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Theoretical Estimation of Mixing Properties of Al-Mg Binary Liquid Alloy at Different Temperatures

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Abstract The Regular Solution model has been used to describe thermodynamic properties, transport properties and structural properties at different temperatures theoretically. The interaction energy is temperature dependent and played an important role to explain the properties of Al-Mg liquid alloy at different temperatures. The theoretical values of interchange energy at different temperatures are obtained by best fit parameter approximation with the help of experimental values at 1073K. The properties have been studied with the help of computed theoretical interchange energy at different temperatures using interchange energy and temperature relation. A comparison of theoretical and experimental values at 1073K shows that they are in good agreement and using this basis we have studied properties at different temperatures.

Keywords Regular solution model, Order energy parameter, Thermodynamic properties, Transport properties, Structural properties, Different temperatures

1. Introduction

Al-Mg has wide range of uses in industries because of its properties like comparatively less inflammable, non-toxic, non-magnetic, good mechanical strength, crack resistance, and low cost maintenance [1-3]. It is also used in automobile engines, construction and shipborne as it has light weight and excellent corrosion resistance [4-6].

We have estimated the mixing properties i.e. thermodynamic, transport and structural properties of the alloy at different temperatures using regular solution model [7]. We have the model to investigate the different properties of the alloy as atoms of Al and Mg are almost same in shape and size i.e. atomic volume of Mg/atomic volume of Al = 16.29/11.4 ≈ 1 [8] which indicates that the alloy is suitable for investigation. In thermodynamic properties, we have calculated free energy of mixing (G_M), heat of mixing (H_M), entropy of mixing (S_M) and activity (a). In transport properties, viscosity (η) and diffusion coefficient ratio (D_m/D_{id}) have been computed. And, in structural properties the long wavelength limit ($S_{cc}(0)$) and chemical short range order parameter (α_1) are studied. Many researchers been working on several models to explain the mixing behavior of binary liquid alloys have been studied binary liquid alloys with different models [9-14]. And, viscosity is with the help of Moelwyn-Hughes equation [15-16]. The theoretical formulation has been presented in section (2), result and discussion in section (3) and conclusion in section (4).

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2. Formalism

Regular alloys constituent atoms A and B are sufficiently similar in size and shape so they are interchangeable on the lattice. The binary liquid alloy A-B of homogenous solution consists of $C_{A\ (A=Al)}$ (≡c) mole of A and $C_{B(B=Mg)}$ {≡(1-c)} mole of B respectively.

2.1. Thermodynamic Properties

The free energy of mixing (G_M) of binary liquid alloy is

$$G_M = G_M^{id} + G_M^{XS} \quad (1)$$

Where, excess free energy of mixing (G_M^{XS}) and ideal free energy of mixing (G_M^{id}) are given by

$$G_M^{XS} = \omega c_A c_B \quad (2)$$

and

$$G_M^{id} = RT\,[clnc + (1-c)ln(1-c)] \quad (3)$$

From equation (1), (2) and (3), we get

$$G_M = RT\,[c\ ln\ c + (1-c)ln(1-c)] + c(1-c).\omega \quad (4)$$

Where, T stands for temperature, ω is interaction energy and R is molar gas constant.

The expression for activities a_A of the elements A and B (a_B) in the binary liquid alloy can be derived from the standard relations

$$lna_A = lnc + \frac{\omega}{RT}(1-c)^2 \quad (5)$$

And

$$lna_B = \ln(1-c) + \frac{\omega}{RT}c^2 \quad (6)$$

The entropy of mixing (S_M) is given by

$$S_M = -\frac{\partial G_M}{\partial T} \quad (7)$$

From equation (4) and (7), we get

$$\frac{S_M}{R} = -[c\ ln\ c + (1-c)ln(1-c)] - c(1-c).\frac{1}{R}\frac{\partial \omega}{\partial T} \quad (8)$$

The importance of ω as temperature dependent has been studied by Bhatia et al. [17], Shrestha et al. [18], Alblas et al. [19]. The interchange energy (ω) is temperature dependent.

