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Viscosity of liquid Na-K alloy

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Abstract

The present work reports a theoretical assessment of the composition dependence of mixing properties of liquid Na-K alloy at 384K at fixed pressure, most likely, at the atmospheric pressure. In this work we have estimated the interaction energy in the alloy at the mentioned temperature on the basis of quasi-chemical approximation for regular alloy and employed it to work out basic thermodynamic properties of mixing such as free energy of mixing, entropy of mixing and enthalpy of mixing as function of composition of the alloy. To understand the dynamic behaviour in the liquid Na-K alloy, we have carried out theoretical investigation of viscosity under consideration of Moelwyn-Hughes equation by using data for enthalpy of mixing obtained from quasi-chemical approximation. Due to scanty of experimental data on viscosity of Na-K alloy at 384K, we have performed computation of viscosity also from Kaptay equation for comparison. The calculations have shown that there is a good match between theoretically computed thermodynamic functions and the available corresponding experimental data. The sets of viscosity values obtained against composition from the two equations show deviations from the ideality and themselves are reasonably comparable to each other.

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1. Introduction

Sodium and potassium occupy large proportion of the mass of the earth. They are respectively the sixth and seventh most abundant elements on the earth's crust. These metals are one of the most reactive and electropositive classes of all metals. Potassium and sodium can be alloyed to form Na-K alloy. Industrially, Na-K is produced in a reactive distillation [1]. Na-K is highly reactive with water and on exposure to air it ignites instantly, so it is stored under hexane or other hydrocarbons or an inert gas such as dry argon or dry nitrogen, for safety reasons [2].

Na-K is low melting eutectic alloy (Eutectic composition: 22% Na & 78% K, by mass; Eutectic temperature =−12.8°C) [3] and the alloy containing 40% to 90% potassium by weight is usually liquid at room temperature. As the eutectic composition of Na-K has wide liquid temperature range at atmospheric pressure (−12.8°C to 785 °C) and has properties such as melting point, density, vapor pressure, viscosity all low but thermal conductivity higher, it has got wide range applications. It is identified as an ideal HTF (heat transfer fluid) candidate in a storage system such as concentrating

solar power (CSP) that uses latent heat storage materials or PCMs (phase changing materials) for storing relatively large amounts of energy in small volumes [3]. The alloy is used as the coolant in experimental fast neutron nuclear reactors; the Soviet RORSAT radar satellites were powered by a Na-K cooled reactor [4]. In the past the Na-K system has been the subject of several other research studies too in different aspects [5-13]. Such potential applications of Na-K alloy are drawing the attention of many researchers towards the investigation of its properties. Here in this work the purpose of this article is to discuss the role of enthalpic and viscous properties in the mixing behaviour of liquid Na-K alloy at 384K.

It is known that most binary alloys are far from ideal solutions and reveal a micro-inhomogeneous atomic distribution and are less easy to understand as compared to crystals. The solubility in a homogeneous metallic mixture is basically governed by the size factor, electrochemical effect and electron concentration [14]. Experience shows that these factors alone cannot be used effectively to understand the alloying of metals in the liquid phase and hence other properties of mixing should be incorporated. The properties of mixing are attributed to short range atomic interactions at the state of disorder in the alloy. Based on the nature of atomic interaction in a liquid alloy, they are generally categorized into two generic types of ordering or phase separating alloys. The phase behaviour of a binary A-B alloy is determined by a balance between the atomic interactions E_{AA}, E_{BB} and E_{AB}. Broadly, the properties of liquid alloys can be studied mainly under two distinct groups of theoretical models, the statistical mechanical model and electronic models. All the statistical theoretical models developed for the alloys are based on some sort of inter-atomic interactions which plays an important role in the formation of an alloy. The accurate determination of the interaction energy terms among species of a disordered system like a liquid alloy is actually a complex problem as compared to those of crystals. Thus there always exists an opportunity for the investigation of the true nature of atomic interaction in a liquid alloy so that the alloying properties could be comprehended on the basis of theoretical analysis. Over the past years, several theoretical models have been developed to assess the properties of liquid alloys and efforts have been on the move for future investigations.

