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## Thermophysical Study of Sodium–Indium Alloy

N. Panthi, I. B. Bhandari\*, and I. Koirala\*

*Patan Multiple Campus, Tribhuvan University,  
44618 Kirtipur, Nepal*

*\*Central Department of Physics, Tribhuvan University,  
44618 Kirtipur, Nepal*

This study explores the mixing nature of sodium–indium liquid alloy at temperatures of 713 K, 850 K, 950 K and 1050 K. It uses quasi-lattice approximation for the thermodynamic analysis of concentration dependent mixing behaviours of sodium–indium liquid alloy under the assumption of  $\text{Na}_3\text{In}$  complex. It compares the obtained theoretical results with the experimental result and result of Redlich–Kister (R–K) equation for the validity. The researchers concentrate on the viscosity and surface tension of the alloy under the modelling equations as suggested by Kaptay and improved derivation of Butler equation, respectively. This paper focuses on the interaction energy parameters among neighbouring atoms of the alloy. It observes that the alloy is moderately interacting and ordering nature at the lower concentration of sodium. The theoretical results of the thermodynamic properties are nearly in agreement with the corresponding experimental data as well as results obtained by R–K equation at 713 K. It claims that the ordering behaviour, viscosity and surface tension of the alloy decreases with the increase in temperature.

**Key words:** liquid alloys, thermodynamic properties, R–K equation, energy parameters, ordering.

Досліджено характер змішування рідкого стопу натрій–індій за температур у 713 К, 850 К, 950 К і 1050 К. Було використано квазіґратницьке наближення для термодинамічної аналізи залежних від концентрації характеристик змішування рідкого стопу натрій–індій за припущенням наяв-

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Corresponding author: Narayan Panthi  
E-mail: [narayan.755711@cdp.tu.edu.np](mailto:narayan.755711@cdp.tu.edu.np)

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ности комплексу  $\text{Na}_3\text{In}$ . Порівняно одержані теоретичні результати з експериментальними та результатами, що дає рівняння Редліха–Кістера. Дослідники переважно зосереджуються на вивченні в'язкості та поверхневого натягу стопу відповідно до рівнянь, запропонованих Каптаєм, і вдосконаленого виведення рівняння Батлера. Цю статтю присвячено дослідженню енергетичних параметрів взаємодії між сусідніми атомами стопу. Зазначається, що взаємодія для цього стопу є помірною; крім того, наявне впорядкування за нижчої концентрації Натрію. Теоретичні результати для термодинамічних властивостей узгоджуються з відповідними експериментальними даними, а також результатами, одержаними за допомогою рівняння Редліха–Кістера для 713 К. Стверджується, що впорядкування, в'язкість і поверхневий натяг стопу зменшуються з підвищенням температури.

**Ключові слова:** рідкі стопи, термодинамічні властивості, рівняння Редліха–Кістера, енергетичні параметри, впорядкування.

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## 1. INTRODUCTION

The properties of liquid alloys mainly depend on composition of constituent elements, temperature and pressure. The alloying phenomena play an important role on stability, strength, electrical character *etc.* of the materials. Thus, the study of mixing nature of elements forming alloys has been given great attention by researchers. However, detailed analysis of different behaviour of the alloys at high temperature and at all compositions of constituent elements becomes strenuous due to inconvenience in experimental task and time limitation. To overcome such difficulties and to speed up the study, many theoreticians have put forward different theoretical models [1–7].

The sodium element is highly reactive and makes complex alloys with other elements like lead, potassium, calcium and so on. Because of the development of such complexes, the thermodynamic properties of binary sodium alloys frequently deviate significantly from those of regular alloys. This makes it fascinating to investigate the properties of various alloys of sodium. Hence, different researchers [8–14] have investigated different properties of sodium alloys. However, the alloying nature of sodium with indium is found lacking till now except few experimental thermodynamic results explored by few experimentalists [15].

The present study aims to study the thermophysical behaviours of sodium–indium alloy at temperatures 713 K, 850 K, 950 K and 1050 K by assuming  $\text{Na}_3\text{In}$  complex. The thermodynamic behaviours of the alloy are analysed by quasi lattice approximation [4]. The validity of this model is tested comparing result obtained with theoretical results of

Redlich–Kister (R–K) equation [2] and experimental results at temperature 713 K. The viscosity and surface tension of the alloy have been studied at aforementioned temperature by Kaptay model [16] and improved Butler equation [17], respectively.

