# Concentration Fluctuations and Interfacial Adhesion at the Solid-Liquid Interface Between $Al_2O_3$ and Al-Sn Liquid Alloys

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## ABSTRACT

A theoretical investigation of the energetics of the bulk as well as the surface of Al-Sn liquid alloys has been made through the study of the concentration dependence of various properties such as concentration fluctuations in the long wavelength limit [$S_{CC}(0)$], CSRO parameter ($\alpha_1$), diffusion ($D_M$), surface tension ($\gamma$) and surface composition ($C_i^s$). Besides the use of statistical mechanical model for the surface tension and surface composition, we have also made an attempt to link the surface tension with the bulk through the sensitive $S_{CC}(0)$ and isothermal compressibility. Our computed results of the surface tension have been further used to compute the work of adhesion (W) and interfacial tension ($\gamma_{SL}$) between solid $Al_2O_3$ and liquid Al-Sn alloys over the whole concentration range. Our study suggests like atoms pairing as nearest neighbours at all concentrations. The maximum tendency for the segregation has been observed around $C_{Sn} = 0.35$. As regards surface tension, the results obtained from $S_{CC}(0)$ are quite different from those obtained from statistical mechanical model in the middle concentration range of Al-Sn alloys. However, both approaches present almost the same trend, i.e. a decrease in $\gamma$ with the addition of Sn to Al, in the experimental concentration range (3.8 % Sn). The surface of the Al-Sn system has been found to be enriched with Sn atoms at all bulk compositions. It is also observed that W decreases with the addition of Sn to Al and increases with the addition of Al to Sn. So far as $\gamma_{SL}$ is concerned, it remains unaffected up to 80 % Sn and shoots up sharply after that – being one of the reasons for the increase of W in the Sn rich end.

**Keywords:**
Al-Sn liquid alloys, Surface tension, Work of adhesion, Interfacial tension, Concentration fluctuations, CSRO, Diffusion

## 1. INTRODUCTION

Wetting and adhesion between metallic melts and ceramic play an important role /1,2/ in the fabrication of metal matrix composites as well as in the joining of ceramics to metals. But the poor wetting /3/ of ceramics, particularly oxides, by liquid metals or alloys has led researchers to explore /4,5/ the factors responsible for better wetting and interfacial adhesion. It has been found that wetting depends not only upon the bonding characteristics of liquid alloys and ceramics but also upon the interactive forces at the interface. So one cannot understand wetting phenomena without having a clear picture of the bulk as well as the surface of the liquid alloys. During the last few years, more attention has been paid to understanding the wetting behaviour of $Al_2O_3$ by aluminum based alloys through the investigation /6,7/ of concentration dependence of the angle of contact. The simple understanding of the force equilibrium (due to surface tension ($\gamma_{LV}$) of the liquid, interfacial tensions ($\gamma_{SL}$ and $\gamma_{SV}$) between solid-liquid and

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solid-vapour) at the interface is not sufficient. One also needs to have knowledge of the energetics of the bulk and its effect on the structural arrangement and the surface properties. That is why a comprehensive study of the bulk as well as surface of liquid alloys is essential for this purpose. Keeping this in mind we have made an attempt to investigate the bulk as well as surface properties of Al-Sn liquid alloys and extend the results to analyse the interfacial adhesion between Al-Sn and $Al_2O_3$ through the study of the concentration dependence of the work of adhesion between solid $Al_2O_3$ and liquid alloys. Recently /9,10/ it has been found that Al, when mixed with the eutectic mixture of Sn-Zn, improves the corrosion and oxidation resistance of Sn-Zn alloys and Al-Sn-Zn can be used as a suitable substitute for the conventional $Sn_{0.6}Pb_{0.4}$ solder alloys. It will be proper to mention that because of health hazards and environmental impacts of Pb, attempts are being made to develop /9-14/ lead free solders through the investigation of various binary and ternary systems. So the investigation of surface properties is important to understand the wetting behaviour of Al-Sn-Zn alloys. In recent years concentration fluctuations in the long wavelength limit /15/ [$S_{CC}(0)$] and chemical short range order (CSRO) parameter ($\alpha_1$) /16,17/ have emerged /18-21/ as powerful microscopic functions to understand the phenomena of compound formation and phase segregation in the bulk liquid. $S_{CC}(0)$ is being utilized to visualize the nature of chemical order and segregation whereas $\alpha_1$ helps in quantifying the degree of order and segregation. $S_{CC}(0)$ has not only been utilized to understand the energetics of the bulk but also for the diffusion properties /21/ of Al-Sn liquid alloys. As regards surface segregation in Al-Sn liquid alloys, we have used statistical mechanical theory based on a grand partition function approach /22,24/. In addition to this we have also tried to correlate the surface tension of Al-Sn liquid alloys with the bulk through $S_{CC}(0)$ and isothermal compressibility ($\chi_T$).