Na-K alloy has an interesting feature that some of its equilibrium properties of mixing, such as free energy of mixing, entropy of mixing and enthalpy of mixing of this alloy at 384 K, are nearly symmetrical [15] about equiatomic composition despite the large atomic size mismatch of K and Na ($\Delta r / \bar{r} = 21.2\%$), where $\bar{r}$ is the mean of the atomic radii of potassium and sodium. The experimental enthalpy of mixing for Na-K is positive, the maximum being 762 J/mole [15] and concentration structure factors computed from the activity data indicate weaker tendency of phase separation in the alloy over whole range of concentration although both sodium and potassium have simple electronic structures and are soluble with each other [11]. From the analysis of the size difference of the constituent atoms as well as the strength of the interatomic interactions, Alblas et al. have considered the Na-K system as a borderline case for treatment as a regular solution [12]. In this analysis we aim to review the concentration dependence of the enthalpy of mixing, H_M of Na-K alloy at 384K by computing free energy of mixing, G_M and entropy of mixing, S_M in the framework of quasi-chemical approximation (QCA) for regular alloy [16,17]. In compound formation model (CFM) for a metallic liquid A-B alloy, appropriate privileged groups or chemical complexes $A_\mu B_\nu$ (μ, ν small integers) are assumed to exist in the melt and the energy of an AB, AA or BB bound depend on whether that bond is part of the complex or not. When no complexes are formed, the CFM model reduces to the QCA for regular alloys [16-18]. Application of the approximation to a segregating alloy in particular makes it

possible to reproduce closely the observed thermodynamic properties. Over time a great deal of attempt has been devoted on this basis of this approximation to the investigation the mixing properties of liquid alloys [19-23].

Viscosity of molten metals and alloys, on the other hand is a very important atomic transport property in metallurgical science which reflects their liquid structure [24]. The viscosity of molten metals and alloys is a structurally sensitive dynamic property which indeed depends on the interactions between cohesive energies of the components giving the cohesive energy of the solution by the arrangement of the solute atoms in the solution. The cohesive interactions in condensed phase, whether liquid or solid, can be approximated by the bonding that occurs between atoms and their nearest neighbors. In order to investigate the dynamic behaviour in the liquid alloys several theoretical methods have been suggested [24-29]. In the current study, we intend to study the viscosity of the liquid Na-K alloy at 384K against concentration by correlating it with the enthalpy of mixing through available theoretical equations, Moelwyn-Hughes equation [29] and the Kaptay equation [29]. Due to scanty of viscosity data of Na-K alloy at hand, we have performed a comparative study in reference to the ideal viscous behaviour by using enthalpy of mixing, deduced theoretically from QCA and those available in the literature.

The layout of this paper is as follows. In section 2, we present the basic theoretical expressions for thermodynamic functions and viscosity of liquid binary alloys used in this work. Section 3 deals with the result and a general discussion of the work. We conclude the paper with a summary of the work in section 4.

2. Formalism

We present the theoretical expressions that we have used in the present analysis for determining the enthalpy of mixing and the viscosity of a metallic binary separately in the following sub-sections.

2.1. QCA for Enthalpy of mixing

The QCA, which is an approximation of compound formation model, in essence, is appropriate for studying liquid alloys having phase separating tendencies. According to this approximation, a binary liquid alloy is assumed to consist of N_1 atoms of component A and N_2 atoms of component B situated at equivalent sites. There is short ranged atomic interaction between the nearest neighbours that forms a polyatomic matrix leading to the formation of like atom clusters or self-associates of the type iA and jB, where i and j are the number of atoms in the clusters of elements A and B respectively. Under the assumption of QCA, working expressions for thermodynamic and microscopic functions of binary liquid alloys have been derived [20].

The enthalpy of mixing, H_M of a liquid binary mixture at a given temperature, T can be calculated theoretically from the knowledge of two important thermodynamic quantities namely the free energy of mixing, G_M and the entropy of mixing, S_M using the standard thermodynamic relation:

$$H_M = G_M + TS_M \tag{1}$$

For one mole of a binary liquid alloy A-B consisting of N_1 ($=xN$) atoms of A-component and N_2 ($=(1-x)N$) atoms of B-component in the bulk phase, N being the total number of atoms and x the mole fraction of A-component, an expression for the free energy of mixing, G_M of a binary liquid alloy at temperature T in QCA can be obtained as [20]:

$$G_M = RT\left[x\ln\frac{x}{x+\gamma(1-x)} + (1-x)\ln\frac{\gamma(1-x)}{x+\gamma(1-x)}\right] + \frac{\gamma x(1-x)}{x+\gamma(1-x)}\,W \tag{2}$$

where, k_B is Boltzmann constant; W is ordering energy parameter and $\gamma = j/i$ is the ratio of the self associates.

