Optimization of thermodynamic and surface properties of ternary Ti–Al–Si alloy and its sub-binary alloys in molten state

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A R T I C L E  I N F O

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A B S T R A C T

Thermodynamic and surface properties of the ternary system were studied at different higher temperatures (1973 K, 2073 K, 2173 K and 2273 K). The thermodynamic properties were studied using the Chou, Kohler and Toop models. Meanwhile, the surface properties were investigated using the same models plus the Butler model. The thermodynamic and surface properties of the ternary alloys computed from different models were compared with one another. The surface concentration of the components of the ternary Ti–Al–Si alloy was computed using Butler’s equation.

1. Introduction

Knowledge of the thermo-physical properties of liquid alloys plays an important role in material design, fabrication and characterization in material science. Moreover, the development of a thermodynamic database by theoretical models serves as a fundamental for experimental procedures. The demand for new ternary alloys is growing day by day because of their wide applicability in different fields. Although alloys are synthesized by experimental methods, knowledge of the thermodynamic or energetic of the initial melts is most essential. This information cannot be entirely obtained from experimental measurements as being cumbersome, expensive, tedious and time-consuming. In addition, the use of computer-based softwares, such as Calphad, Thermocalc and FactSage, etc. for developing a thermodynamic database have proven to be very efficient; however, they also cannot provide all the required information. Therefore, the development and use of different theoretical models [1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11] for the optimization of different reliable data are mandatory.

Titanium alloys have a unique set of physical and mechanical properties, such as high strength-to-weight ratio, very good corrosion resistance and excellent biocompatibility, which make them suitable for use in orthopedic and dental implants [12], automotive industry, power plant turbines and aircraft engines [13]. When titanium aluminide is alloyed with silicon the material shows an interesting characteristic, such as low density, good thermal stability, high specific strength at high temperatures, a favorable ratio of mechanical properties to density and good resistance against oxidation at 600–800 °C [14, 15, 16, 17, 18], as a result of which it is appropriate for high-temperature construction materials. Furthermore, silicon is considered as an important alloying element for titanium aluminides, which can improve creep and oxidation resistance [19]. Commonly, Al–Si is treated as a foundry alloy and Ti is added to the foundry alloy because of its potential grain refining effect [20]. Besides the aforementioned promising features, the Ti–Al–Si alloys have very poor fluidity, so the alloy has many casting defects after casting [21, 22, 23].

In present work, the excess free energy of mixing and surface tension of the ternary Ti–Al–Si alloys in liquid state at different temperature have been computed using the Chou [24, 25], Toop [26] and Kohler models [26]. The activities of the monomers in the ternary Ti–Al–Si alloys have also been studied. The surface concentrations of the components of the ternary Ti–Al–Si alloys have been computed by using Butler’s model [27]. The theoretical outline for this work is provided in Section 2, the results and discussion are presented in Section 3 and the conclusion of this work is furnished in Section 4.

2. Theoretical modeling

The excess Gibbs free energy of mixing of binary liquid alloys can be expressed in terms of the Redlich-Kister (R–K) polynomial as [4, 26, 28, 29, 30].
where $A_i^j$ are the coefficients of the R–K polynomial of the binary systems ‘ij’ for the excess Gibbs free energy of mixing, and $x_i$ and $x_j$ are the mole fractions of the components in the binary alloy.

The partial excess Gibbs free energy ($G_{xs}^i$) of the $i^{th}$ component in binary and multi-component alloys is related to the excess free energy of mixing as [4, 26, 29, 30, 31, 32].

$$G_{xs}^i = G_{xs}^i + \sum_{j=1}^{n} (\delta_j - x_j) \frac{\partial G_{xs}^i}{\partial x_j}$$  \hspace{1cm} (2)

where $\delta_j$ is the Kronecker delta function.

