

Concentration-Concentration Fluctuations and Chemical Ordering in Liquid Ti-Al Alloy at Different Temperatures

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Abstract:- The concentration-concentration fluctuation and chemical ordering in liquid Ti-Al at 1773 K, 1873 K, 1973 K, 2073 K, 2173 K, and 2273 K has been investigated using the quasi-chemical approximation model (QCAM). This analysis shows that chemical ordering leading to complex formation exists in the alloy across the whole concentration range.

I. INTRODUCTION

Alloys represent combinations of one metal with one or more additional elements, which could be either metallic or non-metallic in nature. Essentially, an alloy emerges when two or more elements, one of them being metallic, are melted and fused together. Upon cooling, this blend solidifies into a solid solution, mixture, or an intermetallic compound. Physically disassembling the components of an alloy is impossible. Despite potentially containing non-metals or metalloids, an alloy remains homogeneous and preserves the characteristics of metals. Unalloyed, pure metals may exhibit certain useful traits, like superior electrical conductivity, resistance to heat and corrosion, or impressive strength and hardness.

Additionally, it's been noted that unalloyed metals contain a uniform crystalline structure at the atomic level, stacking layer after layer of identical atoms. However, these layers tend to slide past each other with ease, often making the metal too malleable for practical applications. Take pure gold, for example, it's seldom used in jewelry since it's too delicate to endure much wear and tear. Nevertheless, introducing an element with a distinct atomic size can disrupt the layers at random, reducing their propensity to slide and resulting in a harder alloy. Furthermore, the judicious selection of compounds permits metallurgists to custom design other attributes like resistance to corrosion and the melting point (Bell, 2019; Lim, 2016).

Alloys are commonly categorized as either substitutional or interstitial, a classification dependent on their atomic arrangements. In substitutional alloys, the atoms from each element can substitute for their counterpart in the crystal lattice, whereas, in interstitial alloys, these atoms fill the spaces between the atoms of the host metal. Further classifications of alloys can include homogeneous

(having a single phase), heterogeneous (comprising two or more phases), or intermetallic (characterized by an absence of clear phase boundaries) (Abbott, 1980). The process of alloying involves merging a metal with one or more other elements, either metallic or non-metallic, which typically serves to augment its properties. For instance, steel, primarily composed of iron, is inherently stronger. While an alloy's physical properties like density, reactivity, and conductivity may not markedly differ from its component elements, its engineering properties, such as tensile and shear strength, can vary significantly. Unlike pure metals, most alloys lack a single defined melting point. Instead, they exhibit a melting range where the substance transitions between solid and liquid states. For most alloys, there exists a specific constituent ratio, referred to as the "eutectic mixture," where the alloy exhibits a distinct melting point (Abbott, 1980).

To analyze the alloying behavior of liquid binary alloys, various compound formation models have been proposed (Singh and March 1995). These models encompass the Quasi-Chemical Approximation Model (QCAM), Self-Association Model (SAM), and Complex Formation Model (CFM). The selection of Aluminum-Titanium (Al-Ti) as the focus of our study is spurred by recent predictions of thermodynamic data for liquid Al-Ti at varying temperatures, using the FactSage thermochemical software and database. The FactSage suite comprises several informational, database, computational, and manipulative modules, enabling access and manipulation of pure substances and solution databases as developed by Kostov and team (2006).

For this project, we've chosen the Quasi-Chemical Approximation Model (QCAM) to investigate the energetics of chemical ordering and temperature effects on Titanium-Aluminum (Ti-Al) liquid alloys at various temperatures for which we have experimental data. Both aluminum and titanium have a multitude of significant industrial applications. Aluminium is integrated into a vast array of products, including cans, foils, kitchenware, window frames, and components of doors and airplanes. Titanium, another well-known metal, finds its use in products such as jewelry, tennis rackets, goalkeeper masks, scissors, bicycle frames, surgical tools, and mobile phones. The Ti-Al liquid binary

alloy possesses extensive applications in science and engineering fields. Particularly, Ti-Al alloys exhibit potential for supplanting heavier materials in high-temperature structural applications, such as automotive and aerospace engine components (Kassner, 2015).

The primary objectives of this project are twofold. Firstly, we aim to ascertain the concentration-concentration fluctuations and chemical ordering in liquid Titanium-Aluminum (Ti-Al) alloys at varying temperatures and compare the derived results with corresponding experimental data. The activity of Al and Ti as a function of titanium concentration was calculated in the temperature range between 1773 K and 2273 K. Secondly, we intend to employ the Quasi-Chemical Approximation Model (QCAM) to compute the thermodynamic activities of these binary alloys. The effectiveness of the model will be further evaluated by juxtaposing the calculated outcomes with experimental data.

II. METHODOLOGY

➤ *Description of Quasi-Chemical Approximation*

The Quasi-Chemical Approximation provides a simplified framework for examining liquid alloys. It predicated on the assumption of like-atom pairing at equivalent sites that have short-ranged interactions between nearest neighbors. Such pairing engenders the formation of self-associates within the alloys. A liquid binary A-B alloy comprises N_A atoms of element A and N_B atoms of element B, which collectively form like-atom clusters or self-associates of type μA and νB . Here, μ and ν represent the number of atoms in the clusters within type A and B matrices, respectively. Leveraging this assumption, expressions for thermodynamic and microscopic functions are derived for binary liquid alloys (Koirala et al, 2013).

The grand partition function, Ξ of a binary molten alloys AB, which consist of $N_A = N_c$ and $N_B = N(1 - c)$ atoms of elements A and B, respectively where the total number of atoms N can be expressed as

$$\Xi = \sum_E (q_A(T)N_A(q_B(T))^{NB} \exp[\mu_A N_A + \mu_B N_B - E]/K_B T) \dots\dots\dots(1)$$

where $q_i^N(T)$ and μ_i are atomic partition function and chemical of ith component (i=A,B), K_B is the Boltzmann's constant, T is the absolute temperature and E is the configurational energy of all alloy.

After some little algebra, (Prasad et al, 1998), the solution of (1) is given as the ratio of the activity coefficient γ ($\gamma = \gamma_A/\gamma_B$); γ_A and γ_B are activity coefficients of A and B atoms, respectively for the compound ($A_\mu B_\nu$) forming alloys such as

$$\ln \gamma = \frac{z(1-C)}{2C} \frac{\beta+2c-1}{\beta-2C+1} \dots\dots\dots(2)$$

with

$$\beta = [1 + 4C(1 - C)(\eta^2 - 1)]^{1/2} \dots\dots\dots(3)$$

$$\eta^2 = \exp\left(\frac{2\omega}{2k_B T}\right) \exp(2P_{AB} \Delta\epsilon_{AB} - P_{AA}\Delta\epsilon_{AA} - P_{BB}\Delta\epsilon_{BB}/ K_B T) \dots\dots\dots(4)$$

and the interchange or ordered energy, ω is expressed as

$$\omega = Z[\Delta\epsilon_{AE} - \frac{1}{2}(\Delta\epsilon_{AA} + \Delta\epsilon_{BB})] \dots\dots\dots(5)$$

$\Delta\epsilon_{AB}$, $\Delta\epsilon_{AA}$ and $\Delta\epsilon_{BB}$ are the interaction parameter and $\Delta\epsilon_{ij}$ is the change in the energy of ij bond in the complex $A_\mu B_\nu$, P_{ij} is the probability that ij bond is apart of the complex.

