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Thermodynamic, structural, surface and transport properties of Au-Ni liquid alloy at 1150 K

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ABSTRACT

Thermodynamic, structural, surface and transport properties of Au-Ni liquid alloy at 1150 K were computed using different theoretical approaches. The thermodynamic properties, such as excess Gibbs free energy of mixing, enthalpy of mixing, activity and excess entropy of mixing and structural properties, such as concentration fluctuation in long wavelength limit and Warren-Cowley short range order parameter were computed in the frame work of Flory's model. The effect of positive and negative values of the interchange energy parameter on the excess Gibbs free energy of mixing and concentration fluctuation in long wave length limit was also observed. The surface tension and surface concentration of the system were calculated using Butler's model. In transport property, the viscosity of the system was calculated using Kaptay and Budai-Benko-Kaptay (BBK) models.

1. Introduction

The comprehensive knowledge of the mixing behaviours of binary and multi-component liquid alloys are mandatory in order to develop new materials with desired characteristics. The study and explanation of the hetero-coordinating and homo-coordinating tendencies of the liquid alloys leading to different phase stabilities in the solid states are the most promising research directions in the metallurgical science and engineering. Several researchers [1-16] working in this field have long been attempting to reveal these behaviours by developing different modelling equations or by correlating them. Therefore, an attempt has been made in this work to study and explain the mixing and demixing behaviours of Au-Ni liquid alloy at 1150 K using different theoretical approaches.

The thermodynamics of Au-Ni system have been studied by different authors [17–26]. But the complete set of mixing properties of the system is lacking till date. In this regard, the mixing behaviours of Au-Ni liquid alloy at 1150 K have been explained in terms of the thermodynamic, structural, surface and transport properties. The

thermodynamic and structural properties of the system were computed on the basis of Flory's model [11, 27]. The mixing and demixing behaviours of the binary liquid alloys are generally explained in terms of the thermodynamic and structural properties. The compound forming liquid alloys are characterised by high negative values of excess free energy of mixing (ΔG_M^{xs}) and enthalpy of mixing (ΔH_M). The activities (a) of such systems show negative deviation from the Raoult's law. Meanwhile, the liquid alloys showing demixing behaviours have positive or very low negative values of ΔG_M^{xs} and ΔH_M, and their activities show positive deviation from the Raoul'ts law [4, 28, 29]. Likewise, the study of structural properties has been developed as an important tool to explain and predict the nature of the local arrangements of liquid alloys. If the computed values of concentration fluctuation in long wavelength limit ($S_{CC}(0)$) is less than the ideal value ($S_{CC}^{id}(0)$), i.e., if $S_{CC}(0) < S_{CC}^{id}(0)$, then hetero-coordinating or ordering nature of the system is expected and if $S_{CC}(0) > S_{CC}^{id}(0)$, then homo-coordinating or segregating nature of the system is expected. Consequently, the value of Warren-Cowley short range order parameter (α_1) is found to be non-zero and negative ($\alpha_1 < 0$) for ordering system, $\alpha_1 > 0$ is found for segregating system and $\alpha_1 = 0$ represents random mixing.

The surface properties and the viscosity of the liquid alloys are the direct consequences of their thermodynamic properties. Hence the surface properties, such as surface tension (σ) and the extent of surface segregation (x_i^S) of the system were computed using Butler's model [30-34]. The viscosity (η) of the system was computed on the basis of Kaptay [34, 35] and Budai-Benko-Kaptay (BBK) [36] models. The necessary theoretical formulations of the work are presented in the Section 2, the results and discussion are presented in the Section 3 and the conclusions are outlined in the Section 4.

2. Modeling Equations

Thermodynamic properties

In thermodynamic properties, excess free energy of mixing (ΔG_M^{xs}), activities of monomers Al (a_{Al}) and Ni (a_{Ni}), enthalpy of mixing (ΔH_M) and excess entropy of mixing (ΔS_M^{xs}) have been analysed using Flory's model [11, 22]. This model is fundamentally used to explain the properties of alloys showing like-atoms clusters or self-associates. The preferred liquid alloy, Au-Ni at 1100 K also show the same behaviour. If μ atoms of A (=Al) and v atoms of B (=Ni) are mixed near to their melting temperatures, then there are preferable associations among the like atoms in the metallic mixture of the types $\mu A \leftrightarrows A_\mu$ and $vB \leftrightarrows B_v$, called self-associates. The expression for ΔG_M^{xs} on the basis of the selected model can be given as

