

BIBECHANA

A Multidisciplinary Journal of Science, Technology and Mathematics

ISSN 2091-0762 (Print), 2382-5340 (Online)

Journal homepage: <http://nepjol.info/index.php/BIBECHANA>

Publisher: Research Council of Science and Technology, Biratnagar, Nepal

Theoretical investigation of mixing properties of Sb-Sn binary liquid alloy at 905 K

H. K. Limbu, K. K. Mishra*, A. K. Sah, I. S. Jha, D. Adhikari

Department of physics Mahendra Morang Aadarsh Multiple Campus
Tribhuvan University, Biratnagar, Nepal.

*Email: mailkaushalko@yahoo.com

Article history: Received 05 February, 2017; Accepted 21 August, 2017

DOI: <http://dx.doi.org/10.3126/bibechana.v15i0.18306>

This work is licensed under the Creative Commons CC BY-NC License.

<https://creativecommons.org/licenses/by-nc/4.0/>



Abstract

The thermodynamic, microscopic, surface and transport properties of Sb-Sn liquid alloy at 905K have been studied using regular solution model. In thermodynamic properties, free energy of mixing(G_M), activity(a), entropy of mixing(S_M), heat of mixing (H_M) have been studied. To understand structural behavior of the liquid alloys concentration fluctuations in the long wavelength limit i.e. ($S_{cc}(0)$) and short range order parameter (α_1) have been computed. Surface property is studied with the help of Butler's model while transport property is computed from Moelwyn-Hughes equation. The theoretical and experimental values of thermodynamic and microscopic properties of Sb-Sn liquid alloy at 905K have been compared. In present work the value of interchange energy (ω) is found to be negative suggesting that there is a tendency of unlike atoms pairing (i.e. Sb-Sn) as the nearest neighbor indicating the ordering behavior in Sb-Sn liquid alloy. The symmetric behavior of concentration fluctuations of the liquid alloy has been well explained by the model. The temperature dependence of interchange energy (ω) has been found during the computation of entropy of mixing (S_M) and heat of mixing (H_M) of the liquid alloy.

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

1. Introduction

Alloys of Sn are likely to be very promising as an alternative solder in the electronic industry[1]. The commonly used solder consists of lead which is known to be toxic to the human body and causes serious environmental problems. Antimony compounds have been known since ancient times and were used for cosmetics. Alloying lead and tin with antimony improves the properties of the alloys which are used in solders, bullets and plain bearings. This is the motivation of our theoretical work on Sb-Sn system. The Sb-Sn alloy is of practical importance in view of their use, mainly in optoelectronic devices. The In-Sb compound (diamond type $a = 0.6478\text{nm}$) can be used as a stabilizer for the Sb-Sn phase[2].

The mixing behavior of two elemental metals forming a binary alloy has always been a subject of considerable interest to many researchers[3–11]. 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 phenomenon such as corrosion, wetting characteristics of solders and kinetics of phase transformation. Transport properties are required for many metallurgical process and heterogeneous chemical reactions. The thermodynamic properties are important to understand the behavior of alloys which is studied with the knowledge of free energy of mixing (G_M), activity (a), entropy of mixing (S_M), heat of mixing (H_M).

Structural behavior is interpreted by the concentration fluctuation in long wave limit ($S_{cc}(0)$) and chemical short range order parameters (α_1). Surface tension is computed using Buttler's model[12] and viscosity is calculated with the help of Moelwyn-Hughes equation[13]. Since the atoms Sb and Sn are identical in shape and size (i.e. atomic volume of Sb / atomic volume of Sn =18.703/17.741 ~1), the Sb-Sn liquid alloy is suitable candidate for our investigation on using regular solution model[14]. The theoretical formulation of regular solution model has been presented in section (2), result and discussion in section (3) and conclusion in section (4).

2. Formalism

Regular solution model is based on the approximation that the constituent atoms A and B are sufficiently similar in size and shape so that they are interchangeable on the lattice or quasi-lattice, and the configuration is no longer independent of the mutual disposition of the two or more kinds of molecules. Homogenous solution of binary liquid alloy A-B consists of C_A ($\equiv c$) mole of A and C_B ($\equiv c(1-c)$) mole of B respectively, where c_A and c_B are the mole fractions of A (\equiv Sb) and B (\equiv Sn) in the binary liquid alloy A-B.

2.1. Thermodynamic properties

In the frame work of regular solution model, the expression for the free energy of mixing (G_M) of binary liquid alloy is

$$G_M = G_M^{id} + H_M \quad (1)$$

where, heat of mixing (H_M) and ideal free energy of mixing (G_M^{id}) are given by

$$H_M = \omega c_A c_B \quad (2)$$

$$G_M = RT [c \ln c + (1 - c) \ln(1 - c)] + c(1 - c) \cdot \omega \quad (3)$$

Here ω is interaction energy.

