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Phase segregation and surface properties in liquid Ga-Tl alloys predicted by modified self-association model

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Abstract

Using the self-association model, the effect of temperature on the segregation tendency in the bulk and surface of liquid Ga-Tl alloys was investigated. The ratio of the numbers of two types of atoms in the cluster (n) and the interchange energy (W) in the self-association model were two crucial factors, but it was unknown how these values changed with temperature, which limited the model's capacity for prediction. The current investigation revealed that the exchange energy was linearly related to temperature and that the ratio of the two types of atoms in the cluster was temperature-independent, which was verified by the good agreement of the predicted thermophysical properties, miscibility gap, and surface properties with the available experimental data. This work enhances the theoretical explanation of the self-association model and provides a straightforward method for forecasting liquid immiscible alloys across a wide temperature spectrum.

Key words: liquid Ga-Tl, self-association model, surface property

1. Introduction

As evidenced by lead-free solders [1], superalloys [2–4], and biomedical materials [5, 6], it is essential to comprehend metal characteristics and how they affect alloys to improve current alloys or create new ones. However, experiments on thermodynamic and thermophysical characteristics are challenging or unattainable for metals with high melting points, strong chemical reactivity, and oxygen affinity. In recent decades, appropriate instrumentation and experimental progress have provided reliable property data crucial for microstructure modeling and accurate material design [3, 7, 8]. As a result, the missing data can be inferred using the model predictions or by extrapolating from values obtained through experimentation, combining theory, modeling, and data [3].

Phase separation of liquid immiscible alloys has received considerable interest [9–12]. The core-shell structure is a common solidified microstructure in immiscible liquid alloys, with lower surface tension in the surface phase [9]. Designing core-shell-type phase separation requires a thorough understanding of surface tension and temperature coefficient. Previous studies using thermodynamic information on liquid alloys have connected surface and transport properties to interactions [13–15]. Understanding the temperature-induced demixing tendencies of immiscible alloys helps us comprehend their thermodynamic and thermophysical properties.

The thermodynamic modeling of liquid binary systems is essential to building the thermophysical properties database. Kang and Pelton [16] found that the mixing Gibbs energy of immiscible alloys cannot be described using a regular solution model, suggesting that the modified quasichemical model is more suitable. The self-association model, with two major factors, is useful for revealing the mystery of the segregation of liquid immiscible alloys [17]. However, the temperature dependence of two crucial parameters in the self-association model could not be accurately characterized, making it insufficient for investigating the temperature dependence of physical properties.

The Ga-Tl alloy is a typical example of an immiscible alloy. Krawczyk et al. [18] and Katayama et al. [19] determined by using the electromotive force mea-

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surement the Ga activities (α_{Ga}) in the range of 10 to 90 at.% Tl from 950 to 1050 K and in the range of 5 to 80 at.% Tl from 973 to 1273 K, respectively. Using the same method, Danilin et al. [20] measured the Tl activities (α_{Tl}) in the range of 5 to 90 at.% Tl from 694 to 1032 K. The Tl activities were also determined by Mastromarino et al. [21] using vapor pressure data from Knudsen torsion cell measurements in the composition range of 0.8 to 83 at.% Tl at average temperatures between 838 and 1077 K. The experimental data of [18, 20, 21] were included in [22].

This work studied the thermodynamic features of liquid Ga-Tl alloys at different temperatures using the self-association model and simple predictions about how the ratio of the numbers of the two types of atoms in the cluster and the interchange energy varies with temperature. The calculated thermodynamic information may also predict the surface properties of liquid Ga-Tl alloys. We utilized the discrete experimental information to get continuous thermodynamic data for liquid binary alloys, which could then be compared to future experimental research.

2. Theoretical outlines

Singh and Sommer [17] developed a self-association model for understanding liquid immiscible binary alloys. The model describes short-range interactions forming like-atom clusters or self-associates. The structural and thermodynamic functions of the liquid binary alloy were stated as follows.