The relation for heat of mixing (H_M), entropy of mixing (S_M), and free energy of mixing (G_M) is expressed as

$$\frac{H_M}{RT} = \frac{S_M}{R} + \frac{G_M}{RT} \quad (9)$$

Using equation (4), (8) and (9), we get

$$\frac{H_M}{RT} = c(1-c).\frac{\omega}{RT} - c(1-c)\frac{1}{R}.\frac{\partial \omega}{\partial T} \quad (10)$$

2.2. Transport Properties

The relation between diffusion coefficient and concentration fluctuation derived by Singh and Sommer [20] is given as

$$\frac{D_M}{D_{id}} = \frac{S_{CC}^{id}(0)}{S_{CC}(0)} \quad (11)$$

Where, D_M is the mutual diffusion coefficient and D_{id} is the intrinsic diffusion coefficient for an ideal mixture computed as

$$D_M = c_1 D_2 + c_2 D_1 \quad (12)$$

Where, D_1 and D_2 are the self-diffusivities of pure components A and B respectively.

In term of energy order parameter ω, the diffusion coefficient can be expressed as [21]

$$\frac{D_M}{D_{id}} = [1 - \frac{2\omega}{RT}S_{cc}^{id}(0)] \quad (13)$$

We have used the Moelwyn-Hughes equation [15] to analyze the viscosity of Al-Mg liquid alloy which is given as

$$\eta = (c_1\eta_1 + c_2\eta_2)(1 - c_1c_2.\frac{H_M}{RT}) \quad (14)$$

Where, η_k (k = 1, 2) is the viscosity of pure component K and can be calculated from Arrhenius type equation [22] as

$$\eta_K = \eta_{OK} \exp\left[\frac{E_n}{RT}\right] \quad (15)$$

Where, η_{OK} is constant (in unit of viscosity) and E_n is the energy of activation of viscous flow for pure metal (in unit of energy per mole).

2.3. Structural Properties

The concentration fluctuation in the long wavelength limit ($S_{cc}(0)$) [23] can be calculated as

$$S_{CC}(0) = \frac{RT}{\left(\frac{\partial^2 G_M}{\partial c^2}\right)_{T,P,N}} \quad (16)$$

From equation (4) and (16), we get

$$s_{cc}(0) = \frac{c_A c_B}{1 - 2c_A\ c_B\ .\frac{\omega}{RT}} \quad (17)$$

The experimental values of $S_{cc}(0)$ is derived from experimental values of the activities [27] from the equation

$$s_{cc}(0) = (1-c)a_A\left(\frac{\partial a_A}{\partial c}\right)_{T,P,N}^{-1} = ca_B\left(\frac{\partial a_B}{\partial c}\right)_{T,P,N}^{-1} \quad (18)$$

Where, a_A and a_B are the activities of the component of A and B respectively.

The Warren-Cowley [21,24] short range order parameter (α_1) is used to investigate the arrangement of the atoms in the molten alloys. The theoretical values of these parameters can be evaluated

$$\alpha_1 = \frac{s-1}{s(Z-1)+1} \quad (19)$$

Here, $= \frac{S_{cc}(0)}{S_{cc}^{id}(0)}$, $s_{cc}^{id}(0) = c_A c_B$, and Z is the coordination number = 10 in our investigation.

2.4. Order Energy Parameter at Different Temperatures

The values of free energy of mixing (G_M) of the alloy at different temperatures is computed from equation (4) using the values of order energy parameter (ω) at different temperatures from the relation [25,26]

$$\omega(T) = A + BT \quad (20)$$

Where, *A* and *B* are coefficient constants.

3. Result and Discussion

The values of A and B is calculated using the values of ω/RT and $\frac{1}{R}\frac{\partial \omega}{\partial T}$ at temperature 1073K of the alloy Al-Mg in equation (20). The best fit parameters i.e. ω/RT = -1.122 and $\frac{1}{R}\frac{\partial \omega}{\partial T} = 0.391$ at temperature 1073K by the method of best fit approximation with the experimental values of the alloy from Hultgren et.al. 1973 [27] using equations (4) and (8). The theoretical values of interchange energy (ω) at different temperatures are calculated using eqation (20) which are presented in the table 1.