The expression for activities a_A of the elements A in the binary liquid alloy can be derived from the standard relation, that is

$$RT \ln a_A = G_M + (1 - c) \frac{\partial G_M}{\partial c} \quad (4)$$

$$\frac{\partial G_M}{\partial c} = RT [\ln c - \ln(1 - c)] + (1 - 2c) \cdot \omega \quad (5)$$

Using equation (3) and (5) in equation (4), we get

$$\ln a_A = \ln c + \frac{\omega}{RT} (1 - c)^2 \quad (6)$$

and the activity of the element B (a_B) is given by

$$\ln a_B = \ln(1 - c) + \frac{\omega}{RT} c \quad (7)$$

The temperature derivative of G_M provides an expression for integral entropy of mixing (S_M) which is given by

$$S_M = \frac{\partial G_M}{\partial T} \quad (8)$$

From equation (3) and (8), we get

$$\frac{S_M}{R} = [c \ln c + (1 - c) \ln(1 - c)] - c(1 - c) \cdot \frac{1}{R} \frac{\partial \omega}{\partial T} \quad (9)$$

The first term in the R.H.S of equation (9) is usual ideal of mixing. The last term represents the contributions from the temperature derivatives of the interchange energy. The necessity of taking ω as temperature dependent has been noticed by Bhatia et. al[15], Ratti and Bhatia[7], Alblas et. al[6].

The relation between free energy of mixing (G_M), entropy of mixing (S_M), and heat of mixing (H_M) is given as,

$$\frac{H_M}{RT} = \frac{S_M}{R} + \frac{G_M}{RT} \quad (10)$$

Using equation (3), (9) and (10), we get

$$\frac{H_M}{RT} = c(1 - c) \cdot \frac{\omega}{RT} + c(1 - c) \frac{1}{R} \cdot \frac{\partial \omega}{\partial T} \quad (11)$$

2.2 Structural Properties

The regular solution model has been successfully used to evaluate the value of concentration structure factor or concentration fluctuation in the long wavelength limit ($S_{cc}(0)$) which has been used to study the nature of atomic order in binary liquid alloy [17] by using relation,

$$S_{CC}(0) = \frac{RT}{\left(\frac{\partial^2 G_M}{\partial c^2}\right)_{T,P,N}} \quad (12)$$

From equation (3) and (12), we get

$$S_{CC}(0) = \frac{c_A c_B}{1 - 2c_A c_B} \cdot \frac{\omega}{RT} \quad (13)$$

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

$$s_{cc}(0) = (1 - c)a_A \left(\frac{\partial a_A}{\partial c}\right)_{T,P,N}^{-1} = c a_B \left(\frac{\partial a_B}{\partial c}\right)_{T,P,N}^{-1} \quad (14)$$

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

The Warren-Cowley[15,16] short range order parameter (α_1) is useful parameter to quantify the degree of chemical order in the alloy melt. It provides insight into the local arrangement of the atoms in the molten alloys. Although it is difficult to obtain the experimental values of α_1 , theoretical values of this parameters can be evaluated

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

where, $s = \frac{S_{cc}(0)}{S_{cc}^{id}(0)}$, $S_{cc}^{id}(0) = c_A c_B$

and Z is the coordination number, which is taken Z = 6,8 and 10 for our purpose.

2.3 Surface Property

Butler assumed the existence of mono atomic layer at the surface of a liquid solution, called surface monolayer, as a separate phase that is in thermodynamic equilibrium with the bulk phase and derived an equation[7,18,19]. Butler equation for the surface tension, Γ of a binary A-B solution at temperature T is expressed as

$$\Gamma = \Gamma_1 + \frac{1}{A_1} (G_1^{E,s} - G_1^{E,b}) + \frac{RT}{A_1} [\ln(1 - c_2^s) - \ln(1 - c_2^b)] = \Gamma_2 + \frac{1}{A_2} (G_2^{E,s} - G_2^{E,b}) + \frac{RT}{A_2} [\ln(c_2^s) - \ln(c_2^b)] \quad (16)$$

where, R is universal gas constant. c_i^s and c_i^b are mole fraction of component 'i' in the surface and bulk respectively. 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

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

where, K(= 1.091) is geometrical factor for the liquid alloy, N_A is Avogadro's number, V_i is the molar volume of the component i.

where, $v_{sb} = v_M [1 + \alpha_p (T - T_M)]$ and $v_{sn} = v_M [1 + \alpha_p (T - T_M)]$ (18)
 v_M = atomic volume at melting point
 T_M = melting temperature and
 α_p = volume coefficient at constant temperature.