## 2. THEORETICAL DETAILS

### 2.1. Thermodynamic Properties

Let an alloy of constituent metals X and Y has chemical complexes  $X_\mu Y_\vartheta$  in such a way that  $\mu X + \vartheta Y = X_\mu Y_\vartheta$ , where  $\mu$  and  $\vartheta$  are small integers. The excess Gibbs free energy of mixing  $G_M^{Xs}$  in the case of quasi lattice approximation [4] can be written as:

$$G_M^{Xs} = N(\theta\omega + \theta_{XY}\Delta\omega_{XY} + \theta_{XX}\Delta\omega_{XX} + \theta_{YY}\Delta\omega_{YY}), \quad (1)$$

where N is Avogadro's number,  $\theta_{i,j}$  ( $i, j = X, Y$ ) are simple polynomials in concentration ( $C$ ),  $\omega$  is interchange energy and  $\Delta\omega_{i,j}$  are interaction energy parameters.

The value of  $\theta$  is always  $C_X C_Y$ , where  $C_X$  and  $C_Y$  are concentration of constituent elements X and Y respectively. The sum of concentration of two components is always one (*i.e.*,  $C_X + C_Y = 1$ ). The values of  $\theta_{i,j}$  in the case of  $\mu = 3$  and  $\vartheta = 1$  are found to be [4, 18]:

$$\theta_{XY} = \frac{1}{5}C_X + \frac{2}{3}C_X^3 - C_X^4 - \frac{1}{5}C_X^5 + \frac{1}{3}C_X^6, \quad (2)$$

$$\theta_{XX} = -\frac{3}{20}C_X + \frac{2}{3}C_X^3 - \frac{3}{4}C_X^4 + \frac{2}{5}C_X^5 - \frac{1}{6}C_X^6, \quad (3)$$

$$\theta_{YY} = 0. \quad (4)$$

The Gibbs free energy of complex formation of an alloy is given by standard equation as:

$$G_M = G_M^{Xs} + RT(C_X \ln C_X + C_Y \ln C_Y) = N(\theta\omega + \theta_{XY}\Delta\omega_{XY} + \theta_{XX}\Delta\omega_{XX} + \theta_{YY}\Delta\omega_{YY}) + RT(C_X \ln C_X + C_Y \ln C_Y). \quad (5)$$

The enthalpy of mixing of an alloy is found out from Gibbs free energy by standard thermodynamic equation as:

$$H_M = G_M - T \left( \frac{\partial G_M}{\partial T} \right)_{C,N,P} = G_M - TN \left( \frac{\partial \omega}{\partial T} \theta + \frac{\partial \Delta\omega_{XY}}{\partial T} \theta_{XY} + \frac{\partial \Delta\omega_{XX}}{\partial T} \theta_{XX} \right). \quad (6)$$

The activity  $a_i$  of each constituent element of the alloy is related to Gibbs free energy by standard relation given as:

$$RT \ln a_X = G_M + C_Y \left( \frac{\partial G_M}{\partial C_X} \right)_{T,P,N} \quad \text{and} \quad RT \ln a_Y = G_M - C_X \left( \frac{\partial G_M}{\partial C_Y} \right)_{T,P,N}. \quad (7)$$

Similarly, the partial excess Gibbs free energy is related to activity of each component by the following relation [19, 20]:

$$G_i^{Xs} = RT \ln(a_i / C_i). \quad (8)$$

## 2.2. Structural Properties

For the theoretical study of arrangement of atoms in the binary alloy, we compute concentration fluctuation in long-wavelength limit and Warren–Cowley chemical short-range order parameter.