In order to estimate QCA fitting parameters W and $\gamma = j/i$, the value of the energy parameter W, we also recall the following standard thermodynamic relations for the chemical activities, a_i [20]:

$$\ln a_A = 1 + \ln\frac{x}{x+\gamma(1-x)} - \frac{1}{x+\gamma(1-x)} + \left[\frac{\gamma(1-x)}{x+\gamma(1-x)}\right]^2 \frac{W}{RT} \tag{3a}$$

$$\ln a_B = \ln\frac{\gamma(1-x)}{x+\gamma(1-x)} + \frac{(1-\gamma)x}{x+\gamma(1-x)} + \gamma\left[\frac{x}{x+\gamma(1-x)}\right]^2 \frac{W}{RT} \tag{3b}$$

The entropy of mixing (S_M) of mixture, defined through the standard thermodynamic relation

$$S_M = -\left(\frac{\partial G_M}{\partial T}\right)_P \tag{4}$$

is obtained in QCA for regular alloy as follows:

$$S_M = -R\left[x\ln\frac{x}{x+\gamma(1-x)} + (1-x)\ln\frac{\gamma(1-x)}{x+\gamma(1-x)}\right] + \frac{\gamma x(1-x)}{x+\gamma(1-x)}\frac{\partial W}{\partial T} \tag{5}$$

2.2. Viscosity

2.2.1. The Moelwyn-Hughes equation

The viscous flow of a liquid mixture depends on the cohesive interactions in liquid phase. Cohesive interactions arise due to the bonding that develops between atoms/or molecules and their nearest neighbours and they give rise to the enthalpic effect. In a liquid metal, the variations in the cohesive energies may result from coupling of geometric and electronic shell effects [30]. The Moelwyn-Hughes equation for viscosity of liquid mixture has been formulated to incorporate the cohesion energy in terms of enthalpic effect in order to account for viscous flow in a liquid binary alloy and at a temperature T, it is given as [29]:

$$\eta = [x\eta_1 + (1-x)\eta_2]\,[(1 - 2x + 2x^2)\frac{H_M}{RT}] \tag{6}$$

where η and η_k represent respectively the viscosity of the liquid alloy A-B and of pure component metal k $(k = 1, 2)$; x the mole fraction of component A, H_M the enthalpy of mixing of the alloy and R is the universal gas constant. For most metals the variation of viscosity, η_k with temperature T may be expressed as [31]:

$$\eta_k = \eta_{ok}\exp(E/RT) \tag{7}$$

where, η_{ok} and E are constants for pure metal, in units of viscosity and energy per mole respectively.

2.2.2. The Kaptay equation

In an attempt to give better insight into the viscous nature of a binary liquid alloy, Kaptay unified various thermodynamic models into a single equation. The unified equation, termed as Kaptay equation for viscosity, is in a qualitative conformity with the majority of the thermodynamic models [29]. It takes into account of the theoretical relationship between the cohesion energy of the alloy and the activation energy of viscous flow, stating that in alloys with stronger cohesion energy the viscosity will increase, and not decrease. This equation reads as:

$$\eta = \frac{h N_A}{x V_1 + (1-x) V_2 + V^E} \exp\left(\frac{x G_1^* + (1-x) G_2^* - \alpha . H_M}{R T} \right) \quad (8)$$

where G_k^* is the Gibbs energy of activation of the viscous flow in pure component metal k given by:

$$G_k^* = RT \ln\left(\frac{\eta_k V_k}{h N_A}\right) \quad ; \quad (k = 1,2) \quad (9)$$

Here, H_M is enthalpy of mixing of the alloy; N_A is Avogadro number; V_k represents the molar volume of the component k; V^E is the excess molar volume upon alloy formation which can be neglected for simplicity [29]; η_k is the viscosity of the component at temperature T as given by Eq. (7) and α is a constant whose value is taken to be (0.155 ± 0.015) [29].

3. Results and Discussion

3.1. Enthalpy of mixing

The formation of a metallic solution depends on the inter-atomic interactions that can take place between atoms of the pure components and also between the two different components. The interactions of atoms in solutions give rise to two thermodynamic effects such as enthalpic and entropic. The fundamentals of mixing of liquid metals forming binary alloys are generally discussed in terms of the enthalpic and entropic constructs as they are required to understand the nature and strength of the atomic bonding and the phase stability in the alloy. To determine these functions in the framework of QCA for regular alloy, the fitting parameters namely, the interaction energy parameter, W and the ratio of self-associates γ are needed. In the present work the parameters W and γ for the Na-K alloy at 384K were estimated over the whole range of concentration from the method of iterative procedure by fitting them into the available literature data of free energy of mixing (G_M) and chemical activities (a_i) [15] respectively in Eqs. (2) and (3a & 3b). We have obtained the best fit values of these parameters for the alloy to be $W = +1.106\,RT$ and $\gamma = 0.795$. The positive sign of W is an indication of like atom pairing (homo-coordination) in the liquid Na-K alloy. The estimated value of γ implies that the mixing of the molten metals Na and K is energetically favoured forming like atom clusters of Na atoms and K atoms in the ratio of about 4:5 so that the short range interaction between the nearest atoms results in the equilibrium properties of Na-K liquid alloy at the temperature of investigation. Chemical activity on the other hand is a measure of the tendency of the component to leave the solution. The deviations in the nature of a solution from ideal behaviour can be incorporated into activity. The calculated values of the free energy of mixing (Fig.1) and the chemical activities have found to agree quite satisfactorily with the available literature data [15] over the whole range of concentration.