2.4 Transport Properties

The mixing behavior of an alloy in microscopic level can also be studied by diffusion coefficient. Singh and Sommer[18] have derived a relation between diffusion coefficient and concentration fluctuation as

$$\frac{D_M}{D_{id}} = \frac{S_{CC}^{id}(0)}{S_{CC}(0)} \quad (19)$$

where, D_M is the mutual diffusion coefficient and D_{id} is the intrinsic diffusion coefficient for an ideal mixture given as

$$D_M = c_1 D_2 + c_2 D_1 \quad (20)$$

where, D_1 and D_2 are the self-diffusivities of pure components A and B respectively.

In term of energy order parameter ω , the diffusion coefficient can be expressed as[18]

$$\frac{D_M}{D_{id}} = [1 - \frac{2\omega}{RT} S_{CC}^{id}(0)] \quad (21)$$

The ration D_M/D_{id} indicates the mixing behavior of the alloys i.e. $D_M/D_{id} > 1$ indicates compound formation and if $D_M/D_{id} < 1$ then there is tendency of phase separation. For ideal mixing, D_M/D_{id} approaches 1

Viscosity is one of the transport properties of binary liquid alloy. The Moelwyn-Hughes equation[13] for viscosity of liquid mixture is given as

$$\eta = (c_1 \eta_1 + c_2 \eta_2) (1 - 2c_1 c_2 \frac{H_M}{RT}) \quad (22)$$

where, η_K is the viscosity of pure component K(K= 1, 2), c is the mole fraction of the component K and H_M is the enthalpy of mixing. For most liquid metals, η_K can be calculated at temperature T from Arrhenius type equation[20]

$$\eta_K = \eta_{OK} \exp \left[\frac{E_n}{RT} \right] \quad (23)$$

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

3. Results and Discussion

3.1. Thermodynamic properties: free energy of mixing, activity, heat of mixing and entropy of mixing

The value of energy parameter i.e. interchange energy (ω) used for the calculation of thermodynamic properties of Sb-Sn liquid alloy at 905 K has been estimated from equation (3) by the method of successive approximation, using the observed data of free energy of mixing, G_M [21] over the entire range of concentration. The best fit value is found to be $\frac{\omega}{RT} = -0.8925$.

The sign and size of the energy parameter governs the general character, phase separation or complex formation of the alloys system. The negative sign of energy parameter suggests that there is a general tendency for unlike atoms to pair in the Sb-Sn liquid alloy at 905 K. However, the tendency of pairing is weak as the energy parameter is small. The same value of the energy parameter has been used throughout the calculations in order to maintain the consistency for the other properties of mixing of the liquid alloy at 905 K.

The plot of experimental and computed values of free energy of mixing (G_M) of Sb-Sn liquid alloy with respect to the concentration of Sb is shown in Fig.1.

The computed and experimental values[21] of free energy of mixing are in good agreement in the entire concentration range. The computed value of G_M/RT ($= -0.9163$) is minimum at $c_{Sb}=0.5$ while, experimental value of G_M/RT ($= -0.9163$) at $c_{Sb}=0.5$. The negative small values of free energy of mixing throughout the entire concentration range indicate that the Sb-Sn alloy at 905 K in liquid state is a weak interacting system.

We have computed the activities of Sb and Sn in the liquid Sb-Sn alloy using the same energy parameter (ω) in equation (6) and (7), and then compared them with experimental values as depicted in Fig. 2.

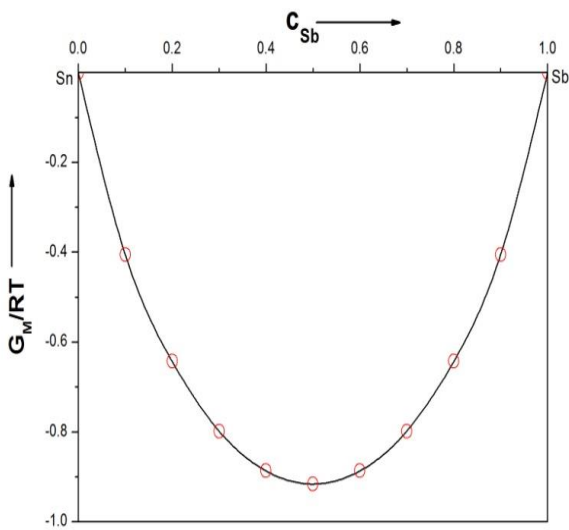


Fig. 1: Free energy of mixing of Sb-Sn liquid alloy at 905 K. The Solid line represents theoretical values and Circle represents experimental values.

