Entropy Change and Free Energy Change in the Liquid-Vapor Phase Transition of Alkali Metals

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Abstract: A three-parameter generalized van der Waals equation of state has been employed to determine free energy change and entropy change in the liquid-vapor transition of fluid alkali metals in the range of temperatures from their boiling point to critical point. This equation differs from the known van der Waals equation of state by the modified expression for molecular pressure. The results yielded by the generalized van der Waals equation of state agree well with those given by Carruth and Kobayashi correlation (CK), Fish and Liemezs correlation (FL) and Watson correlation.

Keywords: Alkali metals, Entropy, Helmholtz free energy, Liquid- Vapor Phase Transition, Equation of state

1. Introduction

The study of the thermodynamic properties of alkali metals significant. scientifically Moreover, numerous is technological applications such as coolant systems for fastneutron breeder nuclear reactors, magnetohydrodynamic energy conversion generators, etc. employ fluid alkali metals. These technological applications require the knowledge of high-temperature properties of fluid alkali metals. Owing to these facts, numerous experimental and theoretical studies have been made [1-29]on the hightemperature properties of fluid alkali metals. However, the experimental studies of alkali metals in the hightemperature region encounter severe difficulties. It is due to the fact that alkali metals are reactive at high -temperatures. This often leads to poor accuracy in high-temperature studies of alkali metals. Hence arises the necessity for further studies on the properties of alkali metals in the hightemperature region. The present work, on the basis of a generalized van der Waals equation of state, deals with the determination of free energy change and entropy change in liquid-vapor transition for fluid alkali metals.

2. Generalized van der Waals equation of state

To improve the accuracy in determining the thermodynamic properties of substances the known two-parameter van der Waals equation of state is modified [30] by introducing the third parameter n in the expression for molecular pressure. The three-parameter generalized van der Waals equation of state for one mole of substance has the form

$$P = \frac{RT}{V-b} - \frac{a}{V^n} \tag{1}$$

Where R is the gas constant, P is the pressure, V is the molar volume, T is the temperature; a, b and n are substance-specific parameters to be determined through experimental data.

Applying the vapor-liquid critical point conditions to the Eq. (1), the generalized van der Waals equation of state may be rewritten in the reduced form as

$$P^* = \frac{4n}{(n-1)^2} \left(\frac{T^*}{NV^* - 1} - \frac{1}{V^{*n}} \right)$$
(2)

Where N = (n+1)/(n-1), $P^* = P/P_c$ is the reduced pressure, $V^* = V/V_c$ is the reduced volume, $T^* = T/T_c$ is

the reduced temperature, P_c is the critical pressure, V_c is the critical volume and T_c is the critical temperature. The reduced equation of state given by Eq. (2) expresses the single-parameter law of corresponding states with the thermodynamic similarity parameter n. That is, the substances with the same value of parameter n are thermodynamically similar. That is, such substances have similar intermolecular force characteristics.

It has been established [30-39] that the generalized van der Waals equation of state given by describes the thermodynamic properties of fluid alkali metals in the range of temperatures from their boiling point to critical point. Hence, it is appropriate to employ this equation of state to determine the properties of fluid alkali metals viz entropy change and free energy change in liquid-vapor phase transition.

For fluid alkali metals, the parameters a, b and n of the generalized van der Waals equation of state have been determined [30] through experimental data on critical point parameters. The obtained values of the parameters a, b and n are presented in Table 1. As seen, the values of the thermodynamic similarity parameter n for fluid alkali metals only slightly differ.

Table 1: Equation of state parameters [30]

Table 1. Equation of state parameters [50]					
ALKALI	а	b	п		
METAL	$\frac{J}{mol} \left(\frac{m^3}{mol}\right)^{n-1}$	$10^{-5}\left(\frac{\mathrm{m}^3}{\mathrm{mol}}\right)$			
Cesium	353.3494	6.7117	1.4831		
Rubidium	237.4616	6.1665	1.5292		
Potassium	606.7640	3.4930	1.4089		
Sodium	3113.1432	0.6986	1.2006		
Lithium	6759.8706	0.4233	1.1452		

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3. Entropy Change in Liquid-Vapor Transition by Generalized van der Waals Equation of State

The Classius – Clapeyron equation $\left[40\right]$ for the saturation vapor pressure is

$$\frac{dP_{sat}}{dT} = \frac{\Delta H_{\nu}}{T(V_g - V_l)} \tag{3}$$

Where ΔH_{v} is the enthalpy of vaporization, V_{g} and V_{l} are molar volumes of vapor and liquid under saturation conditions respectively and P_{sat} is the saturation pressure.

