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Corresponding States Theory and Thermodynamic Properties of Liquid Alkali Metals

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According to phenomenological scaling and the law of corresponding states, reduced coordinates F^*-T^*, where F^* represents the reduced thermodynamic properties (enthalpy of vaporization, speed of sound, surface tension, saturated liquid density) and T^* is the reduced temperature, are introduced for the prediction of the thermodynamic properties of alkali metals. Values of the thermodynamic properties from the melting point up to boiling point are correlated. It has been shown that the correlation between reduced thermodynamic properties, as well as with the reduced temperature, can be expressed as a unique straight-line plot with a linear correlation coefficient of 0.9998. The proposed correlation has a simple form for easy calculation, requires only the melting and boiling point parameters, which are usually easy to acquire, and can predict the thermodynamic properties from the melting temperature up to the boiling temperature accurately.

Keywords: Corresponding states, Alkali metals, Thermodynamic properties

INTRODUCTION

The central idea of the corresponding states theory (CST) is the existence of a universal relation between dimensionless physical quantities formed using the physical quantities of interest. The existence of such a relation may be established by a dimensional analysis or by use of a mathematical equation, if one exists, connecting the relevant quantities [1]. An example of the latter is the van der Waals equation of state from which an explicit expression between the reduced quantities (pressure, volume and temperature) can be obtained [1-3].

The original, two-parameter corresponding-states principle leads to an equation of state which expresses the residual compressibility factor, Z^r, in terms of a universal function of the dimensionless temperature and molar volume (or density):

$$Z^r = \frac{PV}{RT} - 1 = h\,(V^*, T^*) \tag{1}$$

where P is the pressure, R is the gas constant and V^* and T^* are the dimensionless volume and temperature, respectively. Starting from a molecular basis, V^* would be identical to $V/N\sigma^3$ where σ is the intermolecular-potential distance parameter and N is the Avogadro constant. T^* would be given by k_BT/ε where k_B is Boltzman's constant and ε is the intermolecular-potential well depth. If one invokes the stability criteria for a pure-fluid critical point, the dimensionless volume and temperature would be given by V/V_c and T/T_c, respectively, where the subscript c denotes a value at the critical point. We note that this two-parameter corresponding-states principle can be applied to any polynomial equation of state that has a liquid-vapor critical point [4].

In recent years, there has been a spurt of research activity involving the use of the principles of corresponding states for

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the prediction of transport and the other physical properties of ordinary liquids. Indeed, this approach has led to the development of useful predictive expressions for ordinary liquids [5]. However, the applicability of the principles of corresponding states to molten metals has been explored in a rather scant manner [6-8]. In this paper it is demonstrated that an existing formulation for the prediction of the physical properties of liquids can indeed be modified to encompass molten alkali metals. The reliability and utility of the modification proposed herein is established by performing detailed comparisons of the experimental values of the physical properties for alkali metals from the melting temperature up to the boiling temperature.

CORRELATION AND RESULTS

The phenomenological reduced coordinates are defined as:

$$T^* = \left(\frac{T_b - T}{T_b - T_m}\right)\left(\frac{T_b}{T}\right) \tag{2}$$

and

$$F^*(T) = \left(\frac{F(T)}{F_m}\right)\left(\frac{T_b}{T}\right) \tag{3}$$

where T_b, is the boiling temperature. $F(T)$ and F_m are the thermodynamic properties at temperature T and at the melting temperature, T_m, respectively. The experimental data were taken from tabulations by Vargaftik [9] to construct the correlations for alkali metals. Dimensionless quantities were introduced by choosing the corresponding quantities at the melting temperature ($F^* = F(T)/F(T_m)$). When we added the terms, T_m/T, T_b/T, or T_c/T separately to F^*, the equation that gave the best result was

$$F^* = \frac{F(T)}{F(T_m)}\frac{T_b}{T}$$

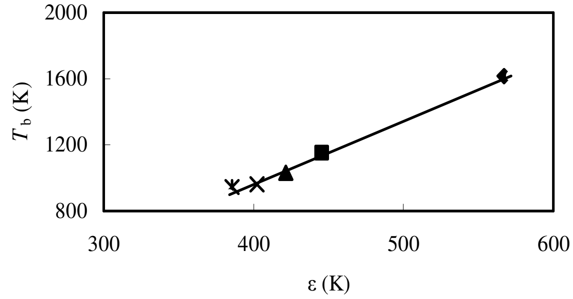

Fig. 1. Plot of T_b with ε for liquid alkali metals: (◆) Li, (■) Na, (▲) K, (×) Rb, (∗) Cs.

