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Thermodynamic Behavior of Cu-Ni Melts at 1823 K

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Abstract

The thermodynamic behavior of Cu-Ni melts at 1823 K has been analyzed by a statistical model known as molecular interaction volume model. The theoretical data have been compared with the correlated experimental data available in the literature. The theoretical results for excess free energy of mixing, free energy of mixing, activity coefficient and thermodynamic activity exhibit well harmony with the experimental data. The theoretical data of the structural properties like concentration fluctuation illustrate excellent similarities with the experimental values. The short- range order parameter, excess stability function and diffusivity ratio of Cu-Ni melts at 1823 K are also analyzed theoretically. The results indicates the segregating nature of Cu-Ni system at 1823 K.

Keywords: *Excess free energy of mixing; activity coefficients; concentration fluctuations; short-range order parameter; MIVM model.*

1 Introduction

Cu-Ni system has various domestic, marine, industrial applications etc [1] due to enhanced corrosion resistance, mechanical strength, fabricability and thermal stability. The phase diagram [2] illustrates that the variation of excess free energy of mixing, G_M^E and free energy of mixing, G_M with respect to concentration of Cu in Cu-Ni molten system are symmetric at concentration, $x = 0.5$. The activity of the constituent metals i.e. a_{Cu} and a_{Ni} possess positive deviations from Raoult's law. The concentration fluctuations, $S_{cc}(0)$, for Cu-Ni melts at 1823

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K is symmetric about $x = 0.5$. The positive values of G_M^E and positive deviations of the activity and $S_{cc}(0)$ from ideal behavior indicate the segregating nature of Cu-Ni liquid alloys at 1823 K [3]. Therefore, Cu-Ni melts has attracted several investigators due to interesting thermodynamic behavior and wide range of applications. In early studies, thermodynamic, structural and physical properties of Cu-Ni and Cu-Ni based ternary alloys have been explored by several investigators [4, 5, 6, 7]. However, there is scarcity of data in the literature related to the thermodynamic behavior of Cu-Ni melts at 1823 K. Therefore, Cu-Ni becomes very important for the further theoretical analysis.

In present work, a statistical mechanical model known as molecular interaction volume model is used to explore the thermodynamic properties and structural properties of Cu-Ni melts at 1823 K. The present model is successfully applied to study the thermophysical properties of various liquid alloys [8, 9, 10, 11]. The investigative equations are deduced in the framework of MIVM model for various thermodynamic and microscopic functions. These expressions are further utilized to determine the theoretical data of various thermodynamic and microscopic functions relative to the composition of Cu.

2 Formalism

2.1 Thermodynamic functions i.e. excess free energy of mixing, G_M^E, free energy of mixing, G_M, activity coefficient, $\gamma_\mu(\mu \equiv i, j)$ and activity, $a_\mu(\mu \equiv i, j)$ of a binary alloy

The MIVM model is a fluid-based model proposed by Tao [11] and worked out from statistical thermodynamics, fluid-phase equilibria and the elementary idea of the non-random interchange of liquid molecules. The molecular excess free energy of mixing of a binary alloy i-j is given by [11]

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

where, x_i and x_j = the concentrations (or mole fractions) of the constituent elements i and j respectively; Z_i and Z_j = first coordination numbers of i and j molecules respectively; A_{ij} and A_{ji} refers to the pair-potential energy interaction parameters respectively, defined as [30]

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

In above equation, ε_{ii}, ε_{jj} and ε_{ji} stands for the $i-i$, $j-j$ and $i-j$ pair potential energies respectively, where $\varepsilon_{ji} = \varepsilon_{ij}$, k = Boltzmann constant, T = absolute temperature of the liquid metal.