P_{ij} may be expressed as

$$P_{AB} = C^{\mu-1}(1 - c)^{\nu-1}[2 - C^{\mu-1}(1 - c)^{\nu-1}] \dots\dots\dots(6)$$

$$P_{AA} = C^{\mu-2}(1 - c)^\nu[2 - C^{\mu-2}(1 - c)^\nu], \mu \geq 2 \dots\dots\dots(7)$$

$$P_{BB} = C^\mu(1 - c)^{\nu-2}[2 - C^\mu(1 - c)^{\nu-2}], \nu \geq 2 \dots\dots\dots(8)$$

From which we can get an expression for the excess Gibbs free energy of mixing G_M^{xs} defined by

$$G_M^{xs} = G_m - RT [c \ln c + (1 - c) \ln(1 - c)] \dots\dots\dots(9)$$

G_m being the free energy of mixing. G_M^{XS} can be obtained from (2) by using the thermodynamic relationship Bhatia *et al.*,(1982).

$$\frac{G_M^{XS}}{RT} = Z \int_0^c [\ln \sigma + (2K_B T) - 1 (P_{AA} \Delta \epsilon_{AA} - P_{BB} \Delta \epsilon_{BB})] dx + \Phi \dots\dots\dots(10)$$

Here, Z is the coordination number, c is the concentration of atom A and R is the universal gas constant and

$$\ln \sigma = \frac{1}{2} \ln \frac{(1-c)(\beta+2c-1)}{c(\beta-2c+1)} \dots\dots\dots(11)$$

The constant Φ in Eq (10) is determined from the requirement that $G_M^{XS} = 0$ at $c = 0$ and 1

➤ *Concentration Fluctuation and Chemical Short Range Order*

The concentration-concentration fluctuations in the long wavelength limit $s_{cc}(0)$ alongside the Warren-Cowley short-range order parameter (α_1) are instrumental properties that provide valuable insights into the structural characteristics of molten alloys. $s_{cc}(0)$ indicates nature of chemical ordering and α_1 represents the degree of ordering in the melt. The standard relation for concentration fluctuation in long wavelength limit ($s_{cc}(0)$) for binary liquid alloys(Zhang *et al.*, 2001).

$$s_{cc}(0) = RT \left(\frac{\partial^2 G_M}{\partial C_A^2} \right)_{T,P,N}^{-1} = C_B a_A \left(\frac{\partial a_A}{\partial C_A} \right)_{T,P,N}^{-1} = C_A a_B \left[\frac{\partial a_B}{\partial (1-C_A)} \right]_{T,P,N}^{-1} \dots\dots\dots(12)$$

For ideal mixing the energy parameters, ω given in (5) is equal to zero and (12) becomes

$$s_{cc}^{id}(0) = C_A C_B \dots\dots\dots(13)$$

$$s_{cc}(0) = \frac{C(1-C)}{1+z/2(\frac{1}{\beta}-1)} \dots\dots\dots(14)$$

(12) is usually utilized to obtain the experimental values of $s_{cc}(0)$ from the measured activity or free energy of mixing data. The mixing behaviour of liquid binary alloys can be inferred from the deviation of $s_{cc}(0)$ from $s_{cc}^{id}(0)$. The presence of chemical order is indicated by $s_{cc}(0) < s_{cc}^{id}(0)$; on the contrary, if $s_{cc}(0) > s_{cc}^{id}(0)$, the segregation and demixing in liquid alloys take place. The Warren-Cowley short-range order parameter, denoted as α_1 , can further provide insights into the local arrangement of atoms in molten alloys. This parameter, α_1 , can be expressed in terms of the β -function (3), allowing a deeper understanding of the atomic organization within these alloys.

The chemical short-range order parameter is given as

$$\alpha_1 = \frac{\beta-1}{\beta+1} \dots\dots\dots(15)$$

The diffusion coefficient is another valuable tool leveraged by both theoreticians and experimentalists to extract information about the composition and characteristics of liquid alloys. The interdiffusion coefficient (D_M) of binary alloys can be expressed in terms of activity (a_i) and self-diffusion coefficient (D_i) of pure component with the help of Darken’s equation:

$$D_M = D_s \frac{\partial \ln a_A}{\partial c} \dots\dots\dots(16)$$

With
 $\dots\dots$
 $D_s = \sum C_i D_i \dots\dots\dots(17)$

If we apply Eq. (12) in Eq. (16), we obtain an expression for D_M in terms of $S_{cc}(0)$, i.e.

$$\frac{D_M}{D_s} = \frac{C_A C_B}{s_{cc}(0)}$$

For ideal mixing, $s_{cc}(0) \rightarrow s_{cc}^{id}(0)$, i.e $D_M \rightarrow D_s$; for ordered alloys, $s_{cc}(0) < s_{cc}^{id}(0)$, i.e $D_M > D_s$ and similarly for segregation, $D_M < D_s$.

III. RESULTS AND DISCUSSION

The quasi-chemical approximation model was applied to calculate Gibbs free energy of mixing G_m , concentration–concentration fluctuation $s_{cc}(0)$, calculated activity C_a , diffusion co-efficient D_M , chemical short range order parameter α_1 . Experimental data were taken from (Kostov *et al*, 2006). The two alloy and their working temperature present in Table 1

Table 1 Activity of Ti And Al in the Temperature Range from 1773 K to 2273 K

X_{Ti}	1773 K		18873 K		1973 K	
	a_{Ti}	a_{Al}	a_{Ti}	a_{Al}	a_{Ti}	a_{Al}
0	0	1	0	1	0	1
0.1	0.015141	0.91191	0.020727	0.91426	0.027485	0.91637
0.2	0.032151	0.79919	0.041609	0.80936	0.052499	0.81860
0.3	0.064489	0.63285	0.078649	0.65378	0.094007	0.67319
0.4	0.12757	0.43767	0.14703	0.46617	0.16703	0.49335
0.5	0.23813	0.26243	0.26123	0.29103	0.28389	0.31938
0.6	0.40118	0.13875	0.42316	0.16141	0.44393	0.18492
0.7	0.59423	0.066993	0.60958	0.082052	0.62371	0.098450
0.8	0.77232	0.030590	0.77915	0.039369	0.78535	0.049389
0.9	0.90274	0.012534	0.90404	0.016800	0.90521	0.021859
1	1	0	0	0	1	0