According to the Chou model, the excess free energy of mixing for ternary systems can be given as [24, 25].

$$G_{xs}^{123} = x_{12}x_{23} \sum_{k=0}^{n} A_{k}^{12}(x_{1} - x_{2})^{k} + x_{2}x_{3} \sum_{k=0}^{n} A_{k}^{23}(x_{2} - x_{3})^{k} + x_{3}x_{1} \sum_{k=0}^{n} A_{k}^{31}(x_{3} - x_{1})^{k} + \delta_{12}x_{1}x_{2}x_{3}$$  \hspace{1cm} (3)

where $f$ is the ternary interaction term and can be given as [24, 25].

$$f = (2\delta_{12} - 1) + A_{1}^{2}(2\delta_{12} - 1)x_{1} + A_{3}^{2}(2\delta_{12} - 1)x_{2} + A_{1}^{3}(2\delta_{12} - 1)x_{3} + A_{1}^{1}(2\delta_{12} - 1)x_{1}x_{2}x_{3}$$  \hspace{1cm} (4)

The terms $x_1$, $x_2$, and $x_3$ are the mole fractions of the components in the ternary alloys. The values of partial excess free energy for binary liquid alloys are obtained using Eq. (2) in Eq. (1) by putting $m = 2$. Similarly, the values of partial excess free energy for ternary alloys are obtained using Eq. (3) in Eq. (1) by putting $m = 3$.

The partial excess free energy of the components in the alloy is related to their respective activities ($a_i$) as [25].

$$G_{xs}^i = RT \ln \left( \frac{a_i}{x_i} \right)$$  \hspace{1cm} (5)

where $R$ is the universal gas constant and $T$ is the absolute temperature. The surface tension of the ternary liquid alloy using the Butler equation is given as [4, 27].

$$\sigma = \sigma_1 + \frac{RT}{\rho_1} \ln \left( \frac{x_1}{\rho_0} \right) + \frac{G_{xs}^1 - G_{xs}^2}{\sigma_1 a_1} (i = 1, 2, 3)$$  \hspace{1cm} (6)

where $\sigma_1$ are the surface tensions of pure components, $a_i$ are the molar surface areas of pure components and $x_1$ and $x_2$ are the surface and bulk concentrations of individual components in the liquid mixture. $G_{xs}^i$ and $G_{xs}^j$ are the partial excess free energy for the surface and bulk phases, respectively, which are related as $G_{xs}^i = \rho G_{xs}^i$ [33]. The value of $\beta$ depends on the crystal structure and is taken as 0.82 [33] for the liquid phase. The molar surface area of the pure component in the alloy is given as [33].

$$a_i = 1.00N_A^{1/3} \left( \frac{M_i}{\rho_0} \right)^{2/3}$$  \hspace{1cm} (7)

where $N_A$ is the Avogadro’s number and $M_i$ and $\rho_0$ are the molar mass and density of the pure components of the liquid alloy. The temperature dependence of the surface tension (\(\sigma\)) and density (\(\rho\)) of the pure element in the liquid alloy are expressed as [34].

$$\sigma = \sigma_0 + \frac{\partial \sigma}{\partial T}(T - T_0) \text{ and } \rho = \rho_0 + \frac{\partial \rho}{\partial T}(T - T_0)$$  \hspace{1cm} (8)

where $\sigma_0$ and $\rho_0$ are the surface tension and density of the pure component at their melting temperature $T_0$ and $T$ is the temperature of interest.

$\frac{\partial \sigma}{\partial T}$ and $\frac{\partial \rho}{\partial T}$ are the temperature-derivative terms of the surface tension and density, respectively.

On the basis of Chou, the excess thermodynamic and surface functions ($z_{123}^e$) of the ternary liquid alloy can be given as [4, 24, 25].

$$z_{123}^e = \frac{x_1x_2}{X_1(1 - X_1)} z_{12}^e(X_1) + \frac{x_2x_3}{X_2(1 - X_2)} z_{23}^e(X_2) + \frac{x_3x_1}{X_3(1 - X_3)} z_{31}^e(X_3)$$  \hspace{1cm} (9)