The partial molar and molar excess free energies are related as

$$\overset{\square}{G}{}_M^E = RT\ln\gamma_i = G_M^E + \delta\left(\frac{\partial G_M^E}{\partial x_i}\right)_{T,P,x[i,N]} - \sum_{j=1}^{N} x_j\left(\frac{\partial G_M^E}{\partial x_i}\right)_{T,P,x[j,N]} \tag{3}$$

It is important to note that the above equation involves the condition like when i ≠ N, $\delta=1$; and i = N, $\delta=0$. Again, $x[i,N]$ defines the variables x_j and x_N where, $x_N = 1-\sum_{j=1}^{N-1} x_j$ .Therefore, the activity coefficients of i and j constituents of the binary alloy are, respectively, given by

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

and

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

Now, the first co-ordination number of ith component may be determined by the relation [11]

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

when ΔH_{mi} and T_{mi} = enthalpy (or heat of formation) at melting temperature and melting temperature respectively; Z_c = 12 for closed packed structure; $\rho_i = N_i V_i = \frac{0.6022}{V_{mi}}$ = molecular number density; r_{mi} and r_{oi} = initial and fist peak values of radial distribution function of the melted metal i close to melting temperature respectively, and R is the molar gas constant. The radial distances are defined as

$$r_{oi} = 0.918 d_{\mathrm{cov}\,i} \text{ and } r_{mi} = \sigma_i \tag{7}$$

where, $d_{\mathrm{cov},i}$ and σ_i = atomic covalent diameter and atomic diameter respectively.

The equations for activity coefficients of the components i and j of the alloy in infinite dilute solution range i.e. x_i or $x_j \rightarrow 0$, are, respectively, given by

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

and

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

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Again, the activity and activity coefficients of the components i and j are related as [3]

$$a_i = \gamma_i x_i \tag{10a}$$

$$a_j = \gamma_j x_j \tag{10b}$$

2.2 Microscopic functions i.e. concentration fluctuations, $(S_{CC}(0))$ and short-range order parameter, α_1 of a binary molten alloy

The concentration fluctuations, $S_{cc}(0)$ and SRO (short-range order parameter), α_1 are very dominant microscopic functions to predict the interatomic interactions in a liquid system. The condition $S_{cc}(0) < S_{cc}^{id}(0)$ refers to ordering alloys and dissimilar atoms or molecules (i.e. i - j) relate together as the adjacent neighbors while $S_{cc}(0) > S_{cc}^{id}(0)$ represents the segregating alloys i.e. similar atoms or molecules (i.e. i-i or j-j) connect together as the closest neighbors [3].

Relation to the free energy of mixing, G_M , $S_{cc}(0)$ is expressed as [3, 12]

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

where, $G_M = G_M^E + G_M^{id}$

(12)

$$G_M = G_M^E + RT\left[x_i \ln x_i + x_j \ln x_j\right] \tag{13}$$

Equations (1), (11) and (13) provide

$$Scc(0) = \frac{x_i x_j}{1 + x_i x_j f(x_i x_j)} \tag{14}$$

where,

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

Since G_M^E = 0 for an ideal mixing, hence equation (12) provides $G_M = G_M^{id}$ .Therefore, equation (14) and (15) yield

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

The concentration fluctuations with respect to activity is expressed as [3, 12 13]

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

The value of $S_{cc}(0)$ computed from equation (17) on employing the experimental data of activity is considered as the experimental value of $S_{cc}(0)$ [3, 12, 13].

The SRO, α_1 described by Warren-Cowley [14, 15] and $S_{cc}(0)$ are related as [3, 16]

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

where, Z stands for coordination number in first neighbor shell.

Equation (18) indicates the positive and negative values of α_1 denote segregating alloys and ordered alloys respectively and $\alpha_1 = 0$ for ideal mixture.

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

The excess stability function, E^{XS} established by Darken [17, 18] may be computed from the relation

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

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

The diffusivity is a transport property suitable for analyzing the mixing behavior of the binary melts at the microscopic scale. The diffusivity ratio for a binary melt is expressed as [3, 19].

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

where, D_M = mutual diffusion coefficient of the alloy and D_{id} = intrinsic diffusion coefficient of ideal alloy.

