

Thermodynamic, Structural, Surface and Transport Properties of In-Tl Liquid Alloy at Different Temperatures

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Abstract

The thermodynamic, structural, surface and transport properties of In-Tl binary liquid alloy are studied on the basis of theoretical analysis using the regular solution model at different temperatures. The properties of the alloy at 723 K have been computed by estimating the best fit value of order energy parameter (ω) in the entire range of concentration to match their observed and theoretical values. The values of order energy parameter at different temperatures have been calculated using the value of order energy parameter at 723 K which played key role to study different properties of the alloy using optimization method. The theoretical analysis gives the positive energy parameter (ω), which is found to be temperature dependent.

Keywords

In-Tl Alloy, Optimization, Segregation, Thermodynamic Properties, Structural Properties

1. Introduction

Various properties, such as hardness, corrosion resistance, wear resistance, mechanical strength, and fatigue strength, melting and boiling temperatures etc. of an alloy are different from its individual components. The properties of alloys in solid state are usually determined by thermodynamic, surface, structural, transport and electrical properties of alloys in liquid state. But there is complexity in determining the properties of liquid alloys due to lack of long range atomic order. Therefore several models [1]-[9] have long been proposed to understand the properties of alloys in liquid state.

The binary In-Tl alloy has widely been studied in the past because of its wide

range of practical applications [10] [11] [12]. It is a shape memory alloy [13] having low melting temperature (fusible alloy). The indium based fusible alloy has the ability to wet and adhere to ceramic and glass surfaces. This characteristic can be useful in sealing and bonding of sensitive electronic devices, glass, ceramic and temperature-sensitive materials.

In this work we have used regular solution model [14] to explain thermodynamic and structural properties of In-Tl at 723 K. In regular solution model, the interaction energy is considered as input parameter and is determined by fitting observed free energy of mixing at different concentrations. Thermodynamic behavior of the alloy is studied by computing free energy of mixing (G_M), activity (a), entropy of mixing (S_M) and heat of mixing (H_M). Structural properties is interpreted by the concentration fluctuation in the long wavelength limit ($S_{cc}(0)$) and chemical short range order parameter (α_1). The surface properties are studied by using Butler's model [15] and viscosity is studied with the help of Moelwyn-Hughes equation [16]. In order to compute all these properties at higher temperatures optimization procedure is used. Basic theoretical formulation of regular solution model has been presented in Section (2), result and discussion in Section (3) and conclusion are dealt in Section (4).

2. Theoretical Formalism

2.1. Thermodynamic Properties

Regular solution model [14] deals with the A-B alloy in liquid state in which 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. We suppose a liquid binary mixture A-B having c_A ($\equiv c$) mole of A and c_B ($\equiv (1-c)$) mole of B respectively, where c_A and c_B are the mole fractions of A (\equiv In) and B (\equiv Tl) in the binary liquid alloy A-B.

The expression for free energy of mixing (G_M) can be derived on the basis of regular solution model. It is given as [14]

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

where R is the universal gas constant, T the temperature, and ω the interaction energy parameter to be determined.

The term $c(1-c) \cdot \omega$ represents the heat of mixing in the frame work of regular solution model, *i.e.*

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

The activity a_A of the element A in the binary alloys A-B is given by the standard relation

$$RT \ln a_A = G_M + (1-c) \frac{\partial G_M}{\partial c} \quad (3)$$

Using Equations (1) and (3) we get the activity of element A as

$$\ln a_A = \ln c + \frac{\omega}{RT}(1-c)^2 \quad (4)$$

Similarly, the activity of the element B is given by

$$\ln a_B = \ln(1-c) + \frac{\omega}{RT}c^2 \quad (5)$$

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

$$S_M = -\frac{\partial G_M}{\partial T} \quad (6)$$

From Equations (1) and (6), we get

$$\frac{S_M}{R} = -[c \ln c + (1-c) \ln(1-c)] - c(1-c) \cdot \frac{1}{R} \frac{\partial \omega}{\partial T} \quad (7)$$

The relation between free energy of mixing (G_M), entropy of mixing (S_M), and heat of mixing (H_M) is given by a standard thermodynamic relation,

$$\frac{H_M}{RT} = \frac{S_M}{R} + \frac{G_M}{RT} \quad (8)$$

Using Equations (1), (7) and (8), we get

$$\frac{H_M}{RT} = c(1-c) \cdot \frac{\omega}{RT} + c(1-c) \frac{1}{R} \cdot \frac{\partial \omega}{\partial T} \quad (9)$$

2.2. Structural Properties

The 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] can also be deduce using regular solution model and is given by using relation,

$$S_{cc}(0) = \frac{RT}{\left(\frac{\partial^2 G_M}{\partial c^2}\right)_{T,P,N}} \quad (10)$$

From Equations (1) and (10), we get

$$s_{cc}(0) = \frac{c_A c_B}{1 - 2c_A c_B} \cdot \frac{\omega}{RT} \quad (11)$$

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

$$s_{cc}(0) = (1-c) a_A \left(\frac{\partial a_A}{\partial c}\right)_{T,P,N}^{-1} = c a_B \left(\frac{\partial a_B}{\partial c}\right)_{T,P,N}^{-1} \quad (12)$$

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

The degree of local order in the liquid mixture is quantified by the Warren-Cowley [18] [19] short range order parameter (α_1). It provides insight into the local arrangement of the atoms in the molten alloys. The theoretical values of

(α_1) can be evaluated as

$$\alpha_1 = \frac{s-1}{s(Z-1)+1} \quad (13)$$

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 10 ($= Z$) for our purpose.

