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Thermodynamic, structural, surface and transport properties of Au-Cu melt

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Abstract

Thermodynamic, structural, surface and transport properties of Au-Cu liquid alloy were calculated on the basis of different theoretical modeling equations. The thermodynamic properties, such as excess Gibbs free energy of mixing, enthapy of mixing and activity were estimated and compared with the available experimental and literature data on the basis of simple theory of mixing (STM). The best fit values of model parameters were estimated using the experimental values of excess Gibbs free energy of mixing and enthalpy of mixing of the system at 1550 K. Using the same model parameters and frame, the concentration fluctuation in long wavelength limit and Warren-Cowley short range order parameter were calculated and analysed to understand the local arrangement of atoms in the liquid mixture. The surface tension and extent of surface segregation of atoms in the initial melt were computed using Butler's model. In transport properties, the ratio of mutual to intrinsic diffusion coefficients was calculated using STM and viscosity was calculated using Kaptay equation. The system showed complete ordering tendency at its melting temperature.

Keywords
Molar surface area, ordering nature, noble metals, transition metals, surface phase.

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1 Introduction

The important information related to the alloy catalytic, wettability, electrical, magnetic, mechanical, etc. behaviours can be obtained by computing and analyzing the thermodynamic, structural, surface and transport properties of liquid alloys. In this regard, several researchers [1–11] working in the field of material science and engineering have made numerous attempts to access the mixing properties of different binary liquid alloys. Therefore, thermodynamic, structural, surface and transport properties of Au-Cu liquid alloy have been studied in this work on the basis of different theoretical approaches.

Topor and Kleppa in 1984 [12] and Okamoto et al. in 1987 [13] have summarized a brief review of the literature related to the thermodynamic properties and phase diagrams of the Au-Cu system. Later, Sundman et al. in 1998 [14] used Calphad approach to access the thermodynamic description of the system using the available experimental results for solid and liquid phases of the system. They successfully reproduced the experimental results by developing a theoretical framework, called Compound Energy Formalism. They further presented optimised values of self-consistent parameters for the excess Gibbs free energy of mixing for the system. Han et al. in 2004 [15] used molecular dynamics simulation techniques to determine the density and specific heat of the system above and below its melting temperature. Their results showed that the density of the system increased linearly with decrease in temperature and specific heat remained constant in the range 900-1900 K. In the same year, Brillo et al. [16] experimentally measured the density and thermal expansion of the system and its pure components. Their results were found to be consistent with those obtained by Han et al. Recently, Kong et al. in 2018 [17] investigated the structural, thermodynamic and elastic properties of the system using first principle calculations based on density functional theory. They found that the metallic complexes, $AuCu$, Au_3Cu and $AuCu_3$ to be mechanically stable.

From the above mentioned literature, it can be stated that their appears immense interest to access the mixing properties of Au-Cu system. Therefore, the thermodynamic, structural, surface and transport properties of the system have been computed using different theoretical approaches in present work.

2 Formulations

2.1 Thermodynamic properties

The thermodynamic properties of the system have been computed using the modeling equations of simple theory of mixing (STM).On the basis of the model, the excess Gibbs free energy of mixing for binary liquid alloy is expressed as [18–20]

$$\frac{\Delta G_M^{\mathrm{xs}}}{RT} = \int_0^c \log \sigma^2 dx = x_A \log \gamma_A + x_B \log \gamma_B \quad (1)$$

where x_A (A=Au) and x_B (B=Cu) are the mole fractions of atoms A and B respectively in the liquid mixture, such that $x_A + x_B = 1$. The terms ϕ, γ_A and γ_B can be given as

$$\phi = (\zeta + 2x_A - 1)\exp(-\omega/zk_BT)/2x_A \quad (2)$$

$$\gamma_A = [(\zeta - 1 + 2x_A)/x_A(1+\zeta)]^{z/2} \quad (3)$$

$$\gamma_B = [(\zeta + 1 - 2x_A)/x_B(1+\zeta)]^{z/2} \quad (4)$$

$$\zeta = 1 + 4x_A x_B[\exp(2\omega/zk_BT) - 1]^{1/2} \quad (5)$$

Herein, ω is called the ordering energy, is the model fit parameter and z is the coordination number. If $\omega < 0$, then complex formation or hetero-coordinating tendency in the initial melt is expected, otherwise homo-coordinating tendency is expected.