## 2. CONCENTRATION FLUCTUATIONS IN THE LONG WAVELENGTH LIMIT [$S_{CC}(0)$]

The number-concentration partial structural factors ($S_{CC}(q)$, $S_{NN}(q)$ and $S_{NC}(q)$), for the binary alloys, A-B, are related /15/ to the intensity, I(q), of the scattered radiation through the relation

$$I(q) = \langle b \rangle^2 S_{NN}(q) + 2 \langle b \rangle S_{NC}(q) + \Delta b^2 S_{CC}(q) \quad (1)$$

with

$$\langle b \rangle = C_A b_A + C_B b_B \quad \text{and} \quad \Delta b = b_A - b_B$$

where $C_i$ and $b_i$ are concentration and coherent scattering amplitude of i-th (A,B) component atom of the alloy respectively. $q = 4\pi \sin\theta / \lambda$; $2\theta$ is the scattering angle and $\lambda$ is the wavelength of the radiation.

These structure factors become more relevant in their long wavelength limit. For $q \rightarrow 0$ limits, the number-concentration structure factors have simple physical meaning for liquid mixtures.

$$S_{NN}(0) = \langle (\Delta N)^2 \rangle / N \quad (2)$$

$$S_{CC}(0) = N \langle (\Delta C)^2 \rangle \quad (3)$$

$$S_{NC}(0) = \langle (\Delta N.\, \Delta C) \rangle \quad (4)$$

where $\langle(\Delta N)^2\rangle$ and $\langle(\Delta C)^2\rangle$ are mean square fluctuations in the number of particle and in the concentration respectively in a volume V. $\langle(\Delta N.\Delta C)\rangle$ describes the correlations between the two fluctuations. All the structure factors in the long wavelength limit are related to each other through isothermal compressibility ($\chi_T$) and dilatation factor, $\delta$ ($V^{-1}$. $(\partial V / \partial C)_{T,P,N}$). Of all structure factors, the most important, the concentration fluctuations in the long wavelength limit [$S_{CC}(0)$], is directly related to free energy of mixing ($G_M$) and activity through the relation

$$S_{CC}(0) = NK_BT\, (\partial^2 G_M / \partial C_A^2)_{T,P,N}^{-1} \quad (5)$$

$$= C_B\, a_A\, (\partial a_A / \partial C_A)_{T\,P\,N}^{-1} \quad (6a)$$

$$= C_A a_B\, (\partial a_B / \partial C_B)_{T,P,N}^{-1} \quad (6b)$$

where $a_A$ and $a_B$ are activities of A and B components respectively. $K_B$ and T refer to Boltzman's constant and absolute temperature respectively.

$S_{CC}(0)$ has presented itself as a most powerful microscopic function to understand the characteristic behaviour of binary liquid alloys. The nature of chemical order (unlike atoms pairing as nearest neighbours) and segregation (due to like atoms pairing as nearest neighbours) can be easily understood by $S_{CC}(0)$. Besides this, many other properties such as glass forming tendency, diffusion, viscosity, surface tension etc., could also be explained by it. The characteristic behaviour of an alloy is usually ascertained through its deviation from ideal values i.e. $S_{CC}(0,id) = C_A C_B$. If at a given composition $S_{CC}(0) > S_{CC}(0,id)$ then there is a tendency for segregation while $S_{CC}(0) < S_{CC}(0,id)$ indicates the presence of chemical order in the melt. On the basis of general characteristics of $S_{CC}(0)$ it is very simple to interpret the behaviour of other structure factors i.e. $S_{NN}(0)$ and $S_{NC}(0)$; shedding light on topological order.

So far as experimental determination of $S_{CC}(0)$ is concerned, it poses a much more difficult problem and has never been accomplished successfully. Owing to this, efforts have been made to compute $S_{CC}(0)$ theoretically. However, we have determined $S_{CC}(0)$ from measured activity data following two equalities of Eqn. (6). Owing to considerable errors faced during numerical differentiation, we have fitted the observed activity data to a high order polynomial such as

$$a_A = a_0 + a_1C + a_2C^2 + a_3C^3 + \text{--------} + a_{10}C^{10} \quad (7a)$$

$$a_B = b_0 + b_1C + b_2C^2 + b_3C^3 + \text{--------} + b_{10}C^{10} \quad (7b)$$

The observed data of activity at eleven different concentrations are used to obtain the coefficients, $a_i$ and $b_i$. Both equations provide $\partial a/\partial C$ of reasonable standard in the intermediate range of concentrations but problems are experienced in the dilute region limit. This is quite natural as one of the activities goes to zero in the dilute region concentrations. In order to solve the problem we have used the activity of the second component to compute $S_{CC}(0)$.