2.3. Surface Property

Butler derived an expression for the surface tension of liquid binary mixture on the basis of the assumption that there exists a 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 [15] [20]. The expression for surface tension, Γ on the basis of Butler equation for binary A-B solution at temperature T is given by

$$\begin{aligned} \Gamma &= \Gamma_1 + \frac{1}{A_1} (G_1^{E,s} - G_1^{E,b}) + \frac{RT}{A_1} [\ln(1-c_2^s) - \ln(1-c_2^b)] \\ &= \Gamma_2 + \frac{1}{A_2} (G_2^{E,s} - G_2^{E,b}) + \frac{RT}{A_2} [\ln(c_2^s) - \ln(c_2^b)] \end{aligned} \quad (14)$$

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

$$A_i = K \cdot N_A^{1/3} \cdot V_i^{2/3} \quad (15)$$

where, $K (=1.091)$ is geometrical factor for the liquid alloy, N_A is Avogadro’s number, $V_i (= V_{M,i}, V_{Tl,i})$ is the molar volume of the component i .

where, $V_{M,i} = V_{M,i} [1 + \alpha_{P,i} (T - T_{M,i})]$ and $V_{Tl,i} = V_{M,i} + [1 + \alpha_{P,i} (T - T_{M,i})]$

$V_{M,i}$ = atomic volume of In at melting point

$V_{M,Tl}$ = atomic volume of Tl at melting point

$T_{M,i}$ = melting temperature of In

$T_{M,Tl}$ = melting temperature of Tl

$\alpha_{P,i}$ = volume coefficient of In at constant temperature

$\alpha_{P,Tl}$ = volume coefficient of Tl at constant temperature

2.4. Transport Properties

The diffusion coefficients also provide the insight into the mixing behavior of an alloy in microscopic level. The relation between diffusion coefficient and concentration fluctuation is given as [21]

$$\frac{D_M}{D_{id}} = \frac{S_{CC}^{id}(0)}{S_{CC}(0)} \quad (16)$$

With

$$D_M = c_1 D_2 + c_2 D_1 \quad (17)$$

where, D_M is the mutual diffusion coefficient and D_{id} is the intrinsic diffusion coefficient for an ideal mixture; 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 [22]

$$\frac{D_M}{D_{id}} = \left[1 - \frac{2\omega}{RT} S_{cc}^{id}(0) \right] \quad (17) \text{ If } D_M/D_{id} > 1, \text{ then it indicates compound formation and if } D_M/D_{id} < 1 \text{ then there is tendency of phase separation. For ideal mixing, } D_M/D_{id} \text{ approaches 1.}$$

The mixing behavior of liquid alloys at microscopic level can also be understood in terms of viscosity. We have employed the Moelwyn-Hughes equation [16] in order to observe the atomic transport behavior of In-Tl alloy. The Moelwyn-Hughes equation for viscosity of liquid mixture is given as

$$\eta = (c_1 \eta_1 + c_2 \eta_2) \left(1 - c_1 c_2 \cdot \frac{H_M}{RT} \right) \quad (18)$$

where, η_K is the viscosity of pure component K and for most liquid metals, it can be calculated from Arrhenius type equation [21] at temperature T as

$$\eta_K = \eta_{OK} \exp \left[\frac{E_n}{RT} \right] \quad (19)$$

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

2.5. Optimization of Free Energy of Mixing (G_M), Activity (a), Heat of Mixing (H_M), Concentration Fluctuations ($S_{cc}(0)$), Surface Tension and Viscosity

The model parameters which can be used to optimize thermodynamic, structural and transport properties of the liquid mixture can be obtained by thermodynamic description based on statistical thermodynamics or polynomial expressions. The adjustable coefficients, used in the process, are estimated by least square method which gives an idea to extrapolate into temperature and concentration region in which the direct observed determination is unavailable.

Redlich-Kister polynomial equation gives the composition dependence of excess free energy of mixing and is given by

$$G_M^{XS}(c, T) = c(1-c) \sum_{l=0}^m K_l(T) [c - (1-c)]^l \quad (20)$$

with

$$K_l(T) = A_l + B_l T + C_l T \ln T + D_l T^2 + \dots \quad (21)$$

The coefficients K_l are the function of the temperature.

The values for the free energy of mixing (G_M) of In-Tl liquid alloy at different temperatures can be calculated from Equation (1) by knowing the values of ordering energy parameter (ω) at different temperatures from the relation [23]

$$\omega(T_K) = \omega(T) + \frac{d\omega}{dT}(T_K - T) \quad (22)$$

where, $\omega(T)$ is the order energy parameter at the temperature 723 K, $\omega(T_K)$ is order energy parameter at required temperature T_K , and $\frac{d\omega}{dT}$ represents temperature derivative of order energy parameter which has already been estimated from observed data of entropy of mixing (S_M) of In-Tl liquid alloys at 723 K from Hultgren *et al.* 1973 [24].

3. Results and Discussion

3.1. Theoretical Investigation of Different Properties of In-Tl Liquid Alloy at 723 K

The value of main input parameters *i.e.* interchange energy (ω) and $\frac{1}{R} \frac{\partial \omega}{\partial T}$ used for the calculation of thermodynamic properties of In-Tl liquid alloy at 723 K were computed from Equations (1) and (9) by using observed values [24] of G_M and H_M respectively. The best fit parameters were found to be

$$\frac{\omega}{RT} = 0.52, \quad \frac{1}{R} \frac{\partial \omega}{\partial T} = 0.15$$

The positive value of interaction energy indicates that In-Tl is segregating in nature. Using above interchange energy (ω), we computed free energy of mixing (G_M), activity (a) entropy of mixing (S_M), heat of mixing (H_M), concentration fluctuation ($S_{cc}(0)$), ratio of diffusion coefficient (D_M/D_{id}), short-range order parameter (α_r), surface tension (Γ_1) and viscosity (η) at 723 K using regular solution model.