Table 1. Order energy parameter (ω) at different temperatures

Temperature (T)	Order energy parameter (ω/RT)
1073K	-1.1220
1350K	-0.9529
1500K	-0.8036
1200K	-0.6841
1650K	-0.5864
1800K	-0.5049
1950K	-0.4360
2100K	-0.3769

Interaction energy is found to be negative at all temperatures which indicate Al and Mg atoms are interacting in nature. Using these values of the interaction energy we have computed free energy of mixing (G_M), entropy of mixing (S_M), heat of mixing (H_M), activity (a), Ratio of

mutual and self-diffusivity (D_M/D_{id}) and viscosity (η) at different temperatures (T) with the help of Regular solution model.

3.1. Free Energy of Mixing

The values of free energy of mixing (G_M) of the alloy at temperatures 1200K, 1350K, 1500K, 1650K, 1800K, 1950K and 2100K have been computed using the values of ω(T) in equation (4) over the entire range of concentration. The free energy of mixing (G_M) of Al-Mg liquid alloy at different temperatures of study in the concentration range $C_{Al} = 0.1$ to 0.9 is shown in figure (1).

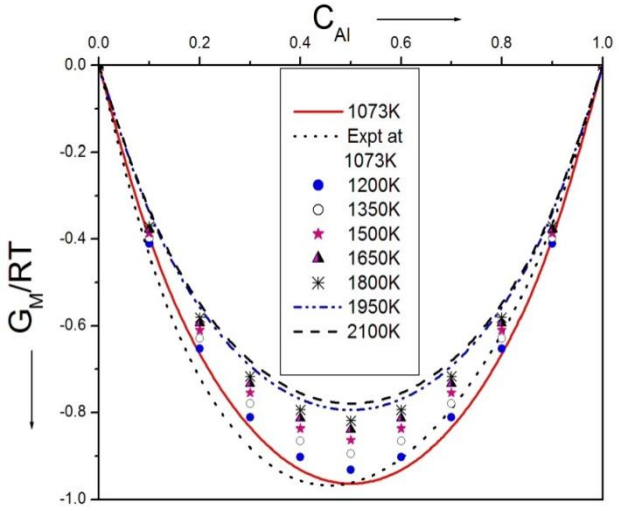

Figure 1. Graph for (G_M/RT) Versus the concentration of C_{Al} of Al-Mg liquid alloy at temperatures 1073K, 1200K, 1350K, 1500K, 1650K, 1800K, 1950K and 2100K

The theoretical and experimental values [27] of G_M/RT of the alloy are in well agreement at 1073K. The values of free energy of mixing are found to be negative which shows weekly interacting nature. At all temperatures of investigations the minimum value is at C_{Al}=0.5 which indicates that the symmetry in free energy of mixing. As the temperature of the alloy increases, the values of G_M/RT increases and vice-versa.

3.2. Entropy of Mixing (S_M)

By best fit method, the theoretical value of $\frac{1}{R}\frac{\partial\omega}{\partial T}$ at 1073K is obtained using equation (8) with the help of experimental values of entropy of mixing (S_M) is taken from Hultgren et. al. [27] i.e. $\frac{1}{R}\frac{\partial\omega}{\partial T} = 0.391$. With the help of this theoretical value of $\frac{1}{R}\frac{\partial\omega}{\partial T}$, entropy of mixing are computed at all temperatures (T) using equation (8) with the help of values of energy order parameters ω(T) at corresponding temperatures presented on the table 1.

The values of S_M/R are found to be positive in the entire concentration range. This explains the symmetry in entropy of mixing of Al-Mg liquid alloy. At all temperatures of study, the values of entropy of mixing remain unchanged as the theoretical value of $\frac{1}{R}\frac{\partial\omega}{\partial T} = 0.391$ remains constant using equation (20).

