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Investigation of some basic thermodynamic properties of Na-K alloy

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ABSTRACT

Investigation of thermodynamic of liquid $Na_{1-X}K_X$ binary alloys using pseudopotential theory is reported. The potential suggested by Fiolhais et al. with its individual parameters is used for entire calculation. A transferability of the potential from the solid to liquid medium is achieved for the presently reported binary alloy. The internal energy components, Helmholtz free energy, entropy and total energy at various proportions of the participating alkali metals are included in the study. The comparison with the other data has been shown in the present article. Exchange and correlation effect is also tested with the help of various local field correction functions.

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1. Introduction

Novelty in materials is key demand of industrial development. The successful synthesis of any material may be useful for the formation of fabrication of parts of various machineries and devices. The choice of the material has direct influence on overall performance of the device. The choice of the material must be on the base of some scientifically obtained physical and chemical properties. As per the present demand of requirement, variety of properties cannot be avail by the conventional metallic materials. The compounds and alloys are the better replacement of the pure metallic material to have combined properties in a single material. Focusing on alloys, the preliminary stage of the formation also requires the exact knowledge of thermodynamical properties of the alloy. The variation of property with respect to a variation of the proportion of each metal used for the formation of alloy is to be obtained to know the required proportion for desired property. Lots of work has been carried out for the thermodynamical properties of the many solid materials [1-3]. But on the other hand, the liquid materials are studied theoretically from the thermodynamical point of view very rarely [4-5]. The liquid alkali metals are widely used in the area of nuclear reactors. As having very fast chemically reactive nature even at room temperature, the study of the thermodynamical properties is of great significance. Binary alloy of alkali metal is able to provide the mix properties of each of the metals of formed alloy. In the present article the focus is made on the one of the binary alkali alloys i.e**.**, $Na_{1-X}K_X$ in liquid state.

2. Computational Methodology

The well-established theory of the pseudopotential is applied for the present calculation. The potential suggested by the Fiolhais et al. [6] is chosen for as the model potentials. The accuracy obtained in previous calculations [9-14] by this potential for various properties is the main reason of the selection of the potential. As the structure factor and the entropy both can easily be characterized by considering the hard sphere system, the well-known Percus-Yevick hard sphere reference system [10] is used. The value of the Hamiltonian of the actual system can be estimated through the perturbation theory. According to the inequality of Gibbs and Bogoliubov [11, 12], the actual Hamiltonian of the reference system is always greater than or equal to the Hamiltonian of the reference system plus the perturbation part. The variational approach along with the Gibbs-Bogoliubov inequality has already been used to approximate the study of thermodynamical study of the metals. The present work extends the computation to the system of binary alkali alloy $Na_{1-X}K_X$. The electronic free energy of the binary alloys with atoms positions $\{R_1\}$ and $\{R_2\}$ can be written as [13, 14],

$$\mathrm{F_{ele}\{R_1, R_2\} = F_{eg} + F_1 F_2\{R_1, R_2\}} \qquad (1)$$

Here, $\mathrm{F_1}$ and $\mathrm{F_2}$ are the first and second order perturbation energy terms, respectively and $\mathrm{F_{eg}}$ is the free energy of the electron gas that can be expressed as,

$$\mathrm{F_{eg}} = \left[\frac{3}{10}\mathrm{k_F}^2 - \frac{3}{4\pi}\mathrm{k_F} + \mathrm{E_c} - \frac{1}{2}\gamma_{\mathrm{eg}}\mathrm{T}^2\right]\bar{\mathrm{Z}}. \qquad (2)$$

To calculate the Helmhöltz free energy $(\mathrm{F_h})$ of the alloy, the effective potential energy of the ions can be obtained by introducing the direct Coulomb interaction between the ions. The expectation value of this effective potential can be given by,

$$\mathrm{F_{ps} = F_{eg} + F_1 + F_2\{R_1, R_2\} + F_M}. \qquad (3)$$

The Madelung contribution $(\mathrm{F_M})$ shown in the above equation (3) can be given as,

$$\mathrm{F_M} = \frac{1}{\pi}\int_0^\infty \left[\mathrm{c_1}^2\mathrm{Z_1}^2(\mathrm{S_{11}} - 1) + 2\mathrm{c_1c_2}(\mathrm{S_{12}} - 1) + \mathrm{c_2}^2\mathrm{Z_2}^2(\mathrm{S_{22}} - 1)\right]\mathrm{dq}. \qquad (4)$$

The $\mathrm{S_{ij}}$ $(\mathrm{i = j; 1,2})$ is the partial structure factors computed from the well-known relations given by as used by Faber [11].

