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Original Research Article

Thermodynamic characteristics of Au-Cu melts over a range of temperature

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ABSTRACT
The composition dependency of the properties of mixing of Au-Cu molten alloys at different temperatures are analyzed in the framework of a theoretical model called molecular interaction volume model (MIVM). The theoretical results for Gibbs excess free energy of mixing, free energy of mixing, activity coefficient, activity, microscopic properties such as concentration fluctuations demonstrate excellent agreement with the correlated experimental results for Ag-Cu melts at 1550 K. The theoretical work is extended for the aforesaid thermodynamic and microscopic properties as well as short range order parameter, excess stability function and diffusivity ratio for Au-Cu melts at 1450, 1550, 1650 and 1750 K. The results reveal that Au-Cu system is an ordered alloy having a likelihood of the formation of chemical complexes in the temperature region 1450 – 1750 K.

1. Introduction

The Au-Cu system is a binary alloy which possesses the capability to form thermodynamically stable super-lattice structure and there exists of temperature dependent order-disorder transformation [1]. The phase diagram demonstrates the presence of intermetallic compounds Au-Cu, Au_3-Cu and Au-Cu_3 in binary Au-Cu system [2]. It is interesting to mention that these intermetallic complexes have wide range of applications in industries due to high melting temperature improved corrosion resistance and enhanced mechanical strength [1]. The properties of mixing like excess free energy of mixing, G_M^E, free energy of mixing, G_M, enthalpy of mixing, H_M and entropy of mixing, S_M for Au-Cu alloys at 1550 K are symmetric relative to the composition about x = 0.5. Again, the thermodynamic activity a_i (i = Au, Cu) represents negative departures from Raoult's law for Au-Cu molten alloys at 1550 K [2]. The negative departures of the activity of both the elements from ideal mixing and negative values of G_M^E of Au-Cu melts at 1550 K refer to the ordering nature of the system. Again, the existence of the chemical complexes as well as symmetric behavior the thermodynamic functions indicate that Au-Cu system at 1550 K is a complex forming regular alloy [3]. Thus, the interesting properties of mixing as well as the extensive applications in industries of Au-Cu alloys have drawn the attentiveness of several researchers [1, 4–9] to explore the thermophysical properties of the alloys. Nonetheless, there is scanty of data related to the microscopic properties namely concentration fluctuations, $S_{cc}(0)$, short-range order parameter, α_1 (SRO) and excess stability function, E^{XS}, and, transport property namely viscosity and diffusivity, D of molten Au-Cu alloys. Therefore, Au-Cu system is an appropriate candidate which requires the further theoretical explanation.

Therefore, the present research work is devoted to the theoretical explanation of the thermodynamic and microscopic properties of Au-Cu liquid alloys at different temperatures in the framework of MIVM model [10, 11]. Earlier, MIVM model has been successfully employed to understand the thermodynamic, microscopic, surface and transport properties of a number of binary alloys such as Ni-Pd, Zn-Bi, Zn-Cd, Pb-Bi etc. [12–15] as well as multicomponent alloys i.e. In-Bi-Sn, C–Fe–Co–Ni [16–18].

2. Formalism

2.1 Thermodynamic properties like excess free energy of mixing, G_M^E, free energy of mixing, G_M, activity coefficient, γ_μ and activity, a_μ (μ = i, j) of binary molten alloys

MIVM model [10] is a fluid-based model which is deduced from statistical thermodynamics on assuming the fundamental motion of non-random interchange of liquid molecules and fluid-phase equilibria. For a binary melt $i-j$, the molecular excess free energy of mixing is expressed as [10]:

$$\frac{G_M^E}{RT} = x_i \ln\left(\frac{V_{mi}}{x_i V_{mi} + x_j V_{mj} A_{ji}}\right) + x_j \ln\left(\frac{V_{mj}}{x_j V_{mj} + x_i V_{mi} A_{ij}}\right) - \frac{x_i x_j}{2}\left(\frac{Z_i A_{ji} \ln A_{ji}}{x_i + x_j A_{ji}} + \frac{Z_j A_{ij} \ln A_{ij}}{x_j + x_i A_{ij}}\right) \quad (1)$$

where, the compositions of the element i and j are respectively represented by x_i and x_j, A_{ij} and A_{ji} refer to pair-potential energy interaction parameters and the first coordination numbers of the constituents i and j are respectively Z_i and Z_j. The pair potential energy interaction parameters may be expressed as [10]:

$$A_{ji} = \exp\left[-\frac{\varepsilon_{ji} - \varepsilon_{ii}}{kT}\right] \text{ and } A_{ij} = \exp\left[-\frac{\varepsilon_{ij} - \varepsilon_{jj}}{kT}\right] \quad (2)$$

where, ε_{ii}, ε_{jj} and ε_{ji} denote the potential energies for the pairs i–i, j–j and i–j respectively, T is the absolute temperature and k is the Boltzmann constant. It is noted that $\varepsilon_{ij} = \varepsilon_{ji}$.