Here, $x_i$ is the mole fraction of element $i$ in the ternary alloy. The values of $X_i$ are obtained using the relations

$$X_1 = x_1 + \xi_{12}x_1, X_2 = x_2 + \xi_{23}x_1, X_1 = x_3 + \xi_{31}x_2$$  \hspace{1cm} (10)

where $\xi_{ij}$ are the similarity indexes and are expressed as
The term $\eta$ is the deviation sum of squares and can be estimated as

$$\eta_i = \int_{0}^{1} (\bar{c}_{ij}^{\eta} - \bar{c}_{ij}^{\eta})^2 \, dX_i, \quad \eta_{ij} = \int_{0}^{1} (\bar{c}_{ij}^{\eta} - \bar{c}_{ij}^{\eta})^2 \, dX_i, \quad \eta_{ij} = \int_{0}^{1} (\bar{c}_{ij}^{\eta} - \bar{c}_{ij}^{\eta})^2 \, dX_i$$

(12)

The expressions for excess thermodynamic and surface functions for ternary liquid alloys on the basis of the Kohler model can be given as [4, 35].

$$\xi_{23} = \frac{\eta_{23}}{\eta_{2} + \eta_{3}}, \quad \xi_{2} = \frac{\eta_{2}}{\eta_{2} + \eta_{3}}, \quad \xi_{3} = \frac{\eta_{3}}{\eta_{2} + \eta_{3}}$$

(11)

The Toop model is an asymmetrical model in which a symmetrical element has to be chosen. According to this model, the expression for the excess thermodynamic and surface function for a symmetrical element $x_1$ is given as [4, 25, 35].

$$\xi_{23} = (x_1^2 + x_3^2)\xi_{23}^{\text{Toop}}(x_1) + (x_2^2 + x_3^2)\xi_{23}^{\text{Toop}}(x_2) + (x_1^2 + x_2^2)\xi_{23}^{\text{Toop}}(x_3)$$

(13)

The excess thermodynamic and surface functions ($\xi_{ij}^{\eta}$) for the binary system $ij$ present in the ternary alloy is expressed in terms of the R–K polynomial as

$$\xi_{ij}^{\eta} = X_i(1 - X_i) \sum_{k=0}^{n} L_{ij}^{k}(X_i - X_j)^k$$

(15)

where $L_{ij}^{k}$ are the optimized values of the coefficients of the R–K polynomial. The values of $X_1$, $X_2$ and $X_3$ are calculated using Eqs. (10), (11), and (12) for the Chou model. For the Kohler model, $X_1 = \frac{x_1}{x_1 + x_2}$, $X_2 = \frac{x_2}{x_2 + x_3}$, and $X_3 = \frac{x_3}{x_1 + x_3}$ and, for the Toop model, $X_1 = x_1$, $X_2 = x_2$, and $X_3 = x_3$ [25, 26].

on the basis of the Chou, Toop and Kohler models, the surface tension of the ternary liquid alloys is expressed as [24].

$$\sigma = \sigma^{\text{ideal}} + \sigma^{\text{mix}} + \sigma^{\text{excess}} + \sigma^{y}$$

(16)

where, $\sigma_1$, $\sigma_2$ and $\sigma_3$ are the surface tensions of the pure components of the liquid mixture.

3. Results and discussion

The temperature dependent coefficients of R–K polynomials for excess free energy of mixing ($G^E_{23}$) of Al–Si and Si–Ti sub-binary systems were optimized using Eq. (1) and their respective experimental data [36, 37]. Due to unavailability of the experimental value of $G^E_{23}$ for Al–Ti sub-binary system, the values of the R–K polynomials from Cost 507 [38] were considered for further calculations. The values of these coefficients are presented in Table 1. The computed and the observed (experimental and literature) values of $G^E_{23}$ of the sub-binary systems were found to be in well agreement (Figure 1). The excess free energy of mixing of all the aforementioned sub-binary systems was found to be negative at all compositions, which indicates that the complex forming tendency is preferred in these sub-binaries. It was clearly observed that the Si–Ti
Figure 3. (a) Variation of activity of Ti, Al and Si from Ti corner at \( \text{Al:Si} = 1:9 \) in Ti-Al-Si alloy at 1973 K. (b) Variation of activity of Ti, Al and Si from Ti corner at \( \text{Al:Si} = 3:7 \) in Ti-Al-Si alloy at 1973 K. (c) Variation of activity of Ti, Al and Si from Ti corner at \( \text{Al:Si} = 5:5 \) in Ti-Al-Si alloy at 1973 K. (d) Variation of activity of Ti, Al and Si from Ti corner at \( \text{Al:Si} = 7:3 \) in Ti-Al-Si alloy at 1973 K. (e) Variation of activity of Ti, Al and Si from Ti corner at \( \text{Al:Si} = 9:1 \) in Ti-Al-Si alloy at 1973 K. (f) Variation of activities of Ti, Al and Si with respect to temperature in Ti-Al-Si at Ti-60at%Al-12at%Si.
sub-binary system was a strongly interacting system followed by the Ti–Al and Al–Si systems.