Now, D_{id} is expressed as

$$D_{id}=x_i D_j + x_j D_i \tag{21}$$

where, D_i and D_j = self-diffusion coefficients of pure constituent i and j respectively.

Equation (20), clarifies that $\frac{D_M}{D_{id}} > 1$ for ordered alloy, $\frac{D_M}{D_{id}} < 1$ for segregating alloy and $\frac{D_M}{D_{id}} =1$ for ideal alloy.

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3. Results and Discussions

3.1 Excess free energy of mixing, G_M^E , free energy of mixing, G_M , activity coefficient , γ_μ , and activity, $a_\mu\,,(\mu = i, j)$ of Cu-Ni melts at 1823 K

The investigative expressions for G_M^E , G_M , γ_μ and a_μ $(\mu = i, j)$ are utilized to compute the thermodynamic properties of Cu-Ni melts at 1823 K. The necessary parameters of pure Cu and Ni are presented in **Table 1** [20] .The coordination number Z_i and Z_j of the constituents i.e. Cu and Ni of alloy are determined from equation (6) which are tabulated in **Table 2**. The parameters A_{ji} and A_{ij} are determined on solving equations (8) and (9) simultaneously by Newton - Rapson method. For this, the infinite dilute activity coefficients i.e. γ_i^∞ and γ_j^∞ are required which have been shown in **Table 2** [2]. The evaluated values of A_{ji} and A_{ij} are slightly altered to obtain a good agreement between the theory and experiment [2] for G_M^E of Cu-Ni melts at 1823 K. The suitable values of A_{ji} and A_{ij} are also depicted in **Table 2**.

Table 1: Input parameters for pure Cu and Ni [20]

Metal,i	ΔH_{mi} (KJ/mol)	σ_i ($\times 10^{-8}$cm)	r_{oi} ($\times 10^{-8}$ cm)	V_{mi} (cm^3/mol)
Cu	13.0	2.50	2.06	$7.94[1+1.0\times 10^{-4}(T-1356)]$
Ni	17.15	2.46	1.86	$7.43[1+1.51\times 10^{-4}(T-1728)]$

Table 2: Values of A_{ij} , A_{ji} , Z_i and Z_j for the constituents of Cu-Ni molten alloys at 1823 K

$i-j$	T(K)	A_{ij}	A_{ji}	Z_i	Z_j	γ_i^∞ [2]	γ_j^∞ [2]
Cu-Ni	1823	0.9959	0.8669	9.47	9.76	2.227	1.906

The theoretical values of G_M^E / RT obtained from equation (1) as a function of concentration of Cu for Cu-Ni alloys at 1823 K represent excellent uniformity with the corresponding experimental values [2] as shown in **Fig 1** . In composition region, x_{Cu} = 0.1 to 0.9, the values of G_M^E / RT are positive with maximum values at x_{Cu} = 0.5 i.e. 0.1947 (Theory) and 0.2002 (Experiment). Thus, the present theoretical investigation successfully reveals the symmetry in G_M^E / RT and segregating behavior of Cu-Ni melts at 1823 K. Again, the values of G_M / RT for Cu-Ni melts at 1823 K calculated from equation (13) is also presented in **Fig. 1** accompanied by experimental data [2]. A well agreement perceived. In the complete composition region, x_{Cu} = 0.1 to 0.9, G_M / RT is negative with minima at x_{Cu} = 0.5 i.e. G_M / RT (min^{m}) = - 0.4985 (Theory) and -0.4933 (Experiment).Thus, the detected symmetry in G_M / RT is successfully described by MIVM.