Figure 1 shows the linear relation between T_b and ε for alkali metals. One distinct advantage of this set is that these are standard parameters that are readily available. The normal boiling temperature, T_b, is a constant that characterizes the vaporization process and the different intermolecular forces. T_b is a corresponding temperature for alkali metals.

Universal Behavior of Enthalpy of Vaporization

We are interested in the general relationship between the enthalpy of vaporization of liquid alkali metals and any of their other properties, which would permit us to predict the enthalpy of vaporization from the melting point up to the boiling point. Because the boiling points of most of the alkali metals have been reliably determined, we feel that this relationship has a definite advantage over that used by Watson [10], Fish and Lielmezs [11], Meyra [12], Cordfunke, or Konings [13], where the critical temperatures must be used. Our equation is able to correlate the enthalpy of vaporization (ΔH_v) with less dispersion than that of Watson, Fish, or Meyra.

The reduced coordinates for the enthalpy of vaporization are defined as:

$$H^*(T) = \left(\frac{\Delta H_v(T)}{\Delta H_m}\right)\left(\frac{T_b}{T}\right) \tag{4}$$

where $\Delta H_v(T)$ and ΔH_m are the enthalpy of vaporization at temperature T and at the melting temperature T_m, respectively. In Table 1 a summary is given of the values of T_m, T_b and ΔH_m at the melting point. Figure 2 shows the correlations suggested in this work. The solid line in Fig. 2 is the linear fit to the experimental data which was obtained by linear regression as:

$$H^*(T) = 0.7586\ T^* + 0.8762 \tag{5}$$

Table 1. Parameters for Alkali Metals in this Work [9]

Substance	T_m (K)	T_b (K)	ΔH_m (kJ kg^{-1})	γ_m (mJ m^{-2})	ρ_m (kg m^{-3})	u_m (m s^{-1})	$^{\#}\varepsilon$ (K)	$^{\#}\sigma$ (Å)
Li	453.7	1615	22556	398.33	398.33	4500	567.1	2.73
Na	371.0	1151	4537	198.02	198.02	2536	445.6	3.33
K	336.4	1032	2233	109.06	109.06	1868	421.4	4.12
Rb	312.7	959	918	87.47	87.47	1255	402.2	4.41
Cs	301.6	943	563	71.34	71.34	946	385.5	4.76

$^{\#}$Data are taken from Ref. [37].

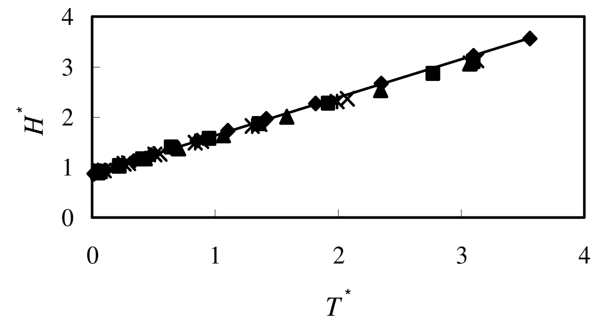

Fig. 2. Correlation results of reduced enthalpy of vaporization, H^*, *vs.* reduced temperature, T^*, for alkali metals. The experimental values are taken from Ref. [9]: (◆) Li, (■) Na, (▲) K, (×) Rb, (∗) Cs.

