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Reassessments of thermo-physical properties of Si-Ti melt at different temperatures

S. K. Yadav[1*], U. Mehta[1, 2], R. K. Gohivar[1, 2], A. Dhungana[1, 2], R. P. Koirala[1], D. Adhikari[1]
[1]Department of Physics, Mahendra Morang Adarsh Multiple Campus, T. U., Biratnagar, Nepal
[2]Central Department of Physics, Tribhuvan University, Kirtipur, Kathmandu, Nepal
*E-mail: yadavshashit@yahoo.com

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ABSTRACT

The compositional dependence of thermo-physical properties of Si-Ti liquid alloy has been reassessed using different modeling equations at temperatures 2000 K, 2400 K and 2473 K. The thermodynamic and structural properties of the system have been computed in the frame work of quasi-lattice test. The extent of surface segregation and surface tension of the liquid mixture have been computed using Butler's equations at afore mentioned temperatures. The results so obtained have been compared with the available literature database. Theoretical investigations show that the compound forming tendency of the system gradually decreases at elevated temperatures and hence it shows ideal behaviours, as expected.

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1. Introduction

The assessment of the thermo-physical properties of metallic solutions are of fundamental importance in the field of metallurgy to characterize, design and fabricate new materials with desired properties. The micro-granules along with the temperature, pressure and atmospheric conditions of the initial melts determine the physical strength, wettability, stability, ductility, electrical resistivity, etc. of the metallic complexes [1]. Therefore, the comprehensive knowledge of the energetic of the liquid mixtures is mandatory. This information cannot be solely obtained from the experimental measurements as they are tedious, expensive and often encounter difficulties to be handled at high temperatures. With these regards, the development of modeling equations to explain and predict the thermo-physical properties of liquid alloys serve as robotic tool in material science and engineering. Several theoretical models [1-8] so far have been developed to access these properties to develop thermodynamic database. In present work, we intend to apply quasi-lattice test [5, 9, 10] to study and predict the thermodynamic, structural and surface properties of Si-Ti liquid alloys at different temperatures.

Alloys of titanium have light weight, high ductility, low thermal conductivity, high melting temperature and high strength due to which they are used as functional materials in aerospace, automotive industries and various high temperature applications [11, 12]. Among which Si-Ti alloy provides useful dispersive strength and improves microstructure stability [11]. These desirable properties have drawn considerable attentions of the researchers [11, 13-17] working in this field to study the thermodynamic properties of liquid Si-Ti alloy. Topor and Kleppa (1986) studied the standard enthalpy of formation of Si_2Ti complex by high temperature calorimetric measurements and have presented the results for the solid state at 298 K. Tabaian et al. (2000) presented the results for the thermodynamic properties of the concerned system by the Langmuir effusion method for the alloy containing 0-58.7 mass% of Ti. Their results constitute the partial enthalpy and entropy of Si and activity coefficient of Ti in the alloy at 1873 K in the above mentioned concentration range. Further, they presented the expression for the integral excess free energy of mixing (G_M^{XS}) in the form of Redlich-Kister (R-K) [18, 19] polynomials and computed the activity of Si (a_{Si}) and G_M^{XS}. Later Tokunaga et al. (2004) used regular solution approximation based on the sub-lattice model to comprehend the Gibbs free for the individual phases in the binary and ternary systems of Ni-Si-Ti alloy on the basis of CALPHAD method and recalculated the phase diagram of Si-Ti alloy. In 2007, Kostov et al. calculated the activities of Si and Ti, partial and integral fee energy of mixing for the alloy at 2000 K, 2400 K and 2473 K using FactSage Thermo-chemical, a computer based software. Taking this database as reference, Awe et al. in 2011 computed the thermodynamic and structural properties of the system at aforementioned temperatures on the basis of statistical mechanical model. Recently, Safarian et al. in 2012 used a quasi-regular solution model to determine the thermodynamic activity of Si in Si-Ti liquid alloy and presented the results so obtained in graphical form. As per the available literatures, the complete dataset of thermo-physical properties of Si-Ti liquid alloy is not yet available till date.