The partial molar energy and excess molar free energy are related as:

$$G_M^E = RT \ln \gamma_\upsilon = G_M^E + \delta\left(\frac{\partial G_M^E}{\partial x_i}\right)_{T,P,x[i,N]} - \sum_{j=1}^{N}\left(\frac{\partial G_M^E}{\partial x_i}\right)_{T,P,x[j,N]} \quad (3)$$

where, $\gamma_\mu\,(\mu = i, j)$ refers to the activity coefficient of binary melts.

It is noted that the equation (3) follows the conditions that for $i \neq N$, $\delta = 1$; for $i = N$, $\delta = 0$. Again, the variables x_j and x_N determined from $x[i, N]$ with $x_N = 1 - \sum_{j=1}^{N-1} x_j$. The activity coefficients of components i and j of a binary melt are respectively, represented by [10]

$$\ln \gamma_i = \ln\left(\frac{V_{mi}}{x_i V_{mi} + x_j V_{mj} A_{ji}}\right) + x_j\left(\frac{V_{mj} A_{ji}}{x_i V_{mi} + x_j V_{mj} A_{ji}} - \frac{V_{mi} A_{ji}}{x_j V_{mj} + x_i V_{mi} A_{ij}}\right) - \frac{x_j^2}{2}\left(\frac{Z_i A_{ji}^2 \ln A_{ji}}{\left(x_i + x_j A_{ji}\right)^2} + \frac{Z_j A_{ij} \ln A_{ji}}{\left(x_j + x_i A_{ij}\right)^2}\right) \quad (4)$$

and

$$\ln \gamma_j = \ln\left(\frac{V_{mj}}{x_j V_{mj} + x_i V_{mi} A_{ij}}\right) - x_i\left(\frac{V_{mj} A_{ji}}{x_i V_{mi} + x_j V_{mj} A_{ji}} - \frac{V_{mi} A_{ij}}{x_j V_{mj} + x_i V_{mi} A_{ij}}\right) - \frac{x_i^2}{2}\left(\frac{Z_j A_{ij}^2 \ln A_{ij}}{\left(x_j + x_i A_{ij}\right)^2} + \frac{Z_i A_{ji} \ln A_{ji}}{\left(x_i + x_j A_{ji}\right)^2}\right) \quad (5)$$

For the component i, the first co-ordination number is determined by the following relation [10]:

$$Z_i = \frac{4\sqrt{2\pi}}{3}\left(\frac{r_{mi}^3 - r_{oi}^3}{r_{mi} - r_{oi}}\right)\rho_i r_{mi} \exp\left(\frac{\Delta H_{mi}(T_{mi} - T}{Z_C R T_{mi}}\right) \quad (6)$$

where, T_{mi} represents the melting temperature in kelvin, ΔH_{mi}, the enthalpy at melting temperature, $\rho_i = N_i V_i = \frac{0.6022}{V_{mi}}$ represents the molecular number density, r_{mi} and r_{oi} are initial value and first peak value for radial distribution function of metal i in molten state near T_{mi}, R stands for molar gas constant and Z_C denotes the co-ordination number for closed packed structure which is usually taken to be 12. The radial distances may be represented by [10]

$$r_{oi} = 0.918 d_{\text{cov},i} \text{ and } r_{mi} = \sigma_i \quad (7)$$

where, d_{cov} represents atomic covalent diameter and σ_i the atomic diameter.

In infinite dilute solution region such as x_i or $x_j \to 0$, the activity coefficients of the constituents i and j may be represented by

$$\ln \gamma_i^\infty = 1 - \ln\left(\frac{V_{mj} A_{ji}}{V_{mi}}\right) - \frac{V_{mi} A_{ji}}{V_{mj}} - \frac{1}{2}\left(Z_i \ln A_{ji} + Z_j A_{ij} \ln A_{ij}\right) \quad (8)$$

and

$$\ln \gamma_j^\infty = 1 - \ln\left(\frac{V_{mi} A_{ij}}{V_{mj}}\right) - \frac{V_{mj} A_{ji}}{V_{mi}} - \frac{1}{2}\left(Z_j \ln A_{ij} + Z_i A_{ji} \ln A_{ji}\right) \quad (9)$$

The activity and activity coefficients for the constituents of the binary melts may be expressed as [3]

$$a_i = \gamma_i x_i \quad (10a)$$

$$a_j = \gamma_j x_j \quad (10b)$$

2.2 Microscopic functions like concentration fluctuations, $S_{cc}(0)$ and SRO, α_1 of a binary molten alloy

The microscopic functions are beneficial to acquire the microscopic knowledge of the complex formation, phase separation, structure etc. of the liquid alloys as well as the metalurgical processes and materials design. The $s_{cc}(0)$ and α_1 are found to be very important microscopic parameters to explore the interatomic interactions in a liquid system. If $S_{cc}(0) < S_{cc}^{id}(0)$ then the system is ordered i.e. dissimilar ($i - j$) atoms or molecules coupled together as the closest neighbors. Similarly, $S_{cc}(0) > S_{cc}^{id}(0)$ describes the segregating character of the binary melt and like (i.e. $i - i$ or $j - j$) atoms or molecules link together as the closest neighbors [3].