The partial molar free energies (G_i and G_j) and activities (α_i and α_j) of the alloy components are:

$$\frac{G_i}{RT} = \ln\alpha_i = \ln\frac{x_i}{x_i + nx_j} + \frac{(n-1)\,x_j}{x_i + nx_j} + \frac{Wn^2x_j^2}{RT\,(x_i + nx_j)^2}, \tag{1}$$

$$\frac{G_j}{RT} = \ln\alpha_j = \ln\frac{nx_j}{x_i + nx_j} - \frac{(n-1)\,x_i}{x_i + nx_j} + \frac{Wnx_i^2}{RT\,(x_i + nx_j)^2}, \tag{2}$$

where n is the ratio of the numbers of two kinds of atoms in the cluster, W is the interchange energy, R is the molar gas constant, T is the Kelvin temperature, and x_i and x_j is the molar concentration of atoms i and j, respectively.

The Gibbs free energy of mixing (G_{M}) has the following expression:

$$\frac{G_{\mathrm{M}}}{RT} = x_i\ln\frac{x_i}{x_i + nx_j} + x_j\ln\frac{nx_j}{x_i + nx_j} + \frac{Wnx_ix_j}{RT\,(x_i + nx_j)}. \tag{3}$$

The partial molar entropy (S_i and S_j) of atoms i and j is expressed as:

$$\frac{S_i}{R} = -\ln\frac{x_i}{x_i + nx_j} - \frac{(n-1)\,x_j}{x_i + nx_j} - \frac{x_j^2}{R\,(x_i + nx_j)^2}\cdot\left(\left((1-n)\,RT + \frac{2Wnx_i}{x_i + nx_j}\right)\frac{\mathrm{d}n}{\mathrm{d}t} + n^2\frac{\mathrm{d}W}{\mathrm{d}T}\right), \tag{4}$$

$$\frac{S_j}{R} = -\ln\frac{nx_j}{x_i + nx_j} - \frac{(n-1)\,x_i}{x_i + nx_j} - \frac{x_i^2}{R\,(x_i + nx_j)^2}\cdot\left(\left(\frac{(1-n)\,RT}{n} + \frac{W\,(x_i - nx_j)}{x_i + nx_j}\right)\frac{\mathrm{d}n}{\mathrm{d}t} + n\frac{\mathrm{d}W}{\mathrm{d}T}\right) \tag{5}$$

The entropy of mixing (S_{M}) has the following expression:

$$\frac{S_{\mathrm{M}}}{R} = -x_i\ln\frac{x_i}{x_i + nx_j} - x_j\ln\frac{nx_j}{x_i + nx_j} - \frac{x_ix_j}{R\,(x_i + nx_j)}\cdot\left(\left(\frac{(1-n)\,RT}{n} + \frac{Wx_i}{x_i + nx_j}\right)\frac{\mathrm{d}n}{\mathrm{d}t} + n\frac{\mathrm{d}W}{\mathrm{d}T}\right). \tag{6}$$

The enthalpy of mixing (H_{M}) has the following expression:

$$\frac{H_{\mathrm{M}}}{RT} = \frac{Wnx_ix_j}{RT\,(x_i + nx_j)} - \frac{x_ix_j}{R\,(x_i + nx_j)} - \left(\left(\frac{(1-n)\,RT}{n} + \frac{Wx_i}{x_i + nx_j}\right)\frac{\mathrm{d}n}{\mathrm{d}t} + n\frac{\mathrm{d}W}{\mathrm{d}T}\right). \tag{7}$$

During the data analysis of the electromotive force measurements, the electromotive force E is usually considered to be linear with temperature:

$$E = A + BT, \tag{8}$$

where A and B are the variables independent of temperature.

The partial molar entropy of element X (S_{X}) in a binary liquid alloy was calculated according to the temperature coefficient of the electromotive force:

$$S_{\mathrm{X}} = NF\frac{\mathrm{d}E}{\mathrm{d}T} = NFB, \tag{9}$$

where N is the number of transferred electrons, and F is the Faraday constant. According to Eq. (9), it should be emphasized that partial molar entropy is independent of temperature within the experimental temperature range. In this research, we propose that W is linearly related to temperature and n is independent of temperature:

$$W = a_0 + a_1T, \tag{10}$$

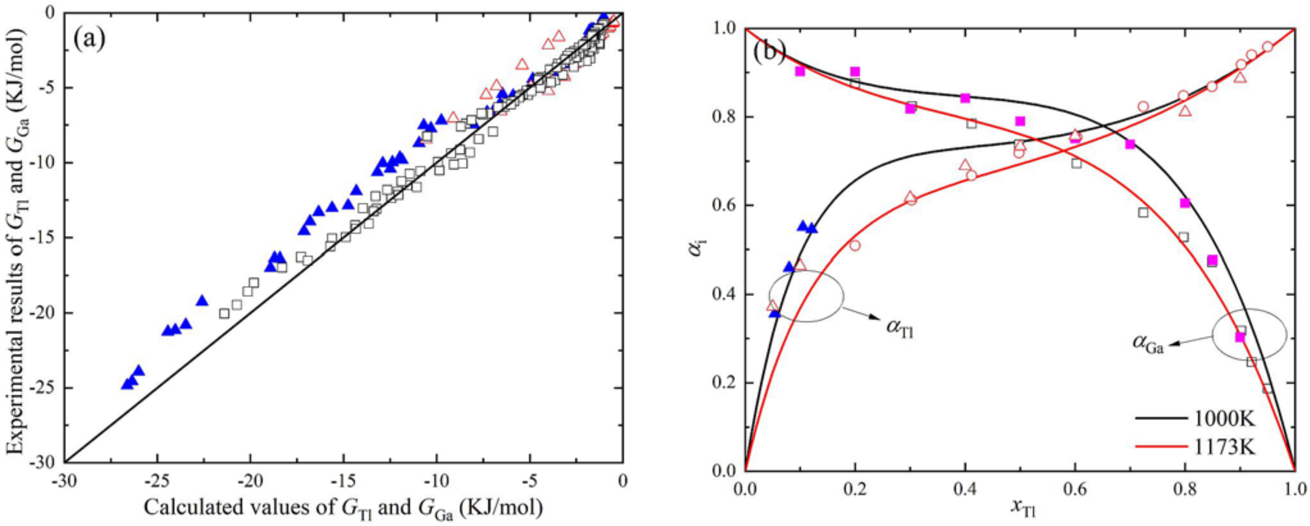

Fig. 1. Comparing experimental data with calculated activities of Ga and Tl in liquid Ga-Tl alloys: (a) Krawczyk et al. [18] (magenta solid squares), Katayama et al. [19] (black open squares), Mastromarino et al. [21] (blue solid triangles), and Danilin et al. [20] (red open triangles). The squares and triangles express the activities of Ga and Tl, respectively. The straight line is the identity where the calculated values equal the experimental values. (b) Component correlation with the activities of gallium and thallium for liquid Ga-Tl alloys at 1000 and 1173 K. The red open circles are labeled as the activities of thallium at 1173 K, which were derived using the α-function [19]. The black and red lines represent the computed values at 1000 and 1173 K, respectively.

where a_0 and a_1 are the variables independent of temperature. Based on this assumption, the expressions of the partial molar entropy and partial molar enthalpy in Eqs. (4)–(7) can be simplified to temperature-independent forms, as expressed in Eqs. (11)–(14). The validity of the assumptions will be demonstrated in subsequent analyses.

$$S_i = -R\ln\frac{nx_i}{x_i+nx_j} - \frac{R\,(n-1)\,x_j}{x_i+nx_j} - \frac{a_1n^2x_j^2}{\left(x_i+nx_j\right)^2}, \tag{11}$$

$$S_j = -R\ln\frac{nx_j}{x_i+nx_j} - \frac{R\,(n-1)\,x_i}{x_i+nx_j} - \frac{a_1nx_i^2}{\left(x_i+nx_j\right)^2}, \tag{12}$$

$$\frac{S_{\mathrm{M}}}{R} = -x_i\ln\frac{x_i}{x_i+nx_j} - x_j\ln\frac{nx_j}{x_i+nx_j} - \frac{a_1nx_ix_j}{R\left(x_i+nx_j\right)}, \tag{13}$$

$$H_{\mathrm{M}} = \frac{a_0nx_ix_j}{x_i+nx_j}. \tag{14}$$

3. Results and discussion

3.1. Assessment of the temperature-independent variables (n, a_0, and a_1)

The experimental partial molar free energies of gallium and thallium in liquid Ga–Tl alloys [19–21] and the experimental enthalpy of mixing at 923 K [23, 24] were simultaneously adopted to fit to calculate n, a_0, and a_1 using Eqs. (1), (2), and (7). The values of the enthalpies of Krawczyk [18], which were quite different from other researchers' results, were rejected. Mean relative percentage error, Er, defined in Eq. (15), is used to quantify the level of agreement between experimental and computed data in this work:

$$Er = \frac{1}{num}\sum_{i=1}^{num}\left|\frac{d_{i,\mathrm{cal}}-d_{i,\mathrm{exp}}}{d_{i,\mathrm{exp}}}\right|, \tag{15}$$

where num denotes the amount of experimental data, $d_{i,\mathrm{cal}}$ and $d_{i,\mathrm{exp}}$ represent the i-th experimental and calculated data for a certain property. The parameter Er is minimized to get the appropriate values of n, a_0, and a_1. In this paper, when $Er = 0.2620$, the optimal temperature-independent values are $n = 0.78$, $a_0 = 20818$, and $a_1 = -3.3$.