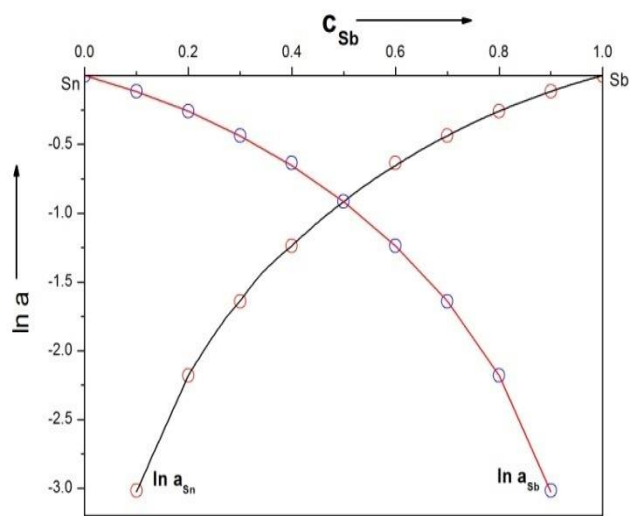


Fig. 2: Activities a_{Sn} and a_{Sb} of Sb-Sn liquid alloy at 905K. The solid line theoretical values and circle represents experimental values.

The plot shows that there is a good agreement between the sets of computed and experimental results. Activity coefficient represents the measure of tendency of a component to leave the solution. The magnitude of activities depends on the bond energies. Activity is very important quantity which can be associated to the free energy of mixing G_M and concentration fluctuation $S_{cc}(0)$. Measurement of activities provides correlation of the behavior of the system.

The heat of mixing and entropy of mixing of the Sb-Sn binary liquid alloy at 905 K has been calculated from equation (11) and (9) respectively. To determine heat of mixing (H_M) and entropy of mixing (S_M) we need temperature derivative of energy parameters. The observed values of heat of mixing[21] were utilized to obtain the temperature derivatives by the method of successive approximation. The best fit value has found to be $\frac{1}{R} \frac{\partial \omega}{\partial T} = -0.18$. Both computed and experimental values of heat of mixing and entropy of mixing are in good agreement at all compositions as shown in figures 3 and 4.

The observed symmetry in the heat of mixing and entropy of mixing about equiatomic composition of Sb-Sn liquid alloy at 905 K is well explained by the present theoretical analysis. The negative value of H_M indicates the ordering nature of the liquid alloy in the entire range of concentration. The theoretical understanding of heat of mixing as a function of concentration is very useful as it directly

leads to the nature of interaction among the constituent's species of the alloys. Though such information is available from free energy of mixing we may emphasize that heat of mixing has some added significance. From the definition of H_M ($H_M = G_M + TS_M$) it is obvious that H_M has greater significance than G_M , as the physical significance of S_M is also locked up in the heat of mixing.

The entropy of mixing S_M is helpful to draw important structural information for binary liquid alloys. It represents the sharing of energy among the atoms of the constituent metals forming the alloys and hence the disorder in the equilibrium state of the alloys. The role of entropy is often complicated for most of the binary liquid alloys. The computed and experimental values[21] of entropy of mixing in Sb-Sn binary liquid alloy are positive in the entire range of concentration.

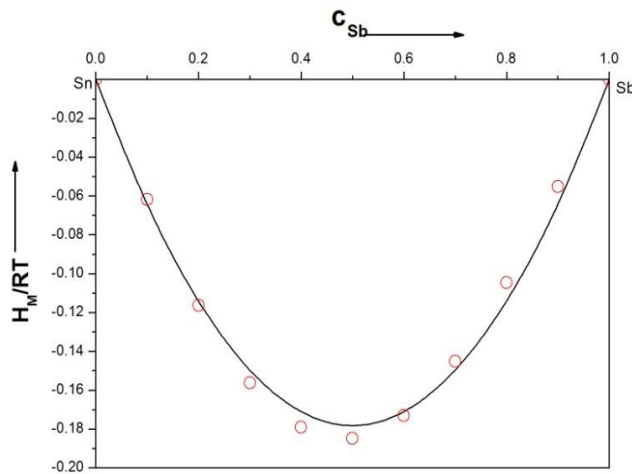


Fig. 3: Heat of mixing of Sb-Sn liquid alloy at 905K. The solid line represents theoretical values and Circle represents experimental values.

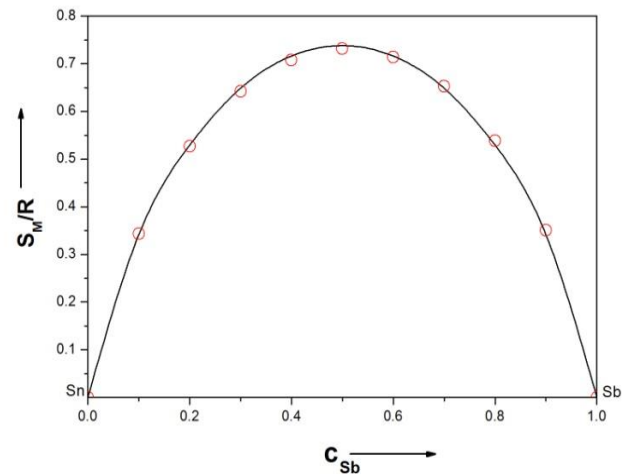


Fig. 4: Entropy of mixing of Sb-Sn liquid alloy at 905K. The solid line represents theoretical values and circle represents experimental values.