The G_M and $s_{cc}(0)$ are related as [3, 19]:

$$s_{cc}(0) = RT\left(\frac{\partial^2 G_M}{\partial x_i^2}\right)_{T,P,N}^{-1} = RT\left(\frac{\partial^2 G_M}{\partial x_j^2}\right)_{T,P,N}^{-1} \quad (11)$$

where, $G_M = G_M^E + G_M^{id}$ (12)

$$G_M = G_M^E + RT\left[x_i \ln x_i + x_j \ln x_j\right] \quad (13)$$

Using equations (1) and (13) in (11), one can obtain

$$S_{cc}(0)=\frac{x_i x_j}{1+x_i x_j f(x_i x_j)} \quad (14)$$

where,

$$f(x_i, x_j) = \frac{V_{mj}A_{ji} - V_{mi}}{x_i V_{mi} + x_j V_{mj} A_{ji}} + \frac{V_{mi}A_{ij} - V_{mj}}{x_j V_{mj} + x_i V_{mi} A_{ij}} + \frac{V_{mj}A_{ji}(V_{mj}A_{ji} - V_{mi})}{(x_i V_{mi} + x_j V_{mj} A_{ji})^2} + \frac{V_{mi}A_{ij}(V_{mi}A_{ij} - V_{mj})}{(x_j V_{mj} + x_i V_{mi} A_{ij})^2} + \left[\frac{Z_i A_{ji}^2 \ln A_{ji}}{(x_i + x_j A_{ji})^3} + \frac{Z_j A_{ij}^2 \ln A_{ij}}{(x_j + x_i A_{ij})^3}\right] \quad (15)$$

For an ideal mixing, $G_M = G_M^{id}$ hence equations (14) and (15) yield

$$S_{cc}^{id}(0) = x_i x_j \quad (16)$$

Again, the activity and $s_{cc}(0)$ are related as:

$$s_{cc}(0) = a_i x_j \left(\frac{\partial a_i}{\partial x_i}\right)^{-1} = a_j x_i \left(\frac{\partial a_j}{\partial x_j}\right)^{-1} \quad (17)$$

The $s_{cc}(0)$ determined from equation (17) on utilizing the experimental data of activity is considered as the experimental data [3, 19, 20].

The SRO, α_1 initiated by Warren-Cowley [21, 22] is related to $s_{cc}(0)$ as [3. 23]

$$\alpha_1 = \frac{S-1}{S(Z-1)+1} \text{ with } S = \frac{S_{cc}(0)}{S_{cc}^{id}(0)} \quad (18)$$

where, Z refers to the coordination number in first neighbor shell.

Equation (18) exhibits that α_1 = positive for segregating system, α_1 = negative for ordered system and α_1 = 0 for ideal mixture.

2.3 Excess stability function, E^{XS} and diffusivity, D of a binary molten alloy

The excess stability function is intimately related to the microscopic function namely $s_{cc}(0)$ and helpful to explain the nature and strength of chemical interactions existing in liquid alloys. The excess stability function, E^{XS} initiated by Darken [24, 25] may be expressed as

$$\frac{E^{XS}}{RT} = Scc(0))^{-1} - (S_{cc}^{id}(0))^{-1} \quad (19)$$

Obviously, E^{XS} = positive for ordering alloy, E^{XS} = negative for segregating alloy and E^{XS} = 0 for ideal alloy depending upon the conditions that $S_{cc}(0) < S_{cc}^{id}(0)$, $S_{cc}(0) > S_{cc}^{id}(0)$, and $S_{cc}(0) = S_{cc}^{id}(0)$ respectively.

The transport property such as diffusivity is very useful to describe the mixing behavior of a binary solution at the microscopic level. For a binary liquid system, the diffusivity ratio is expressed as [3, 26]

$$\frac{D_M}{D_{id}} = \frac{S_{cc}^{id}(0)}{S_{cc}(0)} \quad (20)$$

where, D_M and D_{id} represent the mutual diffusion coefficient of the system and intrinsic diffusion coefficient of ideal system respectively. Now, D_{id} is represented by

$$D_{id} = x_i D_j + x_j D_i \quad (21)$$

where, D_i and D_j are self-diffusion coefficients of pure elements *i* and *j* respectively.

Equation (20) illustrates that $D_M/D_{id} > 1$ for ordered system, $D_M/D_{id} < 1$ for segregating system and $D_M/D_{id} = 1$ for ideal system.

3. Results and discussion

The analytical expressions are utilized to study the temperature and composition dependency of the thermodynamic functions like G_M^E, G_M, γ_μ and a_μ ($\mu = i, j$), microscopic functions namely $s_{cc}(0)$, α_1 and E^{XS} as well as the transport property namely D_M / D_{id} of Au-Cu molten alloys at different temperatures.

3.1 Results for G_M^E, G_M, γ_μ and a_μ ($\mu = i, j$) for Au-Cu melts at 1450, 1550, 1650 and 1750 K

For the calculations of the thermodynamic functions, the needed parameters of the pure Au and Cu are mentioned in Table 1 [27] .The values of Z_i and Z_j for the pure Au and Cu calculated from equation (6) are illustrated in Table 2. The energy interaction parameters A_{ji} and A_{ij} are estimated by Newton - Raphson method on introducing the values of activity coefficients in the range of infinite dilute solution i.e. $\gamma_i^\infty = 0.155$ and $\gamma_j^\infty = 0.155$ [2] in equation (8) and (9). The values of A_{ji} and A_{ij} varied so as to achieve a well harmony between the theoretical and experimental values [2] of G_M^E for Au-Cu melts at 1550 K. The best fit values of A_{ji} and A_{ij} are also cited in Table 2.