X_{Ti}	2073K		2173K		2273K	
	a_{Ti}	a_{Al}	a_{Ti}	a_{Al}	a_{Ti}	a_{Al}
0	0	1	0	1	0	1
0.1	0.0355467	0.91829	0.044707	0.92003	0.055217	0.92163
0.2	0.064677	0.82705	0.078218	0.83479	0.093026	0.84191
0.3	0.11045	0.69121	0.12785	0.70800	0.14610	0.72366
0.4	0.18743	0.51928	0.20810	0.5400	0.22893	0.56756
0.5	0.30605	0.34737	0.32767	0.37489	0.34872	0.40190
0.6	0.46358	0.20910	0.48218	0.23377	0.49978	0.25880
0.7	0.63676	0.11607	0.64885	0.13478	0.66007	0.15446
0.8	0.79099	0.060617	0.79615	0.073008	0.80088	0.086505
0.9	0.90627	0.027728	0.90724	0.034412	0.90812	0.041902
1	1	0	1	0	1	0

➤ *Gibbs Free Energy Of Mixing G_m*

Using equation (9) we calculated the Gibbs free energy of mixing for Ti-Al liquid alloy at temperatures of 1773 K, 1873 K, 1973 K, 2073 K, 2173 K, 2273 K respectively. Figures 1.1-1.6 shows the plots of the concentration dependence of free energy of mixing at different temperatures. These figures show that our interaction parameters give a good representation of the calculated values of free energy of mixing for Ti-Al alloys at respectively temperatures.

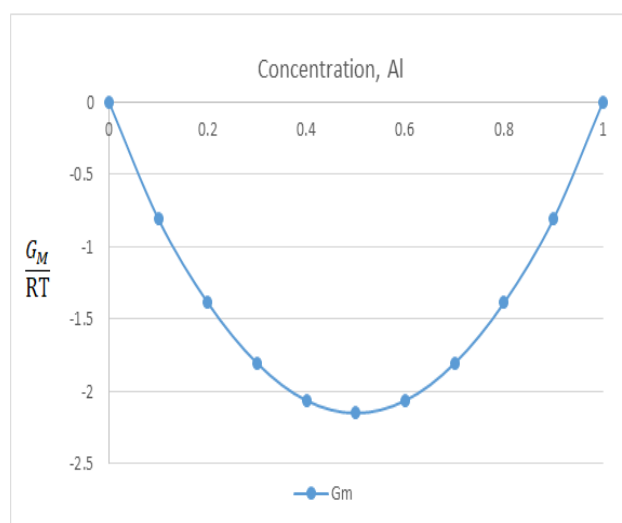


Fig 1 Gibbs Free Energy of Mixing G_M , Versus Concentration of Ti-Al At 1773 K. The Solid Line Represents Calculated Values for Ti-Al Alloy. C_{Al} Is the Al Concentration in the Alloy.

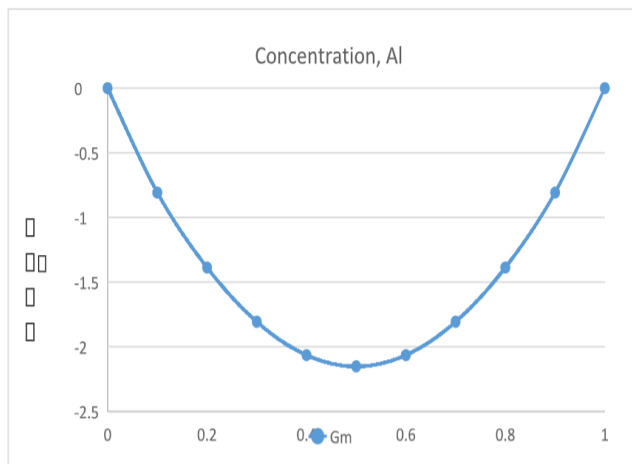


Fig 2 Gibbs Free Energy of Mixing G_M Versus Concentration of Ti-Al At 1873 K. The Solid Line Represents Calculated Values. C_{Al} Is Concentration in the Alloy.

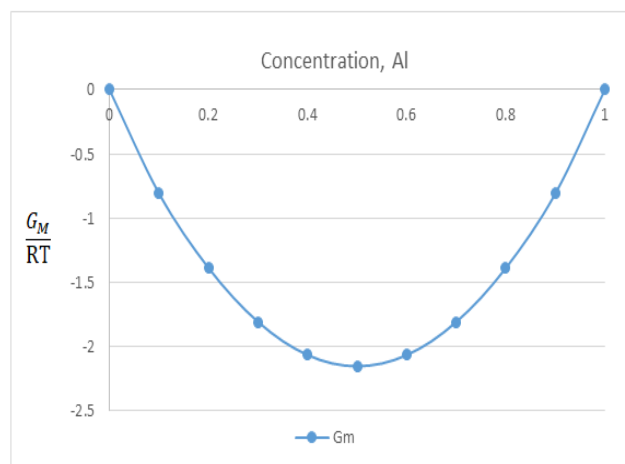


Fig 5 Gibbs Free Energy of Mixing G_M Verse Concentration of Ti-Al At 2173 K. The Solid Line Represents Calculated Values. C_{Al} Is Concentration in the Alloy.

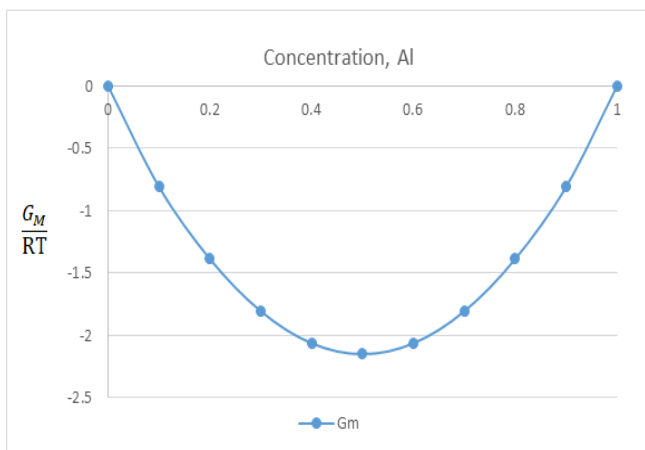


Fig 3 Gibbs Free Energy of Mixing G_M Versus Concentration of Ti-Al At 1973 K. The Solid Line Represents Calculated Values. C_{Al} Is Concentration In The Alloy.