At equiatomic concentration ($x_A = x_B = 0.5$), the expression for ΔG_M^{xs} can be given as [18–20]

$$\frac{\Delta G_M^{\mathrm{xs}}}{RT} = \ln[2^{z/2}(1 + \exp(\frac{-\omega}{zk_BT}))^{-z/2}] \quad (6)$$

Once the value of ω is determined at $x_A = x_B = 0.5$, ΔG_M^{xs} can be computed in the entire composition range using the relation

$$\Delta G_M^{\mathrm{xs}} = Nk_BT(x_A x_B \frac{\omega}{k_BT}) \quad (7)$$

The relation between Gibbs free energy of mixing (ΔG_M) and excess Gibbs free energy of mixing ΔG_M^{xs} is expressed as

$$\Delta G_M = \Delta G_M^{\mathrm{xs}} + RT(x_A \ln x_A + x_B \ln x_B) \quad (8)$$

The enthalpy of mixing (ΔH_M) is expressed in terms of ΔG_M^{xs} as

$$\Delta H_M = \Delta G_M^{\mathrm{xs}} - T\left(\frac{\partial \Delta G_M^{\mathrm{xs}}}{\partial T}\right)_P \quad (9)$$

where P is the pressure of liquid mixture. Using Equations (7) in Equation (9), one can obtain

$$\Delta H_M = Nk_BT[x_A x_B \frac{\omega}{k_BT} - \frac{1}{k_BT} x_A x_B \frac{d\omega}{dT}] \quad (10)$$

where $d\omega/dT$ is the temperature derivative term of ordering energy as it is temperature-dependent and concentration independent. Once the values of ΔH_M and ΔG_M^{xs} are determined, the values of excess entropy of mixing ΔS_M^{xs} can be calculated using the standard thermodynamic relation

$$\Delta G_M^{\mathrm{xs}} = \Delta H_M - T\Delta S_M^{\mathrm{xs}} \quad (11)$$

The activity ($a_i, i = A, B$) of each component of the liquid alloy is expressed in the form as [18–20]

$$\ln a_A = \ln x_A + x_B^2 \frac{\omega}{k_BT} \quad (12)$$

$$\ln a_A = \ln x_A + x_A^2 \frac{\omega}{k_BT} \quad (13)$$

2.2 Structural properties

In structural functions, the expression for concentration fluctuation in long wavelength limit ($S_{\mathrm{CC}}(0)$) in terms of ΔG_M and and a_i can be given as [4, 11, 18, 21–23]

$$S_{CC}(0) = RT\left(\frac{\partial^2 G_M}{\partial x_A^2}\right)^{-1}_{T,P,N} = RT\left(\frac{\partial^2 G_M}{\partial x_B^2}\right)^{-1}_{T,P,N} \qquad (14)$$

and

$$S_{\mathrm{CC}}(0) = x_B a_A(\frac{\partial a_A}{\partial x_A})^{-1} = x_A a_B(\frac{\partial a_B}{\partial x_B})^{-1} \qquad (15)$$

With the aid of Equation (6), Equation (14) becomes

$$S_{\mathrm{CC}}(0) = x_A x_B[1 + \frac{z}{2\zeta}(1-\zeta)]^{-1} \qquad (16)$$

The ideal values of $S_{\mathrm{CC}}(0)$ is calculated using the following equation

$$S^{id}_{\mathrm{CC}}(0) = x_1 x_2 \qquad (17)$$

Another structural functions, Warren-Cowley short range–order parameter (α_1) can be expressed in terms of $S_{\mathrm{CC}}(0)$ as [11, 21, 24, 25]

$$\alpha_1 = \frac{S-1}{[S(Z-1)+1]} \qquad (18)$$

where

$$S = \frac{S_{\mathrm{CC}}(0)}{S^{id}_{\mathrm{CC}}(0)} \qquad (19)$$

2.3 Surface properties

The surface tension (σ) and surface concentration (x_i^S) of the system have been calculated in the framework of Butler's model. According to this model, σ for binary liquid solution is expresses in the form [23, 25–28] as

$$\sigma = \sigma_A + \frac{RT}{\lambda_A} ln\frac{x_A^S}{x_A^b} + \frac{\Delta G^{\mathrm{xs}}_{A,S} - \Delta G^{\mathrm{xs}}_{A,b}}{\lambda_A} = \sigma_B + \frac{RT}{\lambda_B}\ln\frac{x_B^S}{x_B^b} + \frac{\Delta G^{\mathrm{xs}}_{B,S} - \Delta G^{\mathrm{xs}}_{B,b}}{\lambda_B} \qquad (20)$$

where σ_i is the surface tension, λ_i is molar surface area, x_i^S is the surface concentration, x_i^b is the bulk concentration, $\Delta G^{\mathrm{xs}}_{i,S}$ is the molar surface partial excess Gibbs free energy and $\Delta G^{\mathrm{xs}}_{i,b}$ is the molar bulk partial excess Gibbs free energy for the component i in the liquid mixture at the melting temperature of the alloy. $\Delta G^{\mathrm{xs}}_{i,S}$ and $\Delta G^{\mathrm{xs}}_{i,b}$ are correlated by the relation [25, 27, 28]

$$\Delta G^{\mathrm{xs}}_{i,S} = \beta\Delta G^{\mathrm{xs}}_{i,b} \qquad (21)$$

where the value of $\beta = 0.8181$ [25, 28] has been taken for the present calculations. The molar surface area (λ_i) of component i in the liquid mixture can be calculated using the relation