For the computation of the thermodynamic functions of Au-Cu melts at various temperatures, the required input parameters are also presented in Table 1 and Table 2. It is notable that the values of A_{ji} and A_{ij} at temperature, *T* K are determined from the following relation [10]

$$-\frac{\varepsilon_{ji} - \varepsilon_{ii}}{kT} = T \ln A_{ji}(T) = T_0 \ln A_{ji}(T_0)$$

$$A_{ji}(T) = \exp\left(\frac{T_0 \ln A_{ji}(T_0)}{T}\right) \quad (22)$$

and

$$A_{ij}(T) = \exp\left(\frac{T_0 \ln A_{ij}(T_0)}{T}\right) \quad (23)$$

$$-\frac{\varepsilon_{ij} - \varepsilon_{jj}}{kT} = T \ln A_{ij}(T) = T_0 \ln A_{ij}(T_0)$$

where, T_0 = 1550 K.

Table 1: Input parameters for the pure metals [27].

Metal, *i*	ΔH_{mi} (KJ/mole)	σ_i ($\times 10^{-8}$ cm)	r_{oi} ($\times 10^{-8}$ cm)	V_{mi} (cm^3/mole)
Au	12.76	2.80	2.34	11.3[1+0.98×10^{-4}(T-1336)]
Cu	13.0	2.50	2.06	7.90[1+1.0×10^{-4}(T-1356)]

Table 2: Values of A_{ij}, A_{ji}, Z_i and Z_j for the of Au-Cu molten alloys at various temperatures.

i-j	*T* (K)	A_{ij}	A_{ji}	Z_i	Z_j
Au-Cu	1450	1.0763	1.2711	9.69	9.73
	1550	1.0712	1.2516	9.51	9.55
	1650	1.0667	1.2347	9.33	9.36
	1750	1.0628	1.2199	9.15	9.18

The theoretical values of G_M^E / RT calculated using equation (1) as a function of concentration of Au for Au-Cu liquid alloys at the temperatures 1450, 1550, 1650 and 1750 K are depicted in Fig. 1. For comparative study, the experimental data of G_M^E / RT [2] for Au-Cu melts at 1550 K are also included in Fig. 1. An excellent harmony is found between the theory and experiment having minimum values of G_M^E / RT i.e. G_M^E / RT (min^m) = – 0.4669 (Theory) and – 0.4668 (Experiment) both at x_{Au} = 0.5. Again, G_M^E / RT - x_{Au} isotherms indicate the symmetric behavior of Au-Cu melts at 1450 – 1750 K because the minima in G_M^E / RT are found at x_{Au} = 0.5 i.e. G_M^E / RT (min^m) = – 0.5086, – 0.4669, – 0.4307 and – 0.3991 at 1450, 1550, 1650 and 1750 K respectively. The negative values of G_M^E / RT confirm that Au-Cu melt is an ordered alloy at each temperature under consideration. Further, the negative values of G_M^E / RT decrease with the upgradation of temperature from 1450 – 1750 K which advocates the degradation of ordering nature of Au-Cu melts due to rise in temperature from 1450 – 1750 K.

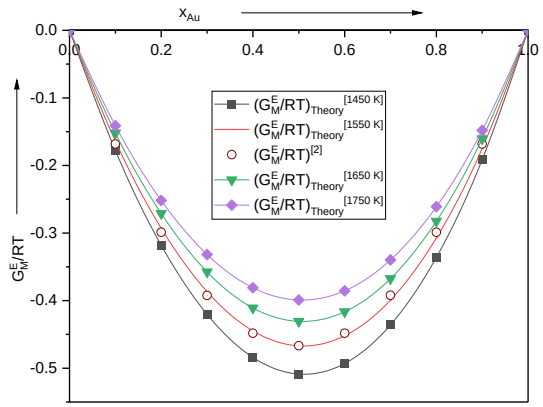

Fig. 1 G_M^E/RT vs x_{Au} for Au-Cu melts at 1450, 1550, 1650 & 1750 K

The temperature dependency of G_M / RT relative to the composition of Au for Au-Cu melts at 1450, 1550, 1650 and 1750 K are presented in Fig. 2. The experimental data of G_M / RT for Au-Cu melts at 1550 K available in literature [2] are also incorporated in Fig. 2 for comparison. An excellent similarity is noticed. The minimum values of G_M / RT are observed at x_{Au} = 0.5 i.e. G_M / RT (min^m) = – 1.1600 (Theory) and – 1.1603 (Experiment) at 1550 K. Again, G_M / RT - x_{Au} isotherms demonstrate the negative values of G_M / RT in the concentration region x_{Au} = 0.1 to 0.9 with minima at x_{Au} = 0.5 i.e. G_M / RT (min^m) = – 1.2018, – 1.1600, – 1.1239 and – 1.0922 at 1450, 1550, 1650 and 1750 K respectively. This exhibits the symmetry in G_M / RT relative to the composition for molten Au-Cu system in the temperature range 1450 – 1750 K. The magnitude of the negative values of G_M / RT indicates that Au-Cu system is a feebly interacting ordered system [20].