The linear correlation coefficient for Eq. (5) is 0.9999 and the standard deviation is 0.0132 in the range of melting to boiling temperatures. From Eq. (5), by using definitions (2) and (4) we can write the ΔH_v as:

$$\Delta H_v(T) = 0.7586\left(\frac{T_b - T}{T_b - T_m}\right)\Delta H_m + 0.8762\left(\frac{T}{T_b}\right)\Delta H_m \tag{6}$$

The values of T_m, T_b, and $\Delta H_v(T)$ depend on the substance, therefore we have a curve $\Delta H_v(T)$ for each substance. The percentage deviations were calculated in the range of melting temperatures to boiling temperatures for potassium. The results are given in Table 2. The average error is within 0.94%.

Universal Behavior of Surface Tension

The surface tension, γ, of a liquid alkali metal is one of the fundamental and most important quantities in the theory and practice of materials processing (crystal growth, welding, sintering). A theoretical treatment of the first principle is unwieldy and does not always enable one to calculate reliable values of γ as a function of temperature. Another active research field deals with semi-empirical predictions based on the correlation between the surface and bulk thermodynamic properties [14-17]. Empirical relations relate γ with the bulk properties of metals (*e.g.*, melting point, critical temperature, isothermal compressibility) using different approaches [8,18-22].

The phenomenological reduced coordinates are defined as

$$\gamma^*(T) = \left(\frac{\gamma(T)}{\gamma_m}\right)\left(\frac{T_b}{T}\right) \tag{7}$$

where γ and γ_m are the surface tension at temperature T and the melting temperature T_m, respectively. Figure 3 shows the correlations suggested in this work. In Fig. 3, the solid line is the linear fit to the experimental data, which was obtained by linear regression as:

$$\gamma^* = 0.8495\,T^* + 0.5574 \tag{8}$$

The linear correlation coefficient and standard deviation for Eq. (8) are 0.9999, and 0.0144, respectively. From Eq. (8), by using definitions (2) and (7) for the surface tension, γ, we can write:

$$\gamma(T) = 0.8495\left(\frac{T_b - T}{T_b - T_m}\right)\gamma_m + 0.5574\left(\frac{T}{T_b}\right)\gamma_m \tag{9}$$

Table 2. Calculated Enthalpy of Vaporization[a] for Potassium at Different Temperatures

T (K)	$(\Delta H_v)_{\mathrm{exp}}$ (kJ kg^{-1})[b]	$(\Delta H_v)_{\mathrm{calc}}$ (kJ kg^{-1})	Deviation (%)[c]
336	2233.0	2269.8	1.65
400	2196.4	2240.0	1.99
450	2180.6	2216.6	1.65
500	2164.9	2193.1	1.30
550	2148.1	2169.7	1.00
600	2129.8	2146.2	0.77
650	2110.0	2122.8	0.61
700	2088.6	2099.3	0.51
750	2066.1	2075.9	0.47
800	2042.6	2052.4	0.48
850	2018.3	2028.9	0.53
900	1993.4	2005.5	0.61
950	1968.2	1982.1	0.71
1000	1942.3	1958.7	0.84
Overall			0.94

[a] $(\Delta H_v)_{\mathrm{calc}}$ calculated with equation (6). [b] $(\Delta H_v)_{\mathrm{exp}}$ are the experimental values [9].

$$^{c}\,Deviation\% = ABS\left[\frac{calc. - \exp.}{\exp.}\right] \times 100$$

Table 3. Calculated Surface Tension[a] for Potassium at Different Temperatures

T (K)	γ_{exp} (mJ m^{-2})[b]	Deviation (%)[c]	Chhabra [23]	Ghatee [24]	Digilov [25]
336	109.07	0.43	11.99	0.03	0.01
400	104.86	0.49	11.34	0.22	0.23
450	101.56	0.54	10.79	0.37	0.37
500	98.26	0.59	10.21	0.54	0.48
550	94.95	0.64	9.58	0.72	0.54
600	91.65	0.70	8.91	0.91	0.56
650	88.35	0.76	8.18	1.11	0.52
700	85.05	0.83	7.41	1.33	0.42
750	81.75	0.91	6.57	1.57	0.26
800	78.45	0.99	5.66	1.83	0.03
850	75.15	1.07	4.67	2.12	0.29
900	71.85	1.17	3.59	2.42	0.71
950	68.55	1.27	2.40	2.76	1.24
1000	65.24	1.38	1.09	3.13	1.91
Overall		0.85	7.31	1.36	1.90

[a] γ_{calc} calculated with Eq. (9). [b] $\gamma_{\mathrm{exp.}}$is the experimental value [9]. [c] This work.