## 2.1 $S_{CC}(0)$ and chemical short range order parameter ($\alpha_1$)

The Warren-Cowley /16,17/ chemical short range order (CSRO) parameter ($\alpha_1$) is a useful parameter to quantify the degree of self coordination and hetero-coordination in the melt. $\alpha_1$, for alloys having components of same size can be expressed /25/ in terms of radial concentration - concentration function, $\rho_{cc}(r)$ as

$$\int_{r_1-\varepsilon}^{r_1+\varepsilon} 4\pi r^2 \rho_{cc}(r) = \alpha_1 Z \quad (8)$$

where $r_1$ is mean distance of first neighbour shell from a given atom. Actual distance is taken between $r_1 - \varepsilon$ and $r_1 + \varepsilon$; Z is the coordination number. As $\rho_{cc}(r)$ may be obtained from the Fourier Transform (FT) of concentration-concentration structure factor, $S_{CC}(q)$, the experimental determination of $\alpha_1$ is possible in principle. But it is very difficult in practice to determine $S_{CC}(q)$ for all kinds of binary liquid alloys. That is why experimental data on $\alpha_1$ are very scarce. However it is interesting to observe that $\alpha_1$ can be evaluated theoretically from the knowledge of $S_{CC}(0)$ through the relation /20,26/

$$\alpha_1 = S_1 (S_{CC}(0) Z - S_1)^{-1} \quad (9)$$

with

$$S_1 = S_{CC}(0) - S_{CC}(0,id)$$

This provides an insight into local arrangements of atoms in the melt. The tendency for like atoms pairing is indicated by $\alpha_1 > 0$ while $\alpha_1 < 0$ suggests the tendency for hetero-coordination. In the case of random distribution $\alpha_1$ becomes equal to zero. From a probabilistic approach one can show the limiting range of $\alpha_1$ as

$$\begin{aligned} -C_A C_B^{-1} &< \alpha_1 < 1 \quad \text{for } C_A \le 0.5 \\ -C_B C_A^{-1} &< \alpha_1 < 1 \quad \text{for } C_A > 0.5 \end{aligned} \quad (10)$$

For equi-atomic composition, one has

$$-1 \le \alpha_1 \le +1$$

($\alpha_1^{min.} = -1$) refers to complete ordering of A-B pairs in the melt whereas phase separation is indicated by ($\alpha_1^{max.} = +1$).

## 2.2 $S_{CC}(0)$ and diffusion

The inter-diffusion coefficient ($D_M$) of the binary system is related to activity through the relation

$$D_M = D_S\,(d\ln a_A / dC_A) \tag{11}$$

where $D_S = \sum C_i D_i$; $D_i$ is the self-diffusion coefficient of the i-th component.

One can easily express $D_M$ /21/ in terms of $S_{CC}(0)$ by using Eqns. (6) and (11)

$$D_M D_S^{-1} = S_{CC}(0,\mathrm{id})\, S_{CC}(0)^{-1} \tag{12}$$

It is obvious from the above equation that in the case of ideal mixing $D_M D_S^{-1} \rightarrow 1$. $D_M D_S^{-1} > 1$ indicates the presence of chemical order while like atoms pairing is represented by $D_M D_S^{-1} < 1$.

### 2.3 $S_{CC}(0)$ and surface tension of binary liquid alloys

One can easily obtain an expression for the surface tension ($\gamma$) in terms of fluctuations by extending the Cahn-Hillard phenomenological /27/ model for pure liquids to the binary mixture. It may be given as

$$\gamma = (2A/l\ \chi_T N^2)\ [\{\Delta N + N\,(\Delta C)\delta\}^2 + (\chi_T N^2/V)\,(\partial^2 G_M/\partial C^2)_{T,P,N}\,(\Delta C)^2] \tag{13}$$

where $l$ is the effective thickness of the interface, $\chi_T$ and $\delta$ are already defined. $\Delta C$ and $\Delta N$ refer to fluctuations in concentration and in number respectively. A is known as accidental fluctuation and taken as equal to ½. One obtains an expression for $\gamma$ of a binary alloy in terms of $S_{CC}(0)$ by minimizing $\gamma$ with respect to $\Delta C$ ($\partial\gamma/\partial\Delta C = 0$)and neglecting number fluctuations ($\Delta N \rightarrow N$) as

$$\gamma = (l/\chi_T)\,[1 + \{\delta^2 S_{CC}(0)/N K_B T \chi_T\}]^{-1} \tag{14}$$

$l$ can be determined by $d\gamma/dl = 0$. For pure liquids, it has been found /28/ that $\gamma_i \chi_T^{\,i} = l_i$. $\gamma_i$, $\chi_T^{\,i}$ and $l_i$ refer to pure component, i.