The plot of observed and computed values of free energy of mixing with respect to the concentration of Tl is shown in **Figure 1**. The computed and observed values [24] of free energy of mixing are in good agreement in the entire concentration range. Both the computed and observed value of free energy of mixing are minimum at $C_{Tl} = 0.5$, which indicates that In-Tl alloys in liquid state at 723 K is symmetric at equiatomic composition.

We computed the activities of In and Tl in the liquid In-Tl alloys using the same energy parameter (ω) as used in Equation (1), and then compared them with observed values as depicted in **Figure 2**. The plot shows that there is a good agreement between the sets of computed and observed results. Activity coefficient represents the measure of tendency of a component to leave the solution and it provides correlation of the behavior of the systems.

The heat of mixing and entropy of mixing of In-Tl binary liquid alloy at 723K were computed from Equations (9) and (7) respectively. To determine heat of mixing (H_M) and entropy of mixing (S_M) we need temperature derivatives of energy parameters. The observed values of heat of mixing [24] were utilized to obtain the temperature derivatives by the successive approximation. The best fit parameters was found to be $\frac{1}{R} \frac{\partial \omega}{\partial T} = 0.15$. Both computed and observed values

of heat of mixing and entropy of mixing are shown in **Figure 3** and **Figure 4**. Both the computed and observed value of heat of mixing and entropy of mixing are maximum at $C_{Tl} = 0.5$. The positive value of heat of mixing indicates the segregating nature of the alloy which is in agreement with the sign of the interaction energy ω . The value of entropy of mixing is positive in whole range of concentration as depicted in **Figure 4**.

We computed concentration fluctuation in the long wavelength limit $S_{cc}(0)$ using Equation (11) with the help of same energy parameter ω for the consistency as used for the computation of free energy of mixing, activity and heat of mixing. The observed $S_{cc}(0)$ were computed from Equation (12). It was found that the computed and observed values of $S_{cc}(0) > S_{cc}^{id}(0)$ at all compositions. This indicates that segregation is favored in the In-Tl alloy at 723 K. The computed values of $S_{cc}(0)$ are in good agreement with the observed values in entire concentration range. The theoretical value is maximum at $C_{Tl} = 0.5$ (i.e. $S_{cc}(0) = 0.3378$) as depicted in **Figure 5**.

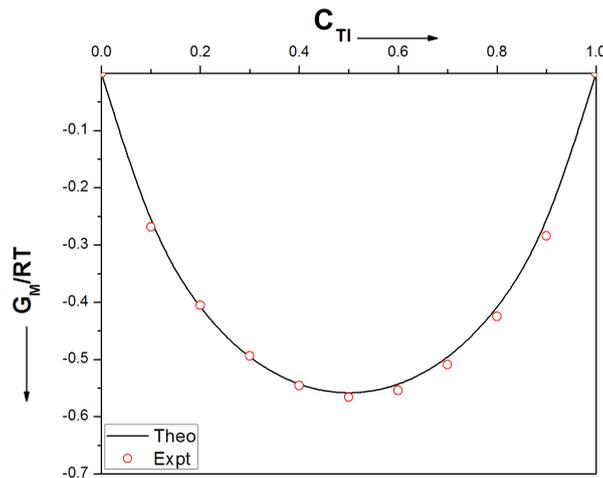


Figure 1. Compositional dependence of $(G_m/RT)_{\text{exp}}$ and $(G_m/RT)_{\text{Th}}$ of In-Tl liquid alloy at 723 K.

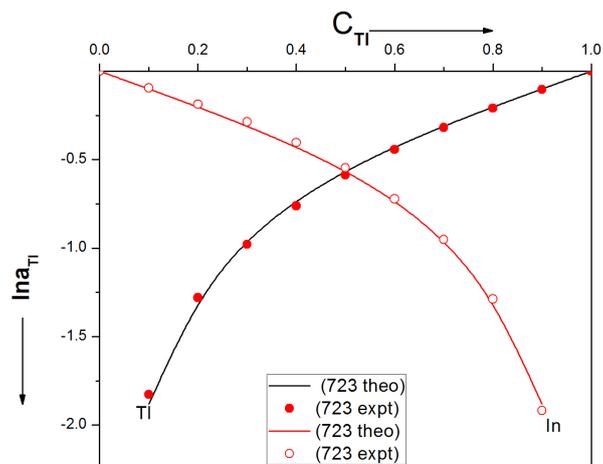


Figure 2. Compositional dependence of $\ln a_{(j)\text{exp}}$ and $\ln a_{(j)\text{Th}}$ of In-Tl liquid alloy at 723 K.

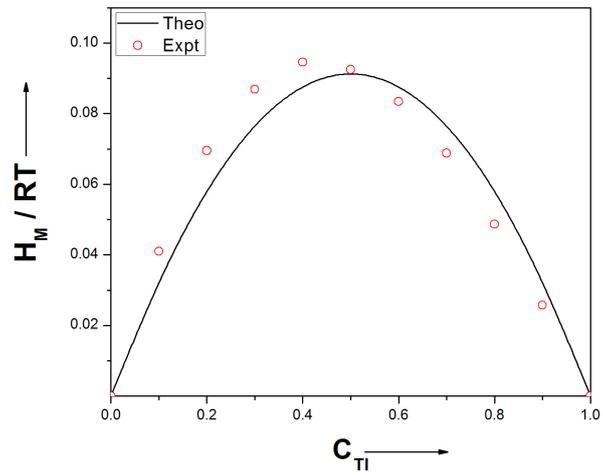


Figure 3. Compositional dependence of $(H_m/RT)_{\text{exp}}$ and $(H_m/RT)_{\text{Th}}$ of In-Tl liquid alloy at 723 K.