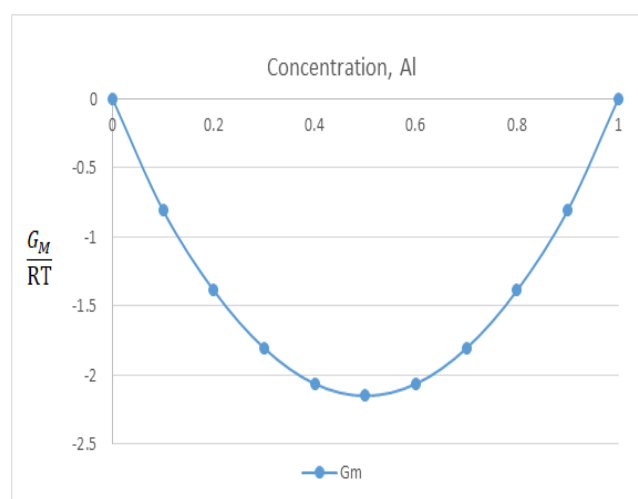


Fig 6 Gibbs Free Energy of Mixing G_M Verse Concentration of Ti-Al At 2273 K. The Solid Line Represents Calculated Values. C_{Al} Is Concentration in the Alloy.

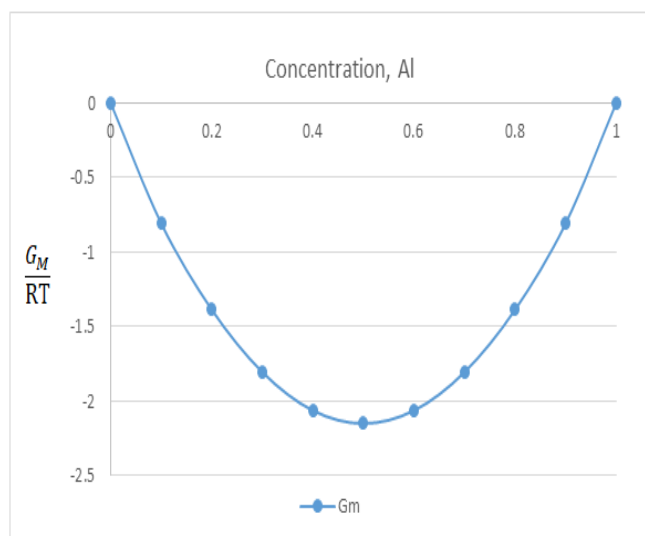


Fig 4 Gibbs Free Energy of Mixing G_M Verse Concentration Ti-Al At 2073 K. The Solid Line Represents Calculated Values. C_{Al} Is Concentration in the Alloy.

➤ *Concentration-Concentration Fluctuations, $s_{CC}(0)$*

From accurate analysis of the nature of ordering in Ti-Al liquid alloys, the first quantity investigated is the concentration- concentration fluctuation in the long wavelength limit $s_{CC}(0)$. The deviation of the $s_{CC}(0)$ from the ideal $s_{CC}^{id}(0)$ is an essential parameters in order to visualize the nature of ordering in liquid alloy. The results are shown in figures 1.7- 1.12. From the figure, it is clear that calculated $s_{CC}(0) < s_{CC}^{id}(0)$ at the six temperatures investigated. This implies there is tendency for unlike atoms in the liquid Ti-Al alloys to come together and form chemical compounds. That's Ti-Al alloy is a compound forming alloys. The degree of compound forming in the liquid Ti-Al alloys does not change much with increasing temperature from 1773 K to 2273 K. However, the G_m suggest that the degree of interaction between the component atoms of the decrease with increase in temperature. That's Ti-Al alloys at 1773 K is more interacting than at 1873 K and in that order.

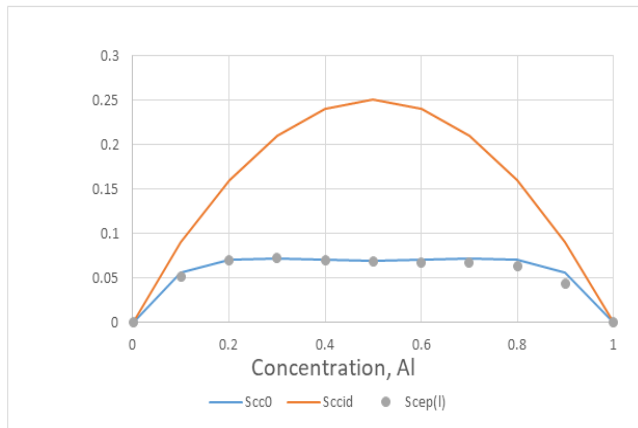


Fig 7 Concentration-Concentration Fluctuation $S_{CC}(0)$ Versus Concentration at 1773 K, for Liquid Ti-Al Alloy. The Red Line Represents Ideal Values, the Blue Line Represents Theoretical Values and the Circle Represents Experimental Values.

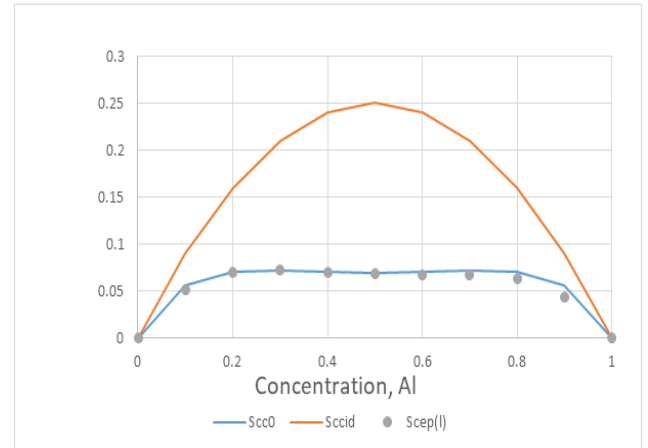


Fig 10 Concentration-Concentration Fluctuation $S_{CC}(0)$ Versus Concentration At 2073 K, for Liquid Ti-Al Alloy. The Red Line Represents Ideal Values, the Blue Line Represents Theoretical Values and the Circle Represents Experimental Values.