In this work, therefore, we have employed the modeling equations of quasi-lattice test to compute the thermodynamic properties, such as excess free energy of mixing (G_M^{XS}) and activity (a_{Ti} and a_{Si}) and structural properties, such as concentration fluctuation in long wavelength limit ($S_{CC}(0)$), chemical short-range order parameter (α_1) and ratio of diffusion coefficients (D_M/D_{id}) of the alloy at different temperatures. Further, the surface properties, such as surface tension (σ) and the extent of surface segregation (x_i^S; $i = Ti$ and Si) have been computed using equations of Butler model [20]. The results so obtained have been compared with the available literature database. We have assumed the metallic complex Ti_3Si as a stable phase [11, 14, 15, 21] and computed the herein required model parameters.

The details of the modeling equations are presented in the Section 2, the results along with the necessary discussions are presented in the Section 3 and the conclusions of the present findings are summarized in the Section 4.

2. Formulations

Thermodynamic properties

Let one mole of the binary liquid solution consisting of $x_1(= c)$ atoms of A (=T_i) and $x_2(= 1 - c)$ atoms of B (=S_i). The quasi-lattice test can explain the thermodynamic properties of binary liquid alloys. According to quasi-lattice test, the expression for the thermodynamic functions having suitable chemical complex of the form $A_\mu B_\vartheta$ (where $\mu = 3$ and $\vartheta = 1$) can be given as [5, 9, 10, 22]

$$G_M^{XS} = Nk_B T\left[\phi \frac{\omega}{k_B T} + \phi_{AB} \frac{\Delta\omega_{AB}}{k_B T} + \phi_{AA} \frac{\Delta\omega_{AA}}{k_B T}\right] \tag{1}$$

where N is Avogardo's number, k_B is Boltzmann constant, T is the temperature and ω's are the ordering energies. $c_1(= c_{Ti})$ is the concentration of A, $c_2(= 1 - c = c_{Si})$ is concentration of B, and ϕ_{ij} $(i, j = A, B)$ are the simple polynomials in c depending upon μ. The ordering energy coefficient $\Delta\omega_{AA} = 0$, if $\mu = 1$ and ordering energy coefficent $\Delta\omega_{BB} = 0$, if $\vartheta = 1$ for the Ti_3Si liquid binary alloy (present case), the values of ϕ's are expressed as [9, 10]

$$\phi = c(1-c) \tag{2}$$

$$\phi_{AB} = (\frac{c}{5} + \frac{2c^3}{3} - c^4 - \frac{c^5}{5} + \frac{c^6}{3}) \tag{3}$$

$$\phi_{AA} = (-\frac{3c}{20} + \frac{2c^3}{3} - \frac{3c^4}{4} + \frac{2c^5}{5} - \frac{c^6}{6}) \tag{4}$$

The free energy of mixing (G_M) then can be expressed as

$$G_M = G_M^{XS} + RT[(c \ln c + (1-c)\ln(1-c)] \tag{5}$$

Activity is one of the thermodynamic functions which are obtained directly from experiment and can be interpreted to describe the stability of the metallic complex of the liquid mixture. Activity depends upon temperature, pressure and concentration of mixture. Although its value depends upon customary choices of standard state for the species, activity is treated as dimensionless quantity. It can be given as [5, 9, 10, 22]

$$\ln a_A = \frac{G_M}{RT} + (1-c)\left[\frac{\omega}{k_bT}\phi' + \frac{\Delta\omega_{AB}}{k_bT}\phi'_{AB} + \frac{\Delta\omega_{AA}}{k_bT}\phi'_{AA} + \ln\frac{C}{1-C}\right] \tag{6}$$

where $\phi' = \frac{\partial\phi}{\partial c}$. In similar manner, the activity of B (a_B) can be obtained from Equation (6) by substituting $\phi' = \frac{\partial\phi}{\partial(1-c)}$ and making the necessary adjustments.