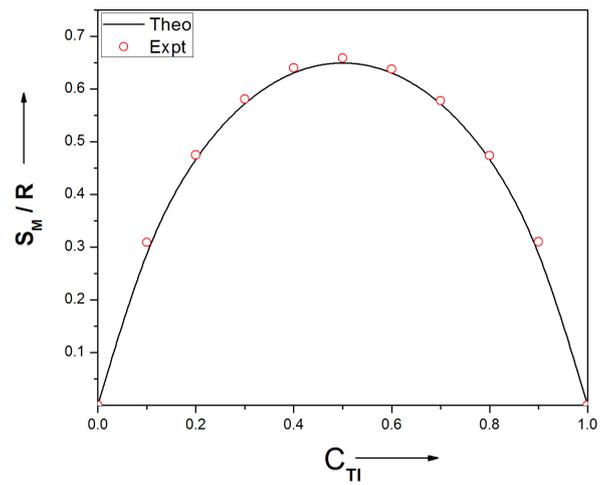


Figure 4. Compositional dependence of $(S_m/R)_{\text{exp}}$ and $(S_m/R)_{\text{Th}}$ of In-Tl liquid alloy at 723 K.

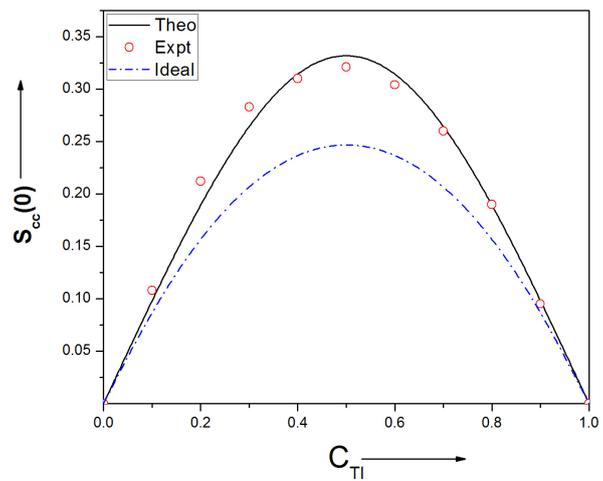


Figure 5. Compositional dependence of $(S_{cc}(0))_{\text{exp}}$ and $(S_{cc}(0))_{\text{Th}}$ of In-Tl liquid alloy at 723 K.

3.2. Theoretical Investigation of In-Tl at 623 K, 723 K, 800 K and 923 K by Optimization Method

By using the best fit value of $\frac{d\omega}{dT}$ and $\omega(T)$ at the given temperature $T = 723$ K in Equation (22), the value of $\omega(T_K)$ at temperature $T_K = 623$ K, 800 K, 923 K are estimated and listed in **Table 1**.

The values of free energy of mixing (G_M) of In-Tl liquid alloy at different temperatures (*i.e.* 623, 723, 800 and 923 K) computed by using the corresponding values of $\omega(T)$ in Equation (22) over the entire range of concentration and then they are used to calculate the corresponding excess free energy of mixing (G_M^{XS}) of the alloy at temperature of study by using Equation (20) conjugation with Equations (21) and (22).

The least-square method was used to calculate the parameters involved in Equation (21) and then the optimized coefficients for the alloy are computed which are listed in the **Table 2**.

We used the parameters *i.e.* k_0 , k_1 , k_2 and k_3 to obtain partial excess free energy. The partial excess free energy of mixing (G_M^{-XS}) of the components A (=In) and B (=Tl) were computed and tabulated below using equations [22]

$$G_{M,A}^{-XS}(C,T) = (1-C)^2 \sum_{l=0}^m K_l(T) [(1+2l)c - (1-c)] (2c-1)^{l-1} \quad (23)$$

and

$$G_{M,B}^{-XS}(C,T) = C^2 \sum_{l=0}^m K_l(T) [c - (1+2l)(1-c)] (2c-1)^{l-1} \quad (24)$$

The partial excess free energy of mixing of both the components In and Tl involved in In-Tl liquid alloy at different temperatures (*i.e.* = 623 K, 723 K, 800 K, 923 K) were calculated separately over the entire concentration range by Equations (23) and (24) with the help of optimized coefficients. With the help of this optimized partial excess free energy of mixing of both the components in the alloy have been used to calculate the corresponding excess free energy of the alloy at different temperatures over the entire range of concentration from the relation

$$G_M^{XS} = CG_{M,A}^{-XS} + (1-C)G_{M,B}^{-XS} \quad (25)$$

The optimized values of excess free energy of mixing for the alloys at different temperatures (*i.e.* = 623 K, 723 K, 800 K, 923 K) over the entire concentration range are shown in **Figure 6**.

Now, the activity coefficients (γ_i), ($i = \text{In or Tl}$) at different temperature over the entire range of concentration for In or Tl components have been computed from the relation $\bar{G}_{M,i}^{XS} = RT \ln \gamma_i$, with $\gamma_i = \frac{a_i}{c_i}$, where, a_i and c_i be the activity and concentration of the component respectively of In-Tl liquid alloy at corresponding temperature.