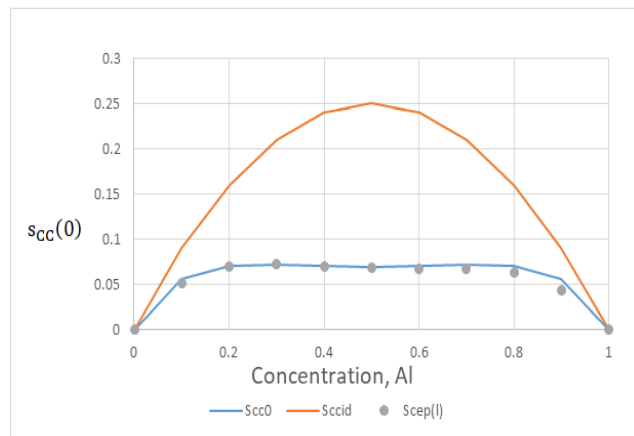


Fig 8 Concentration-Concentration Fluctuation $S_{CC}(0)$ Versus Concentration at 1873 K, for Liquid Ti-Al Alloy. The Red Line Represents Ideal Values, the Blue Line Represents Theoretical Values and the Circle Represents Experimental Values.

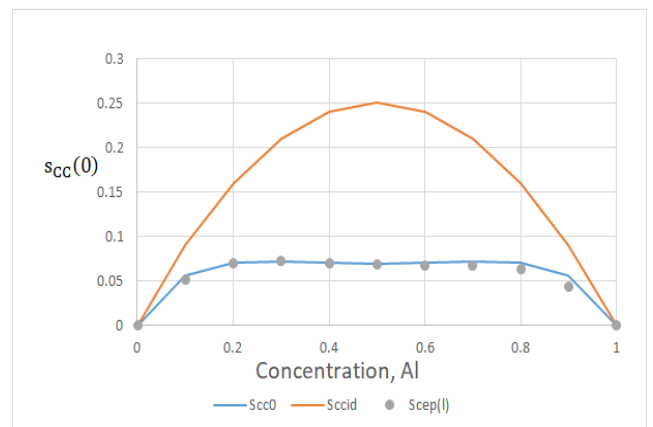


Fig 11 Concentration-Concentration Fluctuation $S_{CC}(0)$ Versus Concentration at 2173 K, for Liquid Ti-Al Alloy. The Red Line Represents Ideal Values, the Blue Line Represents Theoretical Values and the Circle Represents Experimental Values.

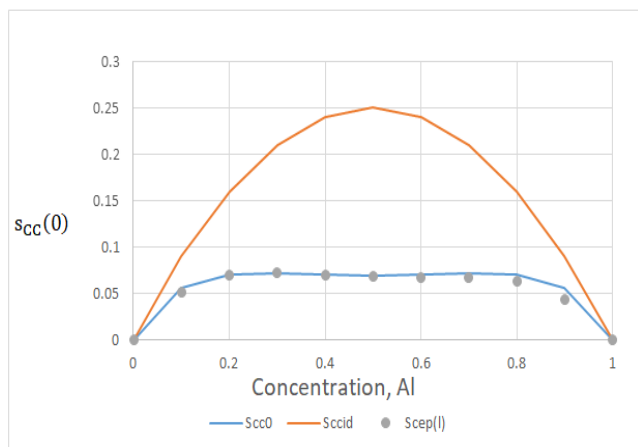


Fig 9 Concentration-Concentration Fluctuation $S_{CC}(0)$ Versus Concentration at 1973 K, for Liquid Ti-Al Alloy. The Red Line Represents Ideal Values, the Blue Line Represents Theoretical Values and the Circle Represents Experimental Values.

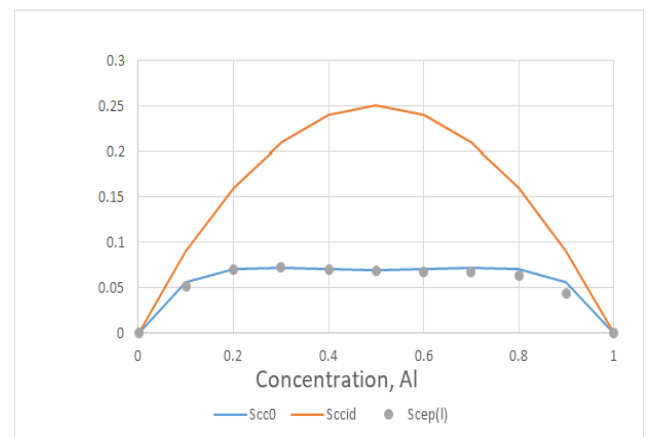


Fig 12 Concentration-Concentration Fluctuation $S_{CC}(0)$ Versus Concentration at 2273 K, for Liquid Ti-Al Alloy. The Red Line Represents Ideal Values, the Blue Line Represents Theoretical Values and the Circle Represents Experimental Values.

➤ Activity Coefficient ($\ln \Gamma$)

Using equation (2) we calculated the activity coefficient for liquid Ti-Al alloys at temperatures of 1773 K – 2273 K. The aluminium activity shows a negative deviation from Raoult’s law while the titanium shows a positive deviation from Raoult’s law. At 0.2 from the negative side of the concentration, there was a positive shift, and at 0.8 from the positive side of the concentration, there was a positive shift.

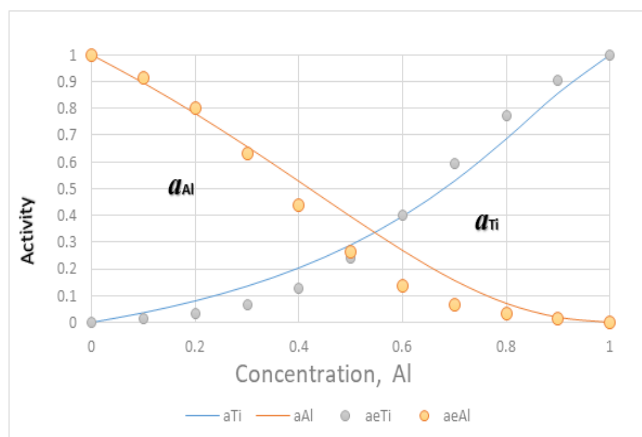


Fig 13 Calculated Activities of Compounds in Liquid Ti-Al Alloys at 1773 K Verse the Concentration of Al

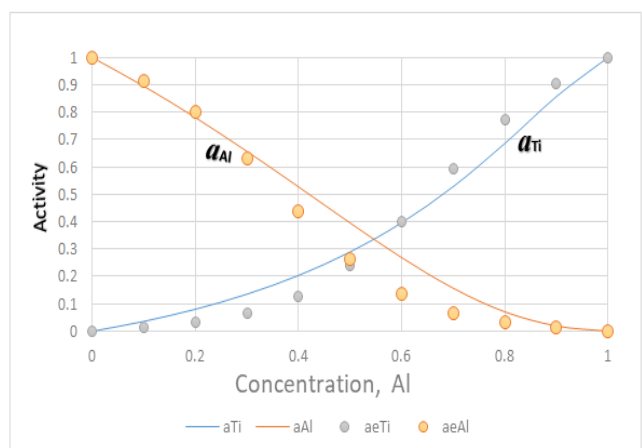


Fig 14 Calculated Activities of Compounds in Liquid Ti-Al Alloys at 1873 K Verse the Concentration of Al