The predictive ability capability of the revised self-association model is shown in Fig. 1. Figure 1a shows the calculated partial molar free energies of gallium and thallium compared with the 215 experimental results [19–21]. Figure 1b shows the component correlation with the activities of gallium and thallium for liquid Ga-Tl alloys at 1000 and 1173 K, respectively. It can be found that there is an excellent agreement between the estimated and actual activities of gallium [19] and thallium [19–21] in liquid Ga-Tl alloys at various temperatures that have been acquired using various techniques, which supports the hypothesis regarding the correlation between the two parameters

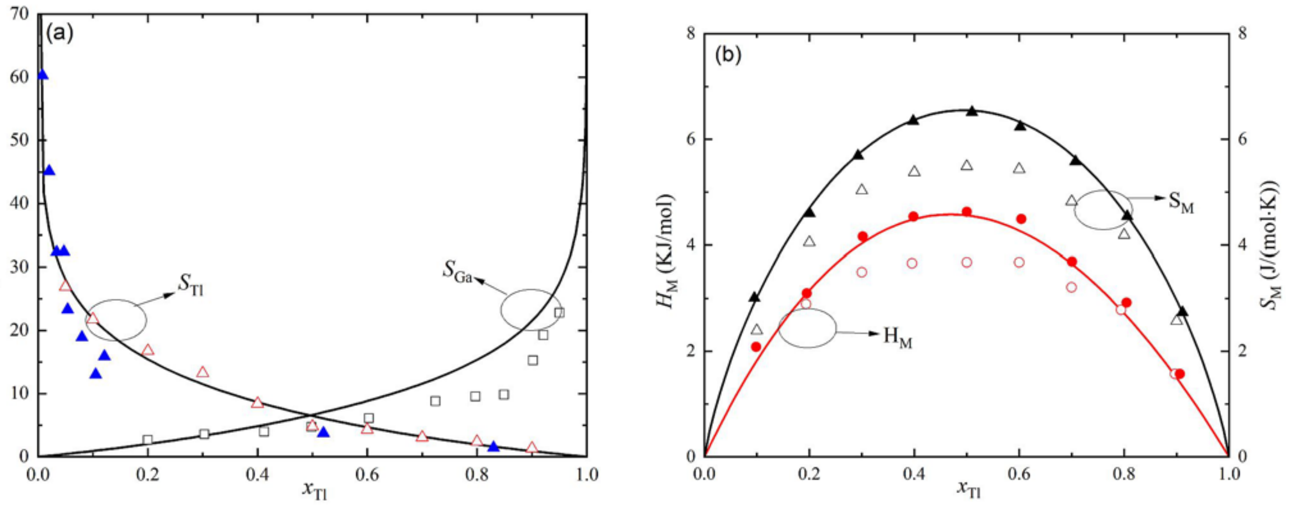

Fig. 2. (a) Comparison of calculated partial molar entropy for liquid Ga-Tl alloys with experimental findings from previous research: Katayama et al. [19] (black open squares), Mastromarino et al. [21] (blue solid triangles), and Danilin et al. [20] (red open triangles). The partial molar entropy of Ga and Tl is represented by squares and triangles. The black lines represent the computed partial molar entropy. (b) Comparison of calculated entropy of mixing and enthalpy of mixing for liquid Ga-Tl alloys with previous findings: the calculated entropy of Predel et al. [23] (black solid triangles), the experimental entropy of Predel et al. [23] (black open triangles), the experimental enthalpy of Danilin et al. [24] (red solid circles), and the experimental enthalpy of Predel et al. [23] (red open circles). The predicted entropy of mixing and the enthalpy of mixing are represented by the black and red lines, respectively.

and temperature. The usefulness of the idea was then further proven by using it to predict various thermodynamic properties.