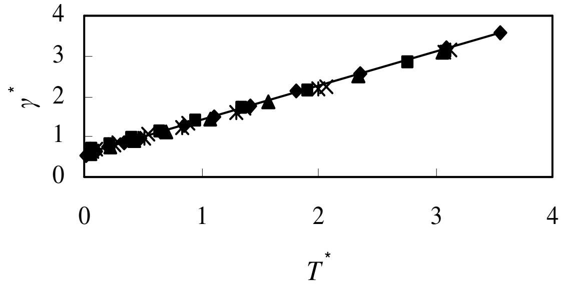

Fig. 3. Correlation results of reduced surface tension, γ^*, *vs.* reduced temperature, T^*, for alkali metals: (◆) Li, (■) Na, (▲) K, (×) Rb, (∗) Cs.

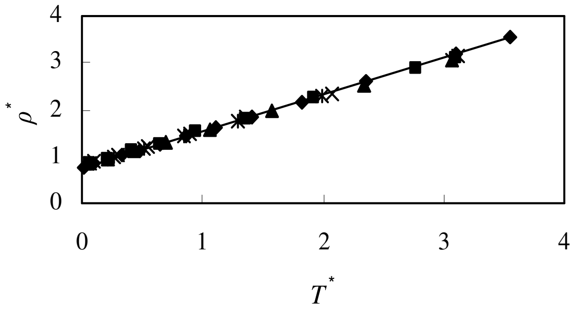

Fig. 4. Correlation results of reduced density, ρ^*, *vs.* reduced temperature, T^*, for alkali metals: (◆) Li, (■) Na, (▲) K, (×) Rb, (∗) Cs.

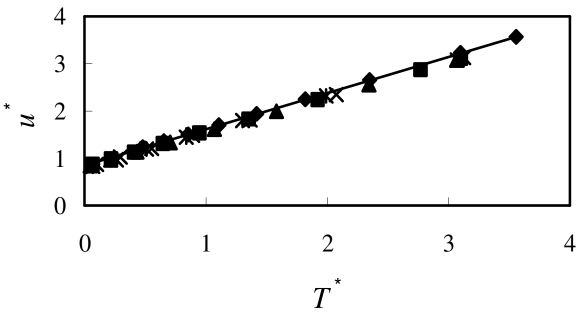

Fig. 5. Correlation results of reduced speed of sound, u^*, *vs.* reduced temperature, T^*, for alkali metals: (◆) Li, (■) Na, (▲) K, (×) Rb, (∗) Cs.

In Table 3 the percentage deviations were calculated in the range from the melting point to the boiling point for potassium. The same calculations were made for the data of Chhabra [23], Ghatee [24], and Digilov [25]. It is apparent from Table 3 that our correlation results are in good overall agreement with the experimental data over the entire temperature range.

Universal Behavior of the Saturation Liquid Density

Alkali metals and alloys have a unique combination of physicochemical properties, such as low densities and viscosities, low melting temperatures and work functions, wide temperature ranges in the liquid state, high values for heat of evaporation, etc. Therefore, they are widely used in modern science and technology [26,27]. The recent assessment of alkali metal liquid density, ρ, by Bystrov *et al.* [28] gives a seven-term polynomial. Figure 4 shows the correlations suggested in our work. In Fig. 4, the solid line is the linear fit to the experimental data which was obtained by linear regression as:

$$\rho^* = 0.7811\, T^* + 0.7609 \tag{10}$$

The linear correlation coefficient and standard deviation for Eq. (10) are 0.9999, and 0.0073, respectively.