## 3. SURFACE TENSION AND GRAND PARTITION FUNCTION

The grand partition function for the surface of a binary system, A-B consisting of $N_A^s$ ($= C_A^s N^s$) number of A- atoms and $N_B^s$ ($= C_B^s N^s$) number of B-atoms may be constructed /21,22/ as

$$\Xi^s = \sum_{E_s} q_{As}(T)^{N_A^s}\, q_{Bs}(T)^{N_B^s} \exp\,[\,(\sum \mu_i^s N_i^s - E_s) / K_B T\,] \tag{15}$$

where $q_{is}$ and $\mu_i^s$ refer to internal partition function and chemical potential of i-th (A or B) atom at the surface respectively. $E_s$ is the surface configurational energy. All the corresponding notations without superscript or subscript 's' refer to the bulk. The importance of the above equation for $\Xi^s$ lies in the fact that it is directly related to the surface tension, $\gamma$, of the binary system through the relation, given below,

$$\Xi^s = \exp\,[-N^s \gamma\, \alpha / K_B T] \tag{16}$$

where $\alpha$ is the mean atomic surface area. In order to obtain an expression for $\gamma$ one needs to have the solution of equation (15), which is not a trivial problem. However, Prasad *et al.* /21,22/ solved it by defining $E_s$ in the framework of quasi-lattice theory and developed expressions for $\gamma$ for different types of alloys. $\gamma$ for simple alloys in the zeroth order approximation can be expressed as

$$\gamma = \gamma_A + (K_B T/\alpha)\ \ln\,(C_A^s/C_A) + [p\,(C_B^s)^2 - (p+q)\,C_B^2]\,(\omega/\alpha) \tag{17a}$$

$$= \gamma_B + (K_B T/\alpha)\ \ln\,(C_B^s/C_B) + [\,p\,(C_A^s)^2 - (p+q)\,C_A^2\,]\,(\omega/\alpha) \tag{17b}$$

where $\gamma_i$ is the surface tension of pure i-th component. $\omega$ is the order energy. p and q are termed as surface coordination fractions and take into account the contribution of various layers. For closed packed structure, p = ½ and q= ¼. The above simultaneous equations can be solved numerically to obtain $C_i^s$ and $\gamma$ as a function of concentration.

### 3.1 Surface tension and work of adhesion

When a liquid drop is in equilibrium state on a smooth horizontal solid surface then all the forces, acting on the point of contact at the interface, are related through the equation i.e.

$$\gamma_{SV} = \gamma_{LV} \cos\theta + \gamma_{SL} \tag{18}$$

where $\gamma_{SV}$ and $\gamma_{SL}$ are interfacial tensions between solid and vapour and solid and liquid phases respectively. $\gamma_{LV}$ (= $\gamma$) is the surface tension of the liquid and $\theta$ is the angle of contact. It is obvious that liquid will spread across the solid surface until the equilibrium contact angle $\theta$ is attained in accord with the above equation. The work of adhesion, W, is given by

$$W = \gamma_{SV} + \gamma_{LV} - \gamma_{SL} \tag{19}$$

Eqns. (18) and (19) give

$$W = \gamma_{LV}\,(1 + \cos\theta) \tag{20}$$

$$\gamma_{SL} = \gamma_{SV} - \gamma_{LV}\cos\theta \tag{21}$$

One can easily calculate W and $\gamma_{SL}$ if one has the value of $\theta$ and $\gamma_{SV}$. As regards $\gamma_{SV}$, it has been found /34/ to be constant. It is natural to say that that interfacial adhesion between solid and liquid depends upon surface tension of the liquid and contact angle; both being a function of concentration.