The first order perturbation term $(\mathrm{F_1})$ is obtained from averaged valence density $\overline{(\mathrm{Z})}$ and the zeroth Fourier component (α_1) of pseudopotential from the following equations (5) and (6), respectively,

$$\mathrm{F_1} = (\mathrm{c_1}\alpha_1 + \mathrm{c_2}\alpha_2)\bar{\mathrm{Z}}\mathrm{n}. \qquad (5)$$

$$\alpha_\mathrm{i} = \lim_{\mathrm{q}\to 0}\left(\mathrm{W_B(q)} + \frac{4\mathrm{Z_i}\pi \mathrm{e}^2}{\mathrm{q}^2}\right). \qquad (6)$$

The band structure energy or second order energy term $(\mathrm{F_2})$ is obtained from,

$$\mathrm{F_2} = \frac{1}{16\pi^3}\int_0^\infty \left\{\begin{matrix}\mathrm{c_1c_2}\left(\mathrm{W_{B_1}} - \mathrm{W_{B_1}}\right)^2 + \mathrm{c_1}^2\mathrm{W_{B_1}}^2\mathrm{S_{11}} \\ +2\mathrm{c_1c_2W_{B_1}W_{B_2}S_{12}} + \mathrm{c_2}^2\,\mathrm{W_{B_2}}^2\mathrm{S_{22}}\end{matrix}\right\}\left\{\frac{1}{\varepsilon(\mathrm{q})} - 1\right\}\mathrm{dq} - \frac{\bar{\mathrm{Z}}}{2}\gamma_2(\mathrm{T})\mathrm{T}^2\,. \qquad (7)$$

Here, $\mathrm{W_i}$ is the screened form factor. $\varepsilon(\mathrm{q})$ is the dielectric screening function. $\gamma_2(\mathrm{T})$ is the second order correction to the usual γ factor describing the low temperature electronic specific heat and is given by,

$$\gamma_2(\mathrm{T}) = \frac{2}{3\pi^2\bar{\mathrm{Z}}}\int_0^\infty \frac{\mathrm{x}^2}{\mathrm{x}^2-1}\mathrm{f(x)}\left\{\begin{matrix}\mathrm{c_1c_2}(\mathrm{W_1} - \mathrm{W_2})^2 + \mathrm{c_1}^2\mathrm{W_1}^2\mathrm{S_{11}} \\ +2\mathrm{c_1c_2W_1W_2S_{12}} + \mathrm{c_2}^2\mathrm{W_2}^2\mathrm{S_{22}}\end{matrix}\right\} \qquad (8)$$

in which, $\mathrm{W_i}$ is the screened form factor. While, the function f(x) is be defined as,

$$\mathrm{f(x)} = \left(\frac{\mathrm{x}^2-1}{4\mathrm{x}}\ln\left|\frac{1+\mathrm{x}}{1-\mathrm{x}}\right|\right)+\frac{1}{2}. \tag{9}$$

The free energy per particle ($\mathrm{F_{mix}}$) can be expressed as,

$$\mathrm{F_{mix}} = \mathrm{c_1\mu_1 + c_2\mu_2} - \frac{\mathrm{P_{hs}}}{\mathrm{n}}. \tag{10}$$

The chemical potential is obtained by following equation,

$$\frac{\mu_\mathrm{i}}{\mathrm{k_BT}} = \ln\left[\mathrm{n_i}\frac{2\pi\hbar^2}{\mathrm{m_ik_BT}}\right]^{\frac{3}{2}} - \ln(\mathrm{l}-\eta) + \ln\left[\frac{3\mathrm{X}\sigma_\mathrm{i}}{\mathrm{l}-\eta}\right] + \frac{3}{2}\left[\frac{3\mathrm{X}^2}{(1-\eta)^2}+\frac{2\mathrm{Y}}{1-\eta}\right]\sigma_\mathrm{i}{}^2 + \left[\frac{\pi\mathrm{P_{hs}}\sigma_\mathrm{i}{}^3}{6\mathrm{k_BT}}\right]. \tag{11}$$

and

$$\frac{\mathrm{P_{hs}}}{\mathrm{K_BT}} = \frac{\mathrm{n}(1+\eta+\eta^2) - \frac{1}{2}\pi\mathrm{n_1n_2}(\sigma_1-\sigma_2)^2(\sigma_1+\sigma_2+\sigma_1\sigma_1\mathrm{X})}{(1-\eta)^3} \tag{12}$$

where,

$$X = \frac{1}{6}\pi(\mathrm{n_1}\sigma_1{}^2 + \mathrm{n_2}\sigma_2{}^2) \tag{13}$$

and

$$\mathrm{Y} = \frac{1}{6}\pi(\mathrm{n_1}\sigma_1 + \mathrm{n_2}\sigma_2). \tag{14}$$

