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Calculation of Darken Stability Functions of Six Binary Alloys

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Abstract

The thermodynamic model based on cluster of two atoms is considered with the view to obtaining Scc(0) and the darken stability function. Concentration-concentration fluctuation, Scc(0)of six binary molten alloys was successfully calculated. Literature data for the thermodynamic properties of these alloys are evaluated based on cluster of two atoms (A & B) or (B & A) for each system with the view to obtaining concentration-concentration fluctuation, Scc(0) enumerating the low order atomic correlation in the nearest neighbour shell of liquid binary alloys with the highlights of reciprocals of Scc(0) of these alloys. The values of Scc(0) for all the alloys throughout the entire concentration are positive and higher than the ideal solution values. The values of darken stability functionof Scc(0)for these alloys fall below the ideal darken stability function with exceptions in some alloys. The indication of the reciprocal of Scc(0) for all the alloys is in support of homocordination in the nearest neighbour shell. The Scc(0) and darken stability function of six binary alloys are presented.

Keywords: Concentration-concentration fluctuation, darken stability function,ordering energy; quasichemical model.

1.0 Introduction

This write-up focuses attention on an aspect of the article written by Singh [1] where concrete attention was not paid. Bhatia and Hargroove [2] had published an article on concentration-concentration fluctuation, Scc(0) and short range order parameter of some binary alloys but did not attach the darken stability functions of the alloys i.e. 1/Scc(0). Awe et al. [3] calculated experimental concentration-concentration fluctuation, Scc (0) using ordering energy from free energy of mixing and experimental activities of eleven binary alloys without attaching their darken stability functions. For these properties, data were generated using inputs such as ordering energy value, coordination number, melting temperature and Boltzmann constant. Thermodynamic expressions as functions of concentration fromquasichemical model were employed in the described programs for generating data for these thermodynamic properties.

Therefore, the observation write up focuses attention on the determination of ordering energy values of six out of eleven binary liquid alloys from values of deviations in Scc(0) [3]. These ordering energy values were used in the calculation of Scc(0) and 1/Scc(0) of six binary liquid alloys. Similar method for generating values was followed by inscribing programs which involves using inputs from Table 1 and the thermodynamic expressions made available by quasichemical model which matches experimental observations. The determined values of ordering energy are displayed in Table 1.

Table 1:Ordering energy (w) in eV of binary alloys

Alloy	Temperature(^oK)	Z	w_1(eV)	w_2(eV)	w_3(eV)
Ag-Ge	1250	10.0	0.0288	0.0281	0.0275
Al-Au	1338	10.0	0.0980	0.1127	0.1130
Bi-Cd	773	10.0	0.0210	0.0206	0.0206
Cd-Ga	700	10.0	0.1133	0.1149	0.1166
Cu-Pb	1473	10.0	0.2260	0.2176	0.2243
Na-Pb	700	10.0	0.0569	0.0558	0.0566

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2.0 Theory

The calculation of Scc(0) is often attracting attachment like $\left(S_{cc}(0)\right)^{-1}$. This view provides additional facts thatshed light on alloying behavior in terms of compound formation [4],self coordination, phase segregation and complex concentration formation [5, 6]. Thermodynamically, the relationship between short range order parameter, SROand concentration-concentration fluctuation,Scc(0) had been sighted in the literature. Moreover, between G_m and 1/Scc(0) it is given below. The following thermodynamic expressions are from quasichemical model.

Quasichemical expressions for various thermodynamic functions

Free Energy of Mixing G_m;

$$G_m = G_m^{id} + G_m^{xs} \quad (1)$$

$$G_m^{id} = RT\{c\ln c + (1-c)\ln(1-c)\} \quad (2)$$

$$G_m^{xs} = RT\{c\ln\gamma_A + (1-c)\ln\gamma_B\} \quad (3)$$

Where G_m^{id} and G_m^{xs} are ideal and excess free energy of mixing.R is molar gas constant, T is temperature, c & 1-crepresent the concentration of A and B atoms in the alloy respectively. γ_A *and* γ_B arethe activity coefficients and stand for

$$\gamma_A = \left(\frac{\beta - 1 + 2c}{c(1+\beta)}\right)^{\frac{z}{2}} \quad (4)$$

$$\gamma_B = \left(\frac{\beta + 1 - 2c}{(1-c)(1+\beta)}\right)^{\frac{z}{2}} \quad (5)$$

where

$$\beta = \left(1 + 4c(1-c)(\eta^2 - 1)\right)^{\frac{1}{2}}, \quad (6)$$

Where $\eta = \exp(w/zk_BT)$ (7)

$$a_A = c\left(\frac{\beta - 1 + 2c}{c(1+\beta)}\right)^{\frac{z}{2}} \quad (8)$$

$$a_B = (1-c)\left(\frac{\beta + 1 - 2c}{(1-c)(1+\beta)}\right)^{\frac{z}{2}} \quad (9)$$

η and β are thermodynamic parameters which are interwoven.