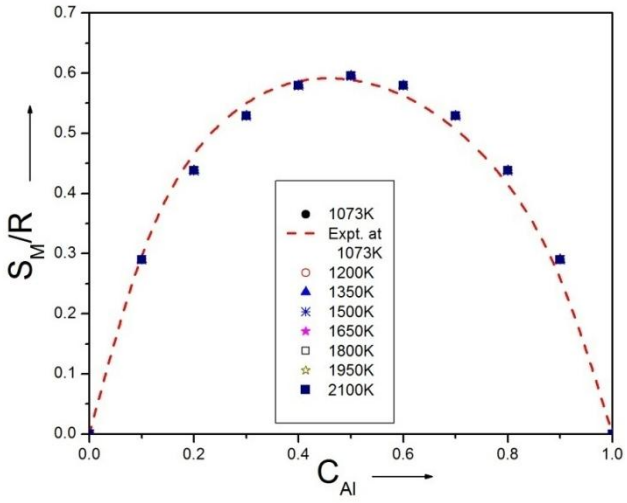

Figure 2. Graph for S_M/R versus the concentration of C_{Al} of Al-Mg liquid alloy at temperatures 1073K, 1200K, 1350K, 1500K, 1650K, 1800K, 1950K and 2100K

3.3. Heat of mixing (H_M)

The heat of mixing (H_M) for the alloy is calculated using equation (10) with the help of theoretical values of order energy parameter which is shown in figure 3.

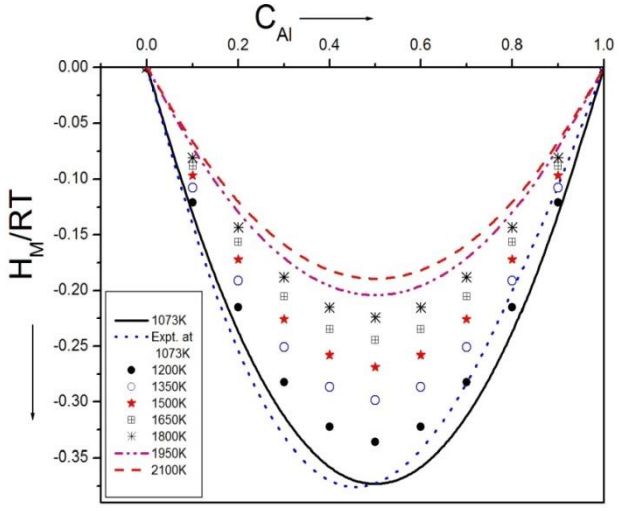

Figure 3. Graph for H_M/RT versus the concentration of C_{Al} of Al-Mg liquid alloy at temperatures 1073K, 1200K, 1350K, 1500K, 1650K, 1800K, 1950K and 2100K

The theoretical and experimental values of H_M/RT are in good agreement at 1073K. The values of H_M/RT are negative in the entire concentration range at all temperatures and increase with increase in temperature from 1073K to 2100K. Thus, the symmetry in heat of mixing is well explained.

3.4. Activity

Activity of Al and Mg atoms of the alloy is Calculated using equations (5) and (6) in entire concentration range. The activity (Ln a) versus the concentration is studied which is shown in figure 4 and figure 5.

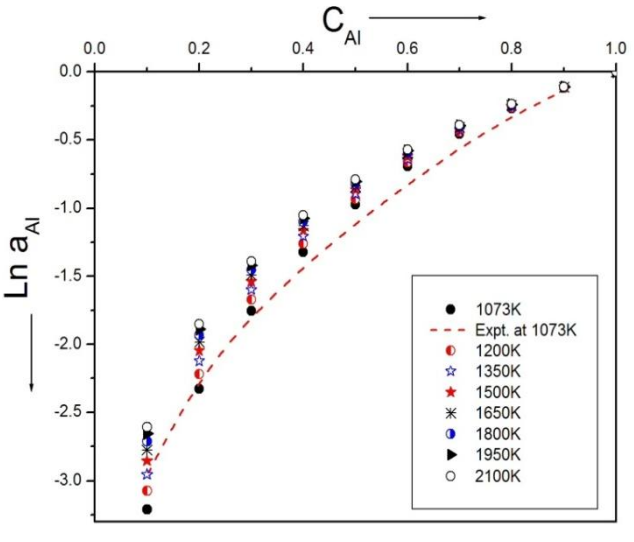

Figure 4. Ln a_{Al} versus C_{Al} of Al-Mg liquid alloy at 1073K, 1200K, 1350K, 1500K, 1650K, 1800K, 1950K and 2100K.