3.2. Prediction of entropy and enthalpy

To further verify the correctness of the temperature-independent variables (n, a_0, and a_1), the entropy and enthalpy were computed and compared with pertinent experimental findings. Figure 2a shows the partial molar entropy (S_i) for liquid Ga-Tl alloys with the aid of Eqs. (11) and (12). The experimental partial molar entropy was usually determined according to the temperature coefficient of the electromotive force measurement, which was derived from the linear connection between the electromotive force measurement and temperature at various concentrations [19–21]. The outcomes of past tests, available at various temperatures, were contrasted in Fig. 2a. The predictions and the experimental results on the partial molar entropy of mixing are found to be rather well-matched.

Figure 2b indicates that the entropy of mixing (S_M) is positive over the whole concentration range. The predicted results of this paper were close to the calculated results of Predel et al. [23] but different from their experimental results [23] (i.e., when $x_{Tl} = 0.5$, the experimental value $S_M = 5.49\,\mathrm{kJ\,mol^{-1}\,K^{-1}}$, while the predicted results $S_M = 6.55\,\mathrm{kJ\ mol^{-1}\,K^{-1}}$). The predicted enthalpy of mixing (H_M) for liquid Ga-Tl alloys agreed well with the experimental values of Danilin et al. [24] while higher than the experimental results of Predel et al. [23]. At $x_{Tl} = 0.5$, the experimental enthalpy of Predel is about $3.68\,\mathrm{kJ\,mol^{-1}}$, whereas the calculated is about $4.56\,\mathrm{kJ\,mol^{-1}}$, which is about 24 % higher.

3.3. Prediction of free energy of mixing (G_M) and miscibility gap

Now that the free energy of mixing at various temperatures can be anticipated, the accuracy of the temperature-independent variables (n, a_0, and a_1) may be checked by contrasting it with the pertinent experimental data. The expected results are shown in Fig. 3 after the free energy of mixing (G_M) was calculated using Eq. (3) at 600, 800, and 1173 K. When the temperature drops, the value of G_M increases and approaches to be positive. The findings of this research agreed with those of the 1173 K experiment [19].

Liquids split into two immiscible phases when cooled through a miscibility gap. Engineering applications rarely investigate stable layer microstructures, but metastable phases can nucleate and grow in rapid solidification processes. Forecasting and interpreting metastable phases is crucial for understanding the kinetics and thermodynamics of phase separation in liquid immiscible alloys [25]. In a liquid alloy, the stability of the microstructure depends on the free energy of mixing. According to Fig. 3, the melt is stable for all x_{Zn} over 1000 K. However, phase separation occurred in the unstable melt at 800 K. The compositions of the two phases (x_1 and x_2) may be determined from the tangent line that joins locations (A and B) on the G_M curve. Equation (3) is used to calculate the slope

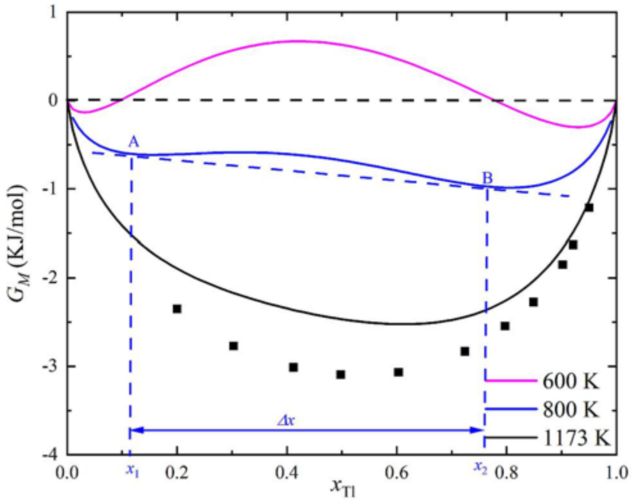

Fig. 3. Estimated free energy of mixing of liquid Ga-Tl alloys (lines) compared with earlier results at 1173 K [19] (solid squares). Blue dash line AB is the tangent line of the curve of G_{M}, where x_1 and x_2 are the compositions of tangency A and B points, respectively.