The relationship between darken stability function; 1/Scc(0) and free energy of mixing $G_m$is given as

$$\left(s_{cc}(0)\right)^{-1} = \frac{1}{N k_B T}\left(\frac{\partial^2 G_m}{\partial c^2}\right)_{T,P,N} \quad (10)$$

Also the relationship between activity (a_A or a_B) and darken stability function is given as

$$\left(S_{cc}(0)\right)^{-1} = \frac{1}{(1-c)a_A}\left(\frac{\partial a_A}{\partial c}\right)_{T,P,N} = \frac{1}{C a_B}\left(\frac{\partial a_B}{\partial(1-c)}\right)_{T,P,N} \quad (11)$$

From Equations (1), (2),(3) to (11) N is the total number of atoms in the alloy, k_B is the Boltzmann constant, T is the temperature, p is the pressure andZ is coordination number of the alloys. The terms a_A and a_B in equation (11) represent the activities of atom A and atom B respectively.

The expression for the ideal darken stability function is given by

$$\left(S_{cc}^{id}(0)\right)^{-1} = \frac{1}{c(1-c)} \quad (12)$$

Scc(0)1, 1/Scc(0)1, Scc(0)2 and1/Scc(0)2are from experimental activities,Scc(0)3 and 1/Scc(0)3 are from experimental free energy of mixing.

3.0 Results and Discussion

Table 2: Calculated experimental concentration-concentration fluctuation and darken stability function of Cu-Pb alloy using experimental activities and free energy of mixing. Ccuis the concentration of copper in the alloy.

Ccu	Scc(0)1	Scc(0)2	Scc(0)3	Scc(0)id	1/Scc(0)1	1/Scc(0)2	1/Scc(0)3	1/Scc(0)id
0.0	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.0000
0.1	0.124	0.255	0.107	0.09	8.0645	3.9216	9.3458	11.1111
0.2	0.302	0.257	0.312	0.16	3.3113	3.8911	3.2051	6.2500
0.3	0.582	0.643	0.620	0.21	1.7182	1.5552	1.6129	4.7619
0.4	0.968	1.031	0.832	0.24	1.0331	0.9699	1.2019	4.1667
0.5	1.357	1.179	1.317	0.25	0.7369	0.8482	0.7593	4.0000
0.6	1.686	1.783	2.672	0.24	0.5931	0.5609	0.3743	4.1667
0.7	1.625	1.797	1.435	0.21	0.6154	0.5565	0.6969	4.7619
0.8	0.552	0.479	0.416	0.16	1.8116	2.0877	2.4038	6.2500
0.9	0.089	0.105	0.171	0.09	11.2360	9.5238	5.8480	11.1111
1.0	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.0000

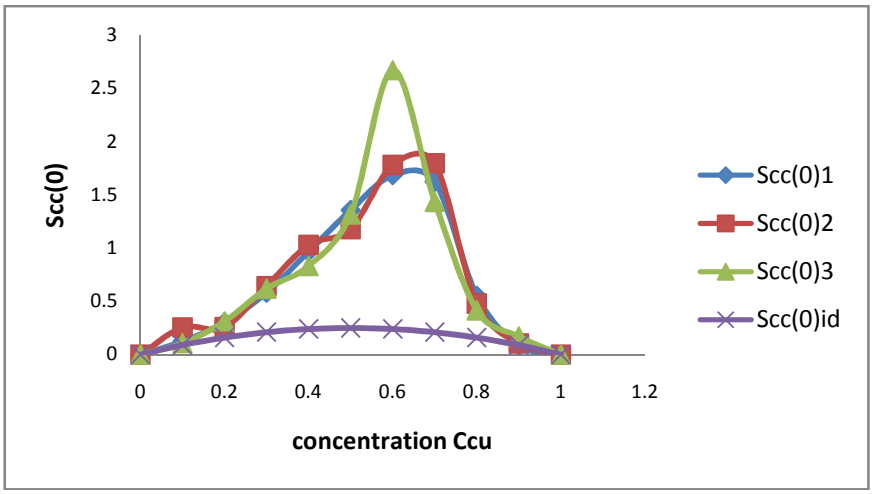

Fig. 1: Graph of concentration-concentration fluctuation Scc(0) versus concentration C_{Cu}

In Figure 1 and Table 2it is observed that, in the range of compositions $0 \le Ccu \le 0.5$ and $0.7 \le Ccu \le 1.0$, the $Scc(0)\exp$ obtained via each of the three methods indicates the alloy is homocoordinated because its values are greater than the ideal values, although in terms of magnitude, the results Scc(0)1 and Scc(0)2 are closer than Scc(0)3. In addition, at the composition Ccu= 0.6, the magnitude of Scc(0)3 is the largest of the three. Hence in the region 0<Ccu<1.0 one can say that the usual Equations (10) and (11) give rise to the expected results in the Cu-Pb liquid alloy. It is also observed from this Figure and the Table that Scc(0)1 appears to be more reliable than the results from Scc(0)2 and Scc(0)3.