Structural properties

The alloys are grown from the liquid state near to the melting temperatures of individual ingredients and hence are assumed to be disorder state exhibiting only short-range order interactions. The study of the structural properties, such as concentration fluctuation in long wavelength limit ($S_{CC}(0)$), Warren-Cowley short-range order parameter (α_1) and ratio of mutual to intrinsic diffusion coefficients (D_M/D_{id}) helps to understand the arrangement of atoms of the liquid mixture in the nearest neighbourhood. According to quasi-lattice model, the expression for $S_{cc}(0)$ can be given as [5, 9, 10]

$$S_{cc}(0) = \frac{c(1-c)}{1+\frac{c(1-c)}{K_BT}[\omega\phi'' + \Delta\omega_{AB}\phi''_{AB} + \Delta\omega_{AA}\phi''_{AA}]} \tag{7}$$

where $\phi'' = \frac{\partial^2\phi}{\partial c^2}$. α_1 and D_M/D_{id} can then be expressed in terms $S_{cc}(0)$ by the following relations [1, 6, 7]

$$\alpha_1 = \frac{S-1}{S(Z-1)+1} \quad \text{with } S = \frac{S_{cc(0)}}{S_{cc}^{id}(0)}; \text{ and } S_{cc}^{id}(0) = x_1x_2 \tag{8}$$

and

$$\frac{D_M}{D_{id}} = \frac{S_{cc}^{id}(0}{S_{CC}(0)} \tag{9}$$

where Z is the coordination number and is takes 10 in present work calculation.

Surface properties

The surface properties of the liquid mixture help to understand the metallurgical phenomenon, such as crystal growth, welding, gas absorption, nucleation of gas bubbles. As mentioned earlier, the Butler's model has been used to compute the surface tension of the concerned alloy. According to this model, the surface tension (σ) of a binary liquid solution at temperature T can be expressed as [20, 23]

$$\sigma = \sigma_1 + \frac{1}{\tau_1}\left(G_1^{E,s} - G_1^{E,b}\right) + \frac{RT}{\tau_1}(\ln x_1^s - \ln x_1) \tag{10}$$

$$\sigma = \sigma_2 + \frac{1}{\tau_2}\left(G_2^{E,s} - G_2^{E,b}\right) + \frac{RT}{\tau_2}(\ln x_2^s - \ln x_2) \tag{11}$$

where σ_1 and σ_2 are surface tensions, and τ 1 and τ 2 are the areas of the hypothetical layer at the surface of the pure components A and B respectively at temperature T. $G_i^{E,s}$and$G_i^{E,b}$, (i =1,2), are partial excess free energies x_i and x_i^s and are mole fractions of component i in the bulk and that in the surface respectively. The area of monatomic surface layer for the component i is obtained from the following relation

$$\tau_i = 1.091\, N_A^{1/3} V_i^{2/3}$$

where NA is Avogadro's constant and V_i represents the molar volume of the component i. The molar volume can be calculated from the atomic mass and density of the pure component at the temperature of investigation. The partial excess Gibbs energy in the bulk and that in the surface are assumed to have the same concentration dependence, they can be related to each other through a parameter λ as

$$\lambda = G_i^{E,s}/G_i^{E,b} \qquad (13)$$

The value of λ is considered to be 0.83 for the calculations.