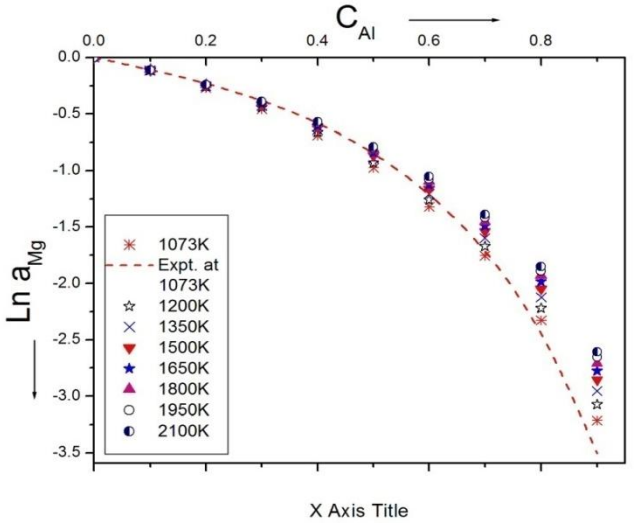

Figure 5. Ln a_{Mg} versus C_{Al} of Al-Mg liquid alloy at 1073K, 1200K, 1350K, 1500K, 1650K, 1800K, 1950K and 2100K

The values of activity (a) of both the components i.e. Al and Mg of the alloy are in good agreement with the experimental values for whole range of concentration at 1073K. The activity increases as temperature increases and vice-versa.

3.5. Transport Properties: Chemical Diffusion and Viscosity

Equation (13) is used to calculate the diffusion coefficient (D_M/D_{id}) with the help of theoretical values of order energy parameters ω(T). The investigation of the diffusion coefficient with the concentration of Al is shown in figure (6).

The value of $\frac{D_M}{D_{id}} > 1$ in the entire range of concentration at all temperatures (T) which shows chemical order in the alloy. The ordering tendency of the atoms in Al-Mg liquid alloys is greater about equiatomic composition as the maximum value of D_M/D_{id} is at C_{Al} = 0.5.

Viscosity of the alloy is calculated using equations (14) and (15). The viscosities of pure components Al and Mg at all temperatures(T) are computed using equation (15) with the help of constants η_{ok} and E for the metals [22] which is used to find the viscosity of the alloy for corresponding temperatures using equation (14).

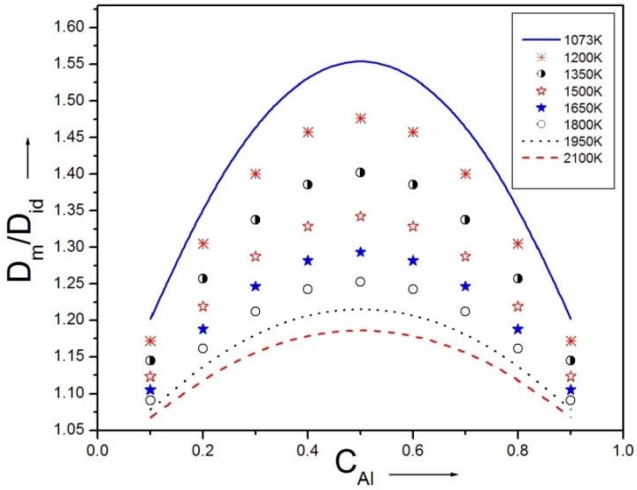

Figure 6. Ratio of mutual and self-diffusivities, D_M/D_{id} for Al-Mg liquid alloy at temperatures 1073K, 1200K, 1350K, 1500K, 1650K, 1800K, 1950K and 2100K versus concentration of Al