$$\Delta G_M^{xs} = RT \left[\ln \frac{(1-\beta)^x}{(1-\beta x)} + \frac{x(1-x)}{(1-\beta x)} \omega \right] \quad (1)$$

where R is the real gas constant, T is the absolute temperature, x is the composition of Al and ω is the interchange energy. The term β is the size factor and is given as

$$\beta = \frac{(\phi-1)}{\phi} \text{ with } \phi = \frac{\Omega_{Ni}}{\Omega_{Au}} \quad (2)$$

where Ω is the atomic size of the atom. The free energy of mixing (ΔG_M) and excess free energy of the mixing (ΔG_M^{xs}) are related as

$$\Delta G_M = \Delta G_M^{xs} + RT[x \ln x + (1-x) \ln(1-x)] \quad (3)$$

The activity ($a_i; i = 1,2$) of the ith component of the binary liquid alloy in terms of ΔG_M is expressed as

$$RT \ln(a_i) = \Delta G_M + (1-x) \left(\frac{\partial \Delta G_M}{\partial x} \right) \quad (4)$$

Using Equations (1) and (3) in Equation (4), the expression for a_i can be obtained as

$$\ln a_i = \ln[(1-\beta)(1-x)\gamma(x)] + \beta(1-x)\gamma(x) + \frac{\omega}{RT}(1-x)^2[\gamma(x)]^2 \quad (5)$$

where $\gamma(x) = 1/(1-\beta x)$.

The standard thermodynamic relation relating enthalpy of mixing, excess entropy of mixing and free energy of mixing are given as

$$\Delta H_M = \Delta G_M - T \left(\frac{\partial \Delta G_M}{\partial T} \right) \quad (6)$$

$$\text{and} \quad \Delta S_M^{xs} = - \left(\frac{\partial \Delta G_M^{xs}}{\partial T} \right) \quad (7)$$

Using Equations (1) and (3) in Equation (7), one can yield the respective expressions for ΔH_M and ΔS_M^{xs} as

$$\Delta H_M = \frac{x(1-x)\omega}{(1-\beta x)} - T\frac{x(1-x)}{(1-\beta x)}\frac{\partial \omega}{\partial T} + RT^2\frac{x(1-x)}{(1-\beta x)}\left[\frac{\beta}{(1-\beta)} - \frac{x}{(1-\beta x)}\frac{\omega}{RT}\right]\frac{\partial \beta}{\partial T}$$

(8)

$$\Delta S_M^{xs} = \frac{RTx(1-x)}{(1-\beta x)}\left[\frac{\beta}{(1-\beta)} - \frac{\omega x}{RT(1-\beta x)}\right]\frac{\partial \beta}{\partial T} - x(1-x)\frac{1}{(1-\beta x)}\frac{\partial \omega}{\partial T} - R[x\ln(1-\beta) - \ln(1-\beta x)]$$

(9)

where $\frac{\partial \omega}{\partial T}$ and $\frac{\partial \beta}{\partial T}$ are the temperature derivative terms of interchange energy and size factor respectively.

Structural properties

To analyse the local arrangements of atoms in the metallic mixture, the concentration fluctuation in long wavelength limit ($S_{CC}(0)$) and Warren-Cowley short range order parameter (WCSROP), α_1, were calculated. $S_{CC}(0)$ in terms of ΔG_M can be expressed by standard thermodynamic relation as [37, 38]

$$S_{CC}(0) = RT\left(\frac{\partial^2 G_M}{\partial x^2}\right)^{-1}$$

(10)

Using Equations (1) and (3), the relation for $S_{CC}(0)$ can be obtained as

$$S_{CC}(0) = \frac{x(1-x)}{1-x(1-x)f(\beta,\omega)}$$

(11)

where $f(\beta,\omega) = \left[\frac{2\omega}{RT}\frac{(1-\beta)}{(1-\beta x)^3} - \frac{\beta^2}{(1-\beta x)^2}\right]$ (12)

Putting $f(\beta,\omega) = 0$ in Equations (11) and (12), the expression for ideal value of $S_{CC}(0)$ can be obtained as

$$S_{CC}^{id}(0) = x(1-x)$$

(13)

The experimental value of $S_{CC}(0)$ can be calculated by the following relation

$$S_{CC}(0) = (1-x)\frac{a_i}{\left(\frac{\partial a_i}{\partial x}\right)} = x\frac{a_j}{\left(\frac{\partial a_j}{\partial(1-x)}\right)}$$

(14)

Herein, a_i and a_j are the experimental values of the activity of ith (a_{Au}) and j^{th} (a_{Ni}) respectively.

