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STRUCTURAL ASYMMETRY IN TWO THALLIUM-BASED ALLOYS: TI-Mg AND TI-Na

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ABSTRACT

Different modelled equations are used to analyze the variation of the thermodynamic, microscopic and surface characteristics between the Tl-Mg and Tl-Na liquid alloys at 1000 K by considering TlNa₂ and TlMg₂ complexes. The Thermodynamic properties are studied under Quasi-Chemical Approximation and the agreement between theoretical and experimental results of thermodynamic properties about their melting temperatures verifies the validity of model. The transport and surface properties of the alloys are studied by Kaptay and Butler models respectively. From the theoretical observations, it is shown that the Tl-Mg alloy is more interacting than that of the Tl-Na alloy and exhibits an ordering nature below 0.3 concentration of Tl, but above this concentration Tl-Na alloy shows more ordering tendency. The Tl-Mg has a higher viscosity than the Tl-Na alloy within whole concentration of Tl. On the basis of surface properties, Na segregates on the surface within entire concentration of Tl in Tl-Na alloy whereas Mg segregates on the surface below 0.35 concentration of Tl in Tl-Mg alloy.

Keywords: Ordering, segregating, surface segregation, thermodynamic properties

INTRODUCTION

The knowledge of molten alloys is crucial in metallurgical research in order to comprehend the nature of solid alloys and for developing novel materials for high-temperature applications. Individual chemical variations are often repressed, and new features emerge during the creation of alloys from the mixing of elemental metals. The atomic size, electrochemical effect and concentration all influence the solubility of metallic mixtures. However, these factors are not easily accessible (Bhandari et al., 2021; Novakovic et al., 2005). As a result, mixing properties are taken into account while evaluating the alloying behavior of liquid alloys that display nonideal behavior. Numerous binary alloys have intriguing behavior in terms of thermodynamic and electrical properties as a function of concentration (Koirala, 2018; Panthi et al., 2020).

Despite the fact that thallium is harmful to human health (Habashir, 1998), it is widely employed in the manufacturing of photoresistors, infrared optical equipment, and low melting glasses. Similarly, thallium oxide is utilized to make a high index of refraction. Likewise, different thallium-based alloys are used for significant industrial applications (Awe, 2018).

Thallium has a lower reactivity than sodium and magnesium (Akhbari & Morsali, 2010). The difference in electronegativity between thallium and magnesium is 0.69, but the difference between thallium and sodium is 0.31. From the phase diagram, the stable compounds of thallium magnesium and thallium sodium are TINa₂ and TIMg₂, respectively. Different thermodynamic, transport and surface characteristics of the two thallium-based

alloys are investigated in this work with the goal of finding the variation of characters as a function of concentration at temperature 1000 K from a theoretical standpoint. Though thermo physical properties some thallium-based alloys have been studied by Awe (2018), but it left out the study of TI-Mg and TI-Na liquid alloys. This paper aims to fulfill such lack too.

The compound forming model proposed by Bhatia and Singh (1982) is utilized to carry out thermodynamic research since it has been proven to be an effective tool in the investigation of various thermodynamic characteristics (Singh et al., 1987; Panthi et al., 2021). Theoretical and experimental findings of thermodynamic properties of the alloys are compared at about their melting temperature in order to validate the model. For this, the interaction energy parameters between the alloy's constituent elements of both the alloys at their melting temperatures are determined and later these are optimized at 1000 K to understand the variation of the properties of the alloys. Similarly, viscosity and surface tension are studied by Kaptay and Butler models respectively (Butler, 1932; Budai et al., 2007).

MATERIALS AND METHODS

Some binary alloys developed from the combination of the two metals X and Y display quick changes in the free energy of mixing for slight compositional alterations and form intermetallic alloys at one or more stoichiometric compositions. Hence, the phases of the alloys in this instance are known as an intermetallic compound and are typically expressed as $X_{\mu}Y_{\nu}$. In this regard, two binary liquid alloys viz. Tl-Na and Tl-Mg have been selected as study materials by assuming TINa₂ and TIMg₂ as most stable intermetallic compounds respectively.

Optimizing the interaction energy parameters of the relevant alloys is crucial for studying thermodynamic properties using the Quasi-Chemical Approximation. Such parameters of each of the alloys are optimized by the method of successive approximation with the aid of corresponding experimental values of Gibbs energy of mixing of the alloys and then used in the mathematical formulation of Gibbs free energy of mixing as derived in Subsection 2.1 to deal the property. Likewise, the experimental heat of mixing is utilized to optimize the temperature derivative interaction energy parameters.