The calculated optimized values partial excess free energy of mixing, the corresponding activity coefficients and corresponding activity of both the compo-

nents involved in In-Tl liquid alloy in the entire concentration range at the temperature $T = 623\text{ K}, 723\text{ K}, 800\text{ K}, 923\text{ K}$ are shown in **Table 3**, **Table 4**, **Table 5** and **Table 6**.

The concentration fluctuations in long wavelength limit $S_{cc}(0)$ of the alloy at different temperatures in entire concentration range have been calculated using Equation (14) with the help of the optimized values of the activity of both the components which is shown in **Figure 7**. The values of concentration fluctuation decreases as the temperature of the alloy increases correspond to the concentration of Tl component. Also, $S_{cc}(0) > S_{cc}^{id}(0)$ at entire concentration range of Tl at all temperature of investigation which indicates segregating nature of the alloy.

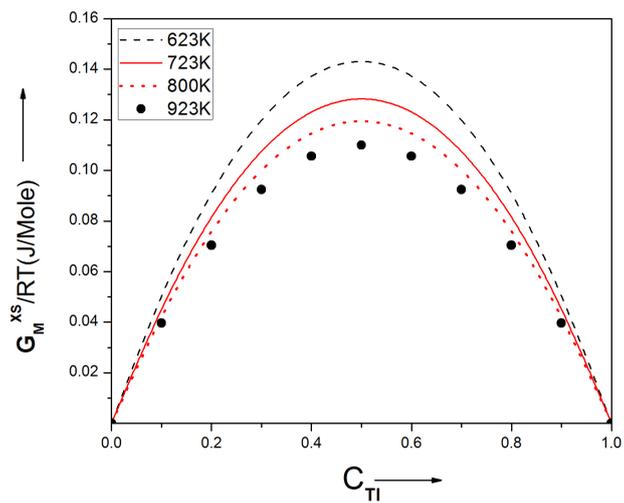


Figure 6. Compositional dependence of $\frac{G_M^{XS}}{RT}$ of In-Tl liquid at different temperatures.

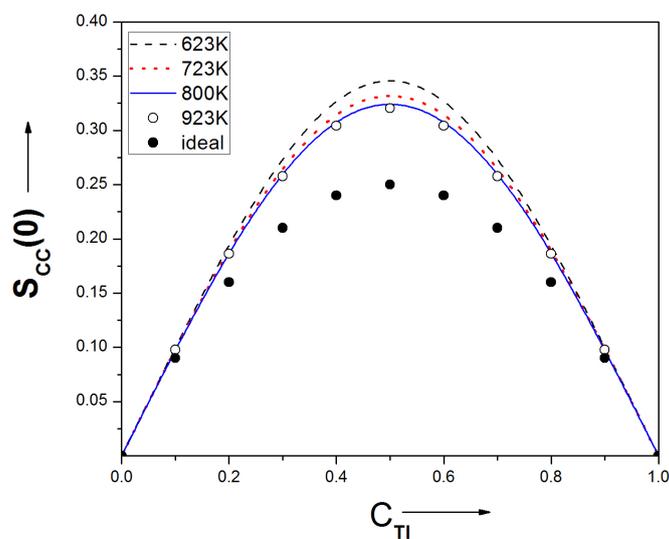


Figure 7. Graph for $S_{cc}(0)$ at different temperatures versus C_{Tl} of In-Tl liquid alloy (the ideal values in the graph are taken at 723 K).

Table 1. Estimated values of order energy parameter at different temperatures in In-Tl liquid alloy.

Temperature (T_K) in K	Order energy Parameter $\omega(T_K)/RT$
623	0.58
723	0.52
800	0.4843
923	0.44

Table 2. Calculated values of optimized coefficients A_i , B_i , C_i and D_i ($i = 0$ to 3) in liquid alloy In-Tl.

Values of i	A_i (Jmole ⁻¹ K ⁻¹)	B_i (Jmole ⁻¹ K ⁻¹)	C_i (Jmole ⁻¹ K ⁻¹)	D_i (Jmole ⁻¹ K ⁻¹)
0	2.345121281	-0.020964158	0.002942825	-1.29189E-06
1	3.88327E-14	-6.62413E-16	9.9353E-17	-6.28848E-20
2	0	0	0	0
3	3.74724E-14	-6.89401E-16	1.04971E-16	-7.35625E-20

Table 3. Optimized values of partial excess free energy of mixing, activity coefficients and activity of both the components involved in In-Tl liquid alloys at 723 K.

C_{Tl}	Tl-Component					In-Component				
	$\frac{\bar{G}_{M,Tl}^{AS}}{RT}$	$\ln Y_{Tl}$	Y_{Tl}	a_{Tl}	$\ln(a_{Tl})$	$\frac{\bar{G}_{M,In}^{AS}}{RT}$	$\ln Y_{In}$	Y_{In}	a_{In}	$\ln(a_{In})$
0.1	0.4212	0.4212	1.5238	0.15238	-1.8814	0.0052	0.0052	1.0052	0.9047	-0.10016
0.2	0.3328	0.3328	1.3949	0.27897	-1.2766	0.0208	0.0208	1.021	0.8168	-0.20234
0.3	0.2548	0.2548	1.2902	0.38706	-0.9492	0.0468	0.0468	1.0479	0.7335	-0.30987
0.4	0.1872	0.1872	1.2059	0.48235	-0.7291	0.0832	0.0832	1.0868	0.6521	-0.42763
0.5	0.1300	0.1300	1.1388	0.56941	-0.5631	0.1300	0.1300	1.1388	0.5694	-0.56315
0.6	0.0832	0.0832	1.0868	0.65206	-0.4276	0.1872	0.1872	1.2059	0.4823	-0.72909
0.7	0.0468	0.0468	1.0479	0.73354	-0.3099	0.2548	0.2548	1.2902	0.3871	-0.94917
0.8	0.0208	0.0208	1.021	0.81681	-0.2023	0.3328	0.3328	1.3949	0.279	-1.27664
0.9	0.0052	0.0052	1.0052	0.90469	-0.1002	0.4212	0.4212	1.5238	0.1524	-1.88139

Table 4. Optimized values of partial excess free energy of mixing, activity coefficients and activity of both the components involved in In-Tl liquid alloys at 623 K.