The Warren-Cowley short range order parameter (α_1) for binary liquid alloys can be given as [37, 38]

$$\alpha_1 = \frac{(S-1)}{[S(Z-1)+1]}$$

(15)

where Z is the coordination number and its value is taken to be 10 in present work and $S = \frac{S_{CC}^{id}(0)}{S_{CC}(0)}$.

Surface properties

According to the Butler model, the surface tension (σ) of the binary liquid alloy in the initial melt can be expressed as [30-32]

$$\sigma = \sigma_1^0 + \frac{RT}{\varphi_1}\left[\ln x_1^S - \ln x_1\right] + \frac{\Delta G_{S,1}^{xs} - \Delta G_1^{xs}}{\varphi_1}$$

(16)

$$\sigma = \sigma_2^0 + \frac{RT}{\varphi_2}\left[\ln x_2^S - \ln x_2\right] + \frac{\Delta G_{S,2}^{xs} - \Delta G_2^{xs}}{\varphi_2}$$

(17)

where σ_i^0 ($i = 1, 2$) is the surface tension of the pure atom i, φ_i is the molar surface area of pure atom i and x_i^S and x_i are the surface and bulk concentrations of atoms i in the surface phase and bulk phase of the liquid alloy respectively. $\Delta G_{S,i}^{xs}$ is the surface partial excess free energy of atom i and ΔG_i^{xs} is the bulk partial excess free energy of atom i and they are related as $\Delta G_{S,i}^{xs} = \beta\Delta G_i^{xs}$, where β is constant and $\beta = 0.83$ [28, 32, 34, 39] is taken for calculations in the present work.

The molar surface area of ith atom is expressed as

$$\varphi_i = 1.091\left(V_i^0\right)^{2/3}(N_A)^{1/3}$$

(18)

where V_i^0 is the molar volume of ith atom and $N_A = N$ is the Avogadro's number. The molar volume (V_i^0) of ith atom is calculated by taking the ratio of its mass (m_i) and density (ρ_i^0) at the required temperature (T_K). The temperature dependent expressions of σ_i^0 and ρ_i^0 can be given as [40]

$$\sigma_i^0 = \sigma_0 + (T_K - T_0)\frac{d\sigma}{dT}$$

and $\rho_i^0 = \rho_0 + (T_K - T_0)\frac{d\rho}{dT}$ (19)

Herein, σ_0 and ρ_0 are the surface tension and density of the pure atom at its melting temperature, T_0 is the melting temperature of the pure atom and $\frac{d\sigma}{dT}$ and $\frac{d\rho}{dT}$ are the temperature derivative terms of surface tension and density respectively.

Transport property

In transport property, the viscosity (η) of the system was calculated using Kaptay equation [34,35] and

Budai-Benko-Kaptay (BBK) model [36]. According to the Kaptay model, the expression for the viscosity of the binary liquid alloys can be given by

$$\eta = \left(\frac{h\,N_A}{\sum_i x_i V_i^0 + V^E}\right) \exp\left(\frac{\sum_i x_i G_i^* - (0.155 \pm 0.015)H_M}{RT}\right)$$
(20)

where h is Planck's constant and V^E is the excess volume of alloy formation which can be neglected. The term G_i^* stands for the Gibb's energy of activation of the viscous flow of ith component and is expressed as

$$G_i^* = RT \ln\left(\frac{\eta_i^0 V_i^0}{hN_A}\right)$$
(21)

where η_i^0 is the viscosity of the pure component of the metallic mixture and can be given as [40]

$$\eta_i^0 = \eta_0 \exp\left(\frac{E}{RT}\right)$$
(22)

where η_0 is the viscosity of the pure atom at its melting temperature and E is the activation energy. The ideal value of the viscosity of the liquid mixture can be expressed as

$$\eta^{id} = \sum_i x_i \eta_i^0$$
(23)