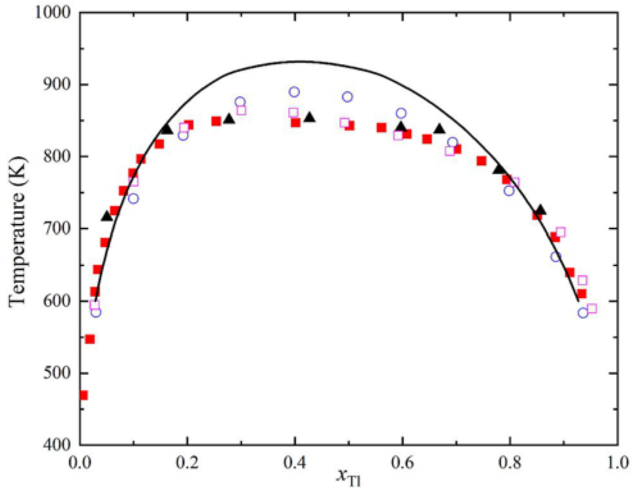

Fig. 4. Comparison of the present miscibility gap (solid line) with the earlier data in the Ga-Tl phase diagram: Predel et al. [26] (red solid squares), Yatsenko et al. [27] (black solid triangles), the Bragg-Williams random mixing model [29] (magenta open squares), and the modified quasichemical model [29] (blue open circles).

of the tangent line on the G_{M} curve, Eq. (16):

$$\frac{\partial G_{\mathrm{M}}}{\partial x_i} = RT\left(\ln\frac{x_i}{nx_j} + \frac{n-1}{x_i+nx_j}\right) + \frac{Wn\left(nx_j^2 - x_i^2\right)}{\left(x_i+nx_j\right)^2}. \qquad (16)$$

The slope of line AB is

$$k = \frac{G_{\mathrm{M}}|_{x_i=x_2} - G_{\mathrm{M}}|_{x_i=x_1}}{x_2 - x_1}. \qquad (17)$$

As

$$k = \left.\frac{\partial G_{\mathrm{M}}}{\partial x_i}\right|_{x_i=x_1} = \left.\frac{\partial G_{\mathrm{M}}}{\partial x_i}\right|_{x_i=x_2}, \qquad (18)$$

it is possible to find the analytical solution of x_1 and x_2. By figuring out x_1 and x_2 at different temperatures, liquid Ga-Tl alloys have been shown to have a miscibility gap. Figure 4 compares the present miscibility gap to earlier data [26–30].

The criteria are used to establish the critical composition x_{c} and the critical temperature T_{c}:

$$\left.\frac{\partial^2 G_{\mathrm{M}}}{\partial x_i^2}\right|_{x_i=x_{\mathrm{c}}} = 0 \quad \text{and} \quad \left.\frac{\partial^3 G_{\mathrm{M}}}{\partial x_i^3}\right|_{x_i=x_{\mathrm{c}}} = 0. \qquad (19)$$

The critical temperature and concentration for the segregation of liquid Ga-Tl alloys are $T_{\mathrm{c}} = 937.7$ K and $x_{\mathrm{c}} = 0.41$, respectively, according to Eqs. (16) and (19). Furthermore, as demonstrated in Fig. 4, the current research is in perfect agreement with the experimental results of Predel et al. [26] and Yatsenko et al. [27]. The Bragg-Williams random mixing model and modified quasichemical model were used by Kim et al. [29] to compute the miscibility gap, and the results are accurately replicated in Fig. 4. Although our critical temperature for segregation is greater than previous experimental and computational results, the expected result for the critical concentration is consistent with experimental evidence and earlier theoretical findings, which makes it logical to presume that the present computation is accurate.

3.4. Surface properties

Surface tension measurements in liquid alloys are time-consuming and costly, leading to the development of thermophysical properties models for multicomponent alloys. The Butler equation [31], subsequently refined by Yeum et al. [32] and Tanaka et al. [8], is one of the early models that links the surface tension of alloys with molar surface areas of components and their activities in a liquid phase. Using this approach, we calculated the surface tension and surface composition of liquid Ga-Tl alloys. The surface tension of a binary alloy may be expressed as Eq. (20):

$$\sigma = \sigma_i + \frac{k_{\mathrm{B}}T}{A_i}\ln\frac{x_i^s}{x_i} - \frac{1}{A_i}\left(G_i^{EXs} - G_i^{EX}\right), \qquad (20)$$

where σ_i is the surface tension of the pure component i of the liquid i-j alloy, k_{B} is the Boltzmann constant, and T is the Kelvin temperature of interest, x_i^s is the mole fraction of component i in the surface layer, and x_i is the mole fraction of component i in the liquid alloy. The surface area of pure component i (A_i) is expressed as:

$$A_i = 1.102 \left(\frac{V_i}{N}\right)^{2/3}, \tag{21}$$

where N is Avogadro's number, and V_i is the atomic volume. The atomic volume of Ga and Tl at different temperatures was derived from [33].

