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# Temperature dependence of entropy of mixing of liquid alkali alloys

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### Abstract

A semi-empirical approach has been considered to study the temperature dependence of entropy of mixing, ( $S^M$ ), for various alkali-alkali alloys using hard-sphere model. The most important physical parameters occurring here is the hard-sphere diameter ( $\sigma$ ) and the packing fraction ( $\eta$ ). For pure liquid metals, this is usually determined empirically from the observed entropy as a function of temperature which in turn are utilised to compute  $S^M$  for Na-K, K-Rb, Na-Rb, Na-Cs, Rb-Cs and K-Cs alloys as a function of concentration at five different temperature ranging from 400°K-800°K. The study reveals that entropy of mixing for Na-K, Na-Rb and K-Rb systems decreases with increasing temperature. But the result for Cs-based alloys exhibit a mixed behaviour.

**Keywords:** Alkali alloys; Semi-empirical approach; Entropy; Helmholtz free energy, Boltzmann Constant; Packing fraction.

### 1. Introduction

Though enormous experimental data exist on entropy of pure liquid metals and alloys [1], the theoretical works lag behind. Recently the hard-sphere (HS) model has been widely used to remedy this lack. The theoretical study of the entropy of mixing is of current interest to unlock the secrets of the structure of liquid alloys on the basis of experimental observations. The most important and basic ingredients are the hard sphere diameter ( $\sigma$ ) and the packing fraction ( $\eta$ ). Several workers [2-5] have computed the HS parameters by minimising the free energy of the system through first principle estimation. In the present work, a semi-empirical technique, without undergoing a rigorous first principle calculation attempt has

been made to determine the HS parameters from the observed entropy of pure liquid metals[6], which in turn are utilised to compute the entropy of mixing for Na-K, Na-Rb, K-Rb, Na-Cs, Rb-Cs, and K-Cs alloys [7] as a function of concentration and temperature. These systems are preferred as their densities as a function of composition are experimentally available [8] and also there are typical representation of simple system where the sizes of constituent atoms differ from a factor of 1 to 3. Out of these, Cs-based binary molten of alloys are of particular interest for both theoretical [9-12] and experimental workers [13-15].

## 2. Theory

We consider a simple binary liquid alloy with components of atomic concentration  $C_1$  and  $C_2$  comprising  $C_1N$  hard spheres with diameter  $\sigma_1$  of species 1 and  $C_2N$  with diameter  $\sigma_2$  of species 2. Following Umar et. al., the expression for the entropy of hard sphere mixture can be expressed as

$$S = S_{gas} + S_c + S_y + S_m \quad (1)$$

where  $S_{gas}$  is the ideal gas entropy,  $S_c$  represents the ideal entropy of mixing,  $S_y$  corresponds to the packing density and  $S_m$  is the entropy contribution due to mismatch of the hard sphere diameters  $\sigma_1$  and  $\sigma_2$ . Explicit expression for the various contributions are

$$\frac{S_{gas}}{K_B} = \frac{5}{2} + \sum_n \left\{ \Omega \left( \frac{m_1^{c_1} m_2^{c_2} K_B T}{2f \hbar^2} \right)^{3/2} \right\} \quad (2)$$

$$\frac{S_c}{K_B} = -(C_1 \ln C_1 + C_2 \ln C_2) \quad (3)$$

$$\frac{S_y}{K_B} = -(\eta - 1)(\eta + s) \quad (4)$$

$$\frac{S_m}{K_B} = A C_1 C_2 + (\eta_2 - \eta_1)^2 \quad (5)$$

The expressions used for  $S_y$  and  $S_m$  which are derived by Umar et. al.[16] from the Helmholtz free energy formulae of Mansoori et. al.[17], the latter work itself providing an accurate analytical fit to the computer Simulation data.