Following Budai-Benko-Kaptay [36], the expression for the viscosity of binary liquid alloy can be given as

$$\eta = A.\frac{(\sum_i x_i M_i)^{1/2}}{(\sum_i x_i V_i + V^E)^{2/3}} T^{1/2} \exp\left[\frac{B}{T}\left(\sum_i x_i T_{m,i}^* - \frac{\Delta H_M}{q.R}\right)\right]$$
(24)

where $q = 25.4$ is the semi-empirical parameter which is related to the cohesion energy of the pure liquid metal and $T_{m,i}^*$ is defined as effective melting point of the pure component. The values of the coefficients A and B can be taken as $(1.8 \pm 0.39) \times 10^8$ and (2.34 ± 0.2) respectively

3. Results and Discussion

The value of $\phi = 1.63$ was first calculated using Equation (2) and then the effect of interchange energy (ω) on free energy of mixing (ΔG_M) was observed. For this purpose, the values of ΔG_M were computed using Equations (1-3) for different values of ω, such as $\pm 3RT, \pm 2RT$ and $\pm 1RT$. The plot of the isotherms of the compositional dependence of ΔG_M for aforementioned values of ω at 1150 K is

portrayed in Fig. 1(a). It can be observed that the negative values ΔG_M gradually increases with the increase in the negative values of ω and decreases with increase in the positive values of ω. The value of ΔG_M is found to be positive for $x_{Au} = x < 0.8$ for $\omega = +3RT$.

The optimised value of $\omega = 15400$ and $\beta = 0.32886$ were then obtained using Equations (1-3) and the experimental value of ΔG_M^{xs} [29] employing the method of successive approximation. The experimental and the calculated values of ΔG_M^{xs} were found to be in well agreement (Fig. 1(b)). Both of these values are found to be positive which correspond that the Au-Ni liquid alloy at 1150 K is a homo-coordinating or segregating system. Hence, the system shows demixing tendency with respect to the thermodynamic functions. The like-atoms clusters or self-associates are favourable in the liquid state and the system exhibit liquid miscibility gaps [4].

The activity of the system was computed using Equations (5) and above determined model parameters. The experimental [29] as well as the theoretical values of activities of the monomers Au (a_{Au}) and Ni (a_{Ni}) were found to be in well agreement (Fig. 1(c)). Both of these values show positive deviation from Raoult's law indicating that the preferred system is segregating in nature. Theoretical investigations show that the model parameters have successfully reproduced excess free energy of mixing and activity of the system.

The temperature derivative terms of model parameters ($\frac{\partial \omega}{\partial T} = -7.2000$ and $\frac{\partial \beta}{\partial T} = -0.00001$) were optimised using Equation (8) and the experimental value of the enthalpy of mixing (ΔH_M) [29]. The enthalpy of mixing (ΔH_M) and the excess entropy of mixing (ΔS_M^{xs}) were then computed using Equations (8) and (9) respectively with the aid of abovementioned model parameters. Both of these values are found to be in good agreement with their respective experimental values (Fig. 1 (b)). Moreover, the positive value of ΔH_M reveals the demixing tendency of the system.
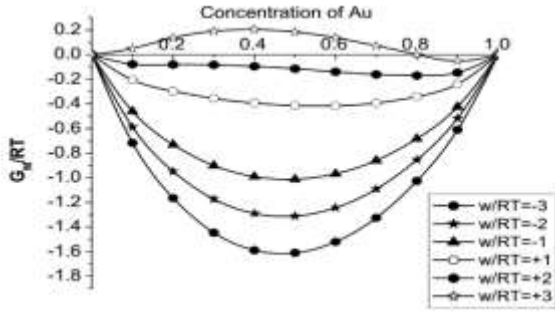
(a)
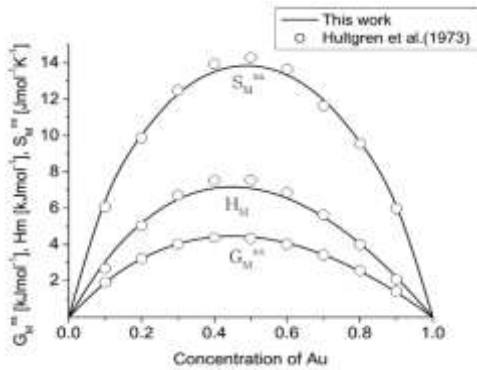
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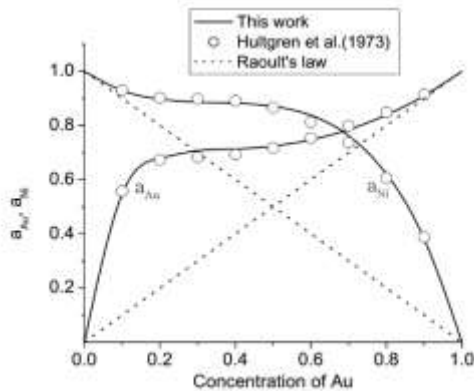
(c)