The internal energy ($\mathrm{F_{int}}$) can be given as,

$$\mathrm{F_{int}} = \frac{3}{2}\mathrm{k_BT} + \mathrm{F_{eg}} + \mathrm{F_1} + \mathrm{F_2\{R_1, R_2\}} + \mathrm{F_M}. \tag{15}$$

Using equations (10)-(15),

$$\mathrm{F_{mix}} = \frac{3}{2}\mathrm{K_BT} - \mathrm{TS_{mix}}, \tag{16}$$

where, $\mathrm{S_{mix}}$ is the total entropy for alloy system and can be divided into following four parts,

$$\mathrm{S_{mix}} = \mathrm{S_{gas}} + \mathrm{S_c} + \mathrm{S}_\eta + \mathrm{S}_\sigma. \tag{17}$$

where, $\mathrm{S_c}$ is the ideal entropy of mixing, $\mathrm{S_{gas}}$ represents the gas term, S_η corresponds to packing density η and S_σ arises due to the difference in diameters of the hard sphere of participating elements of the alloy, respectively.

The various contributions of the entropy can be given as,

$$\frac{\mathrm{S_c}}{\mathrm{k_B}} = -(\mathrm{c_1lnc_1 + c_2lnc_2}), \tag{18}$$

$$\frac{\mathrm{S}_\eta}{\mathrm{k_B}} = \ln(\mathrm{l}-\eta) + \frac{3}{2}\left[\mathrm{l} - \frac{\mathrm{l}}{(\mathrm{l}-\eta)^2}\right], \tag{19}$$

$$\frac{\mathrm{S}_\sigma}{\mathrm{k_B}} = \frac{\pi\mathrm{c_1c_2n}(\sigma_1-\sigma_2)^2\lfloor 12(\sigma_1+\sigma_2) - \pi\mathrm{n}(\mathrm{c_1}\sigma_1{}^4 + \mathrm{c_2}\sigma_2{}^4)^2\rfloor}{24(1-\eta)^2} \tag{20}$$

and

$$\frac{\mathrm{S_{gas}}}{\mathrm{k_B}} = \ln\left[\frac{\mathrm{e}}{\mathrm{n}}\left(\frac{\mathrm{emk_BT}}{2\pi\hbar^2}\right)^{\frac{3}{2}}\right], \tag{21}$$

From all above required contributions, the Helmhöltz free energy ($\mathrm{F_h}$) can be obtained as,

$$\mathrm{F_h} = \mathrm{F_{ps}} + \mathrm{F_{mix}}. \tag{22}$$

To estimate the screening effect over the bare-ion potential the Hartree local field correction function (H) is used [15]. The other local field correction functions used over the bare-ion potential are the functions suggested by Hubbard-Sham (HS) [16, 17], Vashishtha-Shingwi (VS) [18], Taylor (T) [19], Sarkar et al. (S) [20], Ichimaru-Utsumi (IU) [21] and Farid et al. (F) [22] and Nagy (N) [23].

3. Results and Discussion

The individual parameters of the potential of Fiolhais *et al.* [6] and the constants are used as given in the Table 1. The potential parameters are directly adopted from the original work of the Fiolhais *et al.* [6]. Table 2 shows the various parts (F_{eg}, F_1, F_M and F_{mix}) that contributes to the internal energy (F_{int}) of the alloy. Rather than calculating only internal energy (F_{int}) and the Helmhotz free energy (F_h), the present calculation focused on calculation of the microscopic distributions of these energies in further contribution. The present results also compared with the others available theoretical results [14] to validate our results. It can be seen from the Table 2 that the F_{eg} and F_M has excellent agreement with compared results [14].