The partial excess Gibbs free energy of individual components of the sub-binary systems was computed at 1973 K using the values of coefficients of the R–K polynomial [38] and the values from Table 1 for sub-binary systems Ti–Al, Al–Si and Si–Ti in Eqs. (1) and (2). These partial excess free energies were then used to compute the activities of the monomers of the above-mentioned sub-systems in Eq. (5). The computed activities of the monomers of the sub-binary systems are plotted in Figure 2 (a, b, c). It was found that the activities of components

Figure 4. (a) Variation of excess free energy (in KJ/mol) of mixing of Ti–Al–Si alloy from Ti corner at Al : Si = 1 : 9 in Ti–Al–Si alloy at 1973 K. (b) Variation of excess free energy (in KJ/mol) of mixing of Ti–Al–Si alloy from Ti corner at Al : Si = 3 : 7 in Ti–Al–Si alloy at 1973 K. (c) Variation of excess free energy (in KJ/mol) of mixing of Ti–Al–Si alloy from Ti corner at Al : Si = 5 : 5 in Ti–Al–Si alloy at 1973 K. (d) Variation of excess free energy (in KJ/mol) of mixing of Ti–Al–Si alloy from Ti corner at Al : Si = 7 : 3 in Ti–Al–Si alloy at 1973 K. (e) Variation of excess free energy (in KJ/mol) of mixing of Ti–Al–Si alloy from Ti corner at Al : Si = 9 : 1 in Ti–Al–Si alloy at 1973 K. (f) Variation of excess free energy of mixing of Ti–Al–Si alloy with respect to temperature at different corners.

Table 2. Input parameters for surface tension and density of pure elements [34].

| Element | $T_0$ (K) | $\rho_0$ (kg m$^{-3}$) | $\frac{\partial \rho}{\partial T}$ (kg m$^{-3}$K$^{-1}$) | $\sigma$ (Nm$^{-1}$) | $\frac{\partial \sigma}{\partial T}$ (Nm$^{-1}$K$^{-1}$) |
|---------|-----------|------------------|-----------------|----------------|------------------|
| Ti      | 1958      | 4130             | -0.23           | 1.65           | -0.00026         |
| Al      | 933       | 2385             | -0.35           | 0.914          | -0.00035         |
| Si      | 1683      | 2530             | -0.35           | 0.865          | -0.00013         |
of the Si–Ti system deviated most from the Raoult’s law, while those of the Al–Si system deviated least from the Raoult’s law. The activities of the components of the Ti–Al sub-system deviated moderately from the Raoult’s law as compared to other sub-systems. This result supports the conclusion made from the computed values of the excess free energy of mixing of the sub-binary systems regarding their interacting nature. It is evident from Figure 2 (a-c) that there is a negative deviation of the activities of the components of all sub-binary systems from the Raoult’s law at almost all compositions.