C_{Tl}	Tl-Component					In-Component				
	$\frac{\bar{G}_{M,Tl}^{AS}}{RT}$	$\ln Y_{Tl}$	Y_{Tl}	a_{Tl}	$\ln(a_{Tl})$	$\frac{\bar{G}_{M,In}^{AS}}{RT}$	$\ln Y_{In}$	Y_{In}	a_{In}	$\ln(a_{In})$
0.1	0.4698	0.4698	1.5997	0.15997	-1.83279	0.0052	0.0052	1.0052	0.9047	-0.10016
0.2	0.3712	0.3712	1.4495	0.28989	-1.23824	0.0208	0.0208	1.021	0.8168	-0.20234
0.3	0.2842	0.2842	1.3287	0.39861	-0.91977	0.0468	0.0468	1.0479	0.7335	-0.30987
0.4	0.2088	0.2088	1.2322	0.49288	-0.70749	0.0832	0.0832	1.0868	0.6521	-0.42763
0.5	0.1450	0.1450	1.156	0.57802	-0.54815	0.1300	0.1300	1.1388	0.5694	-0.56315
0.6	0.0928	0.0928	1.0972	0.65835	-0.41803	0.1872	0.1872	1.2059	0.4823	-0.72909

Continued

0.7	0.0522	0.0522	1.0536	0.73751	-0.30447	0.2548	0.2548	1.2902	0.3871	-0.94917
0.8	0.0232	0.0232	1.0235	0.81878	-0.19994	0.3328	0.3328	1.3949	0.279	-1.27664
0.9	0.0058	0.0058	1.0058	0.90524	-0.09956	0.4212	0.4212	1.5238	0.1524	-1.88139

Table 5. Optimized values of partial excess free energy of mixing, activity coefficients and activity of both the components involved in In-Tl liquid alloys at 800 K.

C_{Tl}	Tl-Component					In-Component				
	$\frac{\bar{G}_{M,Tl}^{XS}}{RT}$	$\ln Y_{Tl}$	Y_{Tl}	a_{Tl}	$\ln(a_{Tl})$	$\frac{\bar{G}_{M,In}^{XS}}{RT}$	$\ln Y_{In}$	Y_{In}	a_{In}	$\ln(a_{In})$
0.1	0.3923	0.3923	1.4804	0.14804	-1.9103	0.0048	0.0048	1.0049	0.9044	-0.10052
0.2	0.3100	0.3100	1.3634	0.27267	-1.2995	0.0194	0.0194	1.0196	0.8156	-0.20377
0.3	0.2373	0.2373	1.2678	0.38035	-0.9667	0.0436	0.0436	1.0446	0.7312	-0.31309
0.4	0.1743	0.1743	1.1905	0.47619	-0.7419	0.0775	0.0775	1.0806	0.6483	-0.43334
0.5	0.1211	0.1211	1.1287	0.56435	-0.5721	0.1211	0.1211	1.1287	0.5644	-0.57207
0.6	0.0775	0.0775	1.0806	0.64834	-0.4333	0.1743	0.1743	1.1905	0.4762	-0.74194
0.7	0.0436	0.0436	1.0446	0.73119	-0.3131	0.2373	0.2373	1.2678	0.3803	-0.96667
0.8	0.0194	0.0194	1.0196	0.81565	-0.2038	0.3100	0.3100	1.3634	0.2727	-1.29949
0.9	0.0048	0.0048	1.0049	0.90437	-0.1005	0.3923	0.3923	1.4804	0.148	-1.9103

Table 6. Optimized values of partial excess free energy of mixing, activity coefficients and activity of both the components involved in In-Tl liquid alloys at 923 K.

C_{Tl}	Tl-Component					In-Component				
	$\frac{\bar{G}_{M,Tl}^{XS}}{RT}$	$\ln Y_{Tl}$	Y_{Tl}	a_{Tl}	$\ln(a_{Tl})$	$\frac{\bar{G}_{M,In}^{XS}}{RT}$	$\ln Y_{In}$	Y_{In}	a_{In}	$\ln(a_{In})$
0.1	0.3564	0.3564	1.3619	0.13619	-1.9937	0.0044	0.0044	1.0044	0.904	-0.10096
0.2	0.2816	0.2816	1.2764	0.25529	-1.3654	0.0176	0.0176	1.0178	0.8142	-0.20554
0.3	0.2156	0.2156	1.2055	0.36164	-1.0171	0.0396	0.0396	1.0404	0.7283	-0.31707
0.4	0.1584	0.1584	1.1472	0.45886	-0.779	0.0704	0.0704	1.0729	0.6438	-0.44043
0.5	0.1100	0.1100	1.1100	0.55002	-0.5978	0.1100	0.1100	1.1163	0.5581	-0.58315
0.6	0.0704	0.0704	1.0629	0.63775	-0.4498	0.1584	0.1584	1.1716	0.4687	-0.75789
0.7	0.0396	0.0396	1.0349	0.72444	-0.3224	0.2156	0.2156	1.2406	0.3722	-0.98837
0.8	0.0176	0.0176	1.0154	0.8123	-0.2079	0.2816	0.2816	1.3252	0.265	-1.32784
0.9	0.0044	0.0044	1.0038	0.90344	-0.1015	0.3564	0.3564	1.4282	0.1428	-1.94619

The natural logarithms of optimized values of activity of both the components in the alloy at different temperatures in the entire concentration range are shown in **Figure 8**.