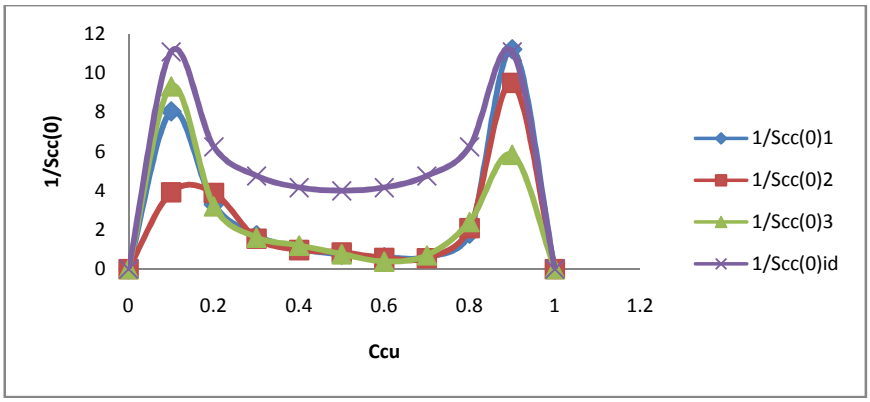

Fig. 2: Graph of darken stability function versus concentration C_{Cu}

In Figure 2 and Table 2 it is observed that, in the range of composition $0.3 \le Ccu \le 0.7$, the 1/Scc(0)exp obtained via each of the three methods is in perfect agreement. In the region $0 \le Ccu \le 0.2$, 1/Scc(0)3 is closest to the ideal darken stability function (1/Scc(0)id) and in the region $0.8 \le Ccu \le 1.0$, 1/Scc(0)1 is the closest to the ideal darken stability function.

Table 3:Calculated experimental concentration-concentration fluctuation and darken stability function of Na-Pb alloy using experimental activities and free energy of mixing. C_{Na} is the concentration of sodium in the alloy.

C_{Na}	Scc(0)1	Scc(0)2	Scc(0)3	Scc(0)id	1/Scc(0)1	1/Scc(0)2	1/Scc(0)3	1/Scc(0)id
0.0	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.0000
0.1	0.218	0.112	0.072	0.09	4.5872	8.9286	13.8889	11.1111
0.2	0.269	0.264	0.251	0.16	3.7175	3.7879	3.9841	6.2500
0.3	0.349	0.362	0.360	0.21	2.8653	2.7624	2.7778	4.7619
0.4	0.428	0.423	0.426	0.24	2.3364	2.3640	7.9365	4.1667
0.5	0.455	0.448	0.453	0.25	2.1978	2.2321	2.2075	4.0000
0.6	0.438	0.437	0.439	0.24	2.2831	2.2883	2.2779	4.1667
0.7	0.382	0.391	0.385	0.21	2.6178	2.5575	2.5974	4.7619
0.8	0.290	0.608	0.290	0.16	3.4483	1.6447	3.4483	6.2500
0.9	0.161	0.179	0.153	0.09	6.2111	5.5866	6.5359	11.1111
1.0	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.0000

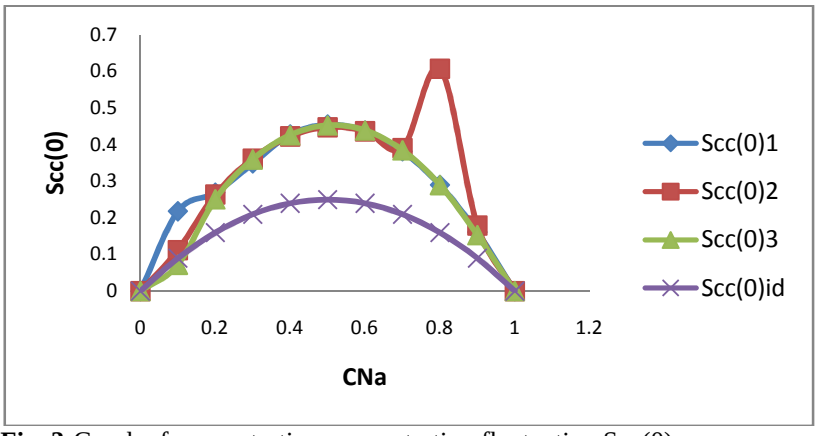

Fig. 3: Graph of concentration-concentration fluctuation Scc(0) versus concentration C_{Na}