Taking into consideration Eq. (3), we get the enthalpy of vaporization for fluids obeying the generalized van der Waals equation of state given by Eq. (1) as

$$\Delta H_{v} = \frac{RT}{V_{l} - b} \left(V_{g} - V_{l} \right) \tag{4}$$

The temperature dependence of the volume constant in the van der Waals type equation of state is given [41] as

$$b = b_c \left(\frac{T_c}{T}\right)^{1/3} \tag{5}$$

Where b_c is volume constant determined through the experimental data on critical volume.

Substituting Eq. (5) into Eq. (4), we get

$$\Delta H_{v} = \frac{RT}{V_{l} - b_{c} \left(\frac{T_{c}}{T}\right)^{1/3}} \left(V_{g} - V_{l}\right)$$
(6)

At the saturated conditions, the entropy change in liquid-vapor transition is given [40] by

$$\Delta S = \frac{\mathrm{d}P_{sat}}{\mathrm{d}T} \left(V_g - V_l \right) \tag{7}$$

Hence, for fluids obeying the generalized van der Waals equation of state, the entropy change in liquid-vapor transition under saturated conditions is

$$\Delta S = \frac{R}{V_l - b} \left(V_g - V_l \right) \tag{8}$$

Substituting Eq. (5) into Eq. (8), we get

$$\Delta S = \frac{R}{V_l - b_c \left(\frac{T_c}{T}\right)^{1/3}} \left(V_g - V_l\right) \tag{9}$$

4. Determination of Entropy Change in Liquid-Vapor Transition of Fluid Alkali Metals

The vapor and liquid volumes of fluid alkali metals at saturation have been determined [42] through the generalized van der Waals equation of state. These data on the vapor and liquid volumes at saturation, the values of the parameter b and n for alkali metals are used to determine the entropy change in liquid-vapor transition through Eq.(8). The entropy change in liquid-vapor transition of fluid alkali metals is also determined using Eq. (9) in which the temperature-dependence of the volume constant b is taken into account. The obtained values are presented in Tables 7-

11. The obtained temperature dependence of entropy change in liquid-vapor transition for fluid alkali metals have been plotted in Figures 6-10. The results yielded by the generalized van der Waals equation of state are compared with those yielded by the known CK correlation, FL correlation and Watson correlation. The CK correlation gives the entropy change in liquid-vapor transition as

$$\Delta S = \frac{RT_c}{T} \left[7.08(1 - T^*)^{0.354} + 10.95\omega(1 - T^*)^{0.456} \right]$$
(13)

The FL correlation gives the entropy change in liquid-vapor transition as

$$\Delta S = \frac{RT_c}{T} \left[\frac{T}{T_b} \left(\frac{X + X^n}{1 + X^m} \right) \right] \tag{14}$$

The Watson correlation gives the entropy change in liquidvapor transition as

$$\Delta S = \frac{RT_c}{T} \left[\Delta H_b \left(\frac{T_c - T}{T_c - T_b} \right)^{0.38} \right]$$
(15)

The temperature-dependence of entropy change in liquidvapor transition of fluid alkali metals is determined by CK correlation, FL correlation and Watson correlation given by Eq. (13),Eq. (14) and Eq. (15), respectively. The results are presented in Tables 7-11.

 Table 2: Entropy change in liquid–vapor phase transition of saturated cesium

	ΔS , 1	kJ/mol/K			
Т, К	$\mathbf{E}_{\alpha}(0)$	$E_{\alpha}(0)$	CK	FL	WATSON
	Eq.(8)	Eq.(9)	Eq.(13)	Eq.(14)	Eq.(15)
1795	0.0223	0.0226	0.0187	0.0185	0.0182
1805	0.0213	0.0216	0.0181	0.0179	0.0176
1814	0.0204	0.0206	0.017	0.0176	0.017
1824	0.0197	0.0196	0.0169	0.0166	0.0163
1834	0.0184	0.0185	0.0163	0.0159	0.0155
1843	0.0173	0.0175	0.0157	0.015	0.0149
1853	0.0162	0.0163	0.0149	0.0145	0.014
1863	0.0151	0.0151	0.0141	0.0137	0.0132
1872	0.0138	0.0139	0.0133	0.0129	0.0124
1882	0.0125	0.0126	0.0124	0.0119	0.0113
1891	0.011	0.011	0.0114	0.0109	0.0103
1901	0.0096	0.0096	0.0101	0.0095	0.0089
1911	0.0079	0.0079	0.0083	0.0078	0.0071
1920	0.0054	0.0054	0.0056	0.0051	0.0045
1930^{*}	0	0	0	0	0