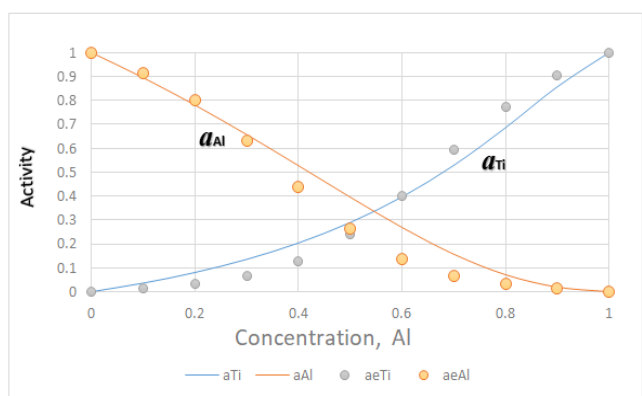


Fig 15 Calculated Activities of Compounds in Liquid Ti-Al Alloys at 1973 K Verse the Concentration of Al.

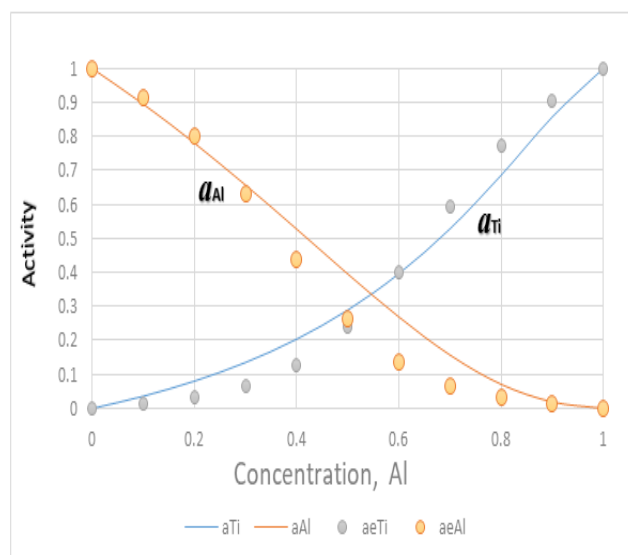


Fig 16 Calculated Activities of Compounds in Liquid Ti-Al Alloys at 2073 K Verse the Concentration of Al.

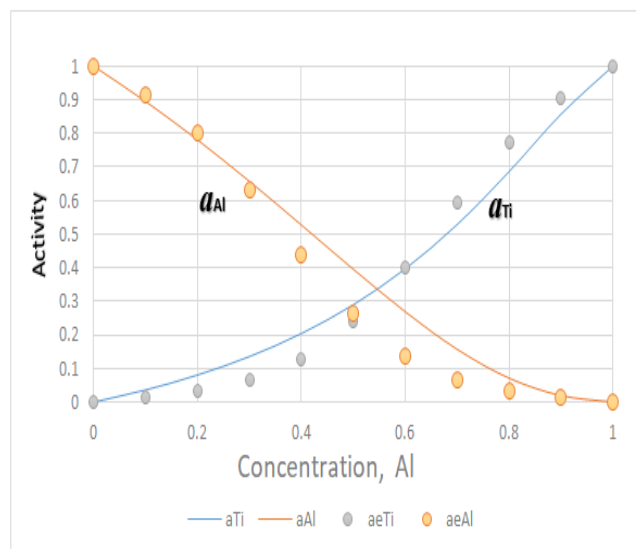


Fig 17 Calculated Activities of Compounds in Liquid Ti-Al Alloys at 2173 K Verse the Concentration Of Al.

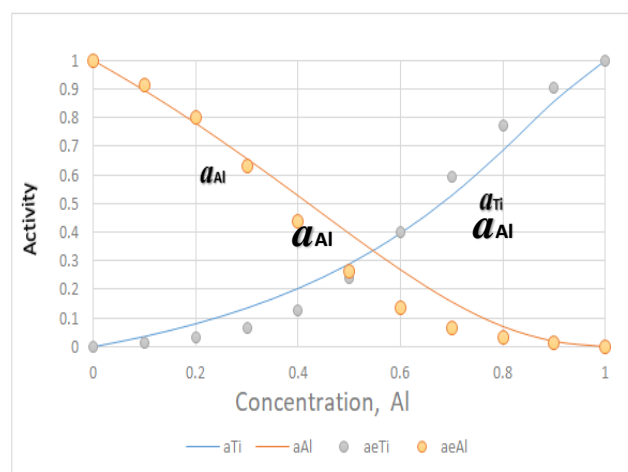


Fig 18 Calculated Activities of Compounds in Liquid Ti-Al Alloys at 2273 K Verse the Concentration of Al.

➤ Diffusion Coefficient D_M

Using equation (16) we calculated the diffusion coefficient for liquid Ti-Al alloys at temperatures of 1773 K- 2273 K respectively. The result are shown in Fig. 1.13 to Fig. 1.18. It can be deduced from the figure that the diffusion coefficient of liquid Ti-Al alloys are greater than unity > 1 , which suggests the presence of compound formation in the liquid Ti-Al alloys.

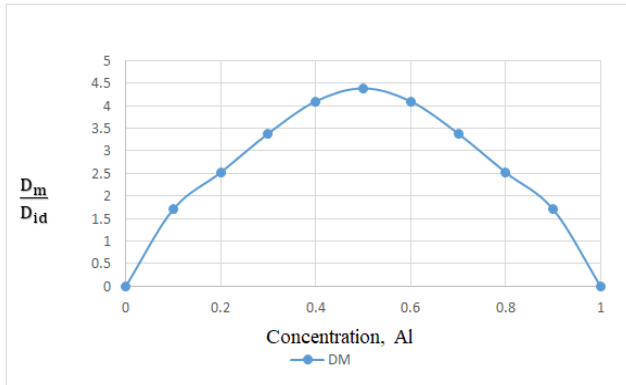


Fig 19 Diffusion Coefficient $\frac{D_M}{D_{id}}$ Verse Concentration at 1773 K. The Solid Line Represents Calculated Values. C_{Al} Is Concentration in the Alloy.