In Figure 3 and Table 3 based on observation, in the range of composition $0.2 \le CNa \le 0.7$, the $Scc(0)\exp$ obtained via each of the three methods are in perfect agreement and indicate the alloy to be homocoordinated because its values are greater than the ideal values. Although in terms of magnitude, the results Scc(0)1 and Scc(0)3 are closer than Scc(0)2 in the range $0.7 \le CNa \le 0.9$. In addition, at the composition C_{Na}= 0.8, the magnitude of Scc(0)2 is the largest of the three. Hence in the region $0<C_{Na}<1.0$it is observed that the usual Equations (10) and (11) give rise to the expected results in the Na-Pb liquid alloy. It is also observed from this Figure and Table that Scc(0)3 appears to be more reliable than the results from Scc(0)1 and Scc(0)2.

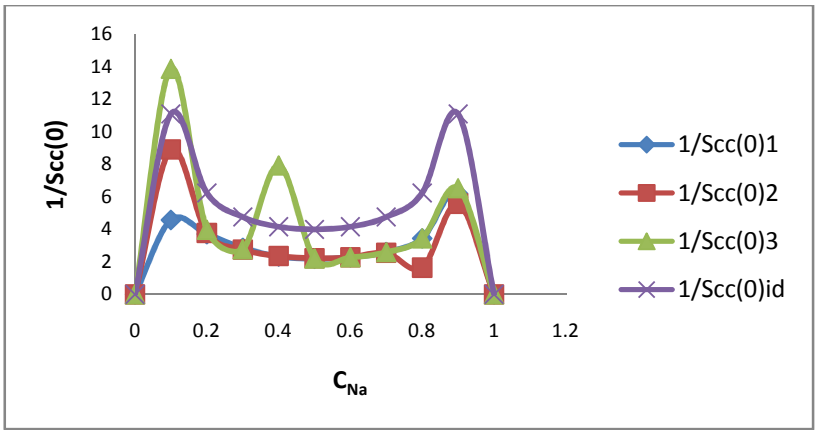

Fig. 4: Graph of darken stability function versus concentration C_{Na}

In Figure 4 and Table 3 in the range of composition $0 \le CNa \le 1.0$, the 1/Scc(0)exp obtained via each of the three methods is not in perfect agreementbecause of the regions $0.3 \le CNa \le 0.5$ and $0 \le CNa \le 0.2$. 1/Scc(0)3 at C_{Na}= 0.1 is above the ideal darken stability function and in the region $0.7 \le CNa \le 0.9$ 1/Scc(0)2 and 1/Scc(0)3 are below the darken stability function for the remain two approaches.

Table 4: Calculated experimental concentration-concentration fluctuation and darken stability function of Al-Au alloy using experimental activities and free energy of mixing. C_{Al} is the concentration of Aluminum in the alloy.

C_{Al}	Scc(0)1	Scc(0)2	Scc(0)3	Scc(0)id	1/Scc(0)1	1/Scc(0)2	1/Scc(0)3	1/Scc(0)id
0.0	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.0000
0.1	0.180	0.167	0.161	0.09	5.5556	5.9880	6.2112	11.1111
0.2	0.313	0.298	0.292	0.16	3.1949	3.3557	3.4247	6.2500
0.3	0.372	0.391	0.392	0.21	2.6882	2.5575	2.5510	4.7619
0.4	0.412	0.448	0.453	0.24	2.4272	2.2321	2.2075	4.1667
0.5	0.422	0.468	0.469	0.25	2.3697	2.1368	2.1322	4.0000
0.6	0.403	0.445	0.440	0.24	2.4814	2.2472	2.2727	4.1667
0.7	0.251	0.348	0.367	0.21	2.8490	2.8736	2.7248	4.7619
0.8	0.268	0.287	0.262	0.16	3.7313	3.4843	3.8168	6.2500
0.9	0.151	0.187	0.137	0.09	6.6225	5.3476	7.2993	11.1111
1.0	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.0000