$$\lambda_i = fN_A^{\frac{1}{3}}V_i^{\frac{2}{3}} \qquad (22)$$

where N_A is the Avogadro's number and $V_i(=\frac{m_i}{\rho_i})$ is the molar volume of element i in the liquid mixture. The values of σ_i and V_i are calculated with the help of the following relations [16, 28–30]

$$\sigma_i = \sigma_i^0 + \frac{d\sigma}{dT}(T - T_0) \qquad (23)$$

and

$$V_i = V_i^0[1 + +\kappa_i(T - T_0)] \qquad (24)$$

where σ_i^0 is the surface tension, V_i^0 is the molar volume and κ_i [28, 29, 31] is the temperature coefficient of volume expansion for i^{th} component at its melting temperature (T_0).

2.4 Transport properties

The viscosity (η) and ratio of mutual to intrinsic diffusion coefficients (D_M/D_{id}) have been computed in order to analyze the transport properties of the system. On the basis of simple theory of mixing, η of binary liquid alloy can be given as [18, 23]

$$\eta = \eta_{\mathrm{id}}[1 - x_A x_B(\frac{2\omega}{k_B T})] \qquad (25)$$

where η_{id} is the ideal value of viscosity calculated using the relation [18, 23, 30]

$$\eta_{\mathrm{id}} = x_A\eta_A + x_B\eta_B \qquad (26)$$

Herein, η_i stands for the viscosity of pure component i at the melting temperature of the liquid alloy and is calculated using the relation

$$\eta_i = \eta_i^0 \exp\left(\frac{E}{RT}\right) \qquad (27)$$

The term η_i^0 is viscosity and E is activation energy of the component i at its melting temperature.

The Kaptay equation for the viscosity of binary liquid alloy can be expressed as [10, 23, 32]

$$\eta = \Big(\frac{hN_A}{x_A V_A + x_B V_B + V^E}\Big)\mathrm{x} \left(\frac{x_A G_A^* + x_B G_B^*(0.155 \pm 0.015)\Delta H_M}{RT}\right) \qquad (28)$$

where G_i^* is the Gibbs energy of activation of the viscous flow for pure component i and is given as

$$G_i^* = RT \ln\left(\frac{\eta_i V_i}{hN_A}\right) \tag{29}$$

where h is the Planck's constant, V^E is excess volume upon alloy formation which can be neglected and the rest of terms carry their usual meanings as stated above.

In the framework of STM, the values of D_M/D_{id} for binary liquid alloy can be estimated using the relation [18, 19, 23]

$$\frac{D_M}{D_{\text{id}}} = \left[1 - x_A x_B\left(\frac{2\omega}{k_B T}\right)\right] \tag{30}$$

$D_M = x_A D_B + x_B D_A$; D_A and D_B are the self-diffusion coefficients of pure components A and B respectively. Equation (30) can also expressed in the form as

$$\frac{D_M}{D_{\text{id}}} = \frac{S_{\text{CC}}^{id}(0)}{S_{\text{CC}}(0)} \tag{31}$$

Herein, the terms carry their usual meanings as stated in the above sub-sections.

3 Results and Discussion

3.1 Thermodynamic properties

The model parameter (ω/k_BT) of simple theory of mixing (STM) is assumed to be temperature-dependent but concentration independent. The effect of model parameter on excess Gibbs free energy of mixing (ΔG_M) was analysed by arbitrarily varying the value of ω/k_BT in the range ±5. The values of ΔG_M were calculated at 1550 K using Equations (7) and (8) in the above mentioned range of model parameter at all concentrations. The compositional dependence of $\Delta G_M/RT$ so estimated are plotted in Figures 1 and 2.

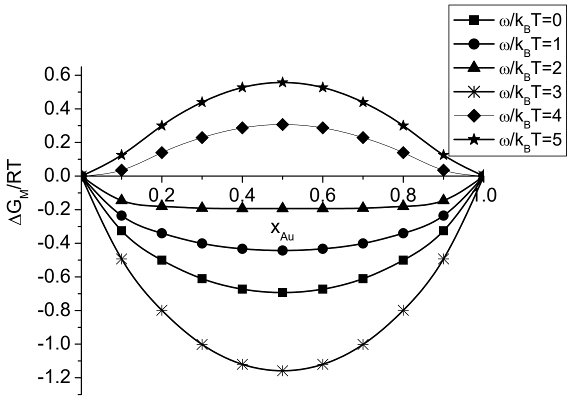

Figure 1: Effect of positive values of ω/k_BT on $\Delta G_M/RT$ versus x_{Au} at 1550 K.