The concentration fluctuation in long-wavelength limit  $S_{cc}(0)$  is given as [21]:

$$S_{cc}(0) = RT \left( \frac{\partial^2 G_M}{\partial C^2} \right)_{T,P,N}^{-1}. \quad (9)$$

$S_{cc}(0)$  can also be found out by observed activities as:

$$S_{cc}(0) = C_Y a_X \left( \frac{\partial a_X}{\partial C_X} \right)_{T,P,N}^{-1} = C_X a_Y \left( \frac{\partial a_Y}{\partial C_Y} \right)_{T,P,N}^{-1}. \quad (10)$$

Thus,  $S_{cc}(0)$  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:

$$S_{cc}(0) = \frac{RT}{(-2\omega + \theta_{XY}'' \Delta\omega_{XY} + \theta_{XX}'' \Delta\omega_{XX} + RT / C_X C_Y)}. \quad (11)$$

The Warren–Cowley chemical short range order parameter  $\alpha_1$  is related to the ratio of concentration fluctuation in long-wavelength limit  $S$  to coordination number  $Z$  [22, 23] as:

$$\alpha_1 = (S - 1)[S(Z - 1) + 1]^{-1}, \quad (12)$$

where

$$S = \frac{S_{cc}(0)}{S_{cc}^{id}(0)},$$

and  $S_{cc}^{id}(0)$  is the concentration fluctuation in long-wavelength limit for an ideal alloy.  $Z$  is co-ordination number and its value is taken 10

[24, 25] for the liquid alloy study.

### 2.3. Transport Property: Viscosity

Kaptay considered the relationship between the cohesive energy and activation energy of the viscous flow and developed an equation of viscosity  $\eta$  for binary alloy as given below [16]:

$$\eta = \frac{hN}{C_X V_X + C_Y V_B + V^E} \exp\left(\frac{C_X G_X + C_Y G_Y - \Phi H_M}{RT}\right), \quad (13)$$

where  $h$  is Planck's constant,  $V_i$  ( $i = X, Y$ ) is the molar volume of pure metal,  $V^E$  is excess molar volume upon alloy formation,  $G_i$  is Gibbs activation energy of viscous flow in pure metals and  $\Phi$  is a constant whose value is  $(0.155 \pm 0.015)$  [26]. The Gibbs energy of activation of pure metal  $i$  is calculated by the following equation:

$$G_i = RT \ln\left(\frac{\eta_i V_i}{hN}\right), \quad (14)$$

where  $\eta_i$  is viscosity of individual elements X and Y, respectively. The variation of viscosity of a metal with temperature is given as [27]:

$$\eta_i = \eta_0 \exp\left(\frac{\varepsilon}{RT}\right), \quad (15)$$

where  $\eta_0$  and  $\varepsilon$  are constants of each metal having units of viscosity and energy per mole respectively.

### 2.4. Surface Properties: Surface Tension

According to this model, there is an existence of a monoatomic layer, called surface monolayer at the surface of the molten alloy as a separate phase and it is in thermodynamic equilibrium with that of the bulk phase. The surface tension  $\sigma$  of binary alloy at temperature  $T$  is given by the improved Butler equation [17] as:

$$\sigma = \frac{\gamma_i^0}{\gamma_i} \sigma_i^0 + \frac{RT}{\gamma_i} \ln \frac{C_i^S}{C_i^b} + \frac{G_i^{S,Xs} - G_i^{b,Xs}}{\gamma_i}, \quad (16)$$

where  $\sigma_i^0, \gamma_i^0, \gamma_i$  are surface tension, molar surface area of each liquid metal and partial molar surface area of  $i^{\text{th}}$  component, respectively.  $G_i^{S,Xs}$  and  $G_i^{b,Xs}$  are partial excess free energy of mixing 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  $\beta$  is taken as

0.818 [28].

The molar surface area of  $i^{\text{th}}$  component is given as [26]:

$$\gamma_i^0 = \chi \left( \frac{M_i^0}{\rho_i^0} \right)^{2/3} N^{1/3}, \quad (17)$$

where  $M_i^0, \rho_i^0, \chi$  are respectively molar mass, density of each constituent element at its melting temperature, and geometrical constant. The value of  $\chi$  is obtained from volume packing fraction  $f_V$  and surface packing fraction  $f_S$  by the expression as [26]:

$$\chi = \left( \frac{3f_V}{4} \right)^{2/3} \frac{\pi^{1/3}}{f_S}. \quad (18)$$

For the liquid metal, the values of both  $f_V$  and  $f_S$  are taken as 0.66 and 0.906, respectively [26, 28].

The density  $\rho_i^0$  and surface tension  $\sigma_i^0$  of each constituent metal of the liquid alloy at any temperature  $T$  are expressed as [27]:

$$\rho_i^0 = \rho_i + \frac{d\rho}{dT} (T - T_0), \quad (19)$$

$$\sigma_i^0 = \sigma_i + \frac{d\sigma}{dT} (T - T_0), \quad (20)$$

where  $\rho_i$  and  $\sigma_i$  are density and surface tension of each component at its melting temperature  $T_0$ . Similarly,  $d\rho/dT$  and  $d\sigma/dT$  are temperature coefficient of density and surface tension, respectively.