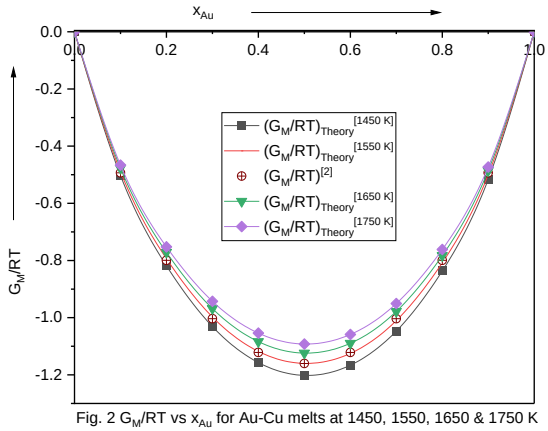

Fig. 2 G_M/RT vs x_{Au} for Au-Cu melts at 1450, 1550, 1650 & 1750 K

The theoretical values of γ_{Au} and γ_{Cu} determined from equations (4) and (5) respectively at the temperatures under considerations are presented in Fig. 3 along with the experimental results of γ_{Au} and γ_{Cu} at 1550 K [2] for Au-Cu molten system. An excellent agreement is observed at 1550 K. It is evident from Fig. 3 that the values of γ_{Au} and γ_{Cu} increase with the rise in temperature from 1450 – 1750 K as well as the addition of Au component in Au-Cu system.

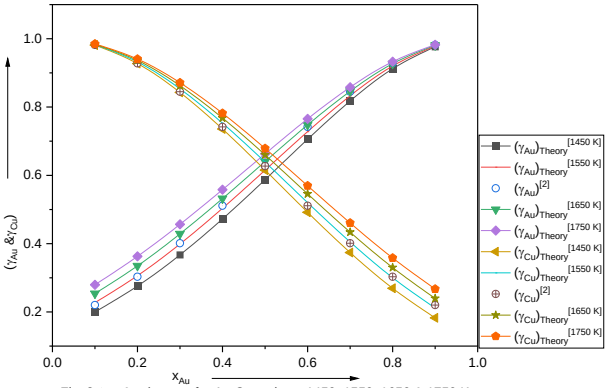

Fig. 3 (γ_{Au} &γ_{Cu}) vs x_{Au} for Au-Cu melts at 1450, 1550, 1650 & 1750 K

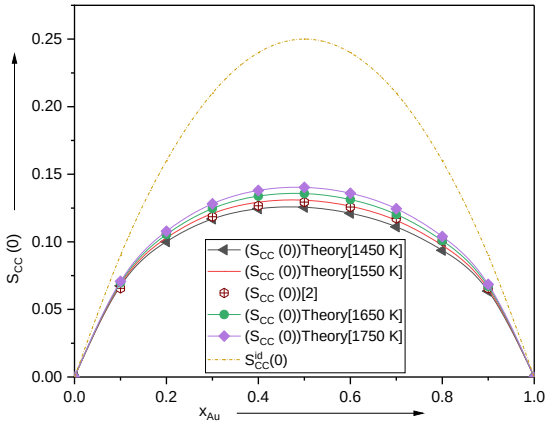

Fig. 5 S_{CC} (0) vs x_{Au} for Au-Cu melts at 1450, 1550, 1650 & 1750 K

The expressions (10a) and (10b) are employed to ascertain the values of a_{Au} and a_{Cu} of Au-Cu melts at 1450, 1550, 1650 and 1750 K relative to the composition of Au. The theoretical results are incorporated in Fig. 4. For comparison, the experimental results for a_{Au} and a_{Cu} at 1550 K [2] are also included in Fig. 4, which demonstrates a well concord. The $a_{\mu}\,(\mu = i, j)$ - x_{Au} isotherms exhibit negative departures from Raoult's law in the entire composition range at each temperature. The variations of a_{Au} and a_{Cu} with respect to temperature are very small. However, the negative departures of a_{Au} and a_{Cu} from linearity degrade with the upgradation of temperatures from 1450 – 1750 K. This justifies the decrease in the ordering characteristics of Au-Cu melts when the temperature is raised from 1450 K to 1750 K.