3.2 Structural properties: concentration fluctuation in the long wavelength limit and chemical short range order parameter

The $s_{cc}(0)$ computed from equation (14) is referred as to the experimental value. The computed and experimental values of $S_{cc}(0)$ are depicted in figure 5 along with the ideal values i.e. $S_{cc}^{id}(0) = c(1 - c)$. liquid alloys. At a given composition, if $S_{cc}(0) < S_{cc}^{id}(0)$ then, ordering in liquid alloys is expected, while $S_{cc}(0) > S_{cc}^{id}(0)$ is the indication of a tendency of phase separation. Figure-5 shows that $S_{cc}(0) < S_{cc}^{id}(0)$ throughout the entire concentration range which indicates that hetero-coordination is favored in the Sb-Sn alloy at 905 K.

The calculated values of $S_{cc}(0)$ are in good agreement with the experimental values[21]. The calculated values of $S_{cc}(0)$ ($= 0.1729$) and experimental values of $S_{cc}(0)$ ($= 0.1729$) are maximum about equiatomic composition. The result can be used to understand the nature of atomic order in binary The Warren-Cowley chemical short-range order parameter α_l is an important microscopic function which is used to predict the degree of local arrangement of atoms in the mixture. The chemical short-range order parameter has been computed from equation (15) on using the theoretical value of $S_{cc}(0)$. The value of coordination number (Z) is taken to be 6,8 and 10. For equiatomic composition the parameter α_l is found to be $-1 \leq \alpha_l \leq +1$. The value of $\alpha_l^{min} = -1$ indicates complete ordering and the value of $\alpha_l^{max} = 1$ indicates complete segregation leading to phase separation and $\alpha_l=0$ corresponds to a

random mixing of atoms. The values of short range order parameter α_1 are negative in the entire concentration range and the minimum value (i.e. $\alpha_1 = -0.0692$ at $z=6, \alpha_1 = -0.0528$ at $z=8, \alpha_1 = -0.0427$ at $z=10$) is at equiatomic composition as shown in figure 6, which indicates the ordering nature of the alloy as evident from the $S_{cc}(0)$.

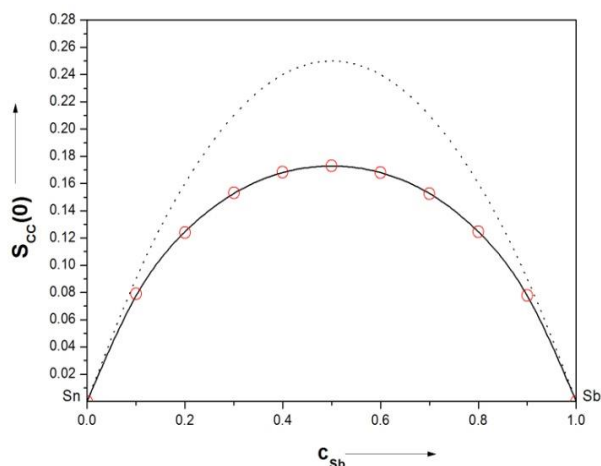


Fig. 5: Concentration fluctuation of Sb-Sn liquid alloy at 905K. The solid line represents theoretical values, circle represents experimental values and dotted line represents ideal values.

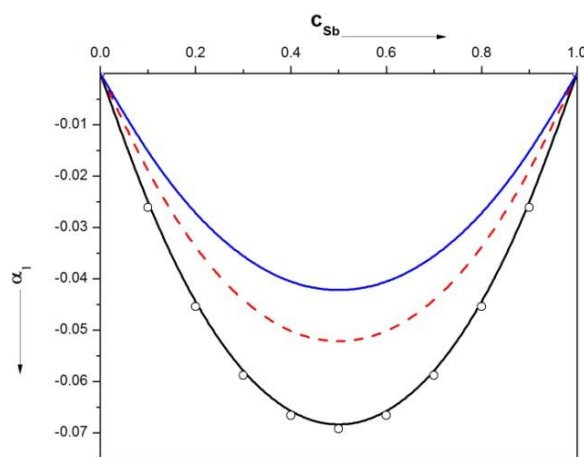


Fig. 6: Chemical short range order parameter (α_1) of Sb-Sn liquid alloy at 905K for different co-ordination number $Z=6, 8$ and 10 . The solid line represents $Z=10$, dotted line represents $Z=8$ and solid line with hollow circle represents $Z=6$.