*-Vapor–Liquid critical point



Figure 1: Entropy change in liquid–vapor phase transition of saturated cesium

Volume 7 Issue 10, October 2018 www.ijsr.net

 Table 3: Entropy change in liquid–vapor phase transition of saturated rubidium

<i>Т</i> , К	ΔS , k	kJ/mol/K			
	Eq.(8)	Eq.(9)	CK	FL	WATSON
			Eq.(13)	Eq.(14)	Eq.(15)
1897	0.0212	0.0215	0.0178	0.0178	0.0171
1907	0.0202	0.0205	0.0172	0.0171	0.0165
1917	0.0191	0.0193	0.0166	0.0165	0.0158
1927	0.018	0.0182	0.0159	0.0158	0.0151
1937	0.0168	0.017	0.0152	0.0151	0.0144
1947	0.0156	0.0157	0.0145	0.0143	0.0136
1958	0.0143	0.0144	0.0137	0.0134	0.0127
1968	0.0129	0.013	0.0128	0.0125	0.0118
1978	0.0113	0.0113	0.0118	0.0115	0.0107
1988	0.0099	0.0099	0.0107	0.0103	0.0095
1998	0.0081	0.0081	0.0093	0.0089	0.0063
2018^{*}	0	0	0	0	0





Figure 2: Entropy change in liquid–vapor phase transition of saturated rubidium

 Table 4: Entropy change in liquid–vapor phase transition of saturated potassium

Т, К	ΔS , kJ/mol/K				
	Eq.(8)	Eq.(9)	CK	FL	WATSON
	_		Eq.(13)	Eq.(14)	Eq.(15)
2118	0.0191	0.0194	0.0195	0.0191	0.0179
2129	0.0184	0.0186	0.0189	0.0185	0.0174
2140	0.0173	0.0174	0.0184	0.0179	0.0168
2152	0.0169	0.017	0.0178	0.0173	0.0161
2163	0.0161	0.0162	0.0172	0.0166	0.0155
2175	0.0152	0.0153	0.0165	0.0159	0.0148
2186	0.0143	0.0144	0.0158	0.0153	0.0142
2197	0.0134	0.0134	0.0151	0.0146	0.0134
2209	0.0123	0.0124	0.0143	0.0137	0.0126
2220	0.0112	0.0113	0.0134	0.0129	0.0117
2232	0.0099	0.0093	0.0124	0.0118	0.0107
2243	0.0087	0.0088	0.0113	0.0108	0.0097
2254	0.0072	0.0072	0.0101	0.0094	0.0084
2266	0.005	0.005	0.0814	0.0077	0.0066
2277^{*}	0	0	0	0	0
0.02 0.0 (MJour/LA) 0.0 0.00	5 2 5 1 5 0				Eq. (8) Eq. (9) CK ←FL ←WATSON
	2100 2	150 2	200 22	.50 230	0