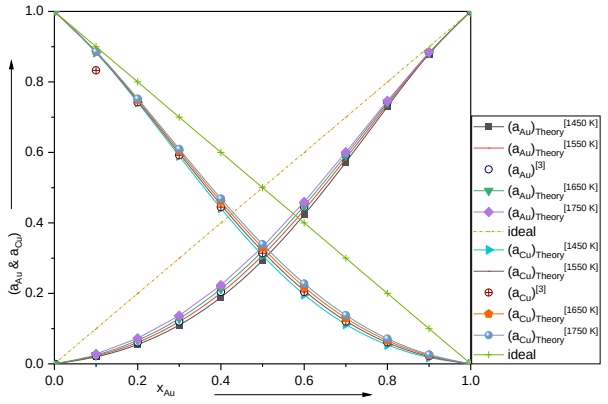

Fig. 4 (a_{Au} & a_{Cu}) vs x_{Au} for Au-Cu melts at 1450, 1550, 1650 & 1750 K

3.2 Results for $S_{cc}(0)$, α_1, E^{XS} and D_M/D_{id} of Au- Cu melts at 1450, 1550, 1650 and 1750 K

For the understanding of microscopic behavior of Au-Cu melts at 1450, 1550, 1650 and 1750 K, the $S_{cc}(0)$ has been theoretically analyzed relative to the composition of Au. The theoretical results for $S_{cc}(0)$ computed using equations (14) and (15) are cited in Fig 5 along with $S_{cc}^{id}(0)$. The experimental data of $S_{cc}(0)$ for Au-Cu melts at 1550 K [2] are also included in Fig. 5 for comparison. A well concord is observed between the theory and experiment.

Obviously, $S_{cc}(0) < S_{cc}^{id}(0)$ in the whole composition region, x_{Au} = 0.1 - 0.9 which justifies the ordering character of Au-Cu melts at 1550 K [3]. The concentration dependence of $S_{cc}(0)$ exhibits the symmetric characteristics of Au-Cu melts at 1550 K having maximum values at x_{Au} = 0.5 i.e. $S_{cc}(0)$ (max^m) = 0.131 (Theory) and 0.129 (Experiment). Again, $S_{cc}(0)$ – x_{Au} isotherms demonstrate the small increase in the values of $S_{cc}(0)$ with the elevation of temperature from 1450 K to 1750 K. The magnitudes of peak values of $S_{cc}(0)$ are 0.126, 0.131, 0.136 and 0.140 respectively at 1450, 1550, 1650 and 1750 K. However, the position of peak values remains same i.e. at x_{Au} = 0.5. Again, the $S_{cc}(0) < S_{cc}^{id}(0)$ in the entire composition range at each temperature under consideration which denotes that Au-Cu melt is an ordered alloy and the degree of ordering degrades with rise in temperature from 1450 - 1750 K.

For the exploration of the atomic arrangement in Au-Cu melts, the SRO, α_1 has been theoretically analyzed at different temperatures relative to the composition of Au. It is notable that there is scarcity of experimental data for Au-Cu melts in the literature. The theoretical results for α_1 of Au-Cu melts at 1450, 1550, 1650 and 1750 K calculated from equation (18) are mentioned in Fig. 6. The α_1 – x_{Au} isotherms for Au-Cu melts at 1450, 1550, 1650 and 1750 K illustrate that α_1 is negative in the complete range of concentration from x_{Au} = 0.1 to 0.9 at each temperature under consideration. The negative values of α_1 predict the ordering nature which refers to the existence of unlike atoms (Au-Cu) interactions as nearest neighbors in Au-Cu melts [2] in the temperature from 1450 K to 1750 K. Again, the smaller magnitudes of the negative values of α_1 i.e. – 0.0902, – 0.0835, – 0.0777 and – 0.0725 at 1450, 1550, 1650 and 1750 K respectively advocate the compound forming tendency of Au-Cu melts in the temperature region 1450 – 1750 K about the x_{Au} = 0.5. Further, the degree of ordering in Au-Cu melts decreases with the elevation of temperature from 1450 K to 1750 K.

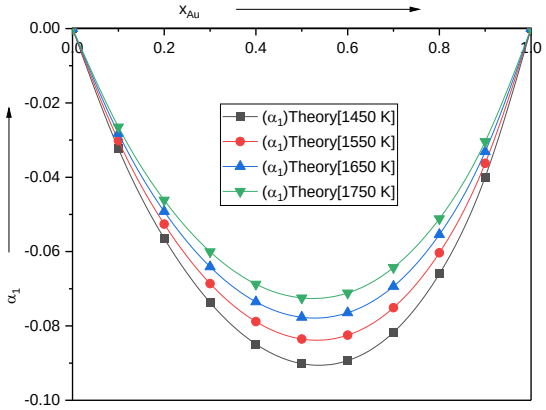

Fig. 6 α_1 vs x_{Au} for Au-Cu melts at 1450, 1550, 1650 & 1750 K

The excess stability function of several binary alloys has been described earlier [24, 28]. Nonetheless, the limited theoretical data of E^{XS} at different temperatures is available [12, 25, 29] on employing the optimization procedure. In present study, MIVM model is applied to explore the stability of Au-Cu melts at 1450, 1550, 1650 and 1750 K. The variation of E^{XS}/RT with respect to the composition and temperature of Au-Cu melts is illustrated in Fig. 7. Obviously, E^{XS}/RT shows positive value at each composition in the region x_{Au} = 0.1 to 0.9 at each temperature under consideration, which manifests the ordering character of Au-Cu melts in the temperature region 1450 – 1750 K. Again, the positive values of E^{XS}/RT decline with the upgradation of temperature. This is a sign of lowering of the ordering behavior of Au-Cu melts [3] in between 1450 K to 1750 K.