## 4. RESULTS AND DISCUSSION

### 4.1. Concentration fluctuations, CSRO and diffusion

The observed activity data /29/ at T= 973K was used in Eqn. (6) to compute $S_{CC}(0)$ of Al-Sn liquid alloys as a function of concentration. The computed values along with ideal values are presented in Fig. 1. It is clear from Fig. 1 that $S_{CC}(0)$ is greater than ideal values at all concentrations; indicating the tendency for like atoms pairing leading to segregation. The peak in $S_{CC}(0)$ vs $C_{Sn}$ around $C_{Sn}$= 0.35 is indicative of maximum segregation around this composition. However at both Sn and Al rich ends, the system behaves ideally. The computed values of $S_{CC}(0)$ have been used in Eqns. (9) and (12) to calculate CSRO ($\alpha_1$) and $D_M/D_S$. Figs. (2) and (3) depict the variation of $\alpha_1$ and $D_M/D_S$ with concentration respectively. For want of experimental values, we could not compare our results. $\alpha_1$ has been found to be positive at all compositions, which suggests the tendency for like atoms pairing. Likewise $S_{CC}(0)$, there is a peak around $C_{Sn}$ = 0.35 - suggesting a maximum tendency for segregation. However, the small

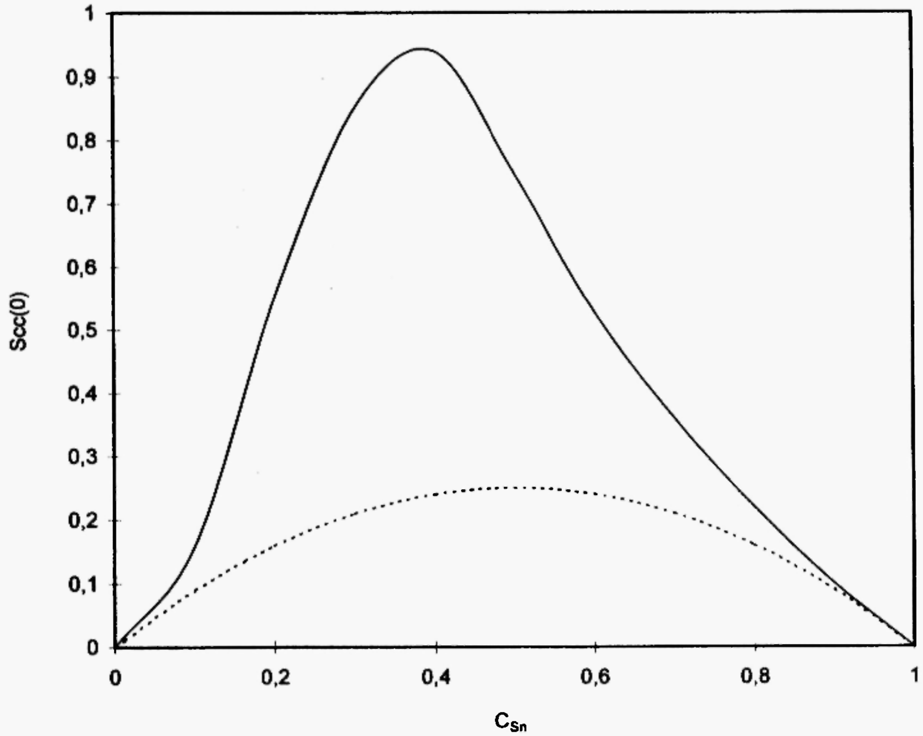

**Fig. 1:** Concentration fluctuations in the long wavelength limit, $S_{CC}(0)$, of Al-Sn system at T= 973K. (—) and (-----) refer to computed and ideal values respectively.