A very good agreement is obtained with the results of Vora [14] for the results of F_{mix}. This means the results generated by the potential used in present calculation and by Vora [14] provides the almost same type of approximation for free electron energy, free energy per particle of the binary mixture and the Madlung static electrical energy. The contributions provided by F_{eg}, F_M and F_{mix} are negative whereas that due to F_1 is positive. Out of these four parts, the F_1 and F_2 depend upon model potential. F_1 is obtained from the zero limit of the potential.

The results of the second order perturbation energy (F_2) for eight different local field corrections functions [15-23] are shown in the Table 3. The comparision of the present result with the other theoretical available data [14] is also shown in the Table 3. From the Table 3, it can be observed that the results due to N-function [23] is highest for all concentration (X). The HS-function [16, 17] provids the minimum exchange and correlation effect with respect to H-function [15].

The total internal energy (F_{int}) of the alloys under study is shown in the Table 4. The minimum deviation from the experimental value [20] is obtained for the N-function [23] and maximum for H-function [15]. However no significant effect is change is observed for any of the local field correction functions. Hence, the suggested bare-ion potential is suffiently efficient to provide the screening and exchange and correlation effect.

It also can b observed from the Table 4 that the variation of F_{int} from the experimental value [13] is maximum at $X = 0$ is and minimum for $X = 1$. Thus, the potential provides the poor results when proportion of Na is more and better results when proportion of K is more.

Various entroy contributions and total entropy S_{mix} are obtained a shown in the Figure 1. The entropy contributes to the Helmholtz free enrgy (F_h) as shown in the equation [3] and [4].

The calculated Helmhotz free energy (F_h) is shown in the Table 6. The results obtained for the F_h are found in well agreement with the experimental data [13] for $X = 1$, better agreement for $X = 0.5$ and reasonable agreement for $X = 0$.

Table 1. Input parameters and constants.

Metal	Ω (au)	k_f (au)	α (au) [6]	R (au) [6]
Na	227.76	0.4742	3.517	0.492
K	528.67	0.3826	3.349	0.679

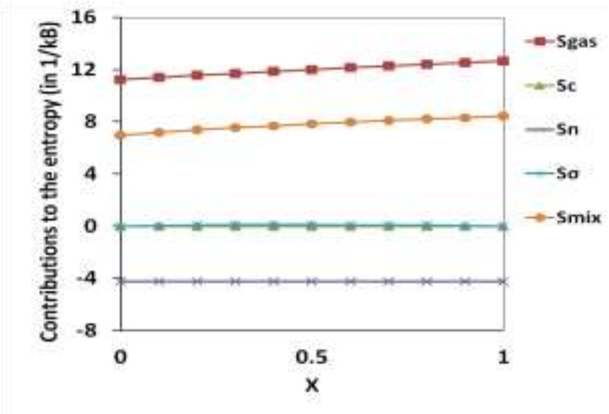

Fig. 1: Various Entropy Contributions.

Table 2: F_{eg}, F_1, F_M and F_{mix} $(*\,10^{-3} in\ au)$

X	F_{eg}	Others [14]	F_1	F_M	Others [14]	F_{mix}	Others [14]
0.0	-81.61	-81.63	78.49	-213.7	-210.17	-6.45	-7.51
0.1	-81.73	-81.74	79.47	-203.1	-210.37	-6.71	-8.12
0.2	-81.73	-81.72	83.30	-194.5	-192.15	-6.93	-8.54
0.3	-81.64	-81.62	86.57	-187.5	-184.55	-7.13	-8.87
0.4	-81.48	-81.45	89.40	-182.1	-178.36	-7.32	-9.12
0.5	-81.28	-81.24	91.87	-178.0	-173.40	-7.49	-9.32
0.6	-81.05	-81.00	94.05	-175.0	-169.53	-7.64	-9.45
0.7	-80.78	-80.73	95.99	-172.9	-166.61	-7.79	-9.53
0.8	-80.50	-80.44	97.71	-171.8	-164.54	-7.92	-9.55
0.9	-80.21	-80.13	99.27	-171.5	-163.20	-8.05	-9.48
1.0	-79.90	-79.82	100.67	-171.8	-162.53	-8.17	-9.23