The partial excess Gibbs energy of the components i and j in the bulk (G_i^{EX}) can be obtained according to Eqs. (1) and (2):

$$\frac{G_i^{EX}}{RT} = \ln\frac{1}{x_i + nx_j} + \frac{(n-1)\,x_j}{x_i + nx_j} + \frac{Wn^2x_j^2}{RT\,(x_i + nx_j)^2}, \tag{22}$$

$$\frac{G_j^{EX}}{RT} = \ln\frac{n}{x_i + nx_j} + \frac{(n-1)\,x_i}{x_i + nx_j} + \frac{Wn^2x_i^2}{RT\,(x_i + nx_j)^2}. \tag{23}$$

Because surface layer atoms have fewer coordination numbers than bulk phase atoms, the partial excess Gibbs energy of a component in the surface layer (G_i^{EX}) is thought to be lower than that in the bulk phase (G_i^{EX}), as expressed in Eq. (24):

$$G_i^{EXs} = \beta G_i^{EX}, \tag{24}$$

where β is an adjustable parameter, we choose $\beta = 0.83$ following Tanaka et al. [8]. However, it should be emphasized that the value of this parameter has no appreciable impact on the estimated surface tension [34]. The surface tension of Ga and Tl was derived from experimental data measured by Kim et al. [29].

Using Eqs. (20)–(24), surface tensions of liquid Ga-Tl alloys were determined as functions of bulk composition at 873 K. Figures 5 and 6 display the findings of our computations. Figure 5 displayed the trend of surface tension decreasing as bulk Tl concentration increased and also included the experimental and computed values at 873 K examined by Kim et al. [29]. The good agreement between the current and prior surface tension results further supports the viability of the idea relating the parameters n and W to temperature.

Figure 6a shows the surface tension of liquid Ga-Tl alloys at 773, 873, 973, and 1073 K. It is noteworthy that the temperature dependence of the surface tension was positive within the composition range from 0.006 to 0.4. However, the experimental results showed negative temperature dependences [29]. According to Lee et al. [35], changing the surface composition may result in a positive temperature dependence of the surface tension when the surface tension difference between pure components is significant enough or the excess Gibbs energy is extremely positive. For liquid Ga-Tl alloys, we found the typical trend of growing surface concentration (x_{Tl}^s) with increasing bulk composition (x_{Tl}) in Fig. 6b. The surface of liquid Ga-Tl alloys segregates Tl-atoms over the whole concentration range. At 773 K, Tl-atoms segregation is more pronounced than at 1073 K. For instance, at a concentration when x_{Tl} is equivalent to 20 %, the surface is enriched with about 87.6 % of Tl atoms at 1073 K, whereas about 97.0 % of Tl atoms segregate at the surface of liquid Ga-Tl alloys at 773 K. Surface tension lowered as Tl concentration at the surface increased. However, the impact of temperature on surface tension is the exact reverse of the impact of Tl concentration. The positive temperature dependence of surface tension, as shown in Fig. 6a, may indicate that the impact of an increase in Tl concentration on surface tension throughout the composition range of 0.006 to 0.4 is greater than the influence of temperature. To fully understand this behavior, more research is necessary for the future.

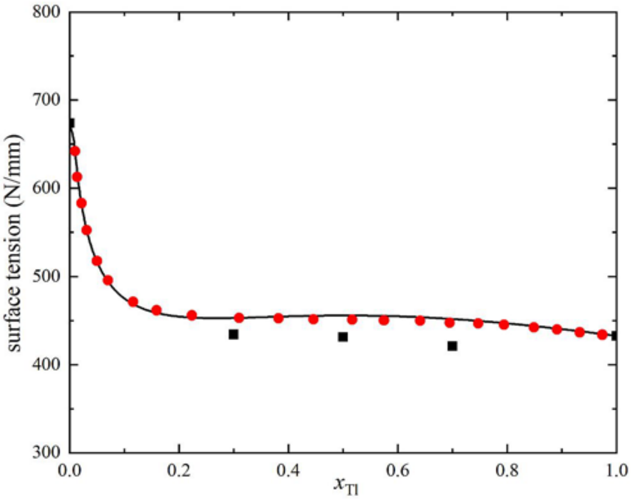

Fig. 5. Predicted surface tension values of liquid Ga-Tl alloys at 873 K, denoted by black lines. The red solid open circles are the calculated values using the modified quasi-chemical model [29]. The black solid squares are the experimental values [29].