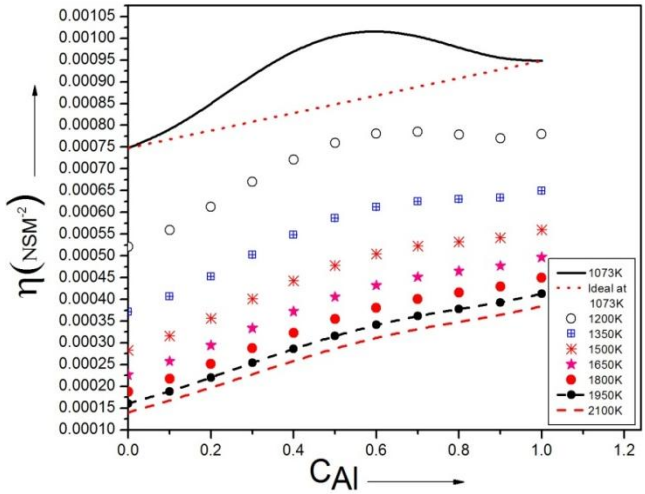

Figure 7. Viscosity of Al-Mg liquid alloy at 1073K, 1200K, 1350K, 1500K, 1650K, 1800K, 1950K and 2100K versus concentration of Al

The viscosity of pure component of Al atom is more than the viscosity of pure component Mg at all temperatures of study. The viscosity of the alloy is temperature dependent and decreases with the increase at each concentration range and maximum at 2100K.

3.6. Structural Properties

The theoretical and experimental values of concentration fluctuation in the long wavelength limit ($S_{cc}(0)$) can be obtained from equations (18) and (19). The values of Short range order parameters (α_1) at different temperatures are calculated from equation (19).

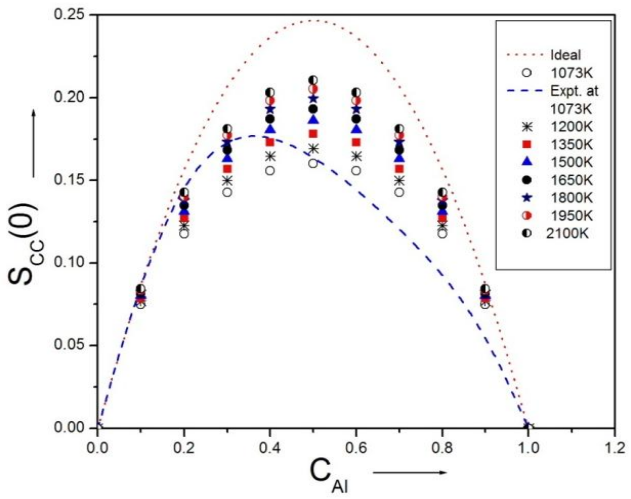

Figure 8. Theoretical values of Concentration fluctuation of Al-Mg liquid alloy at 1073K, 1200K, 1350K, 1500K, 1650K, 1800K, 1950K and 2100K along with experimental values at 1073K and $S_{cc}^{id}(0)$

The concentration fluctuation in the long wavelength limit ($S_{cc}(0)$) is maximum at C_{Al} =0.5 at all temperatures of study which suggests the symmetry in $S_{cc}(0)$ for Al-Mg liquid alloy. It is found that $S_{cc}(0) < S_{cc}^{id}(0)$ throughout the entire concentration range at all temperatures of investigation which implies that ordering is favored in the Al-Mg alloy.

The theoretical values of Warren-Cowley chemical short-range order parameter (α_1) [21,24] is Computed from equation (19) using the theoretical values of $S_{cc}(0)$. We have calculated that the Chemical short range order parameter (α_1) is negative at all the concentrations which indicate ordering nature of the alloy as shown in the figure 9.

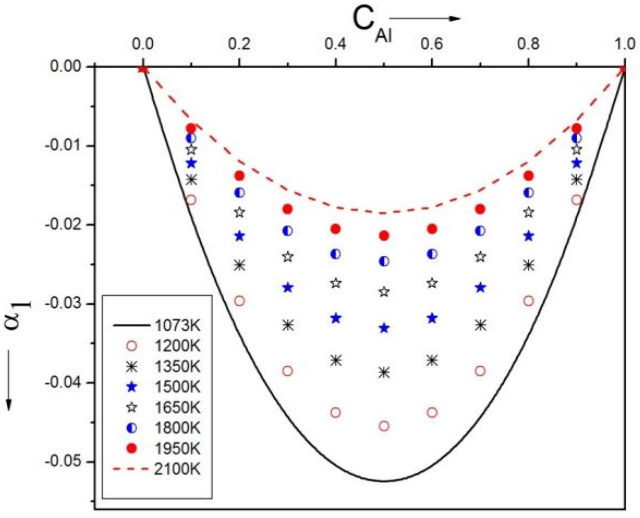

Figure 9. Chemical short range order parameter (α_1) of Al-Mg liquid alloy at 1073K, 1200K, 1350K, 1500K, 1650K, 1800K, 1950K and 2100K with co-ordination number (Z)= 10