### 3. RESULTS AND DISCUSSION

#### 3.1. Thermodynamic and Structural Properties

In order to analyse thermodynamic properties of an alloy under the quasi-chemical treatment, it is necessary to determine the interaction energy parameters and their temperature derivatives. The energy parameters between the atoms of the alloy at a temperature are carried out by successive approximation method using Eq. (1) and experimental results [15] within concentration range 0.1 to 0.9. Now such parameters at high temperatures are obtained using Eq. (21) under the assumption that the parameters are linearly dependent on temperature and independent on the concentration of each component of the alloy:

$$d[\omega_{ij}(T)]_c = \frac{\partial \omega_{ij}(T)}{\partial T} dT, \quad i \neq j; \quad \omega_{ij}(T_K) - \omega_{ij}(T) = \frac{\partial \omega_{ij}}{\partial T} (T_j - T). \quad (21)$$

**TABLE 1.** Interaction energy parameters (J/mol) at different temperatures.

Temperature, K	$\omega$	$\Delta\omega_{XY}$	$\Delta\omega_{XX}$
713	-2641.197	-7800.238	77408.543
850	-2088.8403	-4156.642	65842.259
950	-1685.587	-1497.082	57399.716
1050	-1282.334	1162.477	48957.172

The parameters thus found at different temperature are shown in the Table 1.

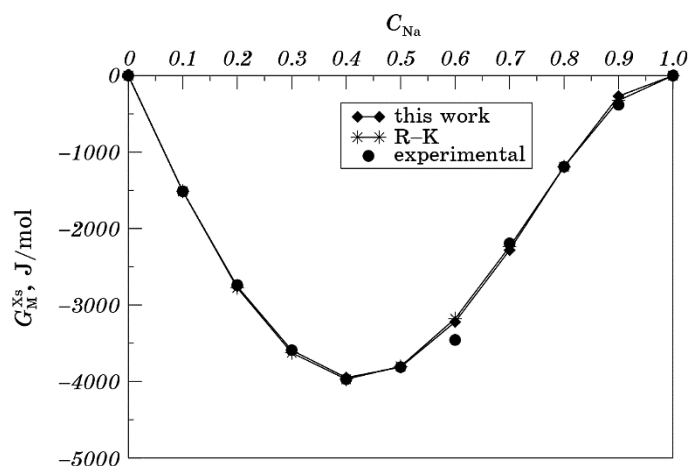
Similarly, we optimized linear temperature dependent R-K polynomials [29] to compare our results with Redlich-Kister equation [2]. The R-K polynomials thus optimized are given below:

$$L_0 = -35087.119 + 27.878T,$$

$$L_1 = 8399.395 - 0.220T,$$

$$L_2 = 16556.200 - 12.267T.$$

The plot of Gibbs free energy versus concentration of Na of the alloy at temperature 713 K is shown in Fig. 1. As clear, the theoretically computed result is in good agreement with experimental result. This proves the validity of model and interaction parameters. Figure 2 is the Gibbs free energy of mixing at higher temperatures at three different compositions, Na<sub>10</sub>In<sub>90</sub>, Na<sub>50</sub>In<sub>50</sub>, and Na<sub>90</sub>In<sub>10</sub> of the alloy. The figure depicts that as temperature increases the Gibbs free energy of mix-

**Fig. 1.** Excess Gibbs energy *vs.* concentration of Na at 713 K.

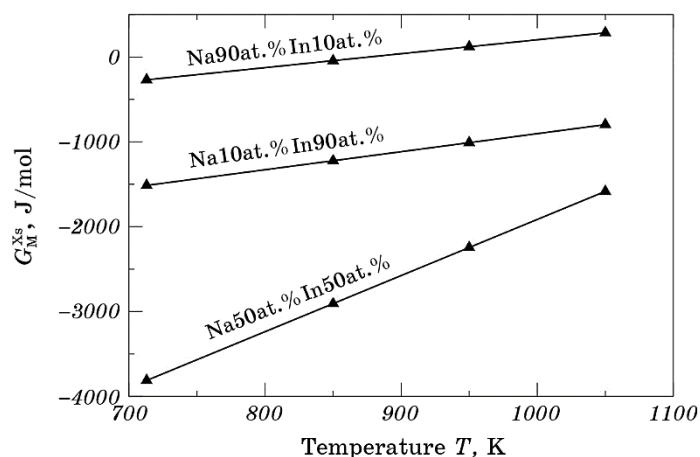


Fig. 2. Excess Gibbs energy *vs.* temperature.

ing becomes less negative indicating that interaction decreases with increase in temperature.