^{* -} Vapor–Liquid critical point

Figure 3: Entropy change in liquid–vapor phase transition of saturated potassium

 Table 5: Entropy change in liquid–vapor phase transition of saturated sodium

	ΔS , kJ/mol/K				
<i>T</i> , K	$\mathbf{E}_{\alpha}(9)$		СК	FL	WATSON
	Eq.(8)	Eq.(9)	Eq.(13)	Eq.(14)	Eq.(15)
2194	0.025	0.0253	0.0272	0.0266	0.0257
2219	0.0242	0.0243	0.0261	0.0254	0.0246
2244	0.0231	0.0232	0.025	0.0243	0.0234
2256	0.0226	0.0227	0.0244	0.0237	0.0228
2269	0.022	0.0221	0.0238	0.0231	0.0222
2281	0.0214	0.0215	0.0232	0.0225	0.0216
2294	0.0208	0.0209	0.0226	0.0219	0.021
2306	0.0201	0.0202	0.022	0.0213	0.0203
2319	0.0195	0.0195	0.0213	0.0206	0.0196
2331	0.0188	0.0188	0.0207	0.0199	0.019
2343	0.018	0.0181	0.02	0.0193	0.0183
2356	0.0172	0.0173	0.0193	0.0185	0.0176
2368	0.0165	0.0165	0.0185	0.0178	0.0168
2381	0.0156	0.0156	0.0177	0.017	0.016
2393	0.0147	0.0147	0.0169	0.0162	0.0152
2406	0.0137	0.0137	0.0159	0.0153	0.0143
2418	0.0127	0.0127	0.015	0.0144	0.0133
2431	0.0105	0.0112	0.0139	0.0132	0.0122
2443	0.0103	0.0103	0.0127	0.0121	0.0111
2456	0.009	0.009	0.0111	0.0106	0.0096
2468	0.0073	0.0073	0.0092	0.0087	0.0078
2481	0.0047	0.0047	0.0057	0.0053	0.0045
2493*	0	0	0	0	0

*- Vapor-Liquid critical point



Figure 4: Entropy change in liquid–vapor phase transition of saturated sodium

Table 6: Entropy change in liquid-vapor phase transition	n of
saturated lithium	

	Saturated Infinant					
<i>T</i> , K	ΔS , k.	ΔS , kJ/mol/K				
	Eq.(8)	Eq.(9)	СК	FL	WATSON	
	_	_	Eq.(13)	Eq.(14)	Eq.(15)	
3215	0.0147	0.0147	0.0158	0.0157	0.015	
3231	0.0133	0.0133	0.0146	0.0145	0.0138	
3248	0.0114	0.0114	0.0131	0.0131	0.0123	
3264	0.0091	0.0091	0.0115	0.0114	0.0107	
3281	0.006	0.006	0.0092	0.0091	0.0083	
3291*	0	0	0	0	0	

*- Vapor–Liquid critical point

Volume 7 Issue 10, October 2018

www.ijsr.net



Figure 5: Entropy change in liquid–vapor phase transition of saturated lithium

5. Free energy Change in the Liquid-Vapor Phase Transition of Alkali Metals

The free energy change in the liquid-vapor phase transition is given by [46]

$$\Delta F = \Delta U - \Delta H_{\nu}$$
(16)
The internal energy change is given by

$$\Delta U = \frac{a}{(n-1)} \left(\frac{1}{V_l^{n-1}} - \frac{1}{V_g^{n-1}} \right)$$
(17)

The enthalpy of vaporization is given by

$$\frac{\Delta H_{\nu}}{T(V_g - V_l)} = \frac{dP}{dT}$$
(18)

Eq. (18) is rewritten as

$$\Delta H_{v} = \frac{RT}{V_{l} - b} \left(V_{g} - V_{l} \right) \tag{19}$$

Finally the free energy is given by

$$\Delta F = \int_{V_l}^{V_s} \frac{a}{V^n} dV - \frac{RT}{V_l - b} \left(V_g - V_l \right)$$
(20)

6. Determination of Free Energy Change in Liquid-Vapor Transition of Fluid Alkali Metals

The vapor and liquid volumes of fluid alkali metals at saturation have been determined [46] through the generalized van der Waals equation of state. These data on the vapor and liquid volumes at saturation, the values of the parameter b and n for alkali metals are used to determine the internal energy change in liquid-vapor transition through Eq. (17). The free energy change in liquid-vapor transition of fluid alkali metals is determined using Eq. (20) in which the temperature-dependence of the volume constant b is taken into account. The obtained values are presented in Tables 7-11. The obtained temperature dependence of the internal energy in liquid-vapor transition for fluid alkali metals have been plotted in Figures 6-10. The results yielded by the generalized van der Waals equation of state of internal energy

 Table 7: Free energy change in liquid–vapor phase transition of saturated cesium

<i>Т</i> , К	ΔF kJ/mol
1795	-21.965
1805	-20.838
1814	-19.915
1824	-18.747
1834	-17.714
1843	-16.259
1853	-15.184
1863	-13.888
1872	-12.389
1882	-11.031
1891	-9.3117
1901	-7.7406
1911	-5.8966
1920	-3.7357
1930*	0





Figure 6: Free energy change in liquid–vapor phase transition of saturated cesium