3. Results and Discussion

The model parameters herein required are optimized using Equations (1-4) with the aid of reference values of G_M^{XS} [11] at temperatures 2000 K, 2400 K and 2473 K. The values so optimized are presented in Table 1. In order to provide validation of the present theoretical reassessment, the computed values of G_M^{XS}/RT were compared with the available literature datasets [11, 14, 16, 21, 24]. Awe et al. [16] obtained the thermodynamic and structural properties of the system considering the dataset of ref. [11] as experimental value, meanwhile, G_M^{XS} is expressed in the form of R-K polynomials [18] in the literatures of ref. [14, 21, 24]. In terms of such polynomials, the integral excess free energy (G_M^{XS}) of the liquid alloy is expressed as

$$G_M^{XS}(x,T) = c_1 c_1 \sum_{z=0}^{n} k_z(T)(c_1 - c_2)^z \qquad (14)$$

with$k_z(T) = A_z + B_z T + C_z \ln T + D_z T^2 + - -$; where A, B, C and D are the coefficient to be determined and k_z has the same temperature dependence as G_M^{XS}. The partial excess free energies of granules of the alloy then can be expressed as

$$G_A^{XS}(x,T) = c_2^2 \sum_{z=0}^{n} k_z(T)\,[(1+2z)c_1 - c_2](c_1 - c_2)^{z-1} \qquad (15)$$

and

$$G_B^{XS}(x,T) = c_1^2 \sum_{z=0}^{n} k_z(T)\,[c_1 - c_2(1+2z)](c_1 - c_2)^{z-1} \qquad (16)$$

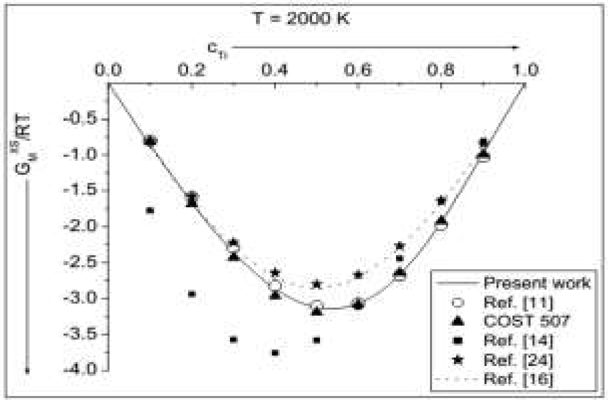
Table 1: Optimized model parameters (present work).

T (K)	$\Delta\omega/k_BT$	$\Delta\omega_{AB}/k_BT$	$\Delta\omega_{AA}/k_BT$
2000	0.67311	0.80990	0.82940
2400	-18.461	-15.383	-14.928
2473	37.609	31.333	30.403

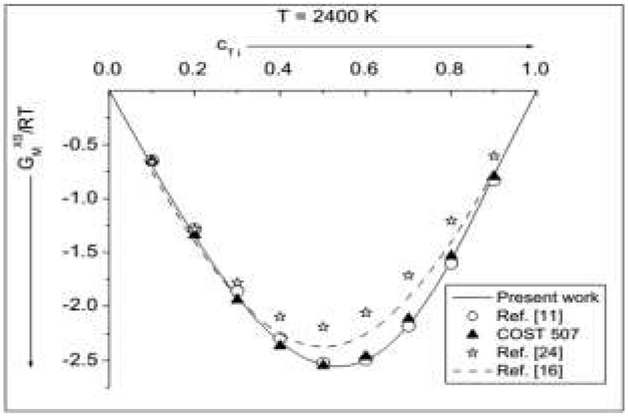
The values of G_M^{XS}/RT have been obtained using Equation (14) with the help of coefficients presented in the above mentioned references. The compositional dependence of the different values of G_M^{XS}/RT at above mentioned temperatures are plotted in Figures 1(a-c). The computed values of the present work is in well agreement with the reference dataset of Kostov et al. [11] and COST 507 [21] throughout the entire concentration range at preferred temperatures. These values slightly deviate with those of Awe at al. [16] and Tokunuga et al. [24] in the concentration range $0.41 < c_{Ti} < 0.86$ whereas that of Tabaian et al. [14] in the concentration range $c_{Ti} < 0.56$. Even though, the reference thermodynamic data is same in the present work and that in ref. 16, the computed values deviate. This may be due the modeling equations used in the present work have three model parameters whereas those used in ref. 16 has only one. Moreover, the values calculated from ref. 14 shows miscibility gap above 2000 K and hence are excluded.