The temperature derivatives of interaction energy parameters are also acquired by successive approximation method by using Eq. (6) and experimental result [15] within concentration range 0.1 to 0.9. The values of such parameters at temperature 713 K are  $\partial\omega/\partial T = 0.485R$ ,  $\partial\omega_{XY}/\partial T = 3.198R$ , and  $\partial\omega_{XX}/\partial T = -10.154R$ . However, for small change in temperature the temperature derivatives of such parameters are considered constant. Figure 3 is the computed enthalpy of mixing of alloy at 713 K, which nearly agrees with experimental results. The computed enthalpy of mixing of alloy at higher temperature and at

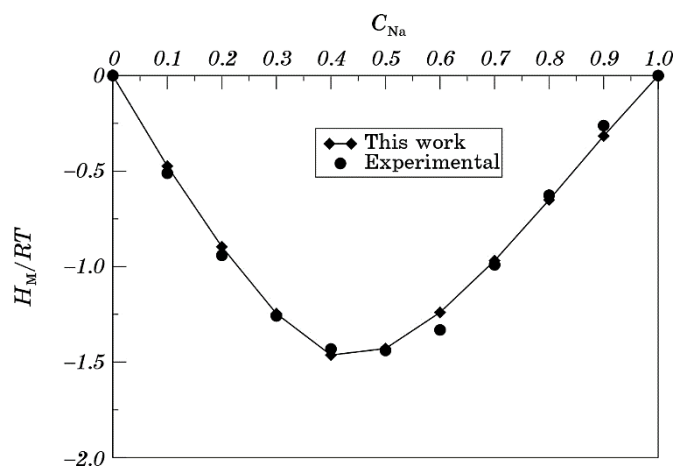


Fig. 3. Enthalpy of mixing *vs.* concentration of Na at 713 K.



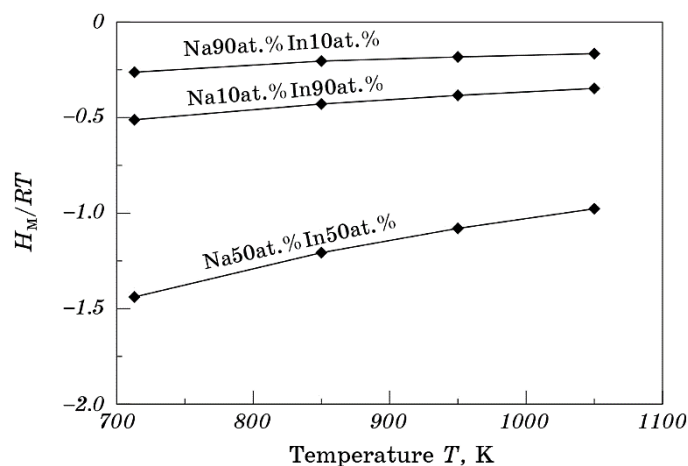


Fig. 4. Enthalpy *vs.* temperature.

$\text{Na}_{10}\text{In}_{90}$ ,  $\text{Na}_{50}\text{In}_{50}$ , and  $\text{Na}_{90}\text{In}_{10}$  compositions of the alloy is shown in the Fig. 4. The less negative values of enthalpy of mixing at higher temperatures indicates that the alloy shows less interacting behaviour with increase in temperatures.