Table 3. $F_2 * 10^{-3}$in au

X	H	HS	VS	T	S	IU	F	N	Others [14]
0.0	-16.09	-15.45	-14.88	-14.63	-15.14	-14.35	-14.33	-13.27	-13.69
0.1	-70.51	-68.68	-67.09	-66.38	-67.83	-65.68	-65.61	-61.83	-
0.2	-74.60	-72.33	-70.23	-69.20	-71.18	-68.30	-68.18	-63.93	-
0.3	-61.07	-58.81	-56.55	-55.33	-57.54	-54.39	-54.24	-50.60	-
0.4	-43.40	-41.39	-39.23	-37.96	-40.14	-37.09	-36.92	-34.29	-
0.5	-27.86	-26.20	-24.30	-23.11	-25.08	-22.37	-22.20	-20.57	-19.20
0.6	-17.44	-16.15	-14.59	-13.56	-15.22	-12.98	-12.83	-11.89	-
0.7	-12.38	-11.40	-10.19	-9.38	-10.67	-8.94	-8.82	-8.17	-
0.8	-12.21	-11.46	-10.56	-9.98	-10.93	-9.66	-9.57	-8.80	-
0.9	-14.43	-13.83	-13.18	-12.83	-13.46	-12.58	-12.53	-11.39	-
1.0	-12.62	-12.22	-11.84	-11.71	-12.03	-11.54	-11.53	-10.30	-14.50

Table : $F_{int} * 10^{-3}$in au

X	**H**	**HS**	**VS**	**T**	**S**	**IU**	**F**	**N**	**Exp [13]**
0.0	-308.75	-308.11	-307.55	-307.29	-307.81	-307.02	-307.00	-305.93	-226.00
0.1	-274.13	-272.30	-270.71	-270.00	-271.45	-269.30	-269.23	-265.45	-
0.2	-265.74	-263.48	-261.38	-260.34	-262.32	-259.45	-259.33	-255.08	-
0.3	-241.92	-239.66	-237.40	-236.18	-238.38	-235.24	-235.09	-231.45	-
0.4	-215.82	-213.80	-211.64	-210.37	-212.55	-209.50	-209.33	-206.71	-
0.5	-193.46	-191.80	-189.90	-188.70	-190.68	-187.97	-187.80	-186.16	-
0.6	-177.62	-176.33	-174.77	-173.73	-175.39	-173.16	-173.00	-172.07	-
0.7	-168.35	-167.37	-166.16	-165.35	-166.64	-164.92	-164.79	-164.14	-
0.8	-165.04	-164.29	-163.39	-162.82	-163.76	-162.50	-162.41	-161.63	-
0.9	-165.06	-164.46	-163.81	-163.46	-164.10	-163.21	-163.17	-162.02	-
1.0	-161.87	-161.48	-161.10	-160.96	-161.28	-160.79	-160.79	-159.55	-190.00

Table 5: $F_h * 10^{-3}$in au

X	**H**	**HS**	**VS**	**T**	**S**	**IU**	**F**	**N**	**Exp [13]**
0.0	-369.62	-368.99	-368.42	-368.16	-368.68	-367.89	-367.87	-366.80	-250.33
0.1	-323.36	-321.53	-319.94	-319.24	-320.68	-318.54	-318.46	-314.68	-
0.2	-320.67	-318.41	-316.30	-315.27	-317.25	-314.37	-314.25	-310.01	-
0.3	-306.24	-303.98	-301.72	-300.50	-302.70	-299.56	-299.41	-295.77	-
0.4	-287.95	-285.94	-283.78	-282.51	-284.69	-281.64	-281.47	-278.84	-
0.5	-269.63	-267.97	-266.06	-264.87	-266.84	-264.14	-263.96	-262.33	-235.90
0.6	-253.39	-252.09	-250.53	-249.50	-251.16	-248.93	-248.77	-247.84	-
0.7	-239.82	-238.84	-237.63	-236.82	-238.12	-236.39	-236.26	-235.61	-
0.8	-229.41	-228.66	-227.76	-227.18	-228.13	-226.86	-226.77	-226.00	-
0.9	-221.56	-220.97	-220.32	-219.97	-220.60	-219.72	-219.67	-218.52	-
1.0	-214.13	-213.74	-213.36	-213.23	-213.54	-213.06	-213.05	-211.81	-218.51

4. Conclusion

The thermodynamical investigation of the liquid alkali $Na_{1-X}K_X$ alloy has been done. The potential of Fiolhais *et al*. [6] is found appropreate to describe the themodynamical properties of the alloy under study. The results for the internal energy (F_{int}) and Helmholtz energy (F_h) are obtained and found in a good agreement at some concentration value. The present results are found to be deviated from the experimental data [13] at lower concentration.

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