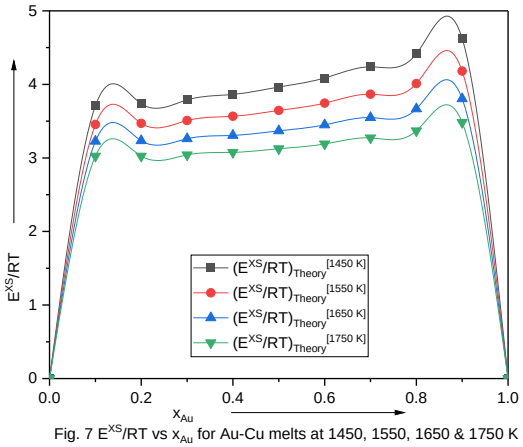

Fig. 7 E^{XS}/RT vs x_{Au} for Au-Cu melts at 1450, 1550, 1650 & 1750 K

The equation (20) is used to compute the values of D_M/D_{id} relative to the concentration of Au for Au-Cu melts at 1450, 1550, 1650 and 1750 K. The theoretically predicted data for D_M/D_{id} for Au-Cu melts at the temperatures under consideration are depicted in Fig. 8. It is evident that $D_M/D_{id} > 1$ at each concentration in the temperature region 1450 – 1750 K, having maxima at x_{Au} = 0.5 i.e. D_M/D_{id} (max^m) = 1.9909, 1.9114, 1.8421 and 1.7814 at 1450, 1550, 1650 and 1750 K respectively. Thus, the theoretical results demonstrate that Au-Cu melt is an ordered system and the degree of ordering in Au-Cu melt decreases when the temperature is raised from 1450 – 1750 K. Again, the maximum values of D_M/D_{id} indicate that there is a likelihood of the maximum ordering in Au-Cu melts about x_{Au} = 0.5 in the temperature region 1450 – 1750 K.

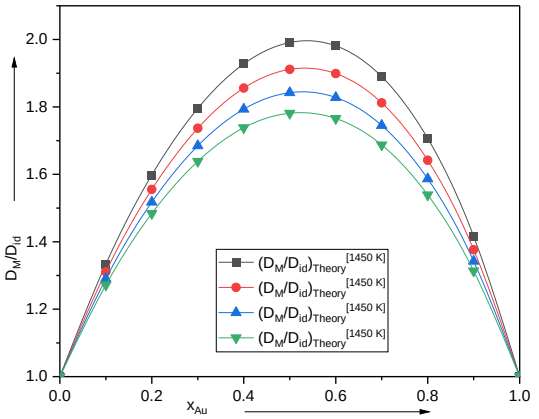

Fig. 8 D_M/D_{id} vs x_{Au} for Au-Cu melts at 1450, 1550, 1650 & 1750 K

4. Conclusions

The present theoretical work reveals that

(a) The theoretical results for G_M^E, G_M, activity coefficient, activity and $S_{cc}(0)$ for Au-Cu melts at 1550 K are in excellent agreement with the correlated experimental result available in the literature.

(b) The negative value of G_M^E, negative departure of the activity of both the components i.e. a_{Au} and a_{Cu}, $S_{cc}(0) < S_{cc}^{id}(0)$, $E^{XS} > 0$, $\alpha_1 < 0$ and $D_M/D_{id} > 1$ refers to the ordering nature of Au-Cu melts in the temperature region 1450 – 1750 K.

(c) The concentration and temperature dependency of the thermodynamic functions G_M^E, G_M, activity, $S_{cc}(0)$, $E^{XS} > 0$, α_1 and D_M/D_{id} justifies that the degree of ordering decreases when the temperature is elevated for Au-Cu melts from 1450 – 1750 K.

(d) The large negative deviation of $S_{cc}(0)$ from $S_{cc}^{id}(0)$ demonstrates that Au-Cu melts is a complex forming alloy in the temperature region 1450 – 1750 K.

(e) The magnitude of negative values of G_M advocates the weakly interacting nature of Au-Cu melts in the temperature region 1450 – 1750 K.

(f) The symmetries in G_M^E, G_M, $S_{cc}(0)$, α_1 and D_M/D_{id} with respect to concentration confirm that Au-Cu melt is a regular alloy at the temperature from 1450 – 1750 K.

(g) Thus, MIVM model is a suitable and reliable model to explain the thermodynamic characteristics of Au-Cu melts at different temperatures.

Authors' contributions

The author read and approved the final manuscript.

Conflicts of interest

The author declares no conflict of interest.

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Data availability

No new data were created.

References

[1] X.J. Han, M. Chen, Z.Y. Guo, Thermophysical properties of undercooled liquid Au–Cu alloys from molecular dynamics simulations, *J. Phys.: Condens. Matter* **16** (2004) 705–713.

[2] R. Hultgren, P.D. Desai, D.T. Hawkins, M. Gleiser, K.K. Kelley, Selected Values of the Thermodynamic Properties of Binary Alloys, ASM, Metal Park, Ohio (1973).

[3] R.N. Singh, F. Sommer, Segregation and immiscibility in liquid binary alloys, *Rep. Prog. Phys.* **60** (1997) 57–150.

[4] J. Cai, Y.Y. Ye, Simple analytical embedded-atom-potential model including a long-range force for fcc metals and their alloys, *Phys. Rev. B* **54** (1996) 8398.