Fig. 1(a-c): Plots of the compositional dependence of thermodynamic functions for Au-Ni liquid alloys at 1150 K. (a) Effect of positive and negative values of ω/RT on ΔG_M, (b) $\Delta G_M^{xs}, \Delta H_M$ and ΔS_M and (c) a_{Au} and a_{Ni}.

The greater insight of the homo-coordinating and hetero-coordinating nature of the liquid alloys can be obtained by computing and analysing the structural

functions. For this purpose, the theoretical, ideal and experimental values of the concentration fluctuation in long wavelength limit ($S_{CC}(0)$) were computed using Equations (11-14) with the help of abovementioned model parameters and the experimental value of activity [29].

The effect of the positive and the negative values of ω on $S_{CC}(0)$ were studied by arbitrarily varying the values of as $\omega = \pm4RT, \pm3RT, \pm2RT$ and $\pm1RT$. It can be observed that with gradual increase in the negative values of ω, the deviation between the ideal and the theoretical values of $S_{CC}(0)$ gradually increases (Fig. 2(a)). These results predict that the preferred system would turn into segregating to ordering nature with increase in the negative value of ω. Likewise, the effect of positive values of ω on $S_{CC}(0)$ as a function of concentration is presented in Fig. 2(b). It indicates that the computed values of $S_{CC}(0)$ gradually increases and reaches the peak value at $x = 0.4$ for $\omega = +1RT$. For $\omega = +2RT$, $S_{CC}(0)$ is found to have negative value in the range $0.1 < x < 0.53$. Likewise, for $\omega = +3RT$, $S_{CC}(0)$ is found to be negative in the range $x < 0.9$ and for $\omega = +4RT$, it is found to be negative in the range $0.1 < x < 0.62, 0.69 < x < 0.9$. As $S_{CC}(0)$ cannot have negative values, the value of interchange energy should be in between 1 and 2, i.e., $+1RT < \omega < +2RT$. The same result ($\omega = +1.6107RT$) was found during the investigation of the thermodynamic properties.

The theoretical as well experimental computed values of $S_{CC}(0)$ are in excellent agreement and are found to be grater than the ideal value at all concentrations (Fig. 2(c)). Therefore, it can be concluded that the liquid Au-Ni at 1150 K shows complete homo-coordinating tendency. The theoretical values of WCSROP (α_1) were computed using Equation (15) and plotted in Fig. 2(d). The value of $\alpha_1 > 1$ at all compositions revealing the similar nature as by $S_{CC}(0)$. Thus, the theoretical findings of thermodynamic and structural properties are in accordance and suggest the demixing behaviour of the system at selected temperature.

The surface tension (σ) and the surface concentrations of the monomers Au (x_{Au}^S) and Ni (x_{Ni}^S) were computed using Butler's model [30]. The surface tensions and densities of the pure components of the system were computed using Equation (19) and the parameters in Table 1. The values of σ, x_{Au}^S and x_{Ni}^S were then computed using Equations (17) and (18). The ideal value of the surface tension (σ^{id}) of the liquid alloy is obtained as the additive sum of the surface tension of the pure atoms in it using the relation $\sigma^{id} = \sum_i x_i \sigma_i^0$. The surface concentration of Au atom is found to be higher whereas that of Ni is found to be lower than their respective ideal values (Fig. 3(a)). Therefore, Au atoms segregates on the surface phase and Ni atoms remains in the bulk phase in the initial melt. The computed values of the surface tension of the system is found to be less than the ideal value at all compositions (Fig. 3(b)).