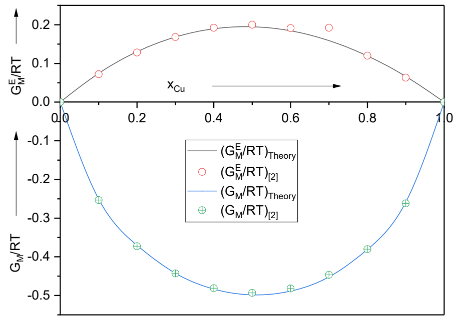

Fig. 1 (G^E_M/RT & G_M/RT) vs x_{Cu} for Cu-Ni Melts at 1823 K

The activity coefficients γ_{Cu} and γ_{Ni} are assessed from equations (4) and (5) respectively relative to the composition of Cu. The theoretical data of γ_{Cu} and γ_{Ni} for Cu-Ni melts at 1823 K are in excellent agreement with the experimental data [2] as shown in **Fig. 2**.

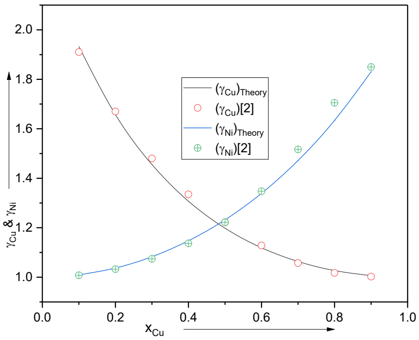

Fig. 2 γ_{Cu} & γ_{Ni} vs x_{Cu} for Cu-Ni melts at 1823 K

The theoretical values of γ_{Cu} and γ_{Ni} are applied to calculate the concentration reliance of a_{Cu} and a_{Ni} respectively from equations (10a) and (10b) for Cu-Ni melts at 1823 K, which are illustrated in **Fig. 3**. A well concord is viewed between theory and experiment [2].The departures of a_{Cu} and a_{Ni} from ideal behavior are positive which indicate the segregating nature of Cu-Ni melts at 1823 K. The harmony between theory and experimental data verifies the reasonable values of A_{ji} and A_{ij}

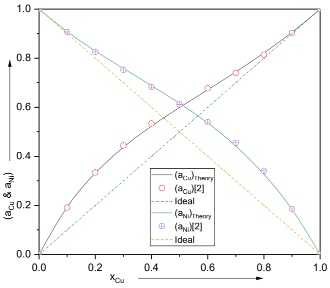

Fig. 3 (a_{Cu} & a_{Ni}) vs x_{Cu} for Cu-Ni melts at 1823 K

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3.2 Microscopic properties i.e. $S_{cc}(0)$, α_1, E^{XS} and D_M/D_{id} of Cu-Ni melts at 1823 K

The concentration dependence of $S_{cc}(0)$ for Cu-Ni melts at 1823 K are ascertained from equation (14) and (15). The theoretical data of $S_{cc}(0)$ are introduced in **Fig. 4** along with $S_{cc}^{id}(0)$ and experimental values of $S_{cc}(0)$ [2]. A well uniformity is found between the theory and experiment with minor discrepancies in the region $0.4 \le x_{cu} \le 0.7$ having maximum error of 14.75% at x_{Cu} = 0.6. It is viewed that $S_{cc}(0) > S_{cc}^{id}(0)$ in the region, $0.05 \le x_{Cu} \le 0.95$ and $S_{cc}(0)$ represents ideal behavior for Cu-Ni melts at 1823 K in the regions $0.0 \le x_{Cu} \le 0.05$ and $0.95 \le x_{Cu} < 1.0$. The maximum values of $S_{cc}(0)$ are found at $x_{Cu} = 0.5$ i.e. $S_{cc}(0)$ (max^{m}) = 0.408 (Theory) and 0.433 (Experiment). Hence, the present exploration reveals that Cu-Ni melt at 1823 K possesses segregating nature.