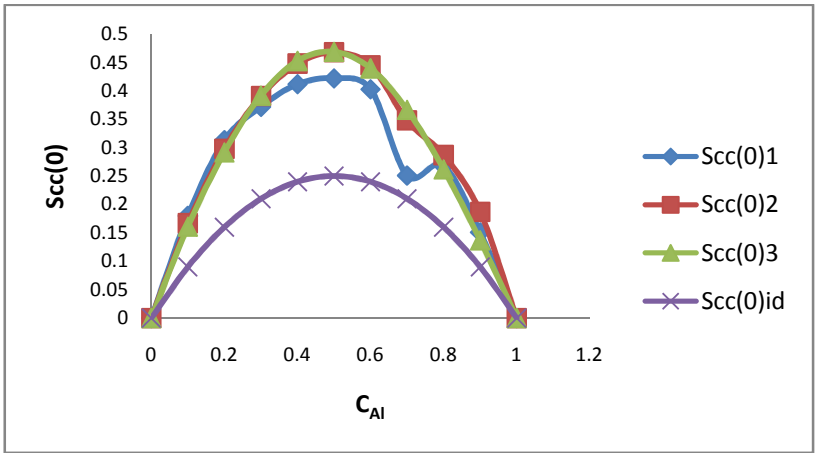

Fig. 5: Graph of concentration-concentration fluctuation Scc(0) versus concentration C_{Al}

In Figure 5 and Table 4 in the range of compositions $0 \le CAl \le 0.5$ and $0.7 \le CAl \le 1.0$, the $Scc(0)\exp$ obtained via each of the three methods indicates that the alloy is homocordinated because its values are greater than the ideal values, although in terms of magnitude, the results Scc(0)2 and Scc(0)3 are closer than Scc(0)1. In addition, at the composition C_{Al}= 0.7, the magnitude of Scc(0)1 is the smallest of the three. Hence, in the region $0<C_{Al}<1.0$ It is also observed from this Figure and Table that Scc(0)2&3 appears to be more reliable than the results from Scc(0)1.

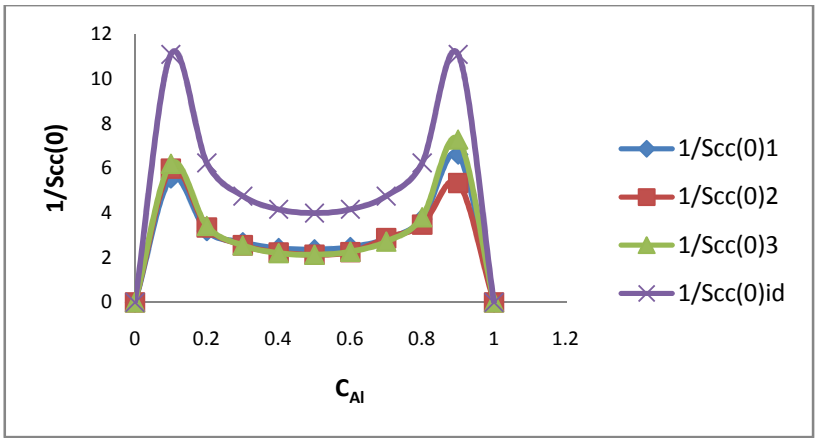

Fig. 6: Graph of darken stability function against concentration C_{Al}

In Figure 6 and Table 4 it is observed that, in the range of composition $0.3 \le C \le 0.7$, the1/Scc(0)exp obtained via each of the three methods is in perfect agreement. In the entire region, Scc(0)1, Scc(0)2 and Scc(0)3 are below the ideal darken stability function and Scc(0)3 is closest to the ideal darken stability function than the remain two approaches.

Table 5: Calculated experimental concentration-concentration fluctuation and darken stability function of Ag-Ge alloy using experimental activities and free energy of mixing. C_{Ag} is the concentration of silver in the alloy.

C_{Ag}	Scc(0)1	Scc(0)2	Scc(0)3	Scc(0)id	1/Scc(0)1	1/Scc(0)2	1/Scc(0)3	1/Scc(0)id
0.0	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.0000
0.1	0.112	0.110	0.124	0.09	8.9286	9.0909	8.0645	11.1111
0.2	0.193	0.191	0.189	0.16	5.1813	5.2356	5.2910	6.2500
0.3	0.262	0.261	0.260	0.21	3.8168	3.8314	3.8462	4.7619
0.4	0.303	0.305	0.305	0.24	3.3003	3.2787	3.2787	4.1667
0.5	0.288	0.287	0.286	0.25	3.4722	3.4843	3.4965	4.0000
0.6	0.222	0.222	0.219	0.24	4.5045	4.5045	4.5662	4.1667
0.7	0.153	0.155	0.152	0.21	6.5360	6.4516	6.5789	4.7619
0.8	0.104	0.102	0.105	0.16	9.6154	9.8039	9.5238	6.2500
0.9	0.076	0.086	0.076	0.09	13.1579	11.6279	13.1579	11.1111
1.0	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.0000