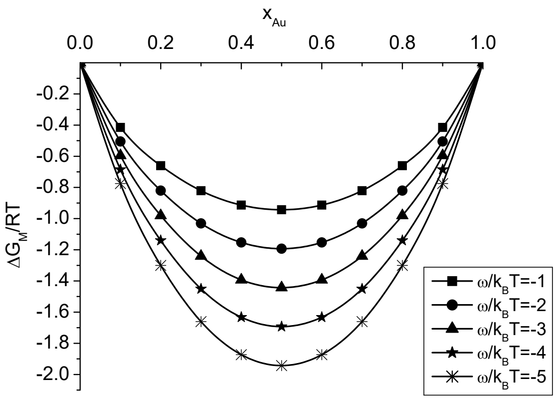

Figure 2: Effect of negative values of ω/k_BT on $\Delta G_M/RT$ versus x_{Au} at 1550 K.

In the preferred model, it is assumed that if $\omega/k_BT < 0$, then ordering tendency among the atoms of liquid alloy is expected and if $\omega/k_BT > 0$, then segregating tendency is expected. It can be observed that the negative values of $\Delta G_M/RT$ gradually decreased with increase in positive values of ω/k_BT, except for $\omega/k_BT = 3$ (Figure 1). Herein, $\omega/k_BT = 0$ represents the ideal values of $\Delta G_M/RT$. The perusal of Figure 2 corresponds that with increase in negative values of ω/k_BT, the negative values of $\Delta G_M/RT$ gradually increased. Thus, the hetero-coordinating tendency or complex forming tendency in the liquid alloy gradually increased with the increase in negative values of ω/k_BT.

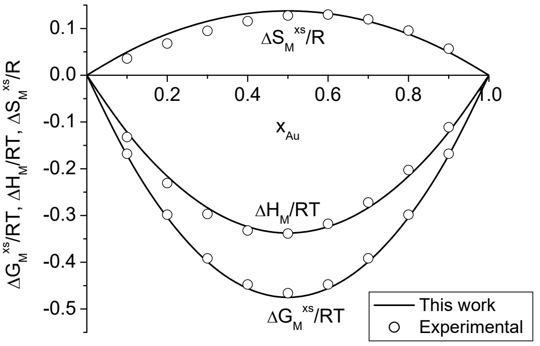

Figure 3: Compositional dependence of $\Delta G_M^{\text{xs}}/RT$, $\Delta H_M/RT$ and $\Delta S_M^{\text{xs}}/R$ for Au–Cu liquid alloy at 1550 K.

The value of ω/k_BT was first obtained with the help of Equation (6) and experimental value of ΔG_M^{xs} [5] at $x_{\text{Au}} = 0.5$ and 1550 K. Equation (7) was then used to calculate ΔG_M^{xs} for the system at all concentrations. In due course, the value of ω/k_BT was slightly adjusted following the method of successive approximation confirming that the obtained values were consistent with those of experimental. The best fit value of ω/k_BT was determined to be -1.900. The compositional dependence of calculated and experimental values of $\Delta G_M^{\mathrm{xs}}/RT$ are depicted in Figure 3.

Both the experimental and computed values of $\Delta G_M^{\mathrm{xs}}/RT$ were found to be consistent with each other at all compositions. The Au–Cu binary system is a regular alloy and the equilibrium values of $\Delta G_M^{\mathrm{xs}}/RT$ are -0.47500 (this work) and -0.46611 (experimental [5]) at $x_{\mathrm{Au}} = 0.5$. Therefore, the system is found to be symmetric with respect to excess Gibbs free energy of mixing and weakly interacting in nature (Figure 3). Moreover, the computed values of $\Delta G_M^{\mathrm{xs}}/RT$ were found to be negative in the entire concentration range indicating the system to be ordering in nature at its melting temperature, 1550 K.

The temperature derivative term of the model parameter $(d\omega/dT)$ was estimated using Equation (10) and experimental data of H_M [5]. The best fit value of $\frac{1}{k_B}\frac{d\omega}{dT}$ was found to be -0.5500 employing iterative process. The values of H_M and S_M^{xs} were then computed at all concentrations using Equations (10) and (11) at 1550 K, and above determined model parameters. The computed and experimental values of these thermodynamic parameters are plotted as a function of concentration in Figure 3.

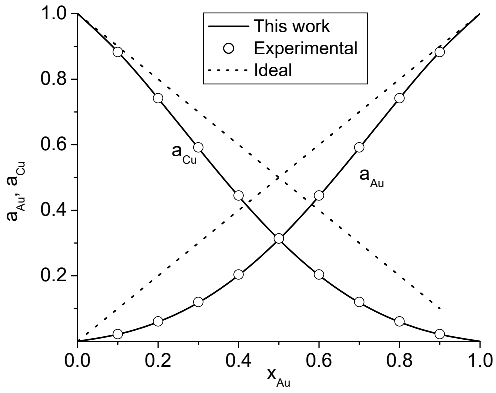

Figure 4: Activities of Au (a_{Au}) and Cu (a_{Cu}) versus x_{Au} of Au–Cu liquid alloy at 1550 K.