3.3. Surface property

The surface tension of a liquid Sb-Sn has been computed by using equation (16). We have used the experimental value of bulk partial excess free energy $G_i^{E,b}$ [21] of the monomers of the liquid alloy at 905 K.

$$\beta = \frac{G_i^{E,s}}{G_i^{E,b}} \quad (24)$$

The value of parameter β has been taken as 0.83 as suggested by different researchers to compute surface tension of liquid alloys [20,22]. For computation of surface tension of Sn-Sn at 905K equation (16) in conjunction with equation (17) and (18) are used. We have calculated the surface tension of pure components (i.e. Sb and Sn) using equation

$$\Gamma(T) = \Gamma_m + \frac{\partial \Gamma}{\partial T} (T - T_m) \quad (25)$$

where, $T = 905$ K, $T_m =$ melting temperature ($T_m = 903.5$ K for Sb, and 505K for Sn) ; $\frac{\partial \Gamma}{\partial T}$ is temperature coefficient of surface tension ($= -0.05 \times 10^{-3} \text{ Nm}^{-1}\text{K}^{-1}$ for Sb, and $-0.07 \times 10^{-3} \text{ Nm}^{-1}\text{K}^{-1}$ for Sn). The partial excess free energy of mixing of the pure components for the systems Sb-Sn at 905 K are taken from the reference [20]. The surface concentration of a component with respect to the bulk concentrations has been calculated from equation (16). The variation of surface concentration with the bulk concentration of Sb component has been shown in Fig. 7.

It is to be noted that the components to the alloy do not mix completely with each other in the bulk because of the difference in the values of surface tension of constituent metals. The calculated values of surface concentration have been used to compute the surface tension of Sb-Sn liquid alloy at 905 K. From Fig. 7 it is clear that the surface concentration of Sb increases with increasing bulk concentration of Sb (i.e. c_{Sb}). This shows that Sb atoms (having lower value of surface tension relative

to Sn- atoms in Sb-Sn system) segregate at the surface of Sb-Sn liquid alloy throughout the entire concentrations. The computed values of surface tension of Sb-Sn liquid alloys at 905 K are depicted in Fig. 8.

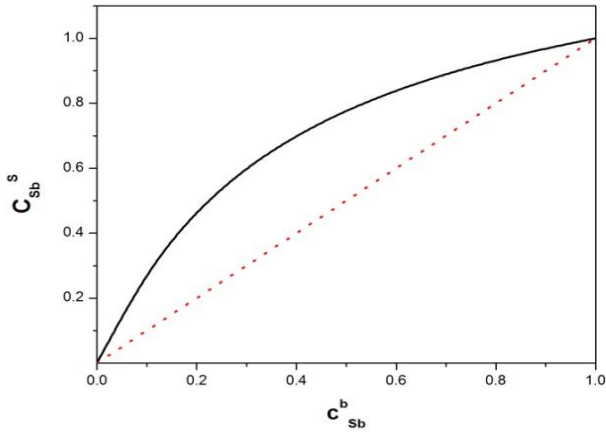


Fig. 7: Surface concentration of Sb versus bulk concentration of Sb in molten Sb-Sn liquid alloy at 905K. The solid line represents theoretical values and the dotted line represents ideal values.

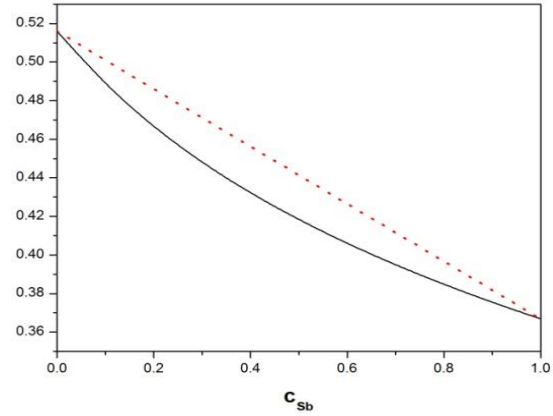


Fig. 8: Surface tension of Sb-Sn liquid alloy at 905K. The solid line represents calculated surface tension and dotted line represents ideal surface tension.

It is obvious from figure 8 that the computed values of surface tension of Sb-Sn liquid alloy at 905 K shows deviation from the ideal values ($\Gamma = c_1\Gamma_1 + c_2\Gamma_2$) i.e. the computed values of surface tension are slightly more than the ideal values at $c_{sb}=0.1$ to 0.9 compositions. It is also found that the surface tension of Sb-Sn liquid alloy decreases with the increase in the concentration of Sb atoms in the alloy as the surface tension of pure component of Sb atom is less than pure component of Sn atom at the temperature of study.