The activities of the monomers of the ternary Ti–Al–Si liquid alloy at five different cross-sections ($x_{Si} = x_{Ti} = 1:9, 3:7, 5:5, 7:3$ and $9:1$) were calculated from Ti corner ($x_{Ti}$) at 1973 K (Figure 3 (a-e)) using Eqs. (2), (3), and (4) and the values from Table 1. It was found that the activity of the Ti component increased with an increase in the concentration of Ti at all considered cross-sections in the ternary alloy, which is obvious. Similarly, the activity of Al was found to be increasing with increase in the concentration of Ti at the lower concentration of Ti. As the concentration of Ti increased, the activity of Al also increased and reached the maximum value. After reaching the peak, the activity of Al was found to be decreasing with increase in the concentration of Ti. As the proportion of Al in the ternary Ti–Al–Si alloys was increased the peak value of the activity of Al shifted towards the lower concentration of Ti. Regarding the activity of Si, it was found to have very small value as compared to that of Ti and Al and the variation with respect to the concentration of Ti was also very small. This result indicates that Si atoms are strongly bonded in the ternary alloy to form a complex. The variation in the activities of the monomers of the ternary Ti–Al–Si alloy with respect to temperature was also studied in the present work (Figure 3(f)).

The compositional dependence of the excess free energy of mixing ($G^{mix}_{mix}$) of the ternary Ti–Al–Si alloys in liquid state was computed at 1973 K.
K from Ti corners at five different cross-sections ($x_{Al}/x_{Si} = 1:9, 3:7, 5:5, 7:3$ and $9:1$) in the framework of Chou, Kohler and Toop models using Eqs. (3), (9), and (13) and values from Table 1. The values so computed were found to be in good agreement with one another (Figure 4(a-e)). The excess free energy of mixing of the ternary Ti–Al–Si alloy in liquid state was found to change significantly as the proportion of Al or Si changes. As the proportion of Al in the ternary Ti–Al–Si alloy was increased, the excess free energy of mixing was found to considerably decrease. For example, the excess free energy of mixing of the ternary alloy at cross-section $x_{Al}/x_{Si} = 1:9$ is about $-48$ KJ/mol while it is about $-13$ KJ/mol for the cross-section $x_{Al}/x_{Si} = 9:1$. However, the peak value of the excess free energy of mixing was found to be at the same concentration ($x_{Ti} = 0.5$) at all cross-sections in all cases. The variation of the excess free energy of mixing of the ternary Ti–Al–Si alloy in liquid state with temperature at different corners has also been studied in the present work (Figure 4(f)). The excess free energy of mixing was found to be changing linearly with temperature at all corners.

The compositional dependence of surface tension and surface concentration for the Ti–Al, Al–Si and Si–Ti sub-binary alloys was computed at 1973 K using Eq. (6) and the values given in Table 2 [34]. The compositional dependence of the surface concentrations of the components of the sub-binary systems are plotted in Figure 5(a-c). The surface concentration of Al was found to be greater than that of Ti in Ti–Al liquid alloy at 1758 K (Figure 5(a)) in the concentration range $x_{Ti} = 0.1 - 0.9$ indicating that Al atoms segregates in the surface phase and Ti atoms remains in the bulk phase in the initial melt. Moreover, the surface concentration of Al was found to decrease and that of Ti was found to increase with the increase in the concentration of Ti (Figure 5(a)). The surface concentration of Si was found to gradually decrease and that of Al was found to gradually increase with gradual increase in the bulk concentration of Al in Al–Si liquid alloy at 1973 K. They crossed each other at around $x_{Al} = 0.32$ (Figure 5(b)) corresponding that Si atoms gradually moved from the surface phase to the bulk phase and Si gradually moved from the bulk phase to the surface phase with increase in the concentration of Al. In Si–Ti liquid alloy, the surface concentration of Si gradually increases and Ti gradually decreases with increase in the concentration of Si (Figure 5(c)). They crossed each other at around $x_{Si} = 0.36$ indicating the gradual transfer of Ti atoms from the surface phase to the bulk phase and Si atoms from the bulk phase to the surface phase. The compositional dependence of the surface tension of the sub-binary systems are plotted in Figure 6. The computed value of the surface tension ($\sigma$) of Al–Si system at 1373 K at $x_{Si}$ ($Al_{0.3}Si_{0.7}$) was found to be in good agreement with available literature data; $\sigma = 0.816$ Nm$^{-1}$ (this work) and $\sigma = 0.800$ Nm$^{-1}$ (literature) [39, 40]. The computed value of the surface tension for Ti–Al system at 1758 K slightly deviated with the results of Xuyang et al. [41] using Butler model (Figure 6). The variation in these results is due to the difference in the input data of the surface tensions of the pure Al ($\sigma_{Al}$) and Ti ($\sigma_{Ti}$) components. Xuyang et al. took $\sigma_{Al} = 0.744$ Nm$^{-1}$ and $\sigma_{Ti} = 1.586$ Nm$^{-1}$ at 1758 K while $\sigma_{Al} = 0.625$ Nm$^{-1}$ and $\sigma_{Ti} = 1.702$ Nm$^{-1}$ [34] were taken in present work at the same temperature. Moreover, the values of the input parameters of Eq. (6), such as $\beta = 0.83$ and $f = 1.091$ were used by Xuyang et al., meanwhile $\beta = 0.82$ and $f = 1.00$ [33] were considered in this work. Even though, the deviation between these two results for the surface tension of the Ti–Al system at 1758 K is the minimum of 2.02% at $x_{Ti} = 0.9$ the maximum of 20.70% at $x_{Ti} = 0.2$, and hence are within considerable range. The surface tension of the Si–Ti system could not be compared due to the unavailability of the experimental data.