Universal Behavior of the Speed of Sound

Bystrov *et al.* [28] represent the speed of sound, u, with a linear equation, which they extrapolate to the critical point. Deviations from Bystrov *et al.* [28] are within 1.3% through 1700 K for sodium. Above 1700 K, deviations increase with temperature. Figure 5 shows the correlations suggested in our work. In Fig. 5, the solid line is the linear fit to the experimental data, which was obtained by linear regression as:

$$u^* = 0.7585\, T^* + 0.8634 \tag{11}$$

The linear correlation coefficient and standard deviation for Eq. (11) are 0.9999, and 0.0056, respectively.

Universal Behavior of Thermodynamic Properties Interacting with Each Other

The enthalpy of vaporization has already been correlated with many physical properties, such as relationships involving liquid volume on the saturation line [29], and surface tension [30]. Informative and exhaustive reviews of the theoretical as well as experimental developments in this area are available in the literature [8,31-34].

Both the process of vaporization of a liquid and the process of surface formation of a liquid are correlated with the process of overcoming the intermolecular forces existing within a liquid. Admixing of liquids is controlled not only by the molecular interactions but also by the interfacial free energy of the mixing partners. The interfacial tension of a solid-liquid determined from the rate of nucleation has a role similar to the activation energy of a chemical reaction [35].

Figure 6 shows the relation between the quantities γ^* and H^* for the liquid alkali metals. As can be seen, the correlation of coordinates is good. The solid line in Fig. 6 is the linear fit to the experimental data, which was obtained by linear regression as:

$$\gamma^* = 1.0889\, H^* - 0.3502 \tag{12}$$

The linear correlation coefficient for Eq. (12) is 0.9999. The percentage deviations calculated for alkali metals are given in Tables 4-7. As is evident from Fig. 6, points for different alkali metals fall well on a common curve. This set of reduced coordinates can predict surface tension well within 2.2%, which is roughly the order of experimental accuracy, *i.e.* the agreement between the predicted and experimental values is regarded as satisfactory and acceptable.

From Eq. (12), and by using definitions (4) and (7), we can write γ as:

$$\gamma = 1.0889\left(\frac{\gamma_m}{\Delta H_m}\right)\Delta H_v - 0.3502\left(\frac{\gamma_m}{T_b}\right)T \tag{13}$$

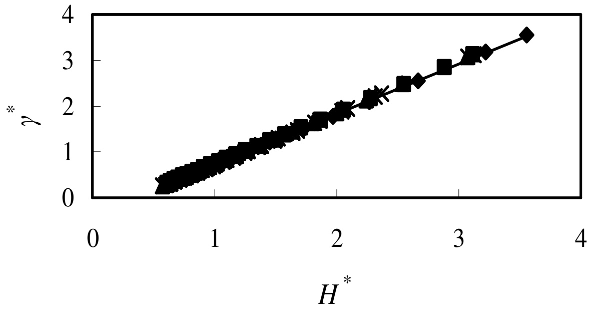

Fig. 6. Correlation results of reduced surface tension, γ^*, *vs.* reduced enthalpy of vaporization, H^*, for alkali metals: (◆) Li, (■) Na, (▲) K, (×) Rb, (∗) Cs.

Table 4. Calculated Surface Tension[a] for Rubidium at Different Temperatures

T (K)	$\gamma_{\text{exp.}}$ (mJ m^{-2}) [b]	$\gamma_{\text{calc.}}$ (mJ m^{-2})	Deviation (%)
313	87.47	86.44	1.17
400	82.41	82.17	0.29
450	79.51	79.65	0.17
500	76.61	77.05	0.58
550	73.71	74.38	0.91
600	70.81	71.61	1.12
650	67.92	68.81	1.32
700	65.02	65.97	1.46
750	62.12	63.08	1.54
800	59.22	60.16	1.59
850	56.32	57.24	1.63
900	53.42	54.33	1.70
950	50.52	51.43	1.79
1000	47.63	48.53	1.89
1100	41.83	42.70	2.09
1200	36.03	36.88	2.35
1300	30.23	31.07	2.75
Overall			1.56

[a] Calculated with Eq. (13). [b] Experimental values are taken from Ref. [9].