Thermodynamic and Microscopic properties

If an alloy of constituent metals X and Y having chemical complexes of type $X_{\mu}Y_{\vartheta}$ (μ and ϑ are small integers) exists then the excess Gibbs free energy of mixing ($G_M^{X_S}$) of such alloy in the case of Quasi Chemical Approximation (Bhatia & Singh,1982) can be written as,

$$G_M = G_M^{AS} + \operatorname{RT}(\operatorname{C}_X \operatorname{lnC}_X + \operatorname{C}_Y \operatorname{lnC}_Y)$$
(1)

Similarly, the Gibbs energy of such complex alloy is given as,

$$\frac{G_M}{RT} = \emptyset \frac{\omega}{K_B T} + \emptyset_{XY} \frac{\Delta \omega_{XY}}{K_B T} + \emptyset_{XX} \frac{\Delta \omega_{XX}}{K_B T} + \emptyset_{YY} \frac{\Delta \omega_{YY}}{K_B T} + C_X \ln C_X + C_Y \ln C_Y \qquad (2)$$

Where $\phi_{i,j}'s(i, j = X, Y)$ simple polynomials in concentration (C), ω is ordering energy and $\Delta \omega_{i,j}$ are interaction energy parameters. The values of $\phi_{i,j}'s$ in the case of $\mu = 1$ and $\vartheta = 2$ are taken from Bhatia & Singh (1982) where $\phi_{XX} = 0$.

The interaction energy parameters at different temperatures are found by following relation: $a_{(1)}$ (T)

$$d[\omega_{ij}(T)]_{c} = \frac{\partial \omega_{ij}(T)}{\partial T} dT, \quad i \neq j$$

$$\omega_{ij}(T_{j}) = \omega_{ij}(T) + \frac{\partial \omega_{jk}}{\partial T} (T_{j} - T)$$
(3)

The enthalpy of mixing is related to the Gibbs free energy by standard thermodynamic equation as; $\begin{bmatrix} \partial G_{M} \end{bmatrix}$

$$H_{M} = G_{M} - T \left[\frac{\partial \omega_{M}}{\partial T} \right]_{C,N,P}$$

$$\frac{H_{M}}{RT} = \emptyset \left(\frac{\omega}{K_{B}T} - \frac{\partial \omega}{K_{B}\partial T} \right) + \emptyset_{XY} \left(\frac{\Delta \omega_{XY}}{K_{B}T} - \frac{\partial \Delta \omega_{XY}}{K_{B}\partial T} \right)$$

$$+ \left(\frac{\Delta \omega_{YY}}{K_{B}T} - \frac{\partial \Delta \omega_{YY}}{K_{B}\partial T} \right) \qquad (4)$$

The activity (a_i) of each component of binary liquid alloy is obtained from the Gibbs free energy by standard relation given as:

$$\ln a_i \left(i = X, Y \right) = \frac{G_M}{RT} + \frac{1 - C_i}{RT} \left[\frac{\partial G_M}{\partial C_i} \right]_{T, P, N}$$
(5)

The partial excess Gibbs free energy is derived from the activity of each component by the following relation (Li *et al.*, 2014).

$$G_i^{Xs} = RTln\left(\frac{a_i}{C_i}\right) \tag{6}$$

The concentration fluctuation in long-wavelength limit $(S_{CC}(0))$ is given as (Bhatia & Thornton, 1970):

$$S_{CC}(0) = RT \left[\frac{\partial^2 G_M}{\partial C^2} \right]_{T,P,N}^{-1}$$
(7)

The $S_{CC}(0)$ can also be found out by observed activities as,

$$S_{CC}(0) = C_Y a_X \left[\frac{\partial a_X}{\partial C_X} \right]_{T,P,N}^{-1} = C_X a_Y \left[\frac{\partial a_Y}{\partial C_Y} \right]_{T,P,N}^{-1}$$
(8)

The $S_{CC}(0)$ thus obtained from observed activities is also called as experimental $S_{CC}(0)$. From equations (5) and (9), the theoretical $S_{CC}(0)$ can be obtained as,

$$S_{CC}(0) = \frac{RT}{\left(-2\omega + \varphi_{XY}^{\prime\prime} \Delta \omega_{XY} + \varphi_{XX}^{\prime\prime} \Delta \omega_{YY} + \frac{RT}{C_X C_Y}\right)} \quad (9)$$

