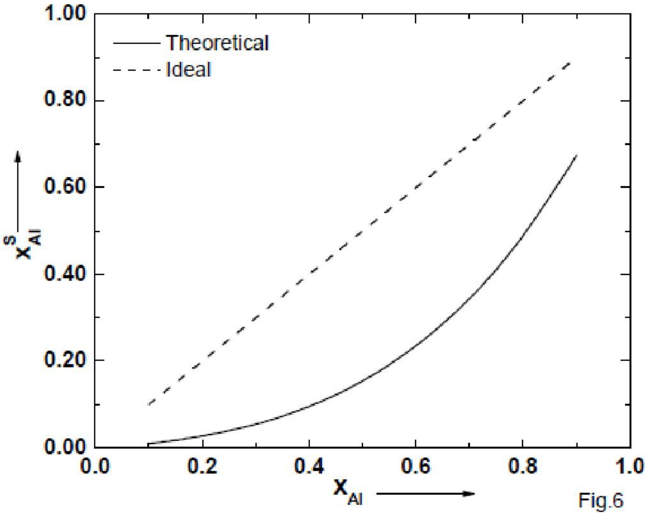

Figure 6 : Surface concentration of aluminium (x^S_{Al}) Vs bulk concentration of aluminium($x_{Al)}$ in liquid Al-Mg alloy at 1073K

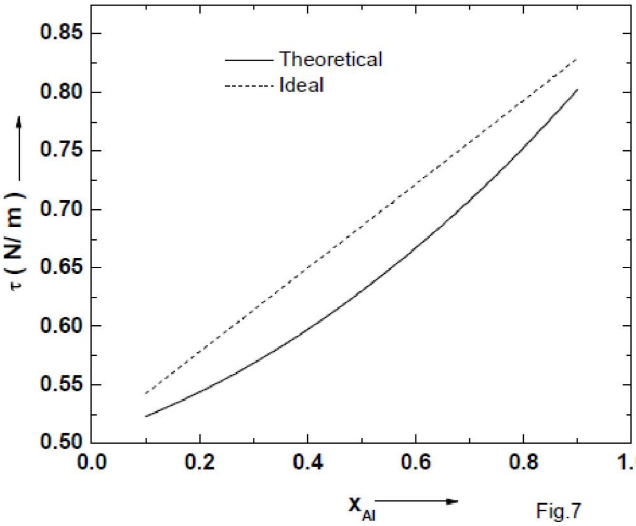

Figure 7 : Surface tension (τ) Vs bulk concentration of aluminium($x_{Al)}$ in liquid Al-Mg alloy at 1073K

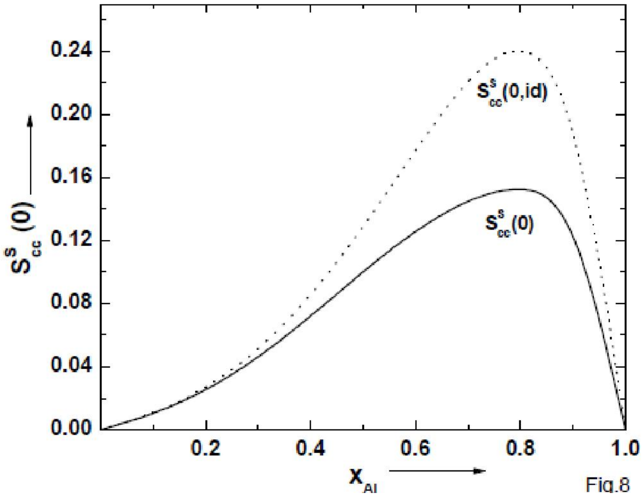

Figure 8 : Surface concentration- concentration fluctuations at long wavelength limit ($S^S_{cc}(0)$)Vs bulk concentration of aluminium($x_{Al)}$ in liquid Al-Mg alloy at 1073K

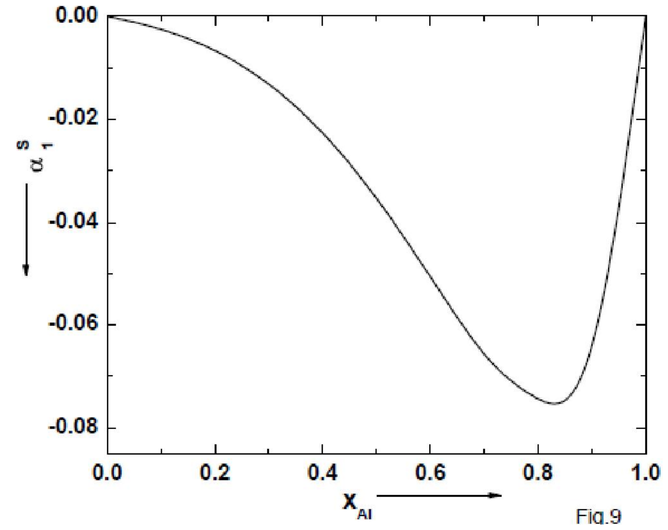

Figure 9 : Surface Warren-Cowely short range order parameter (α_1^s) Vs bulk concentration of aluminium(x_{Al}) in liquid Al-Mg alloy at 1073K

formation in the system. However, going by the phase diagram in ref.[4] intermetallic compound Al_3Mg_2 exist in the solid phase of Al-Mg. To predict the local order on the surface of the liquid alloys, the surface concentration- concentration fluctuations, $S_{cc}^s(0)$ and surface warren-Cowley short range order parameter, α_1^s was computed through eq.(21) and (24) and plotted against the bulk concentrations of aluminium of the alloys in Figures 8 and 9.

The Figure 8 shows that and for the alloys are skewed towards the lower concentrations of magnesium. However just as in Figure 1, Figure 9 shows $S_{cc}^s(0) < S_{cc}^s(0,id)$ throughout the entire composition of aluminium in Al-Mg which indicates the presence of chemical order both in bulk and on the surface of the liquid alloys. In Figure 9, is skewed towards the lower concentrations of magnesium and just like bulk α_1, surface warren-Cowely short range order parameter is negative throughout all concentrations of aluminium.

CONCLUSIONS

The analysis reveals that there is a tendency of unlike atom pairing or hetero-coordination (Al-Mg, Al-Mg) in liquid Al-Mg alloys at all concentrations. The theoretical analysis suggests that this alloy is of a weakly interacting nature. In case of viscosity isotherms, positive deviation from ideality is observed but surface tension isotherms of Al-Mg alloys are deviated negatively from ideal values. Theoretical study proofs that metal with lower surface tension tends to segregate on the surface of molten alloys. The smaller Al-atoms segregate in the bulk but bigger Mg-atoms segregate at the surface of liquid Al-Mg alloys.

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