Table 5. Average and Maximum Errors in the Prediction of Surface Tension

Metal	n[a]	ΔT (K)	Average error[b]
Li	29	453.7-2000	2.36 (6.85)[c]
Na	24	371.0-1500	3.14 (9.22)
K	22	336.4-1400	0.68 (1.78)
Rb	20	312.7-1300	1.56 (2.75)
Cs	20	301.6-1300	3.26 (6.66)
Overall			2.20

[a] Number of experimental points.

[b] $$Average - Error = \frac{1}{n}\sum_{i=1}^{n} ABS\left[\frac{cal.-\exp.}{\exp.}\right]_i \times 100.$$

[c] Maximum error in parentheses.

Table 6. Calculated Enthalpy of Vaporization[a] for Potassium at Different Temperatures

T (K)	$(\Delta H_v)_{\text{exp.}}$ (kJ kg^{-1})^{b}	$(\Delta H_v)_{\text{calc.}}$ (kJ kg^{-1})	Deviation (%)
336	2233.0	2257.0	1.08
400	2196.4	2226.3	1.36
450	2180.6	2202.2	0.99
500	2164.9	2178.1	0.61
600	2129.8	2129.7	0.01
700	2088.6	2081.5	0.34
800	2042.6	2033.2	0.46
900	1993.4	1984.9	0.42
1000	1942.3	1936.7	0.29
1100	1890.0	1912.5	0.19
1200	1836.9	1840.1	0.18
1300	1783.6	1791.8	0.46
1400	1730.8	1743.6	0.74
Overall			0.45

[a] $(\Delta H_v)_{\text{calc.}}$ was calculated with Eq. (13).

[b] $(\Delta H_v)_{\text{exp.}}$ is the experimental value [9].

Table 7. Average and Maximum Errors in the Prediction of Enthalpy of Vaporization

Metal	n[a]	ΔT (K)	Average error[b]	Watson [9]
Li	29	453.7-2000	1.50 (3.51)[c]	1.07 (1.42)
Na	24	371.0-1500	1.88 (4.51)	1.82 (4.82)
K	22	336.4-1400	0.45 (1.36)	1.84 (4.83)
Rb	20	312.7-1300	0.93 (1.10)	2.01 (4.99)
Cs	20	301.6-1300	1.80 (2.54)	2.99 (7.03)
Overall			1.31	1.95

[a] Number of experimental points. [b] This work. [c] Maximum error in parentheses.

The values of γ_m and ΔH_m depend on the substance, therefore we have a curve $\gamma(\Delta H_v, \mathrm{T})$ for each substance. In Fig. 7 we represent Eq. (13) and the corresponding experimental data for alkali metals. There, it can be seen that the transformation does not introduce significant differences, giving a very good correspondence with the experiments.

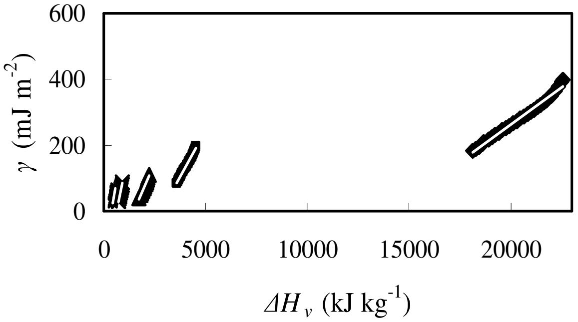

Fig. 7. Surface tension as a function of enthalpy of vaporization. The points and solid lines represent experimental values [9] and values from the correlation, Eq. (14), respectively: (◆) Li, (■) Na, (▲) K, (×) Rb, (∗) Cs.

Tables 6 and 7 show values of ΔH_{v} calculated with the use of Eq. (13), in the temperature range from the melting point to several hundred degrees above the boiling point. Comparison with experimental data shows the good quality of this correlation with an average error within 1.31%.