Entropy of mixing is a measure of disorderness in the local arrangement of atoms in the system. Basically the term entropy represents the sharing of energy between the atoms in the neighbourhood. For the liquid Na-K alloy at 384K, we have computed the entropy of mixing, S_M from Eq. (5) by estimating the temperature dependence of the energy parameter to be $\frac{\partial W}{\partial T} = +0.416\ \text{Jmol}^{-1}\text{K}^{-1}$.

A reasonably good agreement is noticed in the computed values of the entropy of mixing and the corresponding experimental values [15] (Fig. 2). At a given temperature, the atoms in a liquid alloy are assumed to interact only with the limited nearest neighbours (short range interaction). If the temperature of the alloy is increased, the bonding interaction between the atoms is found to increase. The positive value of the temperature derivative of order energy for the Na-K alloy indicates that the interaction strengthens by about 0.42 $\text{Jmol}^{-1}\text{K}^{-1}$ with the increase in temperature of the alloy.
Using the calculated values of the free energy of mixing and the entropy of mixing, we have computed the heat of mixing, H_M throughout the whole concentration range from Eq. (6). The values of H_M predicted from quasi-chemical approximation agree quite satisfactorily with the available experimental data [15], with a small departure (Fig. 3). This disparity may be due to the role of the entropic effect as the behaviour of entropy is often complicated for many of the binary liquid alloys. As segregating systems are generally endowed with positive heat of mixing, the small positive values of H_M for Na-K liquid alloy also suggest that the alloy belongs to a weakly segregating system.

3.2. Viscosity

The local atomic ordering in a liquid A-B alloy is non-periodic, unlike crystals, as the solute atoms in the homogeneous solution can arrange in many ways by diffusion which contrasts with the pure metals A and B, in each of which the atoms can arrange only in a particular way. In the state of disorder in the atomic arrangements, some sort of short range atomic bonding (metallic, ionic, or covalent, or even secondary bonds, such as hydrogen bonding or Vander Waals forces as in the case of molecular crystals or liquids) can be assumed to exist between the nearest neighbours to present cohesive energy of solution. The cohesive energy of solution depends on the size difference of the atomic species and entirely to the size-dependent variations in the latent heats [30]. The cohesive energy may be thought to be responsible for the enthalpic effect and the viscous nature of the liquid alloys. Viscosity of liquid metals/alloys is one of the technologically important transport properties, needed to develop and optimize metallurgical technologies. The analysis of the viscosity gives some insight into the alloying behaviour in the liquid alloys.

In order to study the viscous property of liquid Na-K alloy, we have computed it as function of concentration from the M-H equation, Eqs. (6) and The Kaptay equation, Eq. (8) separately using the viscosities of the component metals at the temperature of investigation, calculated from Eq. (9) and the heat of mixing, H_M of the alloy, calculated from the quasi-chemical approximation. Here the values of the constants η_{0k} and E for the metals Na and K were taken from the reference [31]. Due to unavailability of the experimental data of viscosity for the Na-K alloy at 384K, we could not compare our theoretical result. Alternatively, as a reference for comparison we have computed the viscosity of the alloy at all concentrations from the same equations, Eqs. (6) and (8) but using the experimental data for the enthalpy of mixing [15]. Calculations done from M-H equation shows that the viscosity of Na-K alloy first increases with addition of the Na atoms up to about concentration $x = 0.1$ and then decreases slowly up to the concentration $x = 0.4$, and again increases rapidly for the rest of the concentration (Fig. 4). The Kaptay equation first shows a small decrease in viscosity and then a rapid increase. In both of these cases the viscosity calculated from experimental value of H_M and that from theoretical value of H_M are found to be almost matching (Fig. 4).