3.4 Transport properties: chemical diffusion and viscosity

The calculated values of $S_{cc}(0)$ are used in equation (20) to evaluate the ratio of mutual and self-diffusivities, D_M/D_{id} for Cd-Mg liquid alloy at 923 K. For the consistency of the estimated order energy parameter ω , we have also calculated D_M/D_{id} using equation (21) which predicts $\frac{D_M}{D_{id}} = 1$ at $c_{sb}=0$ and $c_{sb}=1$. Fig. 9 shows plot of D_M/D_{id} against the concentration of Sb calculated from equation (21).

From the figure it is clear that the value of $\frac{D_M}{D_{id}} > 1$ in the entire range of concentration which is indicative for the presence of chemical order in the alloy. The maximum value of D_M/D_{id} is 1.4459 at $c_{sb} = 0.5$, confirms the ordering tendency of the atoms in Sb-Sn liquid alloys is greater about equiatomic composition.

We have used Moelwyn-Hughes equation (22) in conjunction with equation (23). To compute

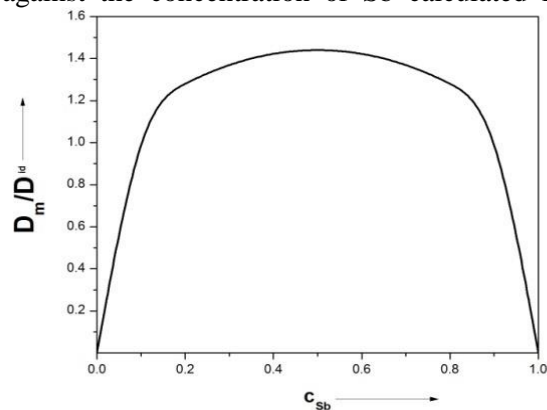


Fig. 9: Ratio of mutual and self-diffusivities, D_M/D_{id} for Sb-Sn liquid alloy at 905K versus concentration of Sb.

viscosity of Sb-Sn alloy at 905 K, the viscosities of pure components Sb and Sn at 905 K are required which is computed using equation (23). The viscosity of pure component can be obtained with the help of constants η_{ok} and E for the metals[20] and is as shown in the Fig.10. The plot shows that viscosity of the alloy is slightly deviated at the concentration $c_{sb}=0.1$ to 0.9. The viscosity of pure component of Sb atom is more than the viscosity of pure component Sn which results increase in the viscosity as the concentration of Sb atom increases in the alloy as shown in the Fig. 10.

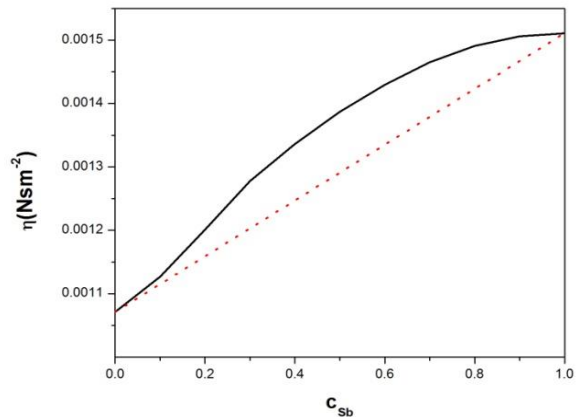


Fig. 10: Viscosity of Sb-Sn liquid alloy at 905K versus concentration of Sb. The solid line represents calculated viscosity and dotted line represents ideal values.

4. Conclusion

The negative interchange energy (ω) suggests that there is a tendency of unlike atoms pairing (Sb-Sn) of the alloy at 905 K. It is a weak interacting system. The interchange energy is found to be temperature dependent and plays important role to explain the alloying behavior of the alloy. All the thermodynamic and microscopic properties of this alloy are symmetric at equi-atomic composition as a function of concentration. The computed values of surface tension show small positive deviations from ideality and surface tension decreases as the concentration of Sb atom increases of the alloy. The study reveals that the viscosity isotherm of Sb-Sn liquid alloy at 905 K is found to deviate positively from ideality and viscosity increases as the concentration of Sb atom increases.