Table 8 shows the relation between other thermodynamic properties (H^*-ρ^*, H^*-u^*, γ^*-ρ^*, γ^*-u^*, u^*-ρ^*). The linear correlation coefficients and standard deviations are within 0.9998, and 0.0153, respectively. Figures 8-12 show the relation between the thermodynamic properties for the liquid alkali metals. As can be seen, the correlation of coordinates in all cases is good.

DISCUSSION

It has been shown that the relationship between reduced thermodynamic properties, as well as with the reduced temperature, can be expressed as a unique straight-line plot with a linear correlation coefficient of 0.9998. In this work we employ $k_{\text{B}}T_{\text{b}}$ as the characteristic energy and no molecular parameters are involved. The proposed correlation has a simple form for easy calculation, requiring only the melting and boiling point parameters, which are usually easy to acquire, and can predict the thermodynamic properties up to the boiling temperature accurately. In the dimensionless representation $F^*(T)$, there is a unique function for alkali metals (Figs. 2-12). With dimensions, $F(T)$ gives a curve for

Table 8. The Linear Regression of Correlation between Thermodynamic Properties

Set	Linear regression	Linear correlation coefficient	Standard deviation
$\gamma^* - H^*$	$\gamma^* = 1.0889\,H^* - 0.3502$	0.9999	0.0145
$H^* - \rho^*$	$H^* = 0.9711\,\rho^* + 0.1374$	0.9998	0.0179
$H^* - u^*$	$H^* = 0.9900\,u^* + 0.0394$	0.9999	0.0126
$\gamma^* - \rho^*$	$\gamma^* = 1.0874\,\rho^* - 0.2699$	0.9998	0.0203
$\gamma^* - u^*$	$\gamma^* = 1.1110\,u^* - 0.3843$	0.9998	0.0161
$u^* - \rho^*$	$u^* = 0.9690\,\rho^* - 0.1298$	0.9999	0.0117

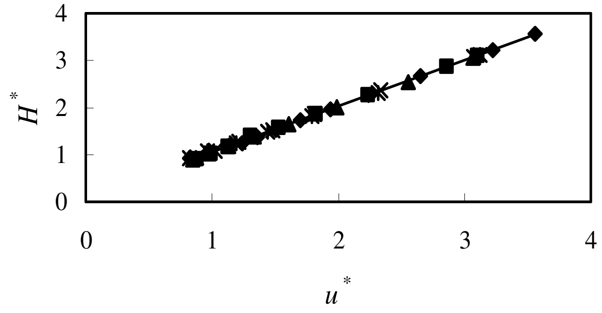

Fig. 8. Correlation results of reduced enthalpy of vaporization, H^*, *vs.* reduced speed of sound, u^*, for alkali metals: (◆) Li, (■) Na, (▲) K, (×) Rb, (∗) Cs.

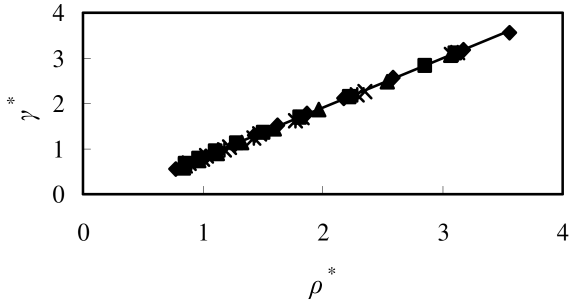

Fig. 10. Correlation results of reduced surface tension, γ^*, *vs.* reduced density, ρ^*, for alkali metals: (◆) Li, (■) Na, (▲) K, (×) Rb, (∗) Cs.