The maximum negative values of H_M/RT were -0.33750 (this work) and -0.33864 (experimental [5]) at $x_{\mathrm{Au}} = 0.5$. Meanwhile, the optimum values of S_M^{xs}/R were 0.13750 (this work) and 0.12770 (experimental [5]) at $x_{\mathrm{Au}} = 0.5$. Additionally, the calculated and experimental values of these functions were found to be consistent with each other (Figure 3). The small negative value of H_M/RT revealed the system to be weakly interacting and ordering in nature at its melting temperature.

The activities of the monomers Au (a_{Au}) and Cu (a_{Cu}) of the system were estimated using Equations (12) and (13) with the aid of determined value ω/k_BT. The experimental and calculated values of a_{Au} and a_{Cu} showed negative deviations from their respective ideal values revealing the system to be hetero-coordinating in nature. They agreed well with each other at all concentrations and 1550 K (Figure 4) thereby validating the present iteration process.

3.2 Structural properties

The knowledge of structural functions helps to understand the nature of arrangement of atoms in the liquid alloy at microscopic level. The concentration fluctuation in long wavelength limit ($S_{\mathrm{CC}}(0)$) and Warren-Cowley short range order parameter (α_1) were calculated in structural properties. At a temperature and concentration, if the calculated values of $S_{\mathrm{CC}}(0) < S_{\mathrm{CC}}^{\mathrm{id}}(0)$, then the alloy shows ordering nature, i.e., there is preferential association among the unlike atoms. Under the similar conditions, if $S_{\mathrm{CC}}(0) > S_{\mathrm{CC}}^{\mathrm{id}}(0)$, then it shows segregating nature, i.e., the association among the same type of atoms is favoured in the liquid state.

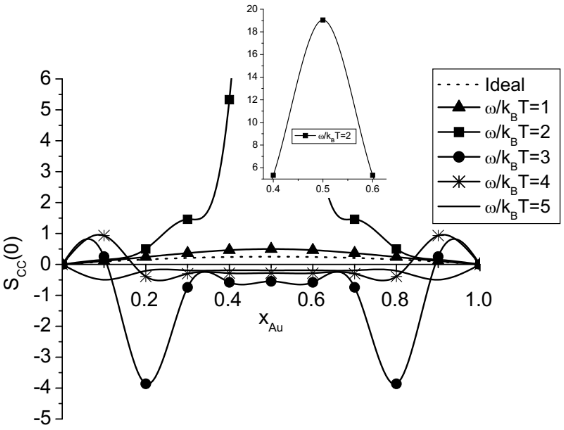

Figure 5: Effect of positive values of ω/k_BT on $S_{CC}(0)$ for Au-Cu liquid alloy at 1550 K.

The effect of ω/K_BT on $S_{\mathrm{CC}}(0)$ was observed by calculating the physical quantity for $\omega/K_BT = \pm 5$ with the help of Equation (16). The value of $S_{\mathrm{CC}}^{\mathrm{id}}(0)$ was estimated using Equation (17) throughout the entire concentration range. It can be observed that the computed values of $S_{\mathrm{CC}}(0)$ showed positive deviation from its ideal values for $\omega/K_BT = 1$ and 2 indicating the increase in segregating tendency in the initial melt (Figure 5). Beyond $\omega/K_BT > 2$, the estimated values of $S_{\rm CC}(0)$ showed unusual trends. But on increasing the negative values of ω/K_BT in the preferred range, the negative deviations of $S_{\rm CC}(0)$ from $S^{\rm id}_{\rm CC}(0)$ increases (Figure 6) revealing the gradual increase in ordering tendency in the alloy. These results are in accordance with those predicted by Gibbs free energy of mixing in the above sub-section.

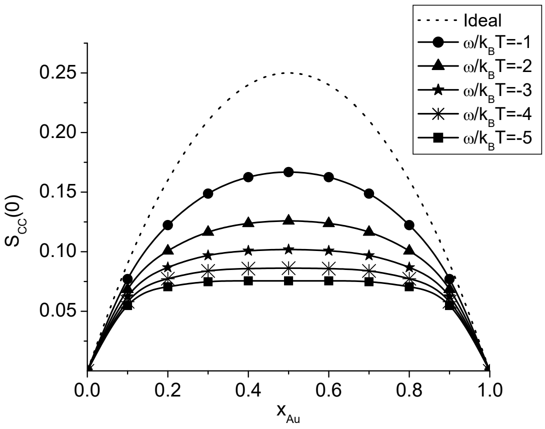

Figure 6: Effect of negative values of ω/k_BT on $S_{CC}(0)$ for Au-Cu liquid alloy at 1550 K.

Theoretical, experimental and ideal values of $S_{\rm CC}(0)$ were estimated using Equations (14-17) with help of determined values of model parameter and experimental value of activity [5]. Both the calculated experimental and theoretical values of $S_{\rm CC}(0)$ showed negative deviation from $S^{\rm id}_{\rm CC}(0)$ at all concentrations revealing the system to be complete ordering in nature at its melting temperature (Figure 7). Both of these values were in excellent agreement with each other.