Here  $\eta = (1 - \alpha)^{-1}$  and  $y = \frac{f}{6}(y_1 \tau_1^3 + y_2 \tau_2^3)$  is the packing fraction.  $\tau$  is the atomic volume of an

alloy for given concentration  $C_1$  and  $C_2$ , ( $C_1 + C_2 = 1$ ).  $m_1$  and  $m_2$  are atomic masses of the constituent species  $n_1 (= c_1 / \tau)$  and  $n_2 (= c_2 / \tau)$  are partial number densities.  $K_B$  is the Boltzmann Constant. In Eq. (5),

$$A = [\alpha(\alpha-1) - Pn\alpha](y_1 + y_2) + 3(\alpha-1)y_1 \quad (6)$$

where

$$y_1 = (\sigma_1 + \sigma_2) \sigma^{-3} \quad (7)$$

$$y_2 = \sigma_1 \sigma_2 (C_1 \sigma_1 + C_2 \sigma_2) \sigma^{-6} \quad (8)$$

where  $\sigma = (C_1 \sigma_1^3 + C_2 \sigma_2^3)^{1/3}$  is the hard-sphere diameter of the alloy. Following Eq.(1), the entropy of mixing for a binary alloy can be written as.

$$\Delta S^M = \Delta S_{gas} + \Delta S_1 + S_2 + S_c \quad (9)$$

with

$$\Delta S_{gas} = S_{gas}^{alloy} - C_1 S_{gas}^{(1)} - C_2 S_{gas}^{(2)} \quad (10)$$

$$\Delta S_y = S_y^{alloy} - C_1 S_y^{(1)} - C_2 S_y^{(2)} \quad (11)$$

Superscript 1 and 2 in Eqs. (10) and (11) refer to pure species 1 and 2 respectively. It is clear that for a given composition and volume of alloy, the entropy expressions contain two unknown parameters namely, hard sphere diameter  $\sigma_1$  and  $\sigma_2$ . The packing fraction  $y_i$  is easily derivable from the value of  $\sigma_i$ . In order to evaluate the value of  $y_i$  at different temperatures, we reduce the expression (2) to (5) to pure elements. For the corresponding pure liquid metals, we merely use  $C_1 = 1, C_2 = 0$  and  $C_2 = 1, C_1 = 0$  above.

The formalism is much simplified and, in particular,  $S_c = 0$  and  $S = 0$  in Eq.(1). Thus

$$S^i = S_{gas}^i - S_y^i \quad (i=1,2) \quad (12)$$

with

$$\frac{S^i}{K_B} = \frac{5}{2} + \ell_n \left\{ \Omega_i \left( \frac{m^i K_B T}{2f \hbar^2} \right)^{3/2} \right\} \quad (13)$$

$$\frac{S^i}{K_B} = \ell_n (1 - y_i) + \{1 - (1 - y_i)^{-2}\} \quad (14)$$

where  $y_i \left( = \frac{2f \hbar^2}{\Omega_i} \right)$  is the packing fraction of pure liquid materials.

It may be mentioned that the well-known Carnahan and Starling [18-22] formula for the entropy of pure element can be obtained from Eq. (14) by expanding  $\ln (1 - y_i)$  and retaining only the terms up to  $y_i^2$ .

The packing fraction and hence the hard sphere diameter has been determined at a given temperature by using Eq. (12) and taking the observed values of entropy from Hultgren et. al., The appropriate density of liquid metals required at a given temperature have been taken from literature [8].

### 3. Results and Discussion

In Table 1. We have listed the computed values of  $\alpha$  and  $\beta$  for Na, K, Rb and Cs at temperatures 400°K, 500°K, 600°K, 700°K and 800°K along with the available experimental values of entropy and density. The result shows that the values of  $\alpha$  vary from metal to metal. The maximum value is obtained for Na and the minimum value is for Cs. The values of  $\beta$  also depend on temperature. As the temperature increases,  $\beta$  decreases. The values of  $(d\alpha/dT)$  for different alkali metals are of the order of:

$$(d\alpha/dT)_{Na} = -2.4 \times 10^{-4} (\text{°K})^{-1}, (d\alpha/dT)_K = -2.7 \times 10^{-4} (\text{°K})^{-1}, (d\alpha/dT)_{Rb} = -2.9 \times 10^{-4} (\text{°K})^{-1} \text{ and } (d\alpha/dT)_{Cs} = -2.9 \times 10^{-4} (\text{°K})^{-1}.$$

Though, these coefficients are very small, but they may affect the properties which are sensitive to temperature. The entropies of the pure alkali metals,  $S$ , are determined by adding ideal gas term ( $S_{gas}$ ) and the packing density term  $S$  through Eq. (12). The former is two to four times greater in magnitude and opposite in sign to  $s$  for all the metals under consideration. The basic parameters required in the calculation are taken from Table 1.