The excess surface tension ($\sigma^{ex}$) and surface concentration for the Ti–Al, Al–Si and Si–Ti sub-binary were computed using their respective partial excess free energy and the coefficients of the R–K polynomials for excess surface tension were optimized as a function of temperature in the range 1973 K–2273 K. The optimized values of the R–K polynomials for the sub-binary alloys are listed in Table 3. The surface concentration of the constituents of the ternary Ti–Al–Si alloy was computed from the corner of Ti at five different cross-sections ($x_{Al}/x_{Si} = 1:9, 3:7, 5:5, 7:3$ and $9:1$) (Figure 7(a-e)) at 1973 K using Eq. (16). The surface concentrations of Si and Ti were found to be increasing or decreasing with the increase or decrease of their respective bulk concentration in the ternary alloy. The surface concentration of Al was found to be increasing with increase in the bulk concentration of Ti at lower concentration region of Ti and reached the maximum value. After this peak value, the surface concentration of Al was found to be decreasing with increase in the concentration of Ti. The unexpected increment of surface concentration of Al with decrease in its bulk concentration was found in the region where interaction between Ti and Si was significantly high (Figure 7(b, c, d)). This is due to the strong ordering nature of the Si–Ti system and hence Al atoms do not take part in bonding with the Si and Ti. As a result of which Al atoms segregate in the surface. In Figure 7(e), surface concentration of Al seemed decreasing continuously due to very low concentration of Si so that interaction between Si and Ti was the minimum. The surface concentration of Al was relatively very high w.r.t. the relative bulk concentration. In equal compositions of the components in the ternary alloy, the surface concentration of Al was found to be about 0.79 while that of Si and Ti were found to be 0.19 and 0.02 respectively at 1973 K. While studying the variation of the surface concentration of the components of the ternary alloys with temperature it was found that the surface concentration of the components changed towards ideal value (bulk concentration) at elevated temperature (Figure 7(f)). The surface concentration of Al was found to be decreasing while that of Ti and Si was found to be increasing with the rise in temperature.

The excess surface tensions obtained for sub-binary were used in Eqs. (9), (10), (11), (12), (13), (14), and (15) to compute excess surface tension of the ternary Ti–Al–Si alloy. The computed excess surface tension for the ternary alloy was used in $\sigma^{ex} = \sigma - \sum_{j} (\sigma x_{j})$ to compute the