Chemical activity of constituent of the alloy is considered another important thermodynamic property of the alloy. It mainly gives the idea about the deviation of constituent element from the ideal behaviour. According to Porter and Easterling [30], the activity informs the tendency of constituents of the alloy whether they are willing to leave the mixture or not. If the activity is high, the atoms show high tenden-

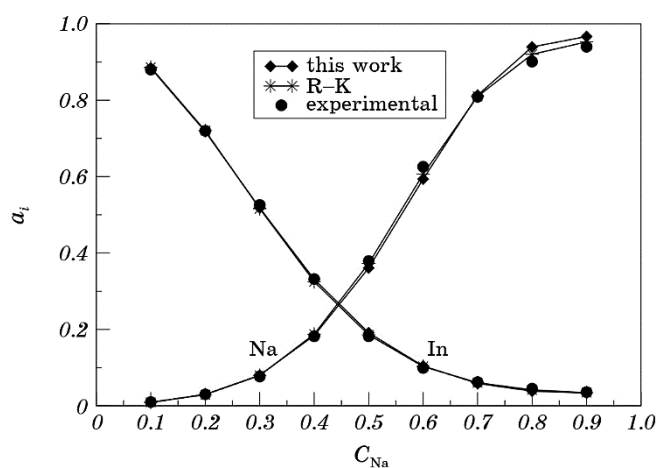


Fig. 5. Activity *vs.* concentration of Na at 713 K.

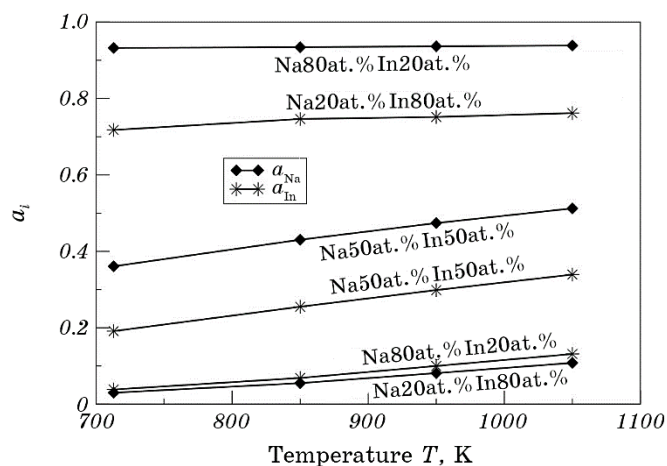


Fig. 6. Activity *vs.* temperature.

cy to leave the mixture and *vice-versa*. Equation (7) is used to calculate the chemical activity of components of alloy NaIn. Figure 5 is the result of experimental, theoretical and R–K equation of the chemical activity of the alloy at 713 K which shows a good agreement between the experimental and theoretical results. It suggests that at lower concentrations of Na, there is probability of pairing of unlike atoms. However, as temperature increases, the activity of each component increases as shown in Fig. 6 indicating that the atoms of constituent elements have less tendency to mix together when temperature of the alloy's increases.

For the theoretical analysis of internal adjustment of atoms in the binary alloy, the concentration fluctuations in the long-wavelength limit  $S_{CC}(0)$  and Warren–Cowley short-range order parameter  $\alpha_1$  are considered important tools. The concept of  $S_{CC}(0)$  removes difficulties on diffraction experiments [21].

The  $S_{CC}(0)$  provides the qualitative information whereas the  $\alpha_1$  provides quantitative information of local arrangement of atoms. For given concentration and temperature, if  $S_{CC}(0) < S_{CC}^{id}(0)$ , then,  $\alpha_1 = -1$ . In this situation the alloy is expected to have ordering nature and if  $S_{CC}(0) > S_{CC}^{id}(0)$ , then,  $\alpha_1 = 1$  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 experimental and theoretical values of  $S_{CC}(0)$  at temperature 713 K is shown in Fig. 7, which suggests that the alloy has ordering tendency up to concentration 0.7 of Na, but at above concentration 0.7, it shows segregating nature as shown in Fig. 8. Accordingly, the value of  $\alpha_1$  is less than zero at 0.7 and more than zero above concentration 0.7 of Na at all temperatures as in Fig. 9.

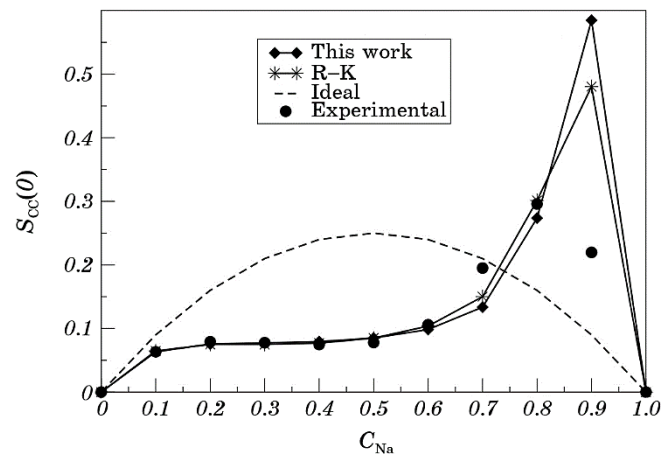


Fig. 7. Concentration fluctuation in long-wavelength limit *vs.* concentration of Na at 713 K.