Hard sphere parameters for pure elements as reported in Table 1, have also been utilised to compute the various contribution i.e.  $S_{gas}$ ,  $S_c$ ,  $S$  and  $S$  and  $S$  respectively from Eqs. (2) to (5) for alloys of interest. The relevant data for the various contribution to the entropy for alloys are listed in Tables 2 to 7 and hence  $S^M$  can easily be evaluated through Eq. (9) for these systems.

The computation of the various contribution to entropy are very important to understand the gross behaviour of alkali-alkali alloys. First we note that the absolute value of  $|S_{gas}|$  are much larger than  $|S|$  but after alloying  $S$  becomes effective in comparison to  $S_{gas}$ . This shows that in alloying the packing of constituent atoms are more important than the behaviour of individual atom in a given volume element. For investigating the effect of temperature on  $S^M$  for all the six systems, the values of entropy of mixing are displayed in Figs. (1) to (6). A perusal of Figs: (1) to (3) reveals that  $S^M$  of Na-K, Na-Rb and K-Rb alloys decreases with increasing temperature. But the result of Na-Cs, Rb-Cs and K-Cs systems exhibit a mixed behaviour. Near the melting point i.e. between 400°K to 600°K for Na-Cs  $S^M$  increases with increasing temperature and above 600°K it starts falling towards the ideal entropy of mixing. But,  $S^M$  for Rb-Cs and K-Cs alloys, increases with increasing temperature between 400° to 500° and above 500°K it starts falling similarly to Na-Cs.

Though the experimental values of temperature dependence of  $S^M$  for the above mentioned systems are not available in literature for an effective comparison, yet good accord with experiment for Na-K and Na-Cs at melting point confirms the validity of results obtained for these system at different temperatures.

Thus, our work provides an additional confidence in the hard sphere formalism for calculating the entropies of liquid metals and alloys. This is the most valuable contribution of semi-empirical approach to study  $S^M$  of alkali-alkali alloys as a function of concentration and temperature. This is not easily accessible from any other source and such investigation to the best of our knowledge are completely lacking.

In conclusion it may be asserted that this paper outlines the general theory of  $S^M$  and the numerical values obtained for alloys form a good set of reliable data.

**Table 1: The hard sphere parameters ( $\sigma\eta$ ) entropy ( $S/K_B$ ) and density of liquid alkali metals at different temperatures.**

Pure liquid Metals	Temp. (°K)	Hard-Sphere parameter		Entropy ( $S/K_B$ ) expt.	Density (a.u) expt.
		Hard-sphere diameter ( $\dagger$ )	Packing factor( $\gamma$ )		
Sodium	400	6.052	0.4143	8.061	0.92038
	500	5.954	0.3835	8.895	0.89438
	600	5.872	0.3572	9.557	0.86838
	700	5.804	0.3346	10.103	0.83238
	800	5.725	0.3111	10.606	0.81638
Potassium	400	7.460	0.4027	9.724	0.812155
	500	7.330	0.3710	10.556	0.788655
	600	7.216	0.3434	11.226	0.765155
	700	7.118	0.3194	11.782	0.741655
	800	7.030	0.2980	12.261	0.718155
Rubidium	400	7.919	0.3928	11.259	1.447653
	500	7.760	0.3578	12.119	1.401553
	600	7.628	0.3287	12.796	1.355453
	700	7.519	0.3041	13.351	1.309353
	800	7.432	0.2833	13.821	1.263253
Caesium	400	8.416	0.3725	12.452	1.77885
	500	8.185	0.3322	13.340	1.72385
	600	8.042	0.3049	13.969	1.6685
	700	7.954	0.2853	14.460	1.61385
	800	7.886	0.2686	14.884	1.55885