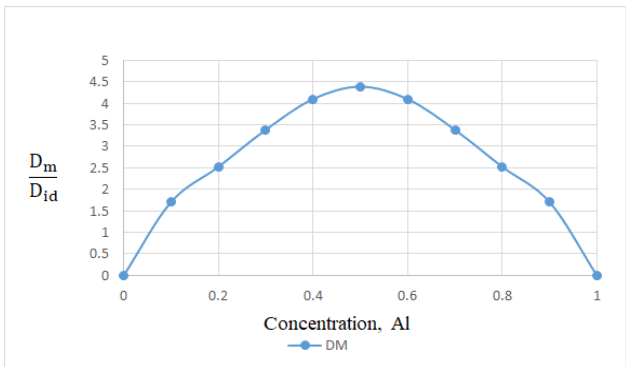


Fig 20 Diffusion Coefficient $\frac{D_M}{D_{id}}$ Verse Concentration at 1873 K. The Blue Line Represents Experimental Values. C_{Al} Is Concentration in the Alloy.

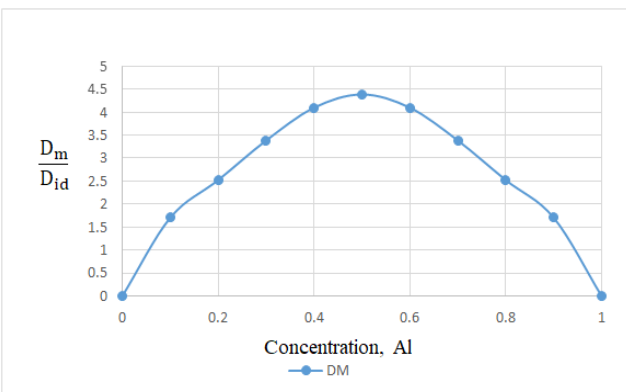


Fig 21 Diffusion Coefficient $\frac{D_M}{D_{id}}$ Verse Concentration at 1973 K. The Blue Line Represents Calculated Values. C_{Al} Is Concentration in the Alloy.

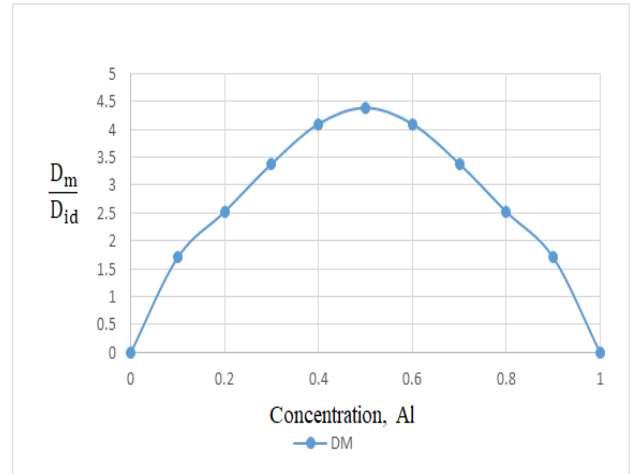


Fig 22 Diffusion Coefficient $\frac{D_M}{D_{id}}$ Verse Concentration at 2073 K. The Blue Line Represents Calculated Values. C_{Al} Is Concentration in the Alloy.

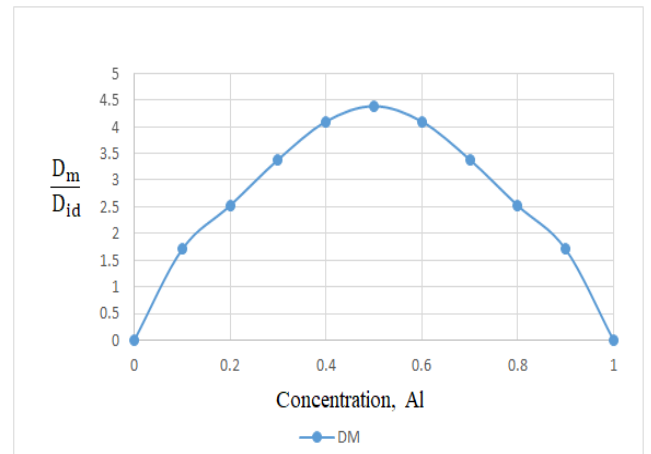


Fig 23 Diffusion Coefficient $\frac{D_M}{D_{id}}$ Verse Concentration at 2173 K. The Blue Line Represents Calculated Values. C_{Al} Is Concentration in the Alloy.

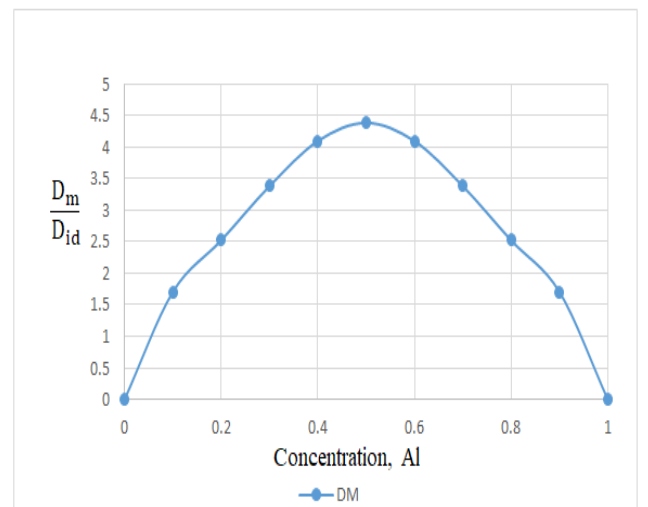


Fig 24 Diffusion Coefficient $\frac{D_M}{D_{id}}$ Verse Concentration at 2273 K. The Blue Line Represent Calculated Values. C_{Al} Is Concentration in the Alloy.

➤ *Chemical Short-Range-Order Parameter α_1*

Equation (15) was used to obtain the Warren-Cowley chemical short-range order parameter (α_1). The plot of this parameter versus concentration are presented in figure 4.19-4.24. The negative values of α_1 over the whole concentration range indicates the degree of ordering in Ti-Al alloys. Chemical short-range order α_1 is much less than 1 in Ti-Al alloys at respective temperatures.