| System | Coefficients (Nm$^{-1}$) |
|--------|--------------------------|
| Ti–Al  | $A^0$ = -9.88 $\times$ 10$^{-7}$ T - 0.966 |
|        | $A^1$ = -1.67 $\times$ 10$^{-4}$ T - 0.240 |
|        | $A^2$ = -6.93 $\times$ 10$^{-5}$ T - 0.759 |
|        | $A^3$ = -2.24 $\times$ 10$^{-6}$ T - 0.890 |
| Al–Si  | $A^0$ = -1.60 $\times$ 10$^{-4}$ T + 0.222 |
|        | $A^1$ = 6.30 $\times$ 10$^{-5}$ T - 0.095 |
|        | $A^2$ = -3.07 $\times$ 10$^{-3}$ T + 0.035 |
|        | $A^3$ = 1.45 $\times$ 10$^{-3}$ T - 0.017 |
| Si–Ti  | $A^0$ = 2.58 $\times$ 10$^{-4}$ T - 0.473 |
|        | $A^1$ = 3.74 $\times$ 10$^{-4}$ T - 2.153 |
|        | $A^2$ = -3.21 $\times$ 10$^{-4}$ T + 0.826 |
|        | $A^3$ = -1.75 $\times$ 10$^{-4}$ T + 1.324 |
Figure 7. (a) Variation of surface concentration of different components with bulk concentration from Ti corners at \( Al:Si = 1:9 \) in Ti–Al–Si alloy at 1973 K. (b) Variation of surface concentration of different components with bulk concentration from Ti corners at \( Al:Si = 3:7 \) in Ti–Al–Si alloy at 1973 K. (c) Variation of surface concentration of different components with bulk concentration from Ti corners at \( Al:Si = 5:5 \) in Ti–Al–Si alloy at 1973 K. (d) Variation of surface concentration of different components with bulk concentration from Ti corners at \( Al:Si = 7:3 \) in Ti–Al–Si alloy at 1973 K. (e) Variation of surface concentration of different components with bulk concentration from Ti corners at \( Al:Si = 9:1 \) in Ti–Al–Si alloy at 1973 K. (f) Variation of surface concentration of the components of Ti–Al–Si alloy with temperature at composition ratio Ti – 60at%Al – 12at%Si.

surface tension of the alloy using Chou, Kohler and Toop models. Eq. (6) was used to compute the surface tension of the ternary alloy using Butler model. The surface tension of the ternary Ti–Al–Si alloy computed from the Ti corner at aforementioned cross-sections at 1973 K is plotted in Figure 8 (a-e). The surface tension computed using different models at all cross-sections were found to agree reasonably with each other. It is also evident from the figures that the surface tension of the alloy decreases with increase in the concentration of Ti. This is expected result because the surface of the pure liquid Ti is much higher than that of Al and Si at concerned temperature. The surface tension of the ternary Ti–Al–Si alloy at different compositions, such as Ti-90at%Al-3at%Si, Ti-56at%Al-20at%Si, Ti-30at%Al-49at%Si and Ti-10at%Al-89at%Si were computed using Chou model in the temperature range 1973 K-2273 K and plotted in Figure 8(f). It was found that the surface tension of the alloy was found to be decreasing linearly with increase in temperature at all compositions.

4. Conclusion

From the present work, the following conclusion can be drawn:

(a) The sub-binaries Ti–Al, Al–Si and Si–Ti of the ternary Ti–Al–Si alloys are all ordering in nature. The Si–Ti sub-binary system was strongly interacting system followed by Ti–Al and Al–Si systems.
(b) The variation of the activities of the monomers Ti, Al and Si of the ternary Ti–Al–Si alloy with respect to temperature was found to be almost linear. The activities of these monomers increase with increase in temperature.

(c) The excess free energy of mixing of the ternary Ti–Al–Si alloy in liquid state changes significantly as the proportion of Al or Si changes.

(d) The surface concentration of Al in the ternary Ti–Al–Si alloys does not vary with respect to the concentration of Ti as expected. The unexpected variation of surface concentration of Al may be due to significantly high interaction between Ti and Si.

(e) The surface tension of the alloy decreases with increase in the concentration of Ti. The surface tension of the alloy decreases linearly with increase in temperature at all compositions.

Declarations

Author contribution statement

S. K. Yadav, D. Adhikari: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Wrote the paper.

U. Mehta: Performed the experiments; Analyzed and interpreted the data.

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