References

- [1] É.B. Hermidaab, F. Povolo, P. Porta, Internal friction and loss tangent of nonlinear viscoelastic materials: different concepts, different results, *J. Alloy. Compd.* 310 (2000) 280–283. [doi.org/10.1016/S0925-8388\(00\)00965-8](https://doi.org/10.1016/S0925-8388(00)00965-8).
- [2] V. Vassiliev, Y. Feutelais, M. Sghaier, B. Legendre, Thermodynamic investigation in In–Sb, Sb–Sn and In–Sb–Sn liquid systems, *J. Alloys Compd.* 314 (2001) 198–205. [doi.org/10.1016/S0925-8388\(00\)01243-3](https://doi.org/10.1016/S0925-8388(00)01243-3).
- [3] N.H. March, J.A. Alonso, Structure, transport and surface properties of dense fluids, especially liquid metals, *Mol. Phys.* 95 (1998) 353–361. doi.org/10.1080/00268979809483167
- [4] R.P. Koirala, B.P. Singh, I.S. Jha, D. Adhikari, Thermodynamic and structural behavior of liquid Al-Ga alloys, *Adv. Mater. Lett.* 4 (2013) 283–287. doi.org/10.5185/amlett.2012.8412
- [5] S. Harada, S. Takahashi, S. Takeda, S. Tamaki, P. Gray, N.E. Cusack, Thermodynamic properties of liquid Na-Cd and Na-In, *J. Phys. F Met. Phys.* 18 (1988) 2559–2567. doi.org/10.1088/0305-4608/18/12/007.
- [6] B.P. Alblas, C. van der Marel, W. Geertsma, J.A. Meijer, A.B. van Oosten, J. Dijkstra, P.C. Stein, W. van der Lugt, Experimental results for liquid alkali-group IV alloys, *J. Non. Cryst. Solids.* 61–62 (1984) 201–206. [doi.org/10.1016/0022-3093\(84\)90551-9](https://doi.org/10.1016/0022-3093(84)90551-9).
- [7] V.K. Ratti, A.B. Bhatia, Number concentration structure factors and their long wave-length limit in multicomponent fluid mixture, *Phys. Chem. Liq.* 6 (1977) 201–213.

- [8] D. Adhikari, B.P. Singh, I.S. Jha, Energetics of Cd-based binary liquid alloys, *J. Non. Cryst. Solids.* 358 (2012) 1362–1367. doi.org/10.1016/j.jnoncrysol.2012.03.008.
- [9] D. Adhikari, B.P. Singh, I.S. Jha, Structural and energetic anomaly in liquid Na–Sn alloys, *J. Mol. Liq.* 167 (2012) 52–56. doi.org/10.1016/j.molliq.2011.12.010.
- [10] L.C. Prasad, R.N. Singh, A Quasi-lattice Model for the Thermodynamic Properties of Au-Zn Liquid Alloys, *Phys. Chem. Liq. An Int. J.* Vol. 22 (1989) 37–41. doi.org/10.1080/00319109008036406.
- [11] M. Hoch, Application of the complex model to liquid metal-salt systems, *Calphad.* 9 (1985) 59–70.
- [12] J.A. V. Butler, The Thermodynamics of the Surfaces of Solutions, *Proc. R. Soc. A Math. Phys. Eng. Sci.* 135 (1932) 348–375. doi.org/10.1098/rspa.1932.0040.
- [13] E.A. Moelwyn-Hughes, *Physical Chemistry*, Oxford, London: Longmans Green and Co., 1974.
- [14] E.A. Guggenheim, *Mixtures*, Oxford University Press, London, 1952.
- [15] J.M. Cowley, An Approximate Theory of Order in Alloys, *Phys. Rev.* 77 (1950) 669–675. doi.org/10.1103/PhysRev.77.669.
- [16] B.E. Warren, *X-ray diffraction*, Addison-Wesley, Reading MA, 1969.
- [17] A.B. Bhatia, R.N. Singh, Thermodynamic properties of compound forming molten alloys in a weak interaction approximation, *Phys. Chem. Liq.* 11 (1982) 343–351. doi.org/10.1080/003191082080755.
- [18] R.N. Singh, F. Sommer, Segregation and immiscibility in liquid binary alloys, *Reports Prog. Phys.* 60 (1997) 57–150. doi.org/10.1088/0034-4885/60/1/003.
- [19] R. Novakovic, Thermodynamics, surface properties and microscopic functions of liquid Al–Nb and Nb–Ti alloys, *J. Non. Cryst. Solids.* 356 (2010) 1593–1598. doi.org/10.1016/j.jnoncrysol.2010.05.055.
- [20] E. A. Brandes and G.B. Brook, ed., *Smithells Metals Reference Book*, 7th editio, Butterworth-Heinemann Linacre House, Jordan Hill, Oxford, 1992.
- [21] R. Hultgren, P.D. Desai, D.T. Hawkins, M. Gleiser, K.K. Kelley, *Selected Values of the Thermodynamic Properties of Binary Alloys*, Metal Park, ASM International, OHIO, 1973.
- [22] K.S. Yeum, R. Speiser, D.R. Poirier, Estimation of the surface tensions of binary liquid alloys, *Met. Trans. B.* B20 (1989) 693–703.