 Table 8: Free energy change in liquid–vapor phase transition of saturated rubidium

Т,К	ΔF kJ/mol
1897	-21.7296
1907	-20.4913
1917	-19.2573
1927	-17.8438
1937	-16.7723
1947	-15.0122
1958	-13.6493
1968	-11.7466
1978	-9.7203
1988	-8.3344
1998	-6.2108
2008	-3.7759
2018*	0

*-Vapor–Liquid critical point



Figure 7: Free energy change in liquid–vapor phase transition of saturated rubidium

Volume 7 Issue 10, October 2018

<u>www.ijsr.net</u>

 Table 9: Free energy change in liquid-vapor phase

 transition of saturated potassium

transition of saturated polassium				
Т, К	ΔF kJ/mol			
2118	-20.9169			
2129	-19.8347			
2140	-18.3465			
2152	-17.9727			
2163	-17.2074			
2175	-15.9819			
2186	-14.9881			
2197	-13.7293			
2207	-13.2424			
2220	-11.0834			
2232	-9.2774			
2243	-7.9394			
2254	-6.2678			
2266	-3.7036			
2277*	0			

*- Vapor-Liquid critical point



Figure 8: Free energy change in liquid–vapor phase transition of saturated potassium

Table 10:	Free energy	change	in lic	quid-vapor	phase
	transition o	f saturat	ed so	odium	

transition of saturated sourum			
<i>Т</i> , к	ΔF kJ/mol		
2194	-29.7224		
2219	-28.7191		
2244	-27.6567		
2256	-27.0437		
2269	-26.428		
2281	-25.6999		
2294	-24.9593		
2306	-24.0218		
2319	-23.2499		
2331	-22.4073		
2343	-21.4127		
2356	-20.0981		
2368	-19.2585		
2381	-18.1363		
2393	-16.7913		
2406	-15.5034		
2418	-13.9919		
2431	-11.9469		
2443	-10.7318		
2456	-9.0093		
2468	-6.8569		
2481	-4.0437		
2493*	0*		

*-Vapor-Liquid critical point



Figure 9: Free energy change in liquid–vapor phase transition of saturated sodium

Table 11: Free energy change in liquid-vapor phas	e
transition of saturated lithium	

Τ, к	ΔF kJ/mol
3125	-20.2447
3231	-18.9019
3248	-15.4235
3264	-11.1156
3281	-6.1286
3297*	0

*-Vapor–Liquid critical point



Figure 10: Free energy change in liquid–vapor phase transition of saturated lithium

7. Results and Discussion

The generalized van der Waals equation of state has been employed to determine the entropy change and free energy change in liquid-vapor transition of fluid alkali metals in the range of temperatures from their boiling point to critical point. Moreover, the entropy change and free energy in liquid-vapor transition of fluid alkali metals has also been determined taking into consideration the temperaturedependence of the volume constant in the generalized van der Waals equation of state.

As seen from Figures 1-5, the volume constant in the generalized van der Waals equation of state weakly depends on temperature for fluid alkali metals. Hence, the temperature-dependence of the volume constant may be neglected for practical purposes. As seen from Tables 2-6 and Figures 1-5, the values of entropy change and free energy in liquid-vapor transition for fluid alkali metals agree well with the values given by CK correlation, FL correlation and Watson correlation in the high temperature region.

Volume 7 Issue 10, October 2018

<u>www.ijsr.net</u>

The generalized van der Waals equation of state has also been established [42] to describe the thermodynamic properties of fluid alkali metals in the temperatures range from critical point to boiling point. Therefore, the values of entropy change and free energy in liquid-vapor transition determined on the basis of the generalized van der Waals equation of state may be considered to be the reliable and recommended values.

8. Conclusion

The entropy change and free energy change in liquid-vapor transition of fluid alkali metals, in the temperatures range from boiling point to critical point have been determined using the generalized van der Waals equation of state. The obtained values of entropy change and free energy change in liquid-vapor transition of fluid alkali metals agree well with the values yielded by CK correlation, FL correlation and Watson correlation in the high temperature region.

As the generalized van der Waals equation of state describes the thermodynamic properties of fluid alkali metals in the temperatures range from boiling point to critical point, the values of entropy change and free energy change in liquidvapor transition determined on the basis of the generalized van der Waals equation of state may be considered to be the reliable and recommended values.

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