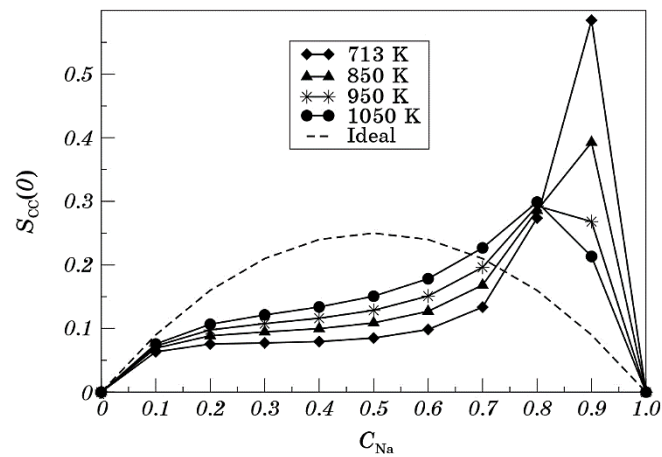


Fig. 8. Concentration fluctuation in long-wavelength limit at different temperatures.

### 3.2. Transport Properties

The concentration and temperature dependent viscosity at temperature 713 K–1050 K is calculated by Kaptay model as shown in Fig. 10. During the calculation, the value of  $V^E$  is taken zero due to the lack of experimental values [16, 31]. As clear, the viscosity of alloy decreases with increase in temperature which is the indication of reduction of interatomic attractive forces with rise in temperature and it is obvious.

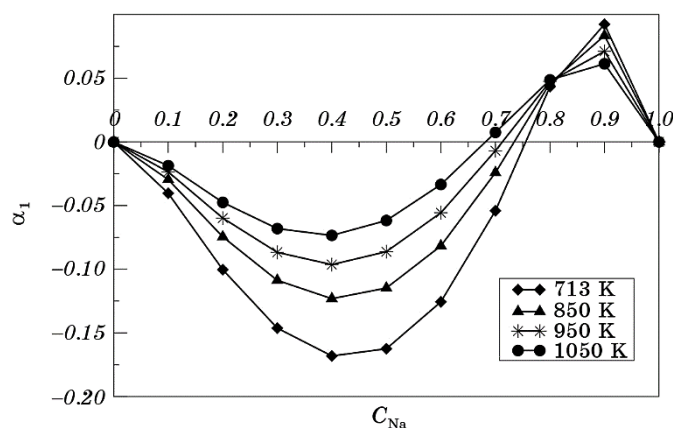


Fig. 9. Warren-Cowley short-range order parameter *vs.* concentration of Na at different temperatures.

### 3.3. Surface Properties

In order to calculate the surface tension of the NaIn liquid alloy, the densities and surface tension required for each metal at temperatures 713 K–1050 K are calculated using equations (19) and (20). Similarly, the partial excess free energy of Na and In at afore-mentioned temperatures are obtained by the theoretical activities obtained from equation (8).

Kaptay [17, 32] suggested that for unknown or negligible excess molar volume of the mixing, the partial molar volume of each component can be replaced by the molar volume of same component. In such situa-

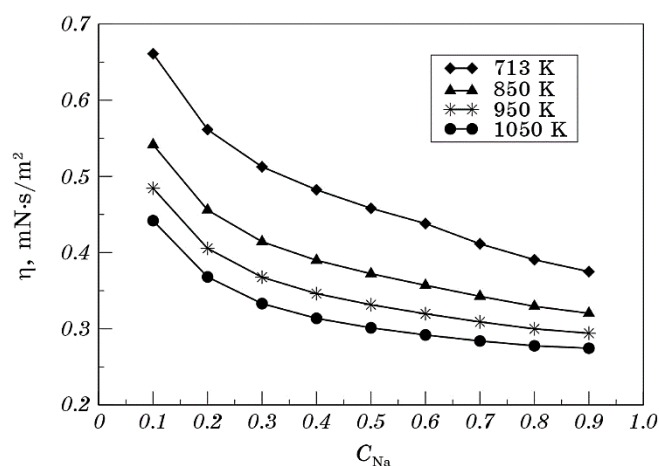


Fig. 10. Viscosity *vs.* concentration of Na at different temperatures.