In our calculation, the value of V^E was taken as zero, due to lack of experimental data. It has been mentioned that the excess molar volume can be neglected for simplicity in the calculation of viscosity [29]. This can be done as this term has too small contribution $(\approx 10^{-7}\,m^3/mol)$, often not significant, in comparison to the mean molar volume of the metallic solution, $(xV_1 + (1-x)V_2 \approx 10^{-5}\,m^3/mol)$ in determining the viscosity. The two sets of the results computed using the theoretical values of H_M are satisfactorily comparable to the calculations based on experimental values of H_M with deviation from the ideal values $\eta_{id} = x\,\eta_1 + (1-x)\,\eta_2$ (Fig. 4).

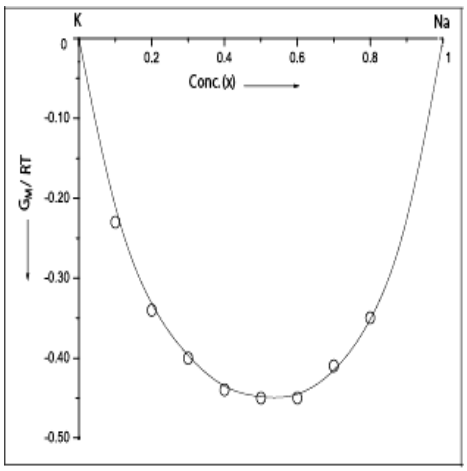

Fig. 1

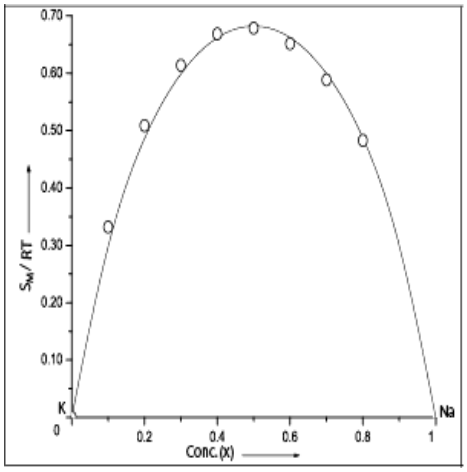

Fig. 2

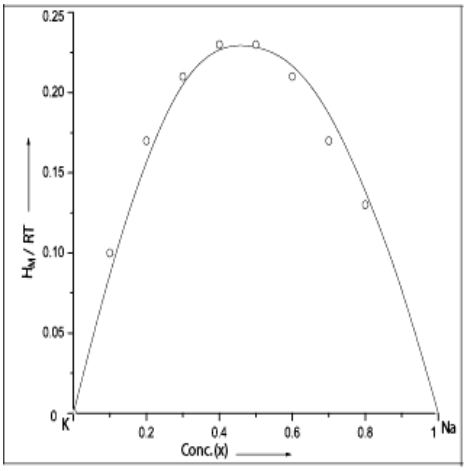

Fig. 3

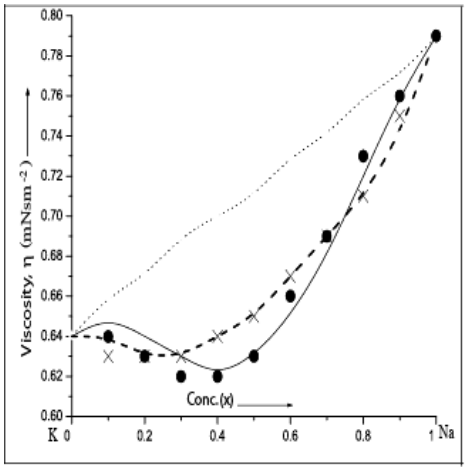

Fig. 4

Fig. 1: Free energy of mixing (G_M) of liquid Na-K alloy versus concentration (x) at 384K: Theory – solid curve; Experiment – circles.

Fig. 2: Entropy of mixing (S_M) of liquid Na-K alloy versus concentration (x) at 384K: Theory – solid curve ; Experiment – circles.

Fig. 3: Enthalpy of mixing (H_M) of liquid Na-K alloy versus concentration (x) at 384K: Theory – solid curve; Experiment – circles.

Fig. 4: Viscosity (η) of liquid Na-K alloy versus concentration (x) at 384K: (i) M-H Equation: (a) With theoretical H_M – solid line; (b) With experimental H_M – solid circles (ii) Kaptay equation: (a) With theoretical H_M – broken line; (b) With experimental H_M – crosses.

5. Conclusion

Calculations of the enthalpy of mixing and the viscosity in liquid Na-K alloy are presented. Our calculations based on Moelwyn-Hughes equation and Kaptay equation predict that liquid Na-K alloy at 384K is non-ideal with respect to the viscosity. There is negative deviation of viscosity from ideal behaviour.

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