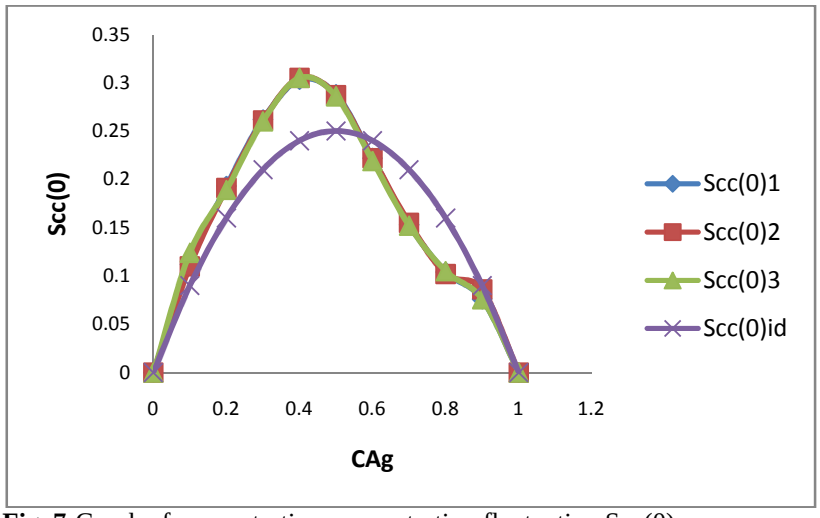

Fig. 7: Graph of concentration-concentration fluctuation Scc(0) versus concentration C_{Ag}

As shown in Table 5 and Figure 7, there is a reasonable level of agreement between the three methods of calculating Scc(0)exp, both in the region $0 \le CAg \le 0.6$ where the segregating nature is exhibited, and in the region $0.6 \le CAg \le 1.0$ where short range order quality is displayed. Hence, it can be said that the usual Equations (10) and (11) lead to the expected results throughout the entire composition of Ag in the Ag-Ge Alloy.

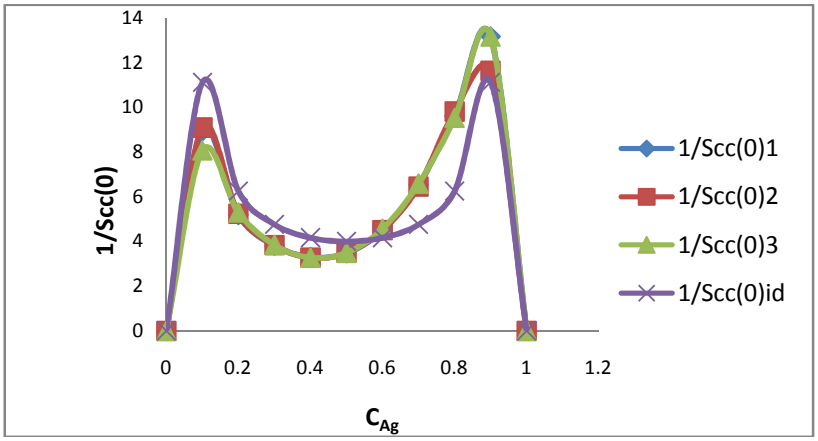

Fig. 8: Graphof darken stability function versus concentration C_{Ag}

In Fig.8, with the exception in C_{Ag} = 0.1 and 0.9 the darken stability function the region $0.2 < CAg < 0.8$ via the three approaches are in perfect agreement only that they do not agree perfectly with ideal darken stability function.

Table 6: Calculated experimental concentration-concentration fluctuation and darken stability function of Bi-Cd alloy using experimental activities and free energy of mixing. C_{Bi} is the concentration of bismuth in the alloy.

C_{Bi}	Scc(0)1	Scc(0)2	Scc(0)3	Scc(0)id	1/Scc(0)1	1/Scc(0)2	1/Scc(0)3	1/Scc(0)id
0.0	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.0000
0.1	0.089	0.090	0.069	0.09	11.2360	11.1111	14.4928	11.1111
0.2	0.166	0.167	0.170	0.16	6.0241	5.9880	5.8824	6.2500
0.3	0.250	0.251	0.251	0.21	4.0000	3.9841	3.9841	4.7619
0.4	0.296	0.296	0.297	0.24	3.3784	3.3783	3.4364	4.1667
0.5	0.296	0.295	0.295	0.25	3.3784	3.3898	3.3898	4.0000
0.6	0.256	0.257	0.257	0.24	3.9063	3.8895	3.8911	4.1667
0.7	0.204	0.204	0.208	0.21	4.9020	4.9020	4.8077	4.7619
0.8	0.156	0.151	0.152	0.16	6.4103	6.6667	6.5789	6.2500
0.9	0.065	0.082	0.061	0.09	15.3846	12.1951	16.3934	11.1111
1.0	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.0000