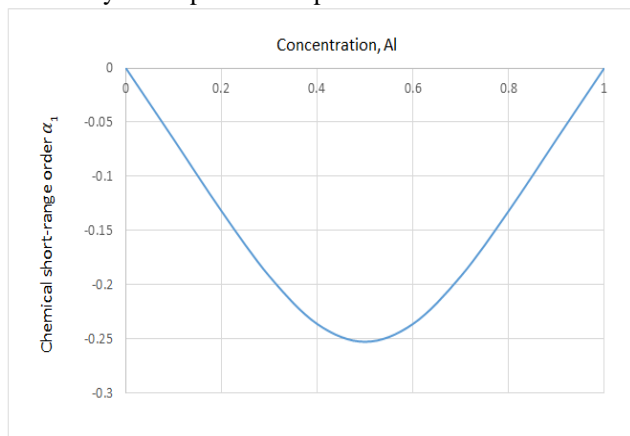


Fig 25 Chemical Short-Range Order Parameter Versus Concentration for Liquid Ti-Al Alloys at 1773 K

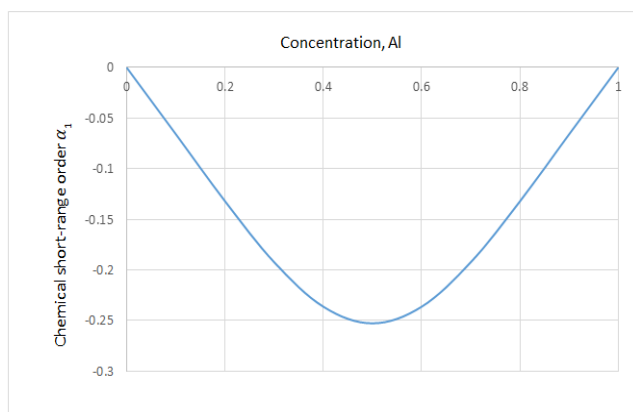


Fig 26 Chemical Short-Range Order Parameter Versus Concentration for Liquid Ti-Al Alloys at 1873 K.

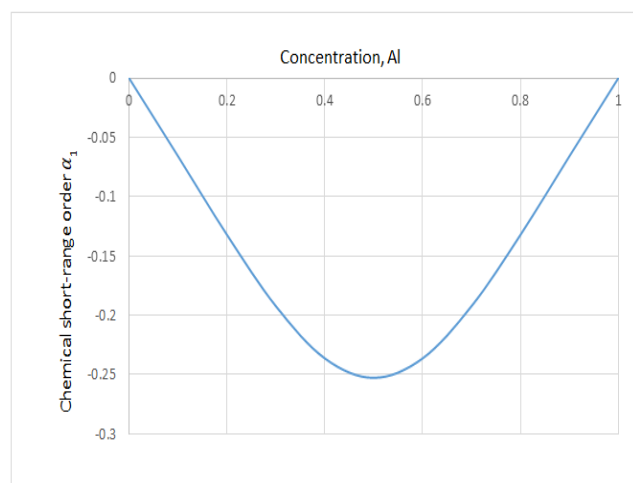


Fig 27 Chemical Short-Range Order Parameter Versus Concentration for Liquid Ti-Al Alloys at 1973 K

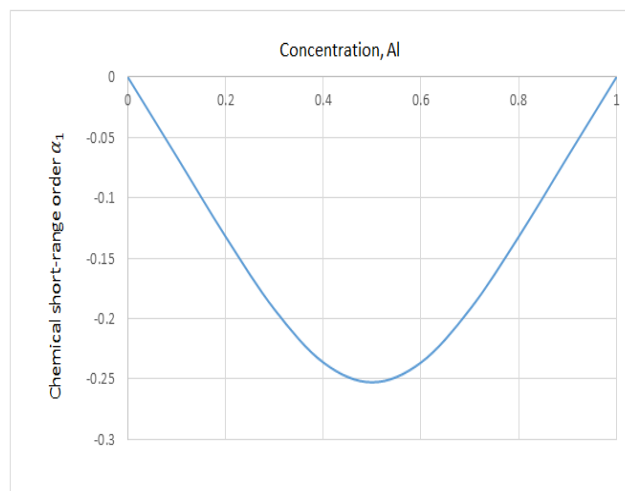


Fig 28 Chemical Short-Range Order α_1 Parameter Versus Concentration for Liquid Ti-Al Alloys at 2073 K

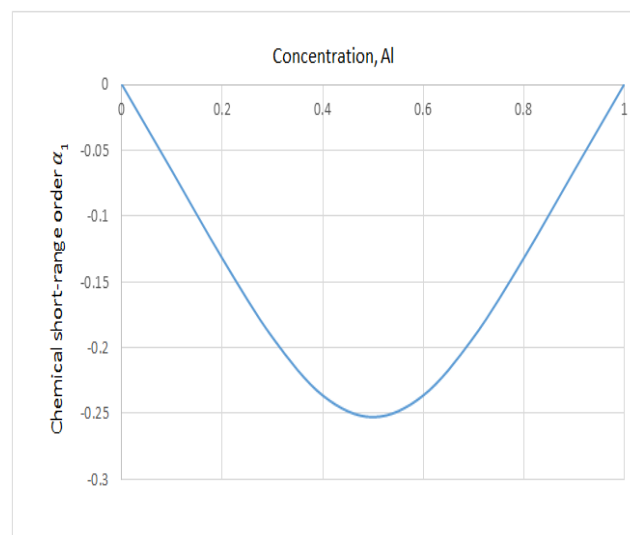


Fig 29 Chemical Short-Range Order Parameter Versus Concentration for Liquid Ti-Al Alloys at 2173 K

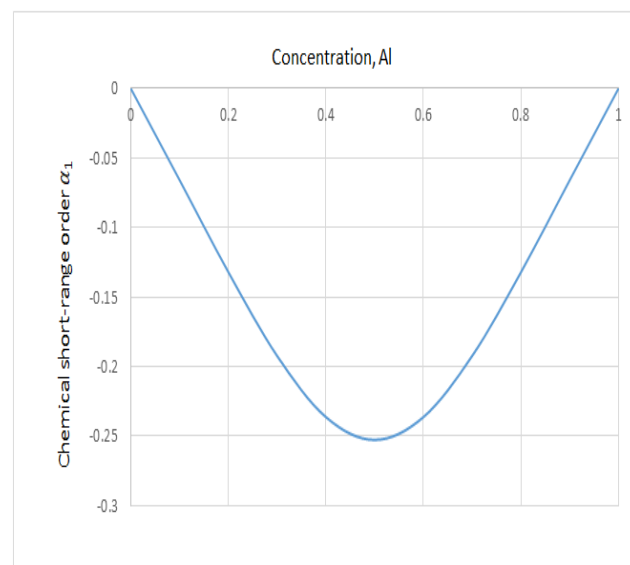


Fig 30 Chemical Short-Range Order Parameter Versus Concentration for Liquid Ti-Al Alloys at 2273 K

IV. CONCLUSION

This is a theoretical approach for the determination of the concentration – concentration fluctuation and chemical ordering in Ti-Al alloy at 1773 K, 1873 K, 1973 K, 2073 K, 2173 K, and 2273 K. The quasi-chemical model (QCAM) was used for the prediction of this work at the temperatures listed above with special emphasis on Gibbs free energy of mixing G_m , concentration- concentration fluctuation in the long-wavelength and $s_{cc}(0)$, chemical short-range order parameter α_1 , diffusion co-efficient D_M and activity coefficient ($\ln \gamma$).

The negative deviation from Raoult's law is noticed which indicates a noticeable tendency for chemical ordering in Ti-Al liquid alloys across the whole concentration range.

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