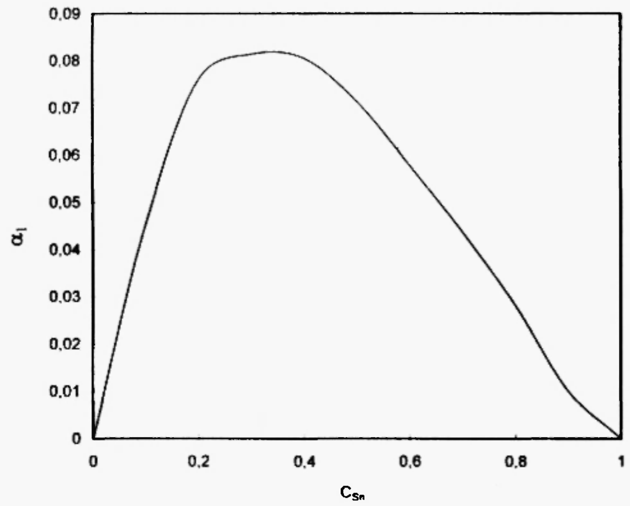

**Fig. 2:** CSRO ($\alpha_l$) vs $C_{Sn}$ of Al-Sn system at T = 973K.

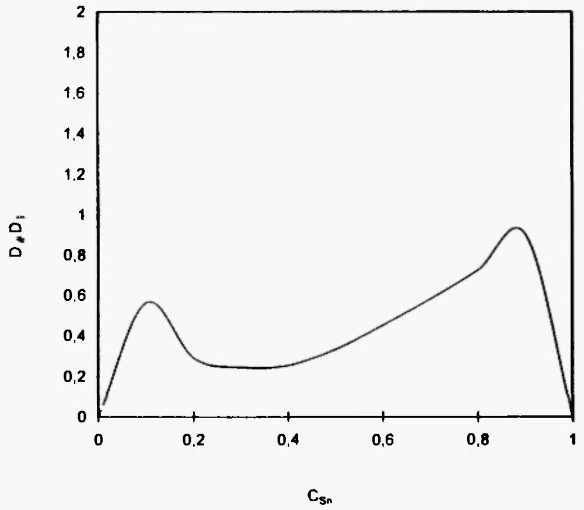

**Fig. 3:** $D_M/D_S$ vs $C_{Sn}$ of Al-Sn system at T = 973K.

value of $\alpha_l$ at all concentrations indicates that the degree of segregation in Al-Sn system is not large. Since the expression for $\alpha_l$ is approximate and magnitude depends upon coordination number, Z, we have tried to see the effect of Z on $\alpha_l$. It increases but not in a dominant way. As regards $D_M/D_S$, it was observed (Fig.-3) that it is below one at all compositions, indicating self-coordination. Our study of $D_M/D_S$ suggests almost ideal behaviour in Sn rich ends. Minimum value around $C_{Sn}$ = 0.35 confirms more tendency for segregation, as observed by the study of $S_{CC}(0)$ and $\alpha_l$.

### 4.2. Surface tension and surface segregation

The computed values of $S_{CC}(0)$ at 973K have been used in equation (14) to calculate surface tension ($\gamma$) of Al-Sn liquid alloys. Required inputs are isothermal compressibility ($\chi_T$), dilatation factor ($\delta$) and thickness ($l$) of the interface. Because of unavailability of the experimental values for $\chi_T$ and volume, we have taken linear values for them. Experimental values for isothermal compressibility /30/ and volume /31/ of pure components are taken. So far as the determination of $l$ is concerned, we have calculated $l_i$ (pure component) through the relation /28/, $\chi_T^{\,1}\gamma_i = l_i$. It would be proper to mention that there is a big difference in the experimental and theoretically /30/ calculated values for $\chi_T^{\,1}$ for Sn. We have used linear values for $l$. The computed values are presented in Fig. 4 along with theoretical values, calculated through Eqn. 17 at 973K and 1273K. It will be proper to mention that experimental values /32/ for Al-Sn exist only up to 3.8 at% Sn. To check our results we have calculated $\gamma$ at 1273K by Eqn. 17 in dilute region limit, which is presented in Fig.-5. The computation of $\gamma$ by Eqn. 17 requires the value of order energy, $\omega$, which has been evaluated from the observed

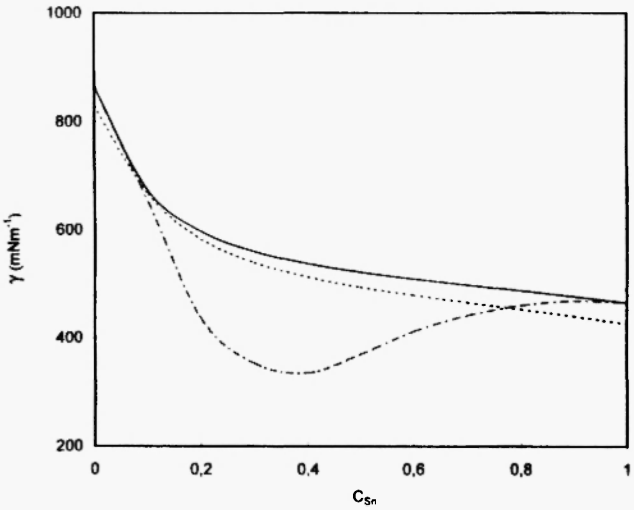

**Fig. 4:** Surface tension ($\gamma$) of Al-Sn liquid alloys. (—) and (----) refer to computed values (Eqn. 17) at 973K and 1273K respectively. (– · – · ) are computed values (Eqn. 14) from $S_{CC}(0)$ at 973K.