Further, the plots of computed values of G_M^{XS}/RT as a function of concentration (c_{Ti}) gradually shallows with increase in temperature of the alloy (Figs. 1(a-c)). It corresponds that the compound forming tendency of the liquid alloy gradually decreases at higher temperatures and is found to be most interacting at its melting temperature (2000 K). The similar tendency has been predicted by several researchers in different alloy systems [1, 11, 16, 19, 25, 26].

The extent of compound forming tendency can further be interpreted in terms of activities of the free monomers (a_{Ti} and a_{Si}) of the liquid alloys. These values for the present work at different temperatures are obtained using Equations (5 and 6) with the help of the parameters from Table 1. To compute these values for the ref. 14, 21 and 24, at first the partial excess free energies of the monomers (G_{Ti}^{XS} and G_{Si}^{XS}) are obtained using the Equations (15 and 16) and the necessary parameters are taken from the respective references. The values so obtained are plotted as function of c_{Ti} in Figs. 2 (a-c).



1(a)



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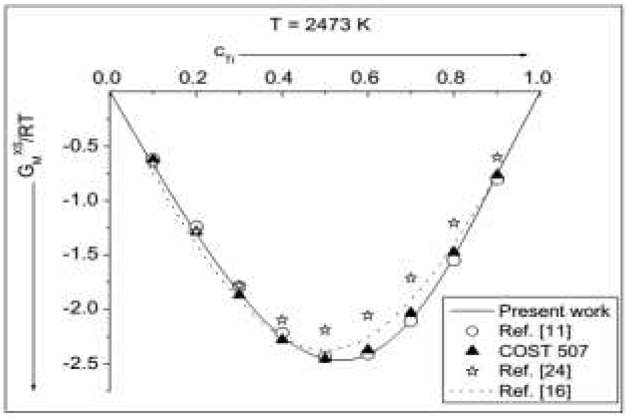
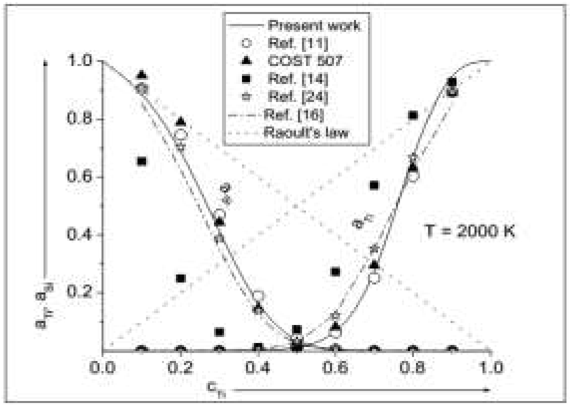
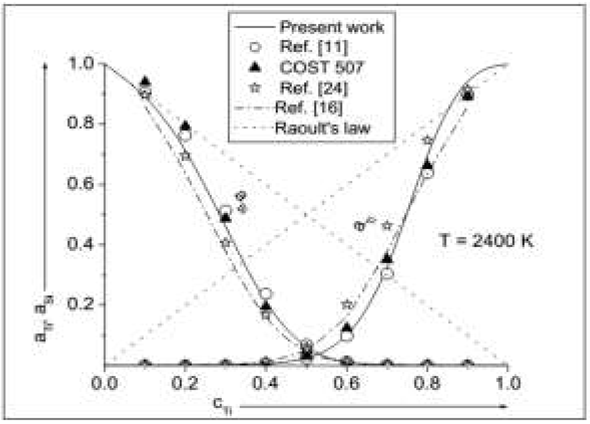


1(c)

Figs. 1(a-c): G_M^{XS}/RT versus c_{Ti} for Si-Ti liquid alloy at different temperatures.