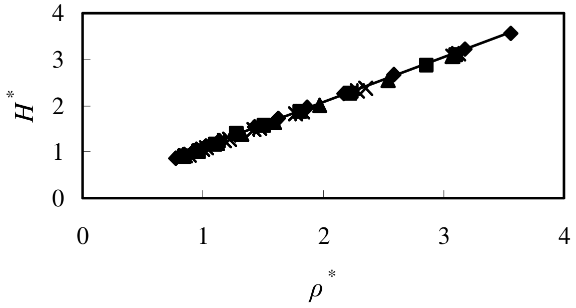

Fig. 9. Correlation results of reduced enthalpy of vaporization, H^*, *vs.* reduced density, ρ^*, for alkali metals: (◆) Li, (■) Na, (▲) K, (×) Rb, (∗) Cs.

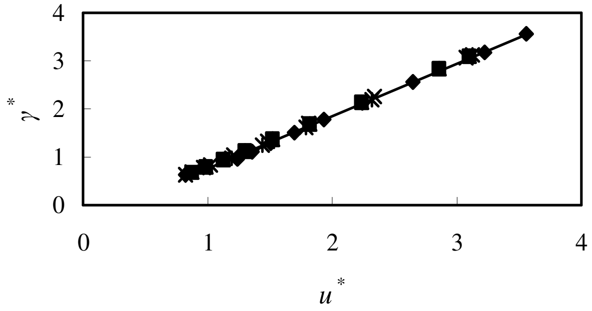

Fig. 11. Correlation results of reduced surface tension, γ^*, *vs.* reduced speed of sound, u^*, for alkali metals: (◆) Li, (■) Na, (▲) K, (×) Rb, (∗) Cs.

each metal resulting in good correlation with the experimental data (Fig. 7). Tables 2-7 show a comparison between experimental results and those calculated.

We have proposed a simple accurate correlation for liquid alkali metals. The correlation uses more readily available input data than that of the critical point data. Most existing correlations for the prediction of the thermodynamic properties of ordinary fluids [2] use the critical point data. For

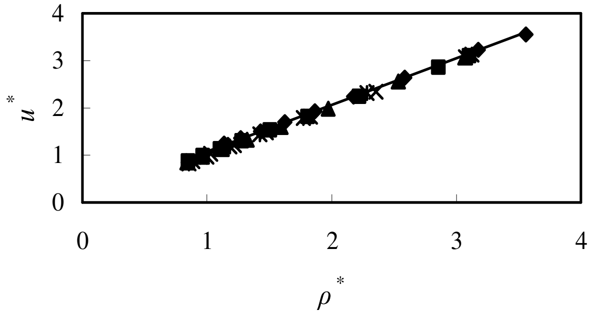

Fig. 12. Correlation results of reduced speed of sound, u^*, *vs.* reduced density, ρ^*, for alkali metals: (◆)Li, (■) Na, (▲) K, (×) Rb, (∗) Cs.

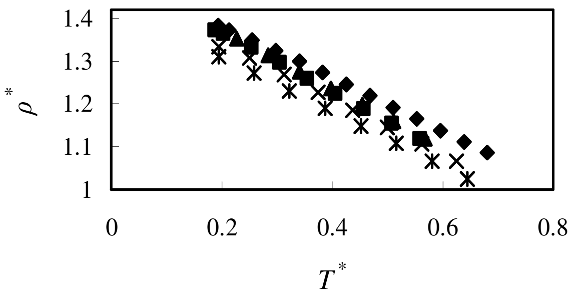

Fig. 13. Correlation results of reduced density, ρ^*, *vs.* reduced temperature, T^*, for alkali metals using Shyu parameters: (◆) Li, (■) Na, (▲) K, (×) Rb, (∗) Cs.

these metals, there is no reasonable consistency for critical parameters between different reported values [36]. Also the correlation shows how the successful empirical regularities can be obtained from a simple equation. As far as our results show, the present correlation could be applicable to other liquid metals, but lack of experimental data hinders us from testing it.

The use of the potential parameters of Shyu *et al.* [37] as characteristic parameters did not give better results for alkali metals (Fig. 13). Shyu's potential parameters provided the best correlation of transport coefficients [2] compared to other sets of parameters. The potential well depth and the inter-ionic distance are not applicable to the CST. First, both of these quantities are temperature dependent. Furthermore, they are very sensitive to the inputs used for the potential calculations.

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