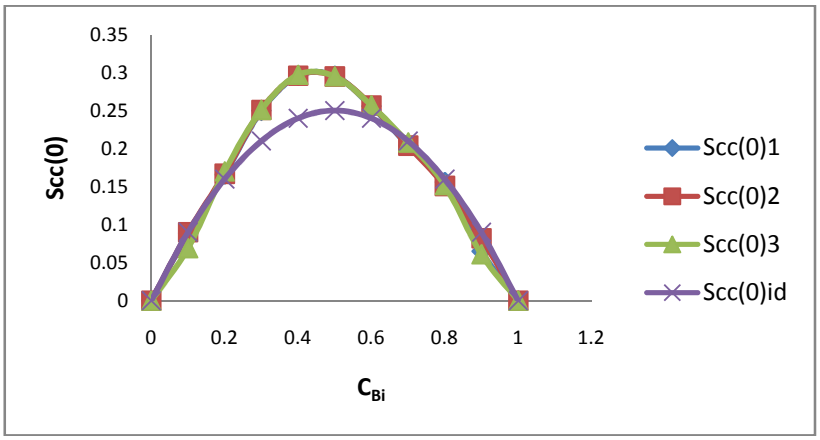

Fig. 9: Graph of concentration-concentration fluctuation Scc(0) versus concentration C_{Bi}

A comparison of the results of Scc(0)exp obtained for Bi-Cd via the three methods as shown in Figure 9 and Table 6 indicates that Equations (10) and (11) give identical results in Bi-Cd throughout the entire composition of Bi in Bi-Cd system. The slightdisparities in the values Scc(0)1,Scc(0)2 and Scc(0)3 at C_{Bi}=0.9 in opinion are ignorable.The values of Scc(0) for the three approaches are also very close to the ideal values except at C_{Bi} = 0.3, 0.4, 0.5 and 0.6 where Scc(0) via the three methods is above ideal solution values i.e. Scc(0)id [7].

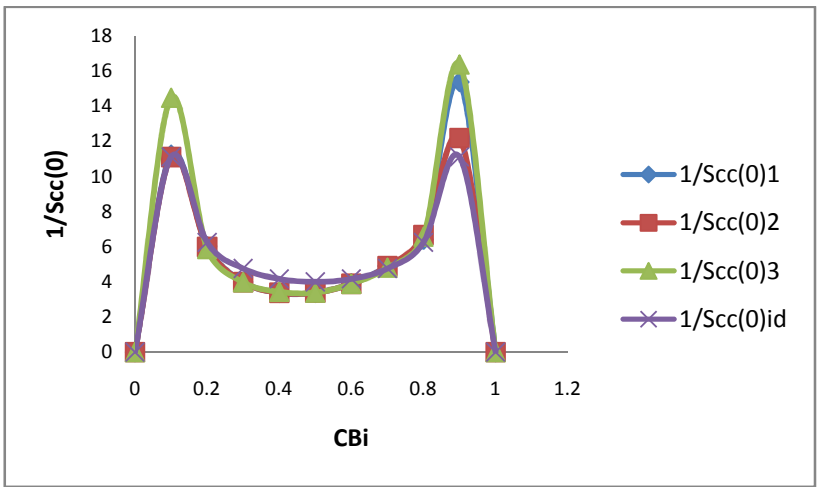

Fig. 10: Graph of darken Stability function versus concentration C_{Bi}

In the region $0.2 \leq CBi \leq 0.8$, the darken stability function via the three methods are in perfect agreement which indicates ordering in these liquid alloys[8]. 1/Scc(0)3 is above ideal mixture only at CBi = 0.1 and the three 1/Scc(0)1, 1/Scc(0)2 and 1/Scc(0)3 are above the ideal darken stability function only at C_{Bi} = 0.9.

Table 7: Calculated experimental concentration-concentration fluctuation and darken stability function of Cd-Ga alloy using experimental activities and free energy of mixing. Ccd is the concentration of cadmium in the alloy.

Ccd	Scc(0)1	Scc(0)2	Scc(0)3	Scc(0)id	1/Scc(0)1	1/Scc(0)2	1/Scc(0)3	1/Scc(0)id
0.0	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.0000
0.1	0.170	0.151	0.207	0.09	5.8824	6.6225	4.8309	11.1111
0.2	0.437	0.450	0.452	0.16	2.2883	2.2222	2.2124	6.2500
0.3	0.976	0.930	0.892	0.21	1.0246	1.0753	1.1211	4.7619
0.4	1.718	1.720	1.486	0.24	0.5821	0.5814	0.6729	4.1667
0.5	1.745	1.886	1.779	0.25	0.5731	0.5302	0.5621	4.0000
0.6	1.143	1.126	1.235	0.24	0.8749	0.8810	0.8097	4.1667
0.7	0.638	0.620	0.642	0.21	1.5674	1.6129	1.5576	4.7619
0.8	0.347	0.363	0.342	0.16	2.8818	2.7548	2.9240	6.2500
0.9	0.228	0.212	0.203	0.09	4.3860	4.7170	4.9261	11.1111
1.0	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.0000