2(a)



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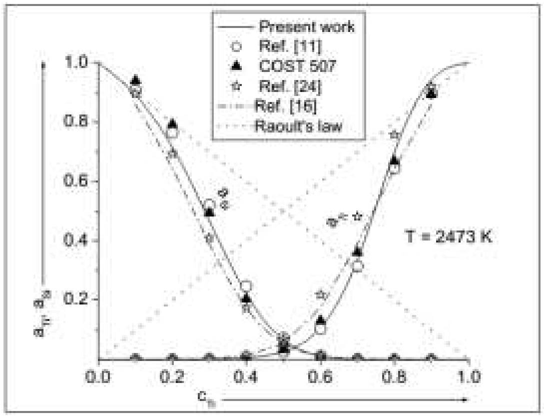


2(c)

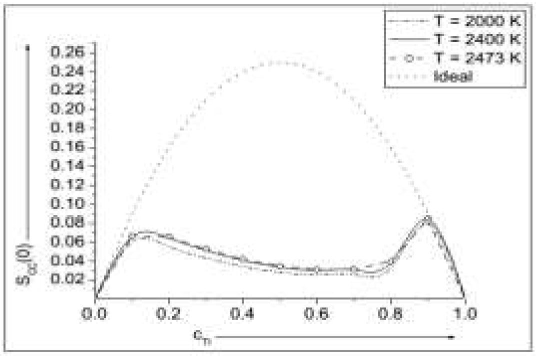
Figs. 2(a-c): a_{Ti} and a_{Si} versus c_{Ti} for Si-Ti liquid alloy at different temperatures.

The computed value of activity of the system for present work are in well agreement with those obtained from the ref. 11 and 21 and is in good agreement with those of ref. 16 and 24. Meanwhile, the activity computed using ref. 14 data are in poor agreement. Therefore, the reassessment of the thermodynamic data in present work is comparatively more reliable. Further, the activity curves show negative deviation from Raoult's law but this deviation gradually decreases with increase in the temperature of the alloy. These findings further strengthen the results obtained from the free energy of mixing as mentioned above.

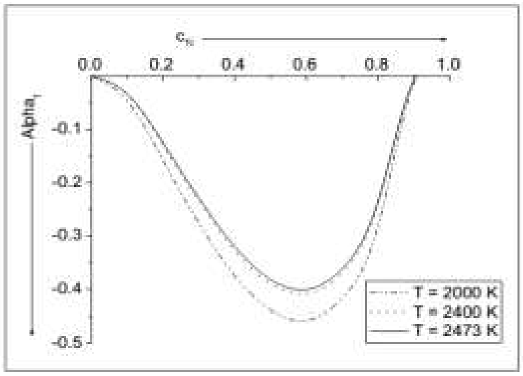
To understand the local arrangement of atoms in the liquid alloy, its structural properties ($S_{CC}(0)$, α_1 and D_M/D_{id}) have been computed using Equations (7-9) and values from Table 1. The compositional dependence of the values so obtained is plotted in Figs. 3(a-c). The computed values of $S_{CC}(0)$ gradually increases and ascends towards ideal values with raise in temperature above 2000 K (Fig. 3(a)). The curve of α_1 gradually shallows up and that of D_M/D_{id} shallows down predicting the similar trends as above (Fig. 3(b and c)). These results are in accordance with those of other researchers [1, 16, 26].

As per the available literatures, the surface properties of Si-Ti liquid alloy have been computed for the first time in present work. For this purpose, we have used Equations (10-13), herein required parameters have been taken from Table 2 (with Tq= 2000 K, 2400 K and 2473 K) and ref. 11, 21 and 24.