4. Conclusions

The self-association model may be quickly and efficiently utilized to anticipate the change in physical and chemical characteristics with temperature due to the presented assumptions regarding the relationship between the ratio of the numbers of two kinds of atoms in the cluster and the interchange energy. By simultaneously fitting the experimental activity of gallium and thallium in liquid Ga-Tl alloys at various tempe-

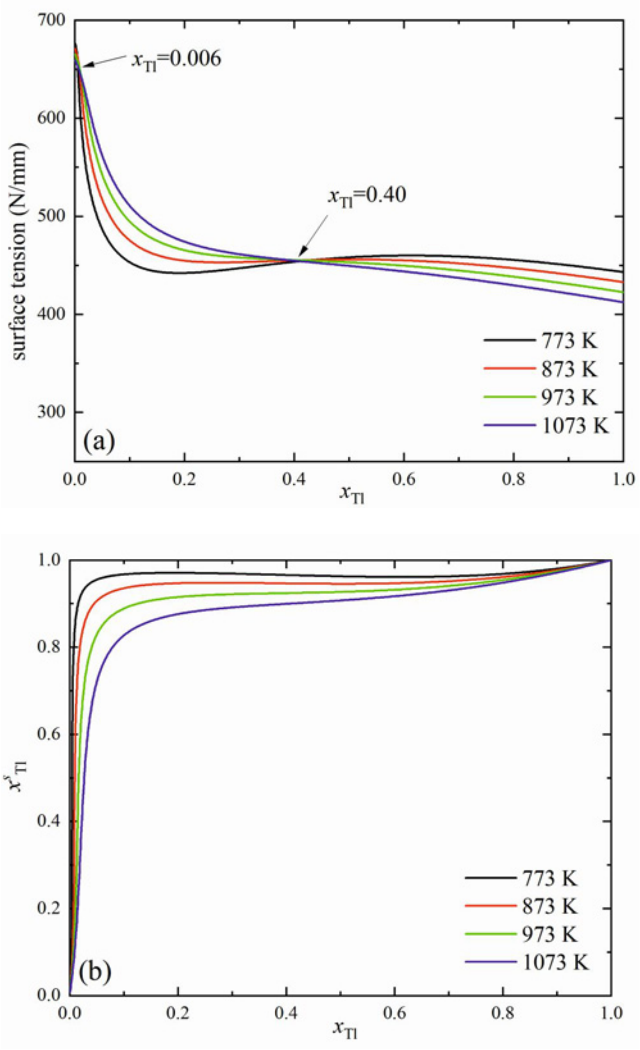

Fig. 6. (a) Surface tension and (b) surface composition of liquid Ga-Tl alloys at different temperatures.

ratures, the values of three temperature-independent coefficients (n, a_0, and a_1) were discovered. When compared to earlier theoretical and experimental discoveries, the results of this study show that:

(1) The various thermodynamic properties (activity, free energy of mixing, and partial molar entropy), miscibility gap, and surface properties are predicted based on the assumptions that the ratio of the numbers of two kinds of atoms in the cluster n is independent of temperature and that the interchange energy W is a linear function of temperature, and they have a great deal in common with prior calculations and experimental findings, which supports the reliability of the premise utilized for the prediction.

(2) Tl-atom segregation is more pronounced at 773 K than at 1073 K. When the temperature dropped from 1073 to 773 K, the surface concentration of Tl rose from 87.6 to 97.0 %. Within the composition range of 0.006 to 0.4, a significant change in the surface concentration of Tl may result in a positive temperature dependence of the surface tension.

The findings of this work contribute to a comprehensive and relevant explanation of the self-association model and provide a simple way for predicting the chemical and physical properties of liquid binary alloys across a broad temperature spectrum.

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