The negative values of Chemical short range order parameter (α_1) at all temperatures indicates ordering nature. The value of α_1 increases as the temperature increases at each concentration range.

4. Conclusions

- The alloy is an ordered system and interacting in nature at all temperatures of investigations.
- The alloy is symmetric in nature at all temperatures.
- The activity of the alloy slightly increases as the temperature increases.
- Viscosity decreases as the temperature of study increases.
- properties can be studied at different temperatures using temperature and interaction energy relation theoretically.

REFERENCES

[1] S. Scudino, M. Sakaliyska, K.B. Surreddi, F. Ali and J. Eckert, "Structure and Mechanical Properties of Al-Mg Alloys Produced by Copper Mold Casting, " Journal of Alloys and Compounds, 504, S483-S486 (2010). https://doi.org/10.1016/j.jallcom.2010.03.208.

[2] M. Krol, T. Tański, P. Snopiński, and B. Tomiczek, "Structure and Properties of Aluminium-Magnesium Casting Alloys after Heat Treatment" Journal of Thermal Analysis and Calorimetry, 127, 299-308 (2017). https://doi.org/10.1007/s10973-016-5845-4.

[3] M. Schoenitz and E. L. Dreizin, "Structure and Properties of Al-Mg Mechanical Alloys." Journal of Materials Research, 18, 1827-1836 (2003). https://doi.org/10.1557/JMR.2003.0255.

[4] L. X. Gao, L.N. Wang, T. Qi,, et al., " Electrodeposition of Aluminum from AlCl3/Et3NHCl Ionic Liquids," Acta Physico-Chimica Sinica, 24, 939-944 (2008). https://doi.org/10.1016/S1872-1508(08)60040-6.

[5] S. Sah, M. Bawase and M. Saraf, "Light-Weight Materials and Their Automotive Applications," SAE Technical Paper 2014-28-0025(2014). https://doi.org/10.4271/2014-28-0025.

[6] Y. Kaushik, A Review on Use of Aluminium Alloy in Aircraft Components. I-Manager 's Journal on Material Science, 3, 33-38(2015). https://www.researchgate.net/publication/319456144.

[7] E.A. Guggenheim, "Mixture" Oxford University Press, Oxford (1952).

[8] E. A. Brandes and G.B. Brook, Ed., Smithells Metals Reference Book, 7th edition, Butterworth-Heinemann Linacre House, Jordan Hill, Oxford, 1992.

[9] P.J. Flory, "Thermodynamics of High Polymer solutions," J. Chem. Phys. 10, 51(1942). https://doi.org/10.1063/1.1723621.

[10] R. N. Singh and F. Sommer, "Segregation and immiscibility in liquid binary alloys," Reports Prog. Phys., vol. 60, no. 1, pp. 57– 150, Jan. 1997. https://doi.org/10.1088/0034-4885/60/1/003.

[11] H. K. Limbu, K. K. Mishra, J. Nirala, I.S. Jha, B.P. Singh, D. Adhikari, "Thermodynamic, microscopic and surface Properties of Zn-In liquid alloy at 700K," IOSR Journal of

applied Physics (IOSR-JAP), 2278- 4861. Volume 9, Issue 1 Ver. I, PP. 58-66. (Jan.– Feb.), 2017. https://doi.org/10.9790/4861-0901015866.

[12] A. Kumar, S.M. Rafique, N.Jha and A. K. Mishra, "Structural, Thermodynamic, electrical, and surface properties of Cu-Mg binary alloy: Complex formation Model," Physica B: Condensed Matter, Volume 357, Issues 3–4, 15 March 2005, Pages 445-451. https://doi.org/10.1016/j.physb.2004.12.031.