The Warren-Cowley chemical short-range order parameter α_i is used to explain the degree of local arrangement of atoms in the mixture. The chemical

short-range order parameter was computed from Equation (13) using coordination number, $Z = 10$ for liquid the alloy at 723 K. The value of short range order parameter has been found positive in all concentration range which indicates that the alloy is segregating at all compositions. The value of short range order parameter has been found maximum at $C_{Tl} = 0.5$ at 723 K as shown in **Figure 9**. We have computed chemical short-range order parameter at temperatures 723 K, 623 K, 800 K and 923 K. The value of short range order parameter has been found maximum at $C_{Tl} = 0.5$ at all temperature of study as shown in **Figure 9**. The plot depicts that α_1 is positive in the entire range of concentration showing that α_1 in In-Tl is segregating.

The surface tension of the liquid alloys can be computed using Equation (14) conjugation with Equations (15) and (16) using Buttler's model [15] [20] [25]. The ratio of partial excess Gibbs energy in the bulk and that in the surface can be expressed as $\beta = \frac{G_i^{E,s}}{G_i^{E,b}}$, β has been taken as 0.83 [26] [27]. We have taken the surface tension of In-Tl and temperature coefficients for pure In and Tl components from the reference [28]. The surface tension of the pure component at temperature of investigation has been calculated using the relation.

$\Gamma(T) = \Gamma_m + \frac{\partial \Gamma}{\partial T}(T - T_m)$, where, $\frac{\partial \Gamma}{\partial T}$ ($= -0.09 \text{ mNm}^{-1}\text{K}^{-1}$ for In, $-0.08 \text{ mNm}^{-1}\text{K}^{-1}$ for Tl) is temperature coefficient of surface tension, T_m ($= 430 \text{ K}$ for In and 577 K for Tl) is melting temperature and $T = 723 \text{ K}$.

The partial excess free energy of mixing of the pure components was taken from Hultgren *et al.* [24]. It was found from the analysis that the computed surface tension for In-Tl system at 723 K is less than ideal value ($= X_1\Gamma_1 + X_2\Gamma_2$) at all the concentration of In *i.e.* there is negative departure of surface tension from ideality at 723 K. It was found that surface tension increases with increase in the concentration of component Tl on the alloy as shown in the **Figure 10**.

To find the surface tension (Γ) of the alloy at different temperature we have used the values of optimized partial excess free energy *i.e.* \bar{G}_M^{XS} from the **Tables 3-6**, and using Buttler's Equation (16) conjugation with Equation (17). The calculated values of surface tension at different temperatures are shown in **Figure 10**. The surface tension of the alloy in the concentration range of Tl component showed that as the temperature increases, the surface tension decreases.

Viscosity of pure component In and Tl at 723 K are calculated using Equation (19) which is required to compute viscosity of In-Tl alloy at 723 K with the help of the value of the constants E and η_{ok} for the metals [28]. To compute viscosity using Moelwyn-Hughes equation [16], heat of mixing H_M is required which is taken from regular solution model calculations. The result of viscosity at 723 K of the alloy with the ideal values ($\eta = c_A\eta_A + c_B\eta_B$) is shown in **Figure 11**. The graph shows that with the increase in concentration of Tl component in the alloy, the viscosity of the alloy decreases (**Figure 11**).

The optimized viscosity of the alloys is calculated by the Moelwyn-Hughes

Equation (22) conjugation with Equation (23). The values of optimized H_M/RT is used in whole concentration range obtained using Equation (11) with the help of optimized values of $\omega(T)$ and $\frac{d\omega}{dT}$ at temperature of study. The values of parameters on Equation (23) is taken from [28]. The plot exhibits that the viscosity show small negative deviation from ideality at all compositions at 723 K. The computed values of viscosity at different temperatures is as shown in **Figure 11**.

The diffusion coefficients $\frac{D_M}{D_{id}}$ of In-Tl liquid alloy at temperatures of study in the entire concentration range can also be calculated using the optimized data from the relation (19) which are shown in the **Figure 12**. The $D_M/D_{id} < 1$ which indicates that there is tendency of phase separation.

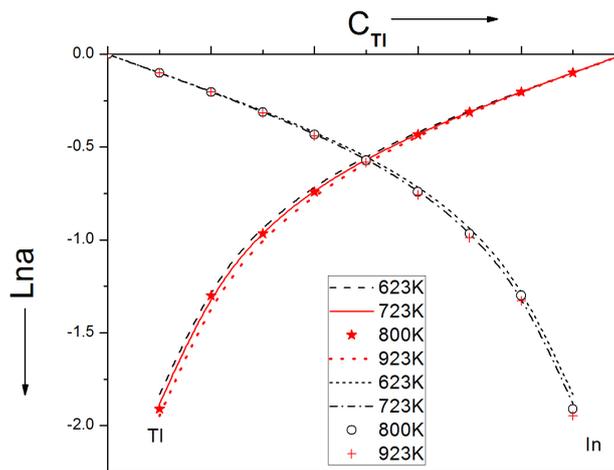


Figure 8. Graph for $\ln a$ at different temperatures versus C_{Tl} of In-Tl liquid.