Where \mathfrak{g}_{ij}'' are second concentration derivatives of \mathfrak{g}_{ij} . The chemical short range order parameter (\mathfrak{a}_1) is related to the ratio of concentration fluctuation in longwavelength limit (L) and coordination number (Z) (Cowley, 1950; Warren. 1969) as:

$$\alpha_1 = (R-1)[R(Z-1)+1]^{-1}$$
(10)

Where

 $R = \frac{S_{CC}(0)}{S_{CC}^{id}(0)}$ and $S_{CC}^{id}(0)$ is the ideal concentration fluctuation in long-wavelength limit for an alloy. *Z* is coordination number and its value is taken 10 (Guggenheim, 1952) for the liquid alloy study.

Transport property: Viscosity

Kaptay established the following viscosity equation for binary alloy after taking into account the relationship between cohesive energy and the activation energy of the viscous flow (Budai *et al.*, 2007).

$$\eta = \frac{hN}{C_X V_X + C_Y V_B + V^{X_S}} \exp\left(\frac{C_X G_X + C_Y G_Y - 0.155 H_M}{RT}\right) (11)$$

Where h and N are Plank's constant and Avogadro's number, V_i (i = X, Y) is the molar volume of pure metal, V^{Xs} is excess molar volume upon alloy formation, G_i is Gibb's activation energy of viscous flow in pure metals. The following equation gives activation energy of a pure metal (Kaptay 2005).

$$G_{i} = RT \ln \left(\frac{\eta_{i} V_{i}}{hN}\right)$$
(12)

Where η and η_i are viscosity of the alloy and viscosity of individual elements A and B respectively. The variation of metallic viscosity with temperature is given as (Brandes & Brook, 2013):

$$\eta_{i} = \eta_{0} \exp\left(\frac{\xi}{RT}\right) \tag{13}$$

Where η_0 and ξ are constants of each metal having units of viscosity and energy per mole respectively.

Surface Properties: Surface segregation and Surface Tension

The surface tension (σ) of binary alloy at temperature T is given by the Butler model (Butler, 1932) as:

$$\sigma = \sigma_i + \frac{RT}{\rho_i} \ln \frac{C_i^S}{C_i^b} + \frac{G_i^{S,XS} - G_i^{\rho,XS}}{\rho_i}$$
(14)

Where σ_i , ρ_i are surface tension, molar area of surface each liquid metal respectively. $G_i^{S,XS}$ and $G_i^{b,XS}$ are partial excess free energies in the surface and bulk of constituent elements of the alloy respectively and are related as $G_i^{S,XS} = \beta G_i^{b,XS}$. For the liquid phase the value of β is taken as 0.818 (Kaptay, 2005).

The molar surface area of i^{th} component is given as (Kaptay, 2005):

$$\rho_i = \chi \left(\frac{M_i}{\delta_i}\right)^{2/3} N^{1/3} \tag{15}$$

Where M_i , δ_i and χ are molar mass, density of constituent element at its melting temperature, and geometrical constant. The value of χ is obtained from volume packing fraction (f_V) and surface packing fraction (f_s) by the expression as:

$$\chi = \left(\frac{3f_V}{4}\right)^{\frac{2}{3}} \frac{\pi^{\frac{1}{3}}}{f_S}$$
(16)

For the liquid metal, the values of both f_V and f_S are taken as 0.66 and 0.906, respectively (Kaptay, 2005).

The density (δ_i) and surface tension (σ_i) of each constituent metal of the liquid alloy at any temperature (T) are expressed as ((Brandes & Brook, 2013):

$$\delta_i = \delta_0 + \frac{d\delta}{dT} \left(\mathbf{T} - T_0 \right) \tag{17}$$

$$\sigma_i = \sigma_0 + \frac{d\sigma}{dT} \left(T - T_0 \right) \tag{18}$$

Where δ_0 and σ_0 are density and surface tension of each component at its melting temperature (T_0) . Similarly, $\frac{d\delta}{dT}$ and $\frac{d\sigma}{dT}$ are respectively temperature coefficient of density and surface tension.