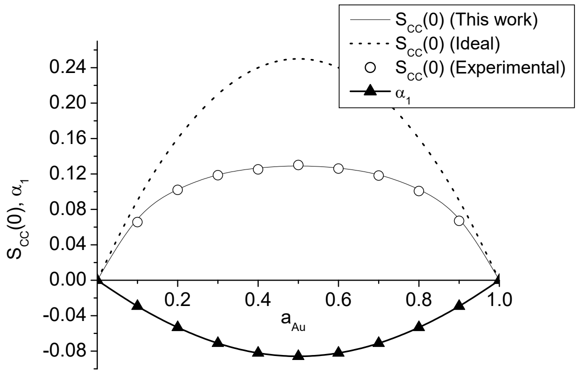

Figure 7: Computed values of $S_{\rm CC}(0)$ and α_1 versus $x_{\rm Au}$ for Au–Cu liquid alloy at 1550 K.

In case of liquid alloys, the value of α_1 can be negative, positive and zero. For a temperature and concentration, the ordering tendency is expected if $\alpha < 0$, segregating tendency is expected if $\alpha_1 > 0$ and ideal mixing tendency is expected if $\alpha_1 = 0$. The values of α_1 were calculated using Equations (17-19) with the aid of estimated values of $S_{\rm CC}(0)$. The calculated values of $\alpha_1 < 0$ at all concentrations revealing the system to be complete ordering in nature.

3.3 Surface properties

The surface tension (σ) and surface concentrations of components Au ($x^s_{\rm Au}$) and Cu ($x^s_{\rm Cu}$) were computed using Equation (20). The required input parameters for the purpose were determined using Equations (21-24) with the help of values in Table 1. The computed values of $x^s_{\rm Au}$, $x^s_{\rm Cu}$ and σ are plotted as a function of concentration in Figures 8 and 9 respectively.

Table 1: Input parameters for surface tension [30]

Atom	T_0 (K)	ρ_0 (kg/m^3)	$\partial\rho/\partial T$ (kg/m^3.K)	σ_0 (N/m)	$\partial\sigma/\partial T$ (N/m.K)
Au	1336	17360	-1.50	1.169	-0.00025
Cu	1356	8000	-0.80	1.303	-0.00023

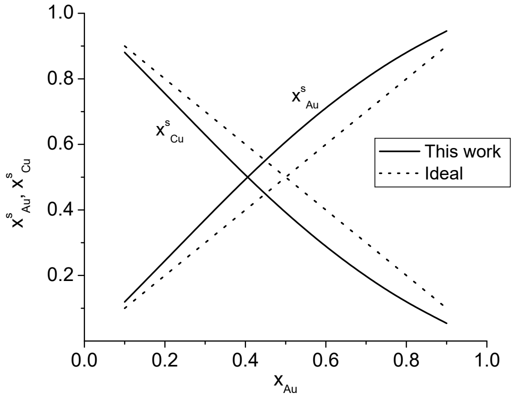

Figure 8: Computed values of $x^s_{\rm Au}$ and $x^s_{\rm Cu}$ versus $x_{\rm Au}$ of Au–Cu liquid alloy at 1550 K.

The computed value of $x^s_{\rm Au}$ showed positive deviation whereas those of $x^s_{\rm Cu}$ showed negative deviation from their respective ideal values (Figure 8). These findings suggested that Au atoms segregate on the surface phase of the liquid mixture, whereas Cu atoms remain in the bulk phase. The computed values of surface tension of individual atoms are $\sigma_{\rm Au} = 1.1155$ N/m and $\sigma_{\rm Cu} = 1.2584$ N/m at the melting temperature of the liquid alloy, 1550 K. As the surface tension of Au is less than that of Cu, Au atom segregated in the surface phase. The computed values of σ is found to be equal to ideal values in the concentration range $x_{\mathrm{Au}} < 0.3$. But it is found to be less than ideal values in the rest of the concentrations (Figure 9). Moreover, the surface tension of the system gradually decreased with the increase in concentration of Au.

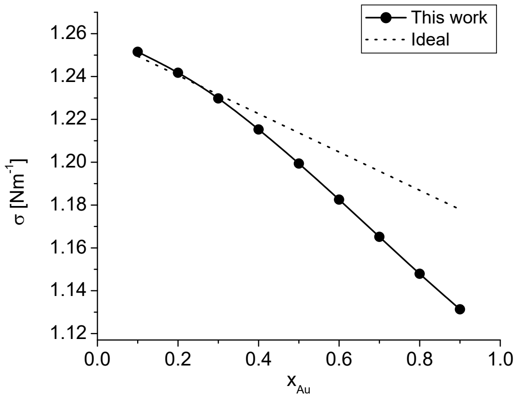

Figure 9: Computed values of σ versus x_{Au} of Au–Cu liquid alloy at 1550 K.