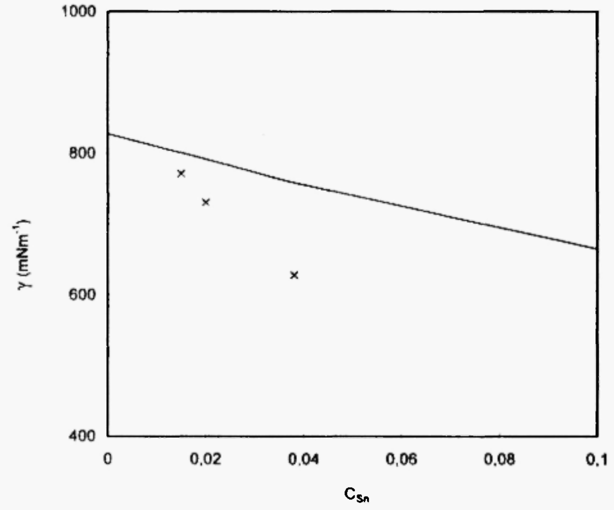

**Fig. 5:** Surface tension of Al-Sn system at T= 1273K in dilute region limit. (—) theory and (× × ×) experimental points / 32 /.

values of excess free energy of mixing ($G_M^{xs}$) at equi-atomic composition through the relation /22/

$$G_M^{xs} = NK_BT \ln [\, 2^{Z/2} \{ 1+\exp(-\omega/ZK_BT) \}^{-Z/2} ]$$

ω has been fixed to be equal to $1.45K_BT$. It is independent of concentration. Linear volume has been used to compute mean atomic surface area and experimental values for $\gamma_i$ have been taken. γ computed from Eqn. 17 is quite different in the middle concentration range from those obtained from $S_{CC}(0)$ (Eqn. 14). The results from Eqn. 14 suggest a sharp decrease in γ up to $C_{Sn} \sim 0.3$, followed by an increase. This is in contrast to those obtained from Eqn. 17, which suggests slow decrease in γ with the addition of Sn over whole concentration range. As experimental values for γ of Al-Sn liquid alloys exist only up to 3.8 % Sn, we are not in a position to check the validity of surface tension results obtained by Eqn. 14 through the use of $S_{CC}(0)$. Our investigation does not allow extrapolation of the surface tension values of Al-Sn alloys over the whole concentration range; as done by Li *et al.* /33/. But both theories suggest a decrease in γ up to 10% Sn (covering the experimental concentration range). Eqn. 17 also suggests an increase in γ with the decrease of temperature. The computed results in dilute region limit from Eqn. 17 are in reasonable agreement (Fig. 5) with the experiment /32/. In both cases there is a decrease, except for some difference in magnitude around 3.8% Sn. The pair of equations (17) have been solved numerically to calculate surface composition. Our investigation (Fig. 6) suggests the enrichment of the surface of Al-Sn alloys by Sn-atoms at all bulk compositions. Around bulk $C_{Sn}$= 0.4, the surface concentration of Sn becomes about 0.938. The extent of surface segregation increases with the decrease of temperature.

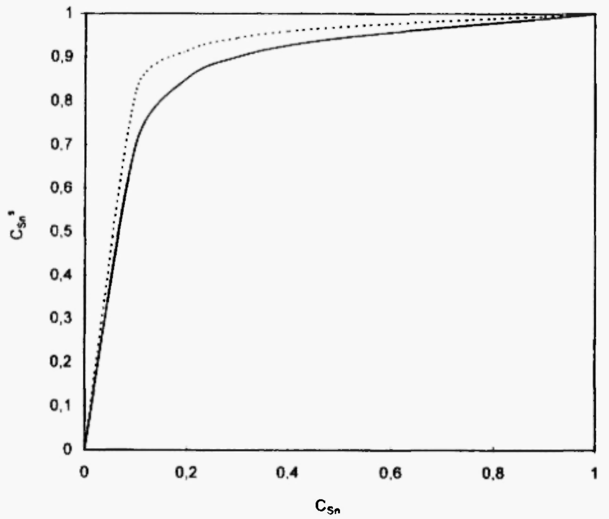

**Fig. 6:** $C_{Sn}^s$ vs $C_{Sn}$ for Al-Sn liquid alloys at 973K (-----) and 1273K (—).

### 4.3. Work of adhesion and interfacial tension with solid $Al_2O_3$

Our computed results of γ through Eqn. 17 have been used in Eqns. (20) and (21) to calculate work of adhesion (W) and interfacial tension ($\gamma_{SL}$) between Al-Sn (l) and $Al_2O_3$ (s) respectively at all compositions. We have used experimentally /33, 34 / measured values for angle of contact, θ, and $\gamma_{SV}$ . It will be proper to mention that Li *et al.* /33/ have used estimated values of γ beyond 3.8% Sn for the computation of W and $\gamma_{SL}$. But our computed values of γ in the middle concentration range might be giving different results. However, it is clear from Fig. 7 that work of adhesion decreases with the addition of Sn to Al and increases with the addition of Al to Sn. Our investigation also presents different