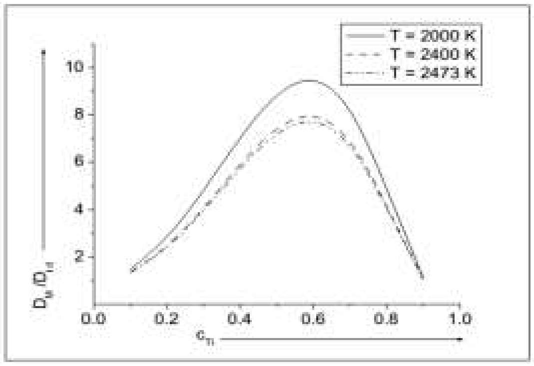
The values of σ, x^S_{Ti} and x^S_{Si} for the concerned liquid alloy are plotted in Figs. 4(a and b). It can be observed that $x^S_{Ti} > x^S_{Si}$ which corresponds that Ti atoms segregate on the surface phase whereas Si atoms remain in the bulk phase of the liquid mixture. With the increase in the temperature of the mixture, Ti atoms gradually move towards the bulk phase and Si atoms move gradually move towards the surface phase in order to maintain equilibrium [1, 16, 25] (Fig. 4(a)). The surface tension of the mixture computed from above mentioned reference data gradually decreases with increase in its temperature (Fig. 4(b)). These findings further stand in favour to the conclusions drawn from the computations of thermodynamic and structural properties of the alloy. Due to unavailability of the experimental and literature data, the present findings of surface properties could not be compared. But the theoretical investigations of the present work are in accordance with that of earlier researches [1, 16, 25].



3(a)



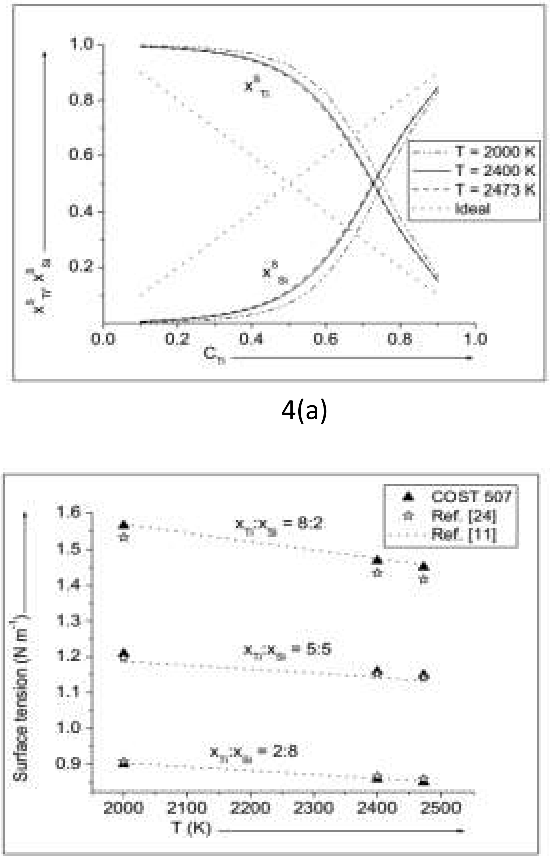
3(b)



Figs. 3(a-c): $S_{CC}(0)$, α_1 and D_M/D_{id}versus c_{Ti} for Si-Ti liquid alloy at different temperatures.

Table 2: Input parameters for surface tension [27].

Metal	Surface tension (Nm^{-1})	Density ($kg\ m^{-3}$)
Si	$0.85-0.13x10^{-3}(T_q-1683)$	$2510-0.32(T_q-1683)$
Ti	$0.165-0.26x10^{-3}(T_q-1958)$	$4110-0.702(T_q-1958)$



4(a)

4(b)

Figs. 4(a, b): Surface tension (σ) and surface concentration (x^S_{Ti} and x^S_{Si}) for Si-Ti liquid alloy at different temperatures.

4. Conclusions

A complete dataset of thermo-physical properties for Si-Ti liquid alloy at different temperatures have been presented in the present work. Theoretical investigations show that the alloy is most interacting and shows greatest mixing tendency at its melting temperature. This tendency, however, gradually decreases at higher temperatures. The preferred liquid alloy shows ideal mixing behavior at elevated temperature.

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