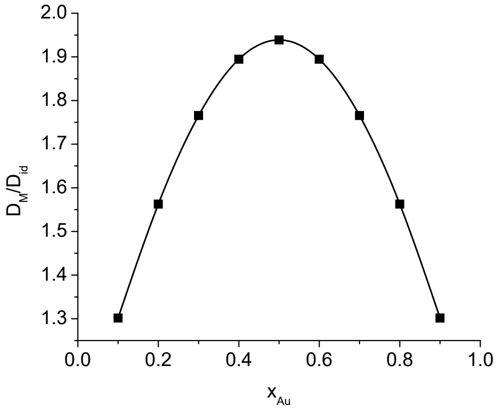

Figure 10: D_M/D_{id} versus x_{Au} of Au–Cu liquid alloy at 1550 K.

3.4 Transport properties

In transport properties, the ratio of mutual to intrinsic diffusion coefficients (D_M/D_{id}) of the system was calculated in the framework of STM. At a constant concentration and temperature, if $D_M/D_{\mathrm{id}} <$ 1, then it indicates homo-atomic pairing tendency and if $D_M/D_{\mathrm{id}} > 1$, then it indicates hetero-atomic pairing tendency among the atoms of liquid mixture. Equation (30) was used to calculate D_M/D_{id} of the system at 1550 K. From Figure 10, it can be observed that the computed values of the physical quantity is found to be greater than 1 throughout the entire concentration range thereby revealing the system to be complete ordering in nature at its melting temperature. Similar results were also obtained from the investigations of thermodynamic and structural functions in the above sub-sections.

Table 2: Input parameters for viscosity [30]

Atom	η_0 (Nsm^{-2})	E (Jmol^{-1})
Au	0.001132	15900
Cu	0.000301	30500

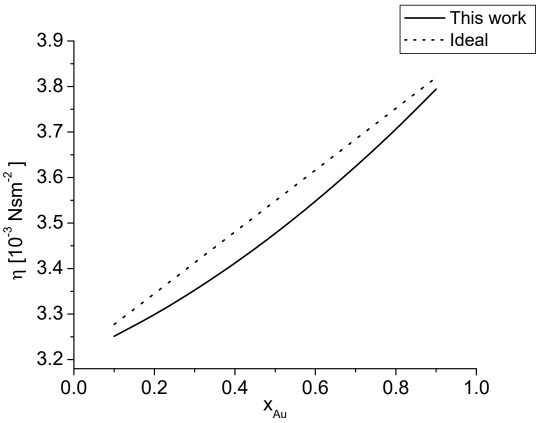

Figure 11: Compositional dependence of η of Au–Cu liquid alloy at 1550 K.

The viscosity (η) of the system was calculated on the basis of Kaptay equation [10] (Equations (27-29)). The necessary parameters required for the process were taken from Table 2. The viscosity of the system showed negative deviation from the ideal value. It increased gradually with the increase in the concentration of Au (Figure 11).

4 Conclusion

The values of the thermodynamic properties computed using the simple theory of mixing were consistent with their respective experimental values. Theoretical investigations showed the Au–Cu liquid alloy to be complete ordering in nature at its melting temperature. The activity of individual components increased with an increase in their respective concentrations. The surface tension of the system decreased, whereas viscosity increased gradually with an increase in the concentration of Au.

References

[1] P. J. Flory. Thermodynamics of high polymer solutions. *The Journal of chemical physics*, 10(1):51–61, 1942.

[2] R. A. Oriani. Thermodynamics of liquid Ag-Au and Au-Cu alloys and the question of strain energy in solid solutions. *Acta Metallurgica*, 4(1):15–25, 1956.

[3] S. Lele and P. Ramachandrarao. Estimation of complex concentration in a regular associated solution. *Metallurgical Transactions B*, 12:659–666, 1981.

[4] A. B. Bhatia and W. H. Hargrove. Concentration fluctuations and thermodynamic properties of some compound forming binary molten systems. *Physical Review B*, 10(8):3186, 1974.

[5] R. R. Hultgren, P. D. Desai, D. T. Hawkins, M. Gleiser, K. K Kelley, and D. D. Wagman. Selected values of the thermodynamic properties of the elements. 1973.

[6] L. S. Darken and R. W. Gurry. Physical chemistry of metals. new york, toronto, london, mcgraw-hill book company, inc. 1953.

[7] R. P. Godbole, S. A. Jha, M. Milanarun, and A. K. Mishra. Thermodynamics of liquid Cu–Mg alloys. *Journal of alloys and compounds*, 363(1-2):187–193, 2004.

[8] R. Novakovic. Thermodynamics, surface properties and microscopic functions of liquid Al–Nb and Nb–Ti alloys. *Journal of Non-Crystalline Solids*, 356(31-32):1593–1598, 2010.