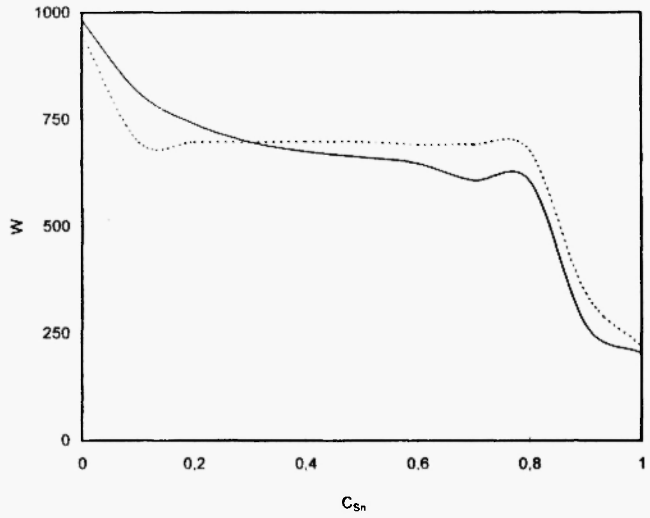

**Fig. 7:** Work of adhesion (W) for Al-Sn(l) / $Al_2O_3$ (s) at 1273K. (—) theory and (-----) expt. values /33/.

results in the concentration range $0.1 < C_{Sn} < 0.8$. We observe slight decrease in W with the addition of Sn while experimental values /33/ are almost constant. As regards $\gamma_{SL}$, it agrees (Fig. 8) well with experimental /33/ values. It remains almost constant up to 90% Sn and then rises sharply in the Sn-rich end; being one of the causes for the increase of W with the addition of Al to Sn.

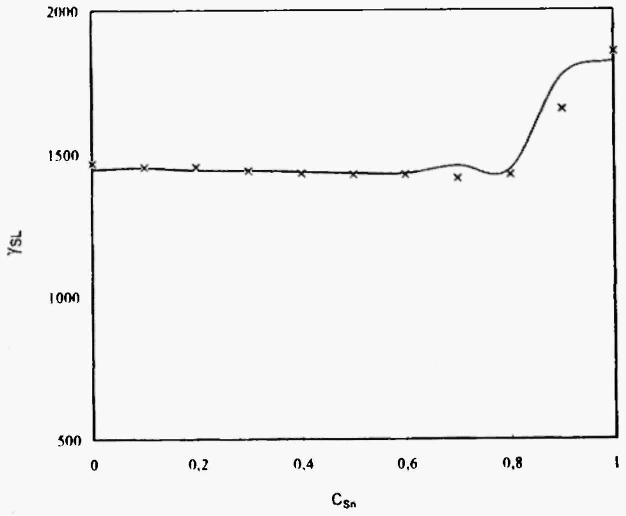

**Fig. 8:** Interfacial tension ($\gamma_{SL}$) between solid $Al_2O_3$ and liquid Al-Sn alloys at 1273K. (—) theory and (×××) expt. points /33/.

## 5. CONCLUSION

Our study of $S_{CC}(0)$, $\alpha_1$ and $D_M/D_S$ suggests like atoms pairing as nearest neighbours in Al-Sn alloys at all compositions; maximum being around $C_{Sn} = 0.35$. The surface tension computed from $S_{CC}(0)$ differs significantly in the middle concentration range from those obtained from grand partition function approach. However, in the experimental concentration range (up to 3.8% Sn) both theories exhibit a decrease in the surface tension of Al-Sn liquid alloys with the addition of tin to aluminium. Sn atoms have been found to segregate to the surface at all bulk compositions. We have also observed a decrease in work of adhesion (W) between Al-Sn and $Al_2O_3$ with the addition of Sn to Al and increase with the addition of Al to Sn. $\gamma_{SL}$ of $Al_2O_3$/Al-Sn has been found to be unaffected up to 80% Sn but it increases sharply after that, causing W to increase with the addition of Al to Sn.

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