In transport property, the viscosity (η) of the system was computed using Kaptay model [34,35] and Budai-Benko-Kaptay (BBK) model [36]. The viscosity is directly related to the enthalpy of mixing of the system and is the measure of the cohesion energy of the metallic solution. The viscosity (η) and its necessary ingredients for the system was computed using Equations (20-24) with the help of the input parameters from Table 1. The computed values of the viscosity of the system from both of the preferred models are found to be less than the ideal value at all compositions (Fig. 4).
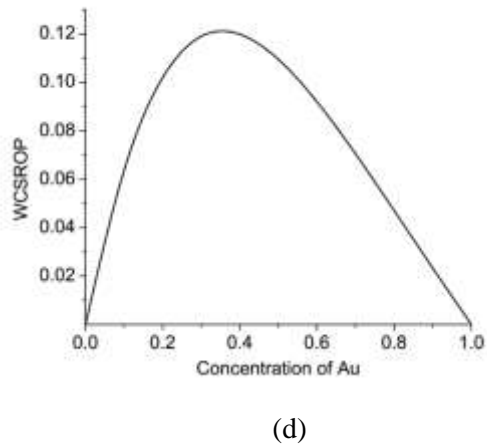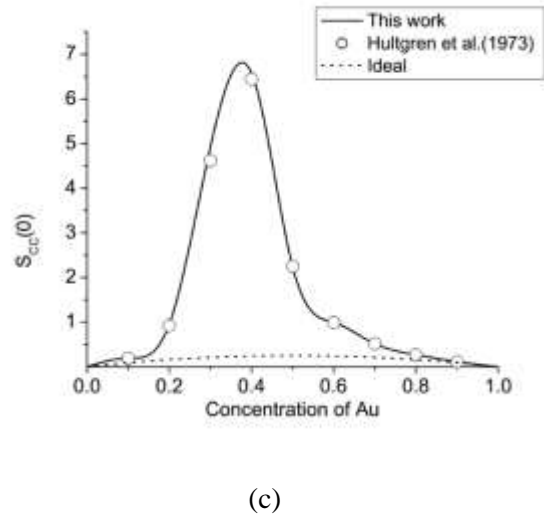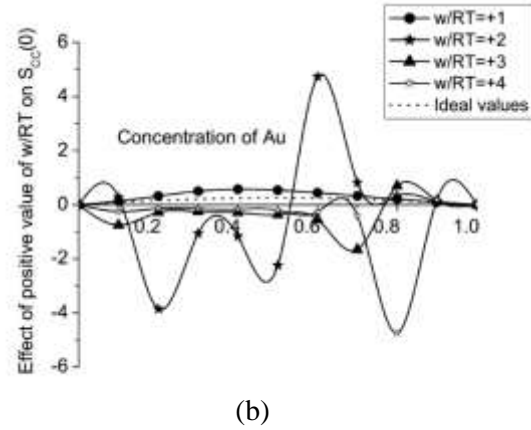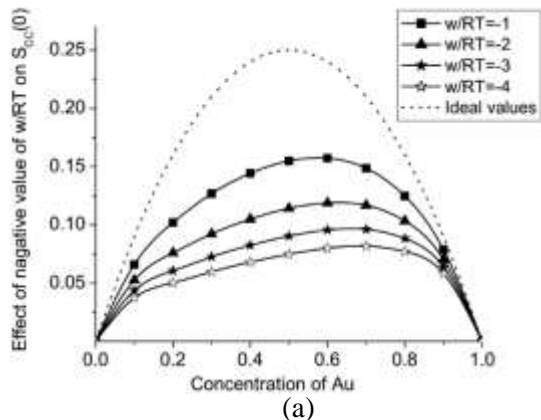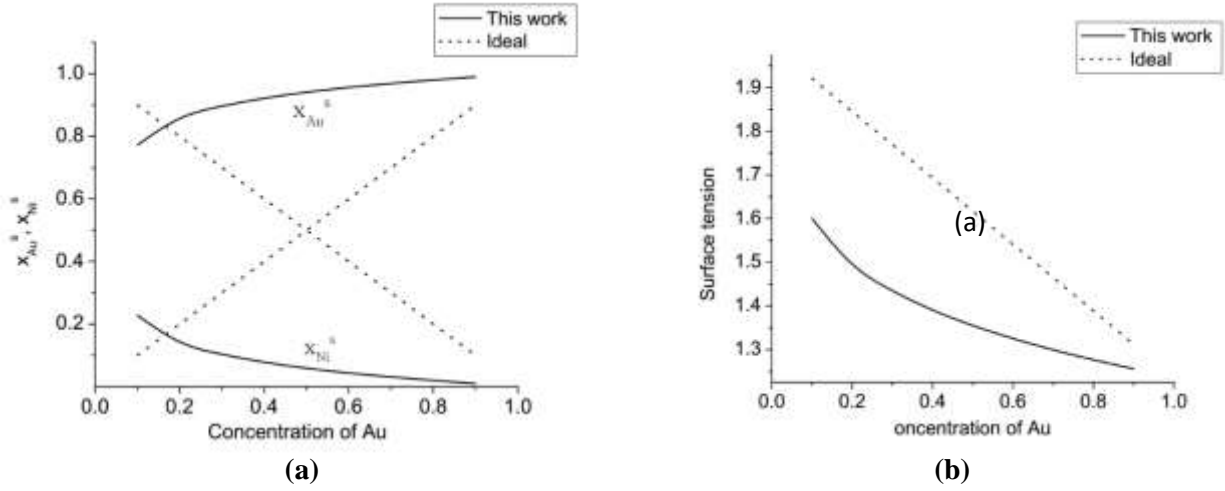