**Table 2 : Relevant data for  $S_{\text{gas}}/K_B$ ,  $S/K_B$  and  $-S_y/K_B$  used in the calculation.**

Alloys	Temp (°K)	$C_{\text{na}}$	$S_{\text{gas}}/K_B$	$-S_y/K_B$	$S/K_B$
Na-K	400	0.8	11.6540	3.3480	0.0520
		0.7017	11.8033	3.3445	0.0632
		0.5001	12.1004	3.3064	0.0650
		0.2997	12.3844	3.2459	0.0475
		0.1999	12.5214	3.2153	0.0339
	500	0.8	12.0158	2.8909	0.0434
		0.7017	12.1649	2.8862	0.0523
		0.5001	12.4624	2.8477	0.0542
		0.2997	12.7471	2.7914	0.0396
		0.1999	12.8842	2.7647	0.0283
	600	0.8	12.3171	2.5463	0.0369
		0.7017	12.4659	2.5406	0.0449
		0.5001	12.7639	2.5015	0.0461
		0.2997	13.0494	2.4478	0.0336
		0.1999	13.1866	2.4237	0.0240
	700	0.8	12.5769	2.2765	0.0318
		0.7017	12.7255	2.2697	0.0387
		0.5001	13.0240	2.2294	0.0398
		0.2997	13.3103	2.1770	0.0289
		0.1999	13.4476	2.1546	0.0207
800	0.8	12.8066	2.0320	0.0284	
	0.7017	13.0347	2.0281	0.0326	
	0.5001	13.2541	1.9951	0.0356	
	0.2997	13.4632	1.9648	0.0289	
	0.1999	13.6786	1.9307	0.0186	

**Table 3: Relevant data for  $S_{\text{gas}}/K_B$ ,  $S /K_B$  and  $S /K_B$  used in the calculation.**

Alloys	Temp (°K)	$C_{\text{Rb}}$	$S_{\text{gas}}/K_B$	$-S /K_B$	$S /K_B$
Na-Rb	400	0.2	13.649	3.036	0.04926
		0.3	13.385	3.054	0.06952
		0.5	12.841	3.099	0.09743
		0.7	12.270	3.163	0.09944
		0.8	11.971	3.206	0.08489
	500	0.2	14.016	2.570	0.03864
		0.3	13.751	2.589	0.0563
		0.5	13.207	2.636	0.0789
		0.7	12.635	2.702	0.0808
		0.8	12.336	2.746	0.0690
	600	0.2	14.322	2.229	0.0347
		0.3	14.058	2.248	0.0471
		0.5	13.512	2.294	0.0661
		0.7	12.940	2.359	0.0676
		0.8	12.640	2.404	0.0578
	700	0.2	14.588	1.968	0.0271
		0.3	14.323	1.987	0.0394
		0.5	13.777	2.031	0.0566
		0.7	13.204	2.059	0.0580
		0.8	12.903	2.138	0.0496
800	0.2	14.824	1.762	0.0256	
	0.3	14.559	1.777	0.0342	
	0.5	14.012	1.815	0.0472	
	0.7	13.438	1.868	0.0519	
	0.8	13.137	1.904	0.0444	

**Table 4: Relevant data for  $S_{gas}/K_B$ ,  $S /K_B$  and  $S /K_B$  used in the calculation.**

Alloys	Temp (°K)	$C_k$	$S_{gas}/K_B$	$S /K_B$	$S /K_B$
K-Rb	400	0.2022	13.8886	3.0419	0.00307
		0.3998	13.6177	3.0711	0.00478
		0.5004	13.4793	3.0864	0.00510
		0.6005	13.3418	3.0980	0.00501
		0.8514	12.9946	3.1353	0.00281
	500	0.2022	14.2519	2.5910	0.00236
		0.3998	13.9817	2.6177	0.00367
		0.5004	13.8431	2.6342	0.00392
		0.6005	13.7068	2.6415	0.01079
		0.8514	13.3579	2.6883	0.00216
	600	0.2022	14.5549	2.2586	0.00191
		0.3998	14.2853	2.2817	0.00297
		0.5004	14.1465	2.2979	0.00317
		0.6005	14.0115	2.3011	0.00310
		0.8514	13.6610	2.3516	0.00175
	700	0.2022	14.8166	2.0044	0.00163
		0.3998	14.5476	2.0229	0.00256
		0.5004	14.4086	2.0380	0.00271
		0.6005	14.2749	2.0369	0.00265
		0.8514	13.9227	2.0873	0.00149
800	0.2022	15.0483	1.8047	0.00148	
	0.3998	14.7800	1.8177	0.00231	
	0.5004	14.6408	1.8309	0.00246	
	0.6005	14.5086	1.8254	0.00239	
	0.8514	14.1544	1.8728	0.00136	