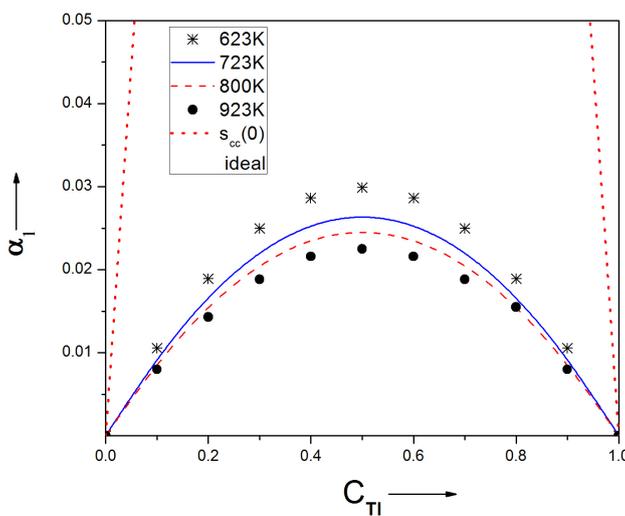


Figure 9. Graph for short range order parameter *i.e.* α_1 at different temperatures versus C_{Tl} of In-Tl liquid alloy (the ideal values in the graph are taken at 723 K).

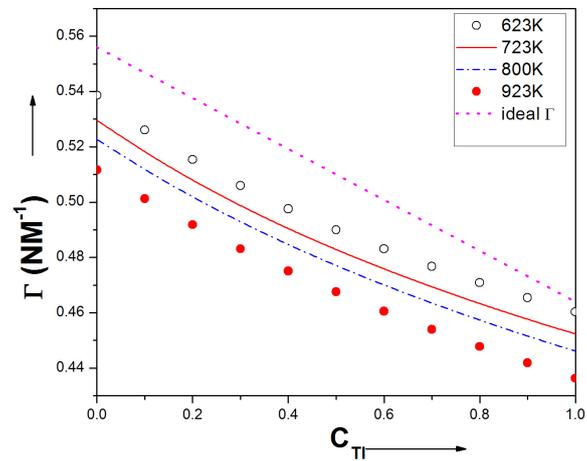


Figure 10. Graph for surface tension *i.e.* Γ at different temperatures versus C_{Tl} of In-Tl liquid alloy (the ideal values in the graph are taken at 723 K).

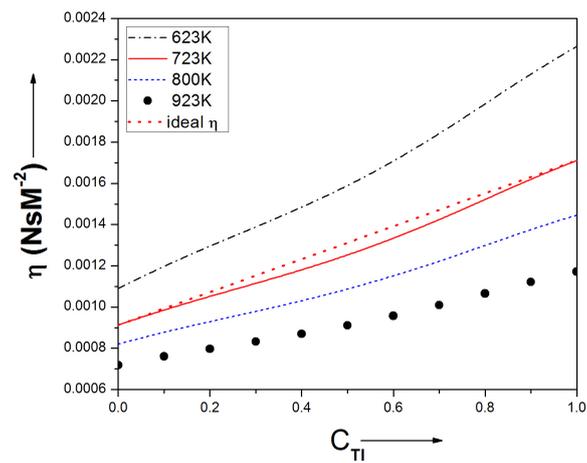


Figure 11. Graph for viscosity *i.e.* η at different temperatures versus C_{Tl} of In-Tl liquid alloy (the ideal values in the graph are taken at 723 K).

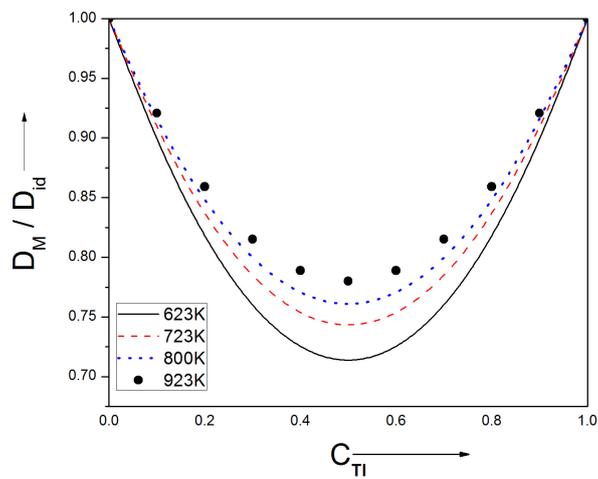


Figure 12. Graph for diffusion coefficient *i.e.* $\frac{D_M}{D_{Id}}$ at different temperatures versus C_{Tl} of In-Tl liquid alloy.

4. Conclusion

From the theoretical investigation following conclusions can be drawn:

- 1) Symmetry is observed in free energy of mixing, heat of mixing and entropy of mixing at all temperatures of investigations.
- 2) Activity decrease with the increase in concentration of Tl component in all temperatures of study *i.e.* 623 K, 723 K, 800 K and 923 K. And, with compare to temperature of study, it decreases as temperatures of investigation increases.
- 3) At all temperatures of investigation, the surface tension decrease with the increase of bulk concentration of Tl in Tl-In liquid alloy. In context of different temperatures, as temperature of study increases, it decreases.
- 4) Viscosity increases as the concentration of Tl component increases in the alloy. And, as temperature of study increases viscosity also increases.
- 5) The diffusion coefficients *i.e.* $D_M/D_{id} < 1$ at all compositions which indicates that there is tendency of phase separation

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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