(b)

(c)

(d)

Fig. 2(a-d): Plots of the compositional dependence of structural functions for Au-Ni liquid alloy at 1150 K. (a) Effect of negative values of ω/RT on $S_{CC}(0)$, (b) Effect of positive values of ω/RT on $S_{CC}(0)$, (c) $S_{CC}(0)$ and (d) WCSROP.

(a)

(a)

(b)

3(a, b): Plots of the compositional dependence of the surface tension (σ) and surface concentrations (x_{Au}^S and x_{Ni}^S) for Au-Ni liquid alloy at 1150 K. (a) Surface tension and (b) Surface concentrations
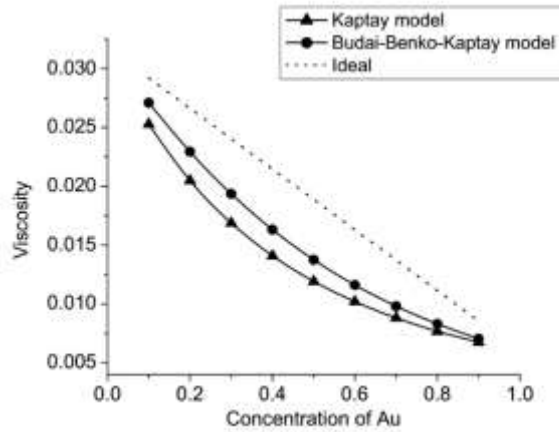

Fig. 4: The compositional dependence of viscosity (η) for Au-Ni liquid alloy at 1150 K.

Table 1: Input parameters for surface tension (σ) and viscosity (η) [40]

Metal	T_0 (K)	ρ_0 (kg m^{-3})	$\partial\rho/\partial T$ (kg m^{-3}K^{-1})	σ_0 (N m^{-1})	$\partial\sigma/\partial T$ (N m^{-1}K^{-1})	η_0 (Nsm^{-2})	E (Jmol^{-1})
Au	1336	17360	-1.50	0.1140	-0.00052	0.001132	15900
Ni	1727	7905	-1.160	0.1778	-0.00038	0.0001663	50200

4. Conclusion

The computed values of the excess free energy of mixing and the enthalpy of mixing were found to be positive and the activity showed positive deviation from the Raoult's law. The value of the concentration fluctuations in long wavelength limit was found to be greater than the ideal value and Warren-Cowley short range order parameter was found to be positive. These results correspond that the liquid Au-Ni alloy at 1150 K showed homo-coordinating behaviours. The computed value of surface tension and viscosity were less than the ideal values. In the initial melt, Au atoms segregates in the surface phase whereas Ni atoms remains in the bulk phase.

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