**Table 5: Relevant data for  $S_{\text{gas}}/K_B$ ,  $S /K_B$  and  $S /K_B$  used in the calculation.**

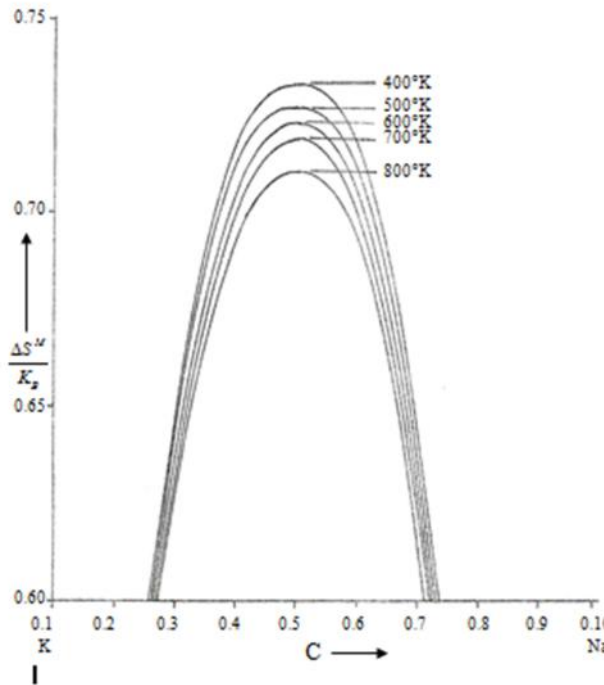
Alloys	Temp (ĒK)	$C_{\text{na}}$	$S_{\text{gas}}/K_B$	$S /K_B$	$S /K_B$
Na-Cs	400	0.1518	14.5366	2.8643	0.0503
		0.3	14.0109	3.0122	0.0969
		0.5023	13.2769	3.1965	0.1470
		0.7014	12.5326	3.3140	0.1579
		0.8	12.1507	3.3468	0.1378
	500	0.1578	14.9034	2.3469	0.0380
		0.3	14.3783	2.4667	0.0731
		0.5023	13.6410	2.6420	0.1113
		0.7014	12.8960	2.7679	0.1201
		0.8	12.5133	2.8206	0.1053
	600	0.1518	15.2099	2.0384	0.0391
		0.3	14.6855	2.1377	0.0611
		0.5023	13.9448	2.3007	0.0933
		0.7014	13.1990	2.4167	0.1006
		0.8	12.8115	2.4703	0.0884
	700	0.1518	15.4754	1.8332	0.0285
		0.3	14.9517	1.9164	0.0543
		0.5023	14.2072	2.0680	0.0832
		0.7014	13.4607	2.1692	0.0896
		0.8	13.0763	2.2176	0.0787
800	0.1578	15.7112	1.6674	0.0263	
	0.3	15.1882	1.7355	0.0501	
	0.5023	14.4397	1.8732	0.0768	
	0.7014	13.6923	1.9547	0.0825	
	0.8	13.3070	1.9932	0.0724	