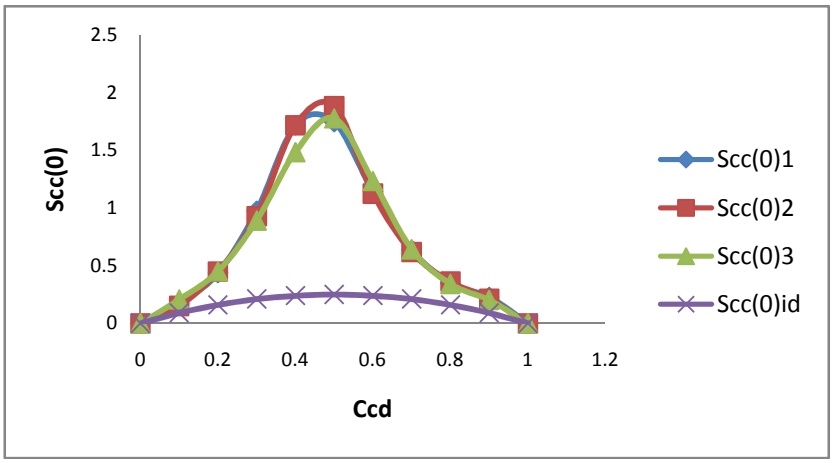

Fig. 11: Graph of concentration-concentration fluctuation Scc(0) versus concentration C_{cd}

As shown in Table7 and Figure 11, we observe that in Cd-Ga, except in the composition Ccd=0.4 and Ccd= 0.5, there is good level of agreement between the results obtained via the three methodsusing the model of approach [9]. This implies the equations (10) and (11) hold significantly in virtually the entire composition of Cd in Cd-Ga alloy.

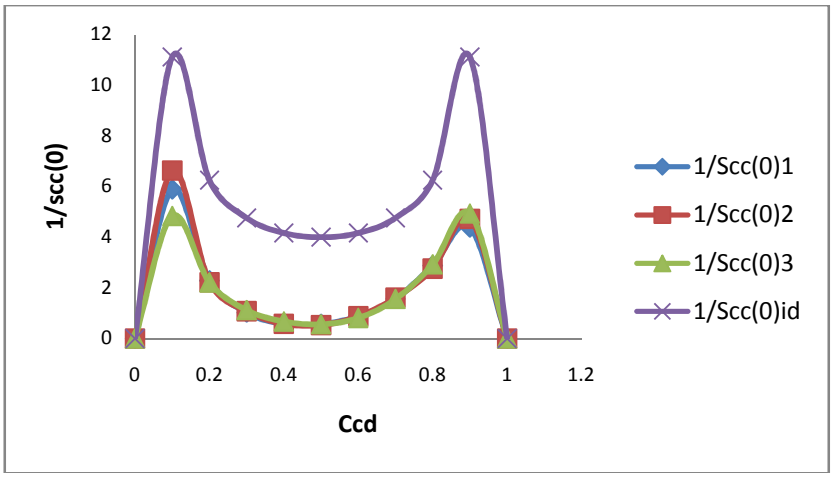

Fig. 12: Graph of darken stability function versus concentration C_{cd}

In Fig. 12 the darken stability functions for the three approaches are in good agreement except at Ccd =0.1. The three 1/Scc(0) fall below the ideal darken stability function. This is in support of homocoordination. The success of Eqs. (10) and (11) depends upon the true knowledge of the ordering energy w [10].

4.0 Concluding Remarks

This study has revealed, contrary to the belief that the Scc(0)exp of liquid binary alloys can be computed via the experimental activities and experimental free energy of mixing using Equations (10) and (11) within a given set of data for experimental a_A , a_B and $G_M$from common source (as different sets of data for a system may not be available, or if available may not necessarily agree throughout the entire composition) that

(1) There are only two instances where the Scc(0)exp obtained via each of the three methods agrees completely throughout the entire composition (these are Ag-Ge and Bi-Cd).

(2) There are four instances where Scc(0)exp obtained via each of the three methods have partial agreement instead of complete agreement (these are Al-Au, Cd-Ga, Cu-Pb, and Na-Pb in Figures 1, 3, 5 and 11 respectively). The equality sign in Equations (10) and (11) is suggested equivalent sign.

(3) There are only two instances where the 1/Scc(0) obtained via each of the three methods agrees almost throughout the entire composition (these are Al-Au and Cd-Ga)

In Conclusion, it is recommend that whenever Scc(0)exp is needed, the common approach of obtaining it via experimental activity a_A should be seen as the best reliable method among the three.

5.0 References

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