[5] J. Brillo, I. Egry, H.S. Giffard, A. Patti, Density and thermal expansion of liquid Au–Cu alloys, *Int. J. Thermophys.* **25** (2004) 1881–1888.

[6] Z. Guo, M. Hindler, W. Yuan, A. Mikula, *Thermochim. Acta* **525** (2011) 183–189.

[7] S. Yadav, U. Mehta, R. Gohivar, Thermodynamic, structural, surface and transport properties of Au–Cu melt, *Bibechana* **21** (2024) 150–158.

[8] K. Fitzner, O.J. Kleppa, Thermochemistry of binary and ternary noble metal alloys: the systems Cu-Ce, Ag-Ce, Au-Ce, Cu-Ag-Au, and Ce-Ag-Au, *Metall. Trans. A* **24** (1993) 1827–1834.

[9] R. Kattel, D. Gadtoula, R.P. Koirala, I.S. Jha, B.P. Singh, D. Adhikari, Mixing behaviour of Au–Cu melt, *J. Chin. Adv. Mater. Soc.* (2015) 1–7.

[10] D.P. Tao, A new model of thermodynamics of liquid mixtures and its application to liquid alloys, *Thermochim. Acta* **363** (2000) 105–113.

[11] J. Jiao, W. Ma, Y. Sun, D. Tao, X. Huang, J. Wu, Y. Dai, Application of molecular interaction volume model for predicting the Ca activity coefficients in Si-Ca binary and Si-Ca-Pb ternary alloys, *Vacuum* **128** (2016) 106–111.

[12] N.K. Roy, N. Vrind, R.K. Sahu, J. Mandal, I.S. Jha, Prediction of the alloying behaviour of Ni-Pd melts by molecular interaction volume model, *Proc. IOP Conf. Ser.: Mater. Sci. Eng.* **1334** (2025) 012005.

[13] G.H. Ding, L.M. Liang, P. Meng, L. Wang, Y.X. Wang, Y.J. Wang, R.X. Gan, Application of the molecular interaction volume model to the assessment of thermodynamic properties of liquid Zn-Bi alloys, *Kovove Mater*. **57** (2019) 55–60.

[14] L.M. Liang, G.H. Ding, Y.X. Wang, Y. Liu, Thermodynamic properties of liquid Zn-Cd alloys investigated by the molecular interaction volume model, *Phys. Chem. Liq.* **59** (2021) 706–715.

[15] L.M. Liang, X. Ren, Y. Wang, C. Zhu, Y.X. Wang, Thermodynamic-guided transport properties of liquid Pb-Bi alloys via molecular interaction modeling, *J. Phase Equilib. Diffus.* **46** (2025) 366–374.

[16] S.K. Shah, I.S. Jha, I. Koirala, Theoretical examination of some thermodynamic properties in In-Bi-Sn liquid alloy and its sub-binary systems, *J. Electron. Mater*. **52** (2023) 6350–6359.

[17] L.X. Kong, B. Yang, B.Q. Xu, Y. Li, D.C. Liu, Y.N. Dai, Application of MIVM for Pb-Sn-Sb ternary system in vacuum distillation, *Vacuum* **101** (2014) 324–327.

[18] D.P. Tao, Prediction of thermodynamic properties of the C-Fe-Co-Ni solid solution by binary infinite dilute activity coefficient, *Mater. Sci. Eng. A* **390** (2005) 70–75.

[19] A.B. Bhatia, D.E. Thornton, Structural aspects of the electrical resistivity of binary alloys, *Phys. Rev. B* **2** (1970) 3004–3012.

[20] A.B. Bhatia, W.H. Hargrove, Concentration fluctuations and thermodynamic properties of some compound forming binary molten systems, *Phys. Rev. B* **10** (1974) 3186–3196.

[21] B.E. Warren, X-ray Diffraction, Addition-Wesley Publishing Company, Reading, MA (1969).

[22] J.M. Cowley, An approximate theory of order in alloy, *Phys. Rev*. **77** (1950) 667–675.

[23] R.N. Singh, Short-range order and concentration fluctuations in binary molten alloys, *Can. J. Phys*. **65** (1987) 309–325.

[24] L.S. Darken, Thermodynamics of binary metallic solutions, *Trans. Metall. Soc. AIME* **239** (1967) 80.

[25] A.P. Singh, R.P. Chaudhary, R. Kumari, I.S. Jha, J. Mandal, Stability of NaK liquid alloys at different temperatures, *Mater. Today: Proc*. **66** (2022) 2251–2258.

[26] L.S. Darken, Diffusion, mobility and their interrelation through free energy in binary metallic systems, *Trans. AIME* **175** (1948) 184.

[27] T. Iida, R.I.L. Guthrie, The Physical Properties of Liquid Metals, Clarendon Press, Oxford (1988).

[28] O.W. Abodunrin, A.A. Ajayi, Calculation of excess stability function of four binary alloys, *Jordan J. Phys.* **14** (2021) 193–200.

[29] A.P. Singh, R.P. Chaudhary, M. Hussain, J. Mandal, I.S. Jha, Theoretical study of the stability of Cd-Sn liquid alloys at different temperatures, *Diffus. Found. Mater. Appl*. **37** (2025) 37–51.