**Table 6: Relevant data for  $S_{\text{gas}}/K_B$ ,  $S /K_B$  and  $S /K_B$  used in the calculation.**

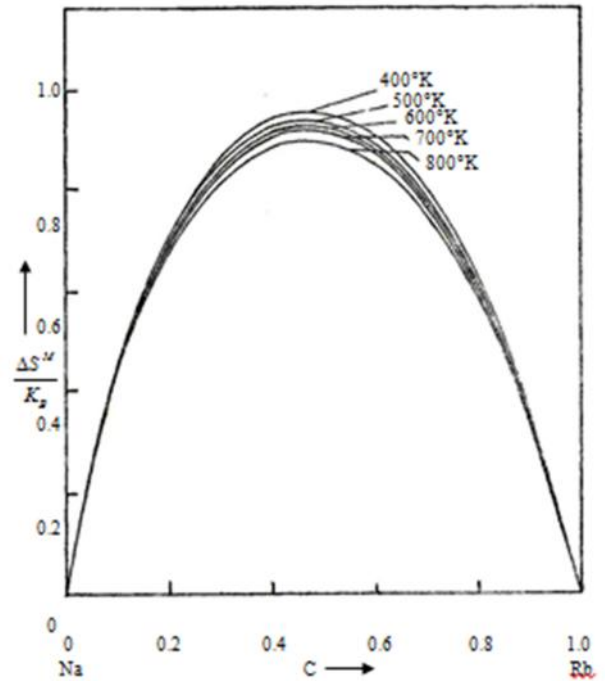
Alloys	Temp (°K)	$C_{Rb}$	$S_{\text{gas}}/K_B$	$S /K_B$	$S /K_B$
Rb-Cs	400	0.2	14.888	2.775	0.0028
		0.3	14.799	2.795	0.0038
		0.5	14.621	2.848	0.0048
		0.7	14.441	2.906	0.0043
		0.8	14.350	2.936	0.0032
	500	0.2	15.254	2.285	0.00177
		0.3	15.166	2.311	0.00232
		0.5	14.988	2.367	0.00303
		0.7	14.808	2.429	0.00271
		0.8	14.717	2.464	0.00213
	600	0.2	15.560	1.987	0.0015
		0.3	15.472	2.009	0.0021
		0.5	15.294	2.056	0.0025
		0.7	15.114	2.108	0.0023
		0.8	15.023	2.137	0.0018
	700	0.2	15.825	1.785	0.00149
		0.3	15.737	1.802	0.00201
		0.5	15.559	1.836	0.00253
		0.7	15.379	1.874	0.00225
		0.8	15.289	1.894	0.00169
800	0.2	16.061	1.626	0.00149	
	0.3	15.973	1.637	0.00201	
	0.5	15.795	1.662	0.00252	
	0.7	15.615	1.689	0.00225	
	0.8	15.525	1.705	0.00176	

**Table 7. Relevant data for  $S_{\text{gas}}/K_B$ ,  $S /K_B$  and  $S /K_B$  used in the calculation.**

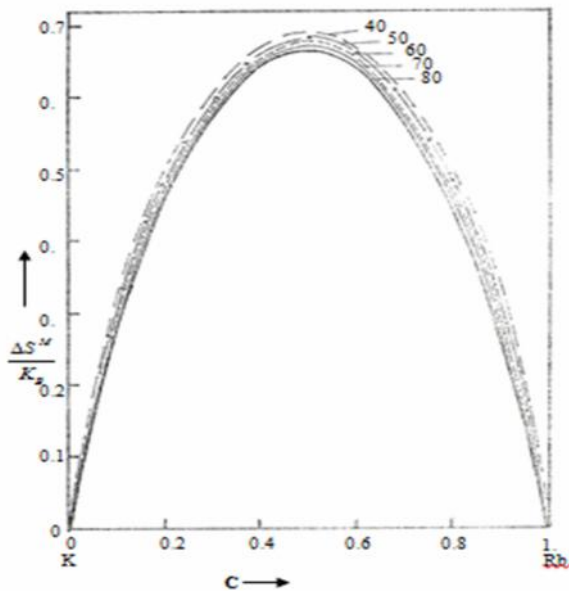
Alloys	Temp (°K)	$C_k$	$S_{\text{gas}}/K_B$	$S /K_B$	$S /K_B$
K-Cs	400	0.2	14.632	2.783	0.0106
		0.3	14.400	2.815	0.0105
		0.5	13.951	2.887	0.0191
		0.7	13.493	2.975	0.0179
		0.8	13.261	3.027	0.0145
	500	0.2	14.988	2.298	0.00721
		0.3	14.766	2.333	0.00993
		0.5	14.315	2.413	0.01311
		0.7	13.857	2.511	0.01234
		0.8	13.625	2.568	0.01001
	600	0.2	15.294	2.002	0.00598
		0.3	15.071	2.032	0.00822
		0.5	14.620	2.104	0.01085
		0.7	14.162	2.191	0.01021
		0.8	13.929	2.242	0.00827
	700	0.2	15.558	1.801	0.00557
		0.3	15.335	1.827	0.00767
		0.5	14.884	1.884	0.01009
		0.7	14.425	1.955	0.00950
		0.8	14.193	1.997	0.00766
800	0.2	15.793	1.640	0.00536	
	0.3	15.570	1.660	0.00736	
	0.5	15.118	1.708	0.00967	
	0.7	14.659	1.765	0.01552	
	0.8	14.426	1.799	0.00732	