RESULTS AND DISCUSSION Thermodynamic properties

Equations (2), (4), and (5) are used to determine thermodynamic properties using a Quasi-Chemical Approximation. Initially the interaction parameters are determined at about melting temperature of the alloys (923 K for Tl-Mg and 673 K for Tl-Na) by successive approximation method using observed values in the concentration range 0.1 to 0.9 (Hultgren et al., 1973) in order to study the Gibbs energy of mixing. Now using Equation (2), the interaction parameters at 1000 K are determined for both the alloys. The interaction parameters thus determined are given in the Table 1.

Alloy system	Temperatures	Interaction parameters				
		ω/k_BT	$\Delta \omega_{XY}/k_BT$	$\Delta \omega_{YY/KBT}$		
	923 K	-1.415	-3.173	6.936		
Tl-Mg	1000 K	-1.371	-2.888	6.107		
	673 K	-1.185	-7.122	5.221		
Tl-Na	1000 K	-1.099	-3.514	0.573		

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The plot of Gibb's free energy versus concentration of Tl for both the alloys Tl-Mg and Tl-Na at their melting temperatures is given in Figure 1. From the figure, it is clear that the theoretical results of both the alloys are in good agreement with their respective experimental results. This proves the validity of the model. Similarly, the Tl-Na liquid alloy is more interacting than Tl-Mg liquid alloy. Figure 2 is the Gibbs free energy of both the alloys at 1000 K. The figure depicts that the Tl-Mg is more interacting up to 0.6 concentration of Tl but above this concentration the Tl-Na alloy is more interacting.

The temperature derivative interaction energy parameters required for enthalpy of mixing are acquired by successive approximation method by using Equation (4) and experimental results (Hultgren et al., 1973) within concentration range 0.1 to 0.9 as shown in the Table 2.

But such parameters are considered constant for small changes in temperature.

Figure 3 is the computed enthalpy of mixing of the alloys at their respective melting temperatures that nearly agree with respective experimental results. The more negative enthalpy of mixing of Tl-Na indicates that it releases more energy in comparison to Tl-Mg alloy during mixing of the components of the alloys. Similar results are observed at temperature 1000 K as shown in the Figure 4.

The knowledge of the deviation of constituent element from the ideal behavior can be studied by another thermodynamic property named as 'Chemical activity'. Equation (5) is used to calculate the chemical activities of components of the alloys. Figure 5 is the theoretically

computed activities of the components of the liquid Tl-Mg and Tl-Na alloys. The figure depicts that all the components of the alloys are negatively deviated from ideal behavior and the thallium present in the Tl-Na alloy deviates more than it present in the Tl-Mg alloy whereas Mg gets deviated more than Na.



Figure 1. Gibbs free energy vs. concentration of Tl for Tl-Mg and Tl-Na liquid alloys



Figure 2. Gibbs free energy vs. concentration of Tl for Tl-Mg and Tl-Na liquid alloys at 1000 K

Table 2. Temperature derivative Interaction energy parameters at different temperatures

		temperature derivative of interaction parameters			
Alloy system	temperatures				
		∂ω	$\partial \Delta \omega_{XY}$	$\partial \Delta \omega_{YY}$	
		$\overline{K_B \partial T}$	$K_B \partial T$	$K_B \partial T$	
Tl-Mg	923 K	-0.839	0.543	-3.828	
Tl-Na	673 K	-0.921	3.910	-8.994	

Theoretically concentration fluctuations in longwavelength limit ($S_{CC}(0)$) and chemical short range order parameter (α_1) are widely used to study the structural arrangement of the atoms in the binary alloy. The ($S_{CC}(0)$) provides the qualitative information where as α_1 provides quantitative information of local order of atoms in the alloys. The values of α_1 lie between -1 to + 1. For given concentration and temperature if $S_{CC}(0)$ < $S_{CC}^{id}(0)$ then α_1 is negative. In this situation the alloy is expected to have ordering nature and if $S_{CC}(0) >$ $S_{CC}^{id}(0)$, then α_1 is positive and expected nature of the alloy is segregating. The value of $S_{CC}(0)$ goes to be zero for strong interacting alloys. The graph of observed and theoretical values of $S_{CC}(0)$ for the alloys at their melting temperatures is shown in Figure 6 which suggests that both the alloy have ordering tendency within whole concentration of Tl. However, at 1000 K, the Tl-Na shows segregating tendency up to 0.1 concentration of Tl and above 0.1 to 0.65 concentration, it shows less ordering than Tl-Mg liquid alloy but above 0.6 concentration of Tl, it is more ordering than Tl-Mg as shown in the Fig. 7. Accordingly, the value of α_1 is positive for Tl-Na liquid alloy.