[9] D Adhikari, BP Singh, IS Jha, and BK Singh. Thermodynamic properties and microscopic structure of liquid Cd–Na alloys by estimating complex concentration in a regular associated solution. *Journal of Molecular Liquids*, 156(2-3):115–119, 2010.

[10] G. Kaptay. A new equation to estimate the concentration dependence of the viscosity of liquid metallic alloys from the heat of mixing data. In *Proceedings of microCAD 2003 Conference. Section Metallurgy, University of Miskolc*, pages 23–28, 2003.

[11] S. K. Yadav, L. N. Jha, and D. Adhikari. Thermodynamic and structural properties of Bi-based liquid alloys. *Physica B: Condensed Matter*, 475:40–47, 2015.

[12] L. Topor and O. J. Kleppa. Thermochemistry of binary liquid gold alloys: The systems gold-copper and gold-silver at 1379 K. *Metallurgical Transactions A*, 15:203–208, 1984.

[13] H. Okamoto, D. J. Chakrabarti, D. E. Laughlin, and T. B. Massalski. The Au- Cu (gold-copper) system. *Journal of Phase Equilibria*, 8:454–474, 1987.

[14] Bo. Sundman, S. G. Fries, and W. A. Oates. A thermodynamic assessment of the Au-Cu system. *Calphad*, 22(3):335–354, 1998.

[15] X. J. Han, M. Chen, and Z. Y. Guo. Thermophysical properties of undercooled liquid Au–Cu alloys from molecular dynamics simulations. *Journal of Physics: Condensed Matter*, 16(6):705, 2004.

[16] J. Brillo, I. Egry, H. S. Giffard, and A. Patti. Density and thermal expansion of liquid Au-Cu alloys. *International journal of thermophysics*, 25:1881–1888, 2004.

[17] Ge-Xing Kong, Xiao-Juan Ma, Qi-Jun Liu, Yong Li, and Zheng-Tang Liu. Structural stability, elastic and thermodynamic properties of Au–Cu alloys from first-principles calculations. *Physica B: Condensed Matter*, 533:58–62, 2018.

[18] S. K. Yadav, P. Sharma, R. P. Koirala, A. Dhungana, and D. Adhikari. Mixing properties of Ni-Al liquid alloys at different temperatures. *Bibechana*, 16:106–121, 2019.

[19] I Koirala, IS Jha, BP Singh, and D Adhikari. Thermodynamic, transport and surface properties in In–Pb liquid alloys. *Physica B: Condensed Matter*, 423:49–53, 2013.

[20] RN Singh, IK Mishra, and VN Singh. Local order in Cd-based liquid alloys. *Journal of Physics: Condensed Matter*, 2(42):8457, 1990.

[21] D. Adhikari, S. K. Yadav, and L. N. Jha. Thermo-physical properties of Al–Fe melt. *Journal of the Chinese Advanced Materials Society*, 2(3):149–158, 2014.

[22] S. K. Yadav, L. N. Jha, and D. Adhikari. Segregating to ordering transformation in In–Sn melt. *Physics and Chemistry of Liquids*, 53(4):443–454, 2015.

[23] S. K. Yadav, L. N. Jha, and D. Adhikari. Thermodynamic, structural, transport and surface properties of Pb-Tl liquid alloy. *Bibechana*, 13:100–113, 2016.

[24] D. Adhikari, S. K. Yadav, and L. N. Jha. Thermo-physical properties of Mg-Tl melt. *Journal of Basic and Applied Research International*, 9:103–110, 2015.

[25] S. K. Yadav, M. Gautam, and D. Adhikari. Mixing properties of Cu–Mg liquid alloy. *AIP Advances*, 10(12):125320, 2020.

[26] J. A. V. Butler. The thermodynamics of the surfaces of solutions. *Proceedings of the Royal Society of London. Series A, Containing Papers of a Mathematical and Physical Character*, 135(827):348–375, 1932.

[27] George Kaptay. Partial surface tension of components of a solution. *Langmuir*, 31(21):5796–5804, 2015.

[28] George Kaptay. A unified model for the cohesive enthalpy, critical temperature, surface tension and volume thermal expansion coefficient of liquid metals of bcc, fcc and hcp crystals. *Materials Science and Engineering: A*, 495(1-2):19–26, 2008.

[29] George Kaptay. A coherent set of model equations for various surface and interface energies in systems with liquid and solid metals and alloys. *Advances in colloid and interface science*, 283:102212, 2020.

[30] Eric Adolph Brandes and G. B. Brook. *Smithells metals reference book*. Elsevier, 2013.

[31] Shashit Kumar Yadav. Thermodynamic, structural and surface properties of rare earth metallic alloys: Au-La liquid system. *BIBECHANA*, 20(3):316–325, 2023.

[32] S. K. Yadav, N. Chaudhary, and D. Adhikari. Thermodynamic, structural, surface and transport properties of Au-Ni liquid alloy at 1150 K. *Bibechana*, 18(1):184–192, 2021.