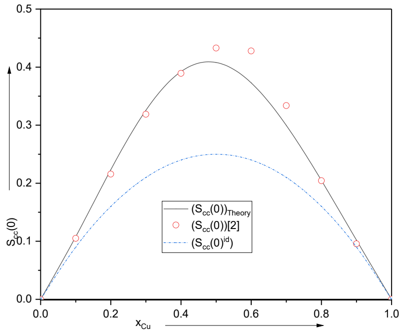

Fig. 4 $S_{cc}(0)$ vs x_{Cu} for Cu-Ni melts at 1823K

Equation (18) is utilized to obtain the composition dependent values of α_1 for Cu-Ni melts at 1823 K on considering Z = 10. The theoretical values of α_1 relative to the concentration of Cu are shown in **Fig 5**. In the complete range of composition, x_{Cu} = 0.1 to 0.9, α_1 is positive at each composition and have maximum value i.e. 0.0404 at x_{Cu} = 0.4, which validates the segregating character of Cu-Ni melts at 1823 K [21].

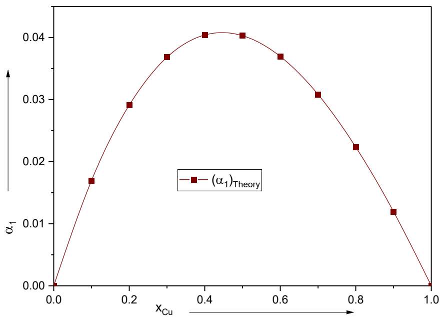

Fig. 5 α_1 vs x_{Cu} for Cu-Ni melts at 1823 K

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The excess stability function, E^{XS}/RT with respect to the concentration of Cu for Cu-Ni alloys at 1823 K found out from equation (19) are depicted in **Fig. 6**. Clearly, E^{XS}/RT exhibit negative value at each composition of Cu in the region x_{Cu} = 0.10 to 0.90 for Cu-Ni melts at 1823 K. This confirms the segregating nature of Cu-Ni melts at 1823 K [21, 22].

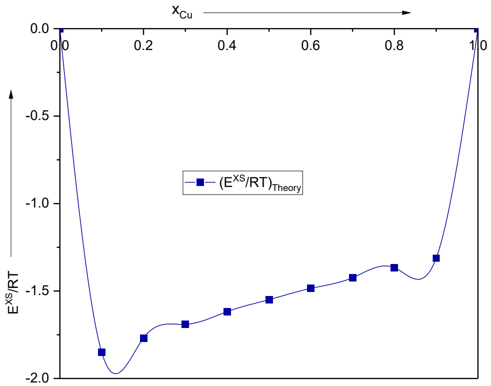

Fig. 6 E^{XS}/RT vs x_{Cu} for Cu-Ni melts at 1823 K

The diffusivity ratio, D_M/D_{id} for Cu-Ni melts at 1823 K is calculated from equation (20) relative to the concentration of Cu. The theoretical values of D_M/D_{id} presented in Fig. 7. It is seen that diffusivity ratio, D_M/D_{id} possesses minimum value i.e. 0.6117 at x_{Cu} = 0.4. This verifies the segregation in Cu-Ni melts at 1823 K [3].

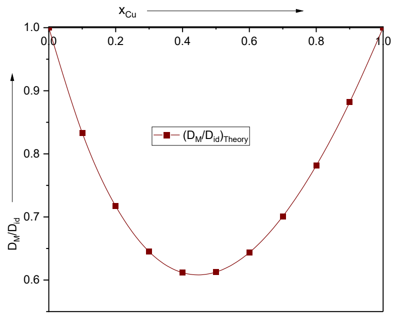

Fig. 7 D_M/D_{id} vs x_{Cu} for Cu-Ni melts at 1823 K

4. Conclusions

The present theoretical model successfully describes the alloying behavior of Cu-Ni melts at 1823 K. The observed symmetries in G_M^E , G_M and $S_{cc}(0)$ are successfully explained. The positive deviations of the activity and $S_{cc}(0)$ from linearity confirm the segregating nature of Cu-Ni melts at 1823 K. The positive values of α_1 and E^{XS}/RT , and $D_M/D_{id} < 1$ point out the segregating character of Cu-Ni melts at 1823 K.

Therefore, MIVM model is a reliable model for the theoretical exploration of the thermodynamic behavior of the binary melts.

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