Figure 4. enthalpy of mixing vs. concentration of Tl for Tl-Mg and Tl-Na liquid alloys at 1000 K



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Figure 6. concentration fluctuation in long wavelength limit vs. concentration of Tl for Tl-Mg and Tl-Na liquid alloys



Figure 7. concentration fluctuation in long wavelength limit vs. concentration of Tl at 1000 K

Up to 0.1 concentration of Tl and its value is more negative at 0.5 concentration of Tl for Tl-Mg liquid alloy showing its maximum ordering tendency at such concentration as depicted in Fig. 8.



Viscosity

The concentration dependent of viscosity at temperature 1000 K is obtained by Kaptay model as shown in Figure 9. The value of V^Xs is considered zero due to the lack of experimental values (Budai et al., 2007; Jha et al.,

2016). From the figure it is clear that the viscosity of Tl-Mg alloy is higher than viscosity of Tl-Na alloy. This infers that more interatomic attractive forces between the atoms of Tl-Mg alloy exists than that of the atoms of Tl-Na alloy.



Figure 9. viscosity vs. concentration of T1 at 1000 K

Surface Segregation and Surface Tension

The surface properties (surface segregation and surface tension) of molten alloy are regarded as a crucial aspect in metallurgical research and industry for the processing as well as the development of advanced materials since these are concerned with problems related to the surface and the interface in the liquid metal process (Iida, 1994; Singh et al., 2014). The surface and interfacial features of molten metals are considered to be of wide importance in metallurgical processes in the processes of welding, casting, and solidification (Brackbill et al., 1992). To estimate the surface tension of the alloys from Butler model, the densities and surface tensions of each metal needed at 1000 K are determined using Equations (17) and (18) respectively whereas densities and surface tensions of each metal at their melting temperatures are taken from Brandes & Brook (2013). The bulk partial surplus free energy of mixing individual components of the alloys 1000K are taken from the reference (Hultgren

et al., 1952). The geometrical constant and the ratio $(G_i^{S,XS}/G_i^{b,XS})$ respectively are 1.061 and 0.8181 (Kaptay, 2005, Kaptay 2008). The computed surface segregation of Tl and surface tension of both the liquid alloys against bulk concentration of Tl are shown in Figures 10 and 11, respectively.

From the Figure 10, it is clear that the sodium atoms seem to segregate on the surface of liquid Tl-Na liquid alloy within whole bulk concentration of thallium but for the Tl-Mg liquid alloy, the magnesium atoms segregate on the surface below 0.35 concentration of thallium but above this concentration , the thallium atoms segregate on surface of the alloy. Similarly, Figure 11 shows that the surface tension of Tl-Mg alloy is greater than Tl-Na liquid alloy and remains nearly constant within entire concentration of Tl but for Tl-Na, it increases with increase in concentration of Tl.



Figure 10. surface segregation vs. concentration of Tl at 1000 K



Figure 11. surface tension vs. concentration of T1 at 1000 K

CONCLUSIONS

The present study is the theoretical comparision of structural, transport thermodynamic, and surface properties of two thallium-based binary liquid Tl-Mg and Tl-Na alloys at 1000 K temperature. The interaction parameters required for the thermodynamic and structural analysis of the alloys at 1000 K are determined from the optimised parameters by using experimental values at about the melting temperatures of the alloys. The study shows that the Tl-Mg alloy is more interacting at lower concentration of thallium. The more negative value of enthalpy of mixing of Tl-Na alloy indicates the release of more energy during the mixing of comoinents of the alloy. The Tl-Mg alloy seems to be more ordering than Tl-Na at lower concentration of Thallium. The sodium atoms of the Tl-Na alloy segregates on the surface over whole concentration of Tl but Mg atoms of Tl-Mg alloy segregates on the surface up to 0.35 concentration of thallium. The viscosity and surface tension of Tl-Mg alloy are more than that of Tl-Na.

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AUTHOR CONTRIBUTIONS

NP: conceptualization, analysis and interpretation of results; IBB: draft manuscript preparation; IK: review and final approval.

CONFLICT OF INTEREST

No conflicts of interest exist in relation to the research work presented in this study.

DATA AVAILABILITY STATEMENT

Data will be made available on request with a suitable reason.

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