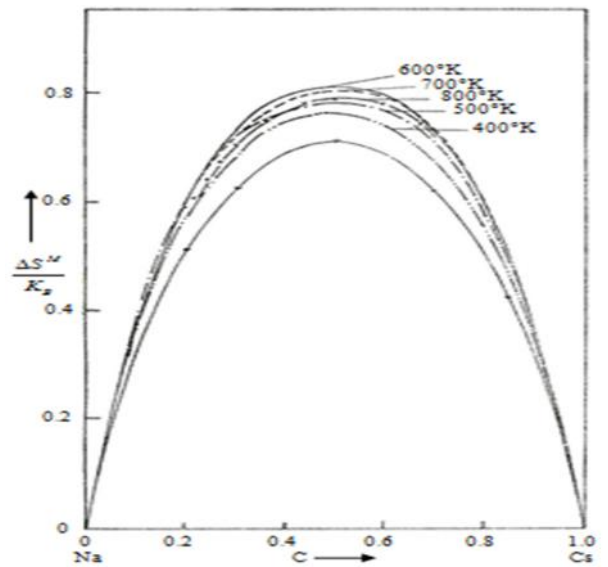
**Fig. 1:** Entropy of mixing ( $\Delta S^M/K_B$ ) of Na-K as a function of concentration and temperature.



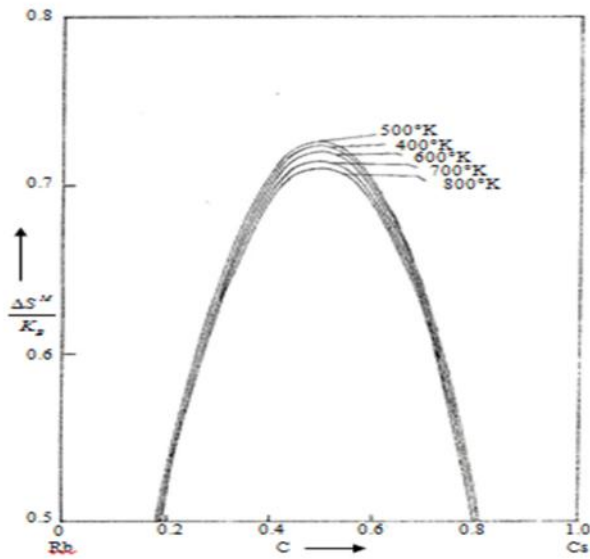
**Fig. 2:** Entropy of mixing of ( $\Delta S^M/K_B$ ) Na-Rb alloys function of concentration and temperature.



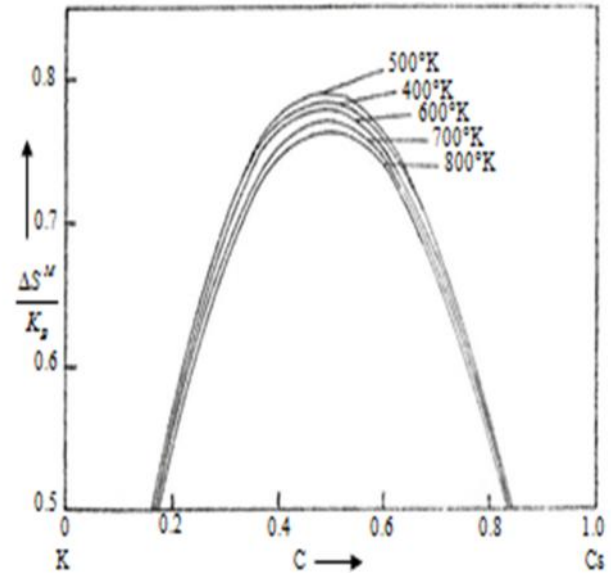
**Fig. 3:** Temperature dependence of the entropy of mixing ( $\Delta S^M/K_B$ ) for liquid K-Rb alloys.



**Fig. 4:** Temperature dependence of the entropy of mixing ( $\Delta S^M/K_B$ ) for liquid Na-Cs alloys. corresponds to the experimental value at 380°K of Neale & Caoack (1982).



**Fig. :** Entropy of mixing ( $\Delta S^M/K_B$ ) of (Rb-Cs) alloys as a function of Temperature.



**Fig. 6:** Entropy of mixing ( $\Delta S^M/K_B$ ) for liquid (K-Cs) alloys as a function of temperature & concentration.

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