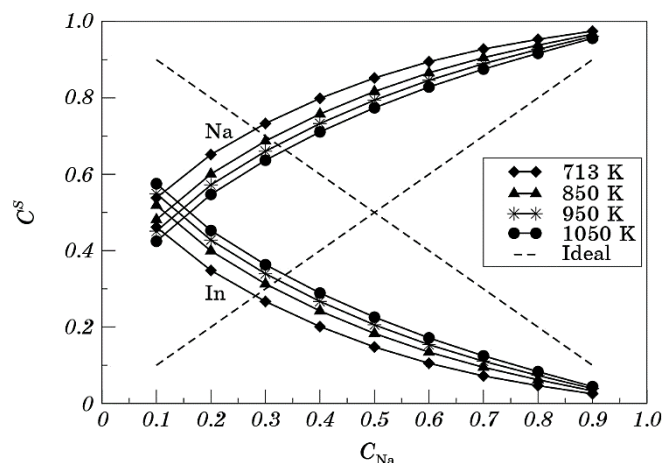


Fig. 11. Surface segregation *vs.* concentration Na at different temperatures.

tion, the surface area  $\gamma_i^0$  of each component replaces the partial surface area  $\gamma_i$ . Now, using above-input parameters and solving Eq. (16), we find surface concentration of each component. The surface concentration of Na at afore-mentioned temperature is shown in the Fig. 11. The figure indicates that the sodium atoms prefer to stay on the surface. Similarly, at higher concentration of Na, the surface segregation of both components shifts towards the ideal value to revoke the temperature effect. Thus, it can be said that at higher concentration of Na, there appears phase separation in the alloy. Similarly, the surface segregation of Na decreases with increase in temperature. The computed values of surface tension obtained at different temperatures are shown in Fig. 12. The figure suggests that the surface tension of the alloy reduces with increase in temperature.

#### 4. CONCLUSION

The present study is the theoretical investigation of thermodynamic, structural, transport and surface behaviours of binary liquid NaIn alloy at 713 K, 850 K, 950 K and 1050 K under the assumption of existence of  $Na_3In$  complex in the binary liquid mixture. From the thermodynamic study, we got the information that the alloy is moderately interacting and exhibits asymmetric behaviour as a function of concentration. The less negative values of thermodynamic properties at higher temperature indicate the weak tendency of compound forming alloy. The study also insights ordered tendencies of the alloy at about 0.7 concentration of Na at 713 K, but it becomes weaker with increase in temperature. As also observed, the surface segregation of sodium in-

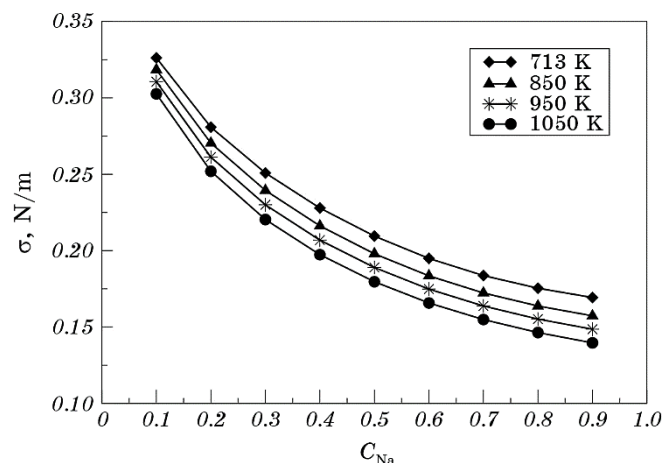


Fig. 12. Surface tension  $\nu s.$  concentration Na at different temperatures.

creases and that of indium decreases with increase in concentration of Na, which is the proof of segregating nature of alloy beyond 0.7 concentration of sodium. The thermophysical properties, viscosity and surface tension both decrease with rise in temperature.

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