[13] T. Tanaka, K. Hack, T. Iida and Hara, "Application of thermodynamic databases to the evaluation of surface tensions of molten alloys, salt mixtures and oxide mixtures," Zeitschrift Fuer Met., vol. 87, pp. 380–389, 1996. http://hdl.handle.net/11094/26520.

[14] H. K. Limbu, K. K. Mishra, A.K. Sah, I. S. Jha, D. Adhikari, " Theoretical investigation of mixing properties of Sb-Sn binary liquid alloy at 905 K " BIBECHANA, pp.1-10, 15 (2018). https://doi.org/10.3126/bibechana.v15i0.18306.

[15] E. A. Moelwyn - Hughes, "Physical Chemistry," Oxford, London: Longmans Green and Co., 1974.

[16] H.K. Limbu, R.P. Ghimire, A.K. Joshi and G.P. Adhikari, " Assessment of mixing properties of Bi-In liquid alloy at different temperatures," SSRG International Journal of applied Physics, Vol. 6, Issue 2 (2019), PP. 14-21. https://doi.org/10.14445/23500301/IJAP-V6I2P103.

[17] A. B. Bhatia, W. H. Hargrove, and N. H. March, "Concentration fluctuations in conformal solutions and partial structure factor in alloys," J. Phys. C: Solid State Phys., vol. 6, no. 4, pp. 621–630, Feb. 1973. http://dx.doi.org/10.1088/0022-3719/6/4/008.

[18] G. K. Shrestha, I.S. Jha, and B.K. Singh, "Theoretical investigations on temperature dependence of thermodynamic properties and concentration fluctuations of In-Tl binary liquid alloys by optimization method" Bibechana, pp. 11- 23, 15 (2018). http://dx.doi.org/10.3126/bibechana.v15i0.18470.

[19] B. P. Alblas, C. van der Marel, W. Geertsma, J. A. Meijer, A. B. van Oosten, J. Dijkstra, P.C. Stein, and W. van der Lugt, "Experimental results for liquid alkali-group IV alloys," J. Non. Cryst. Solids, vol. 61–62, pp. 201–206, Jan. 1984. https://doi.org/10.1016/0022-3093(84)90551-9.

[20] R. N. Singh and F. Sommer, "Thermodynamic investigation of Viscosity and diffusion in binary liquid alloys". Phys. Chem. Liq. Vol. 36, pp. 17- 28, 2006.https://doi.org/10.1080/00319109808035917.

[21] J. M. Cowley, "An Approximate Theory of Order in Alloys," Phys. Rev., vol. 77, no. 5, pp.669–675, Mar. 1950. https://doi.org/10.1103/PhysRev.77.669.

[22] E. A. Brandes and G.B. Brook, Ed., "Smithells Metals Reference Book, 7th edition," Butterworth-Heinemann Linacre House, Jordan Hill, Oxford, 1992.

[23] A. B. Bhatia and R. N. Singh, "Thermodynamic Properties of Compound Forming Molten Alloys in a Weak Interaction Approximation," Phys. Chem. Liq., vol. 11, no. 4, pp. 343–351, 2007. https://doi.org/10.1080/00319108208080755.

[24] B. E. Warren, "X-ray diffraction," Addison-Wesley, Reading MA, 1969.

[25] R.N. Singh and A.B. Bhatia, "Flory's formula for the entropy of mixing of NaCs alloy," J. Phys. F: Met. Phys. Vol. 14, 2309-2314, 1984. https://doi.org/10.1088/0305-4608/14/10/009.

[26] F. E. Neale and N. E. Cusack, "Thermodynamic properties of liquid sodium-caesium alloys " J. Phys. F: Met. Phys. 12, 2839, 1982. https://doi.org/10.1088/0305-4608/12/12/016.

[27] R. Hultgren, P. D. Desai, D. T. Hawkins, M. Gleiser and K. K. Kelley, " selected Values of the Thermodynamic Properties of Binary Alloys," Metal Park, ASM International, Ohio, 1973.