

# New Method of Correlating the Vapor Pressures of Alkali Metals over the Temperature Range 400° to 2500° F.

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Existing vapor pressure values of four alkali metals—sodium, potassium, cesium, and rubidium—below their normal boiling points are shown to be inconsistent and of questionable reliability, while those at higher temperatures are in good agreement. The high-temperature values are used to generate more acceptable values at lower temperatures by the technique of computing and correlating saturation pressures for the hypothetical ideal gas.

IN RECENT YEARS a flood of vapor-pressure measurements for the alkali metals has rather accurately defined saturation pressures from the normal boiling points to about 2500° F. In contrast, the existing vapor-pressure information at lower temperatures is still scarce and/or inconsistent. Reliable saturation data below the normal boiling points are needed to define thermodynamic properties for use in the design of heat pipes, turbines, and other heat-transfer and power-generation systems.

The saturated vapor of an alkali metal approaches ideality only at temperatures below 400° F. Spectroscopic information (12) and compressibility studies (16) have shown that the saturated vapor at higher temperatures consists of a mixture of monomeric and higher molecular weight species. This association in a vapor is reflected in its saturation pressure and must be taken into account in any correlation or comparison of vapor-pressure information over an extended temperature range.

The authors have recently published equations of state for four alkali metals—sodium (14), potassium (13), cesium (15), and rubidium (50). With this information it is possible to interrelate the saturation pressures for these metals above and below the normal boiling points, and to use the vapor-pressure measurements at higher temperatures as a guide to select and define reliable values at lower temperatures.

## ANALYSIS OF VAPOR-PRESSURE DATA

In recent years, investigators (29, 47) have studied the temperature dependency of the saturation pressure of the alkali metals and other liquids which exhibit an association of atoms in the vapor state. Because of the complexity of the pressure-temperature relationships derived in these studies, an alternative method was selected for the present analysis. This involved a conversion of saturation pressures for the real gas to those for a hypothetical perfect gas; an analysis and correlation of these ideal quantities with a single pressure-temperature relationship; a selection of best values over the temperature range; and, finally, a reconversion of the selected ideal quantities to those for the real gas.

With imperfection information from an equation of state, the pressure,  $p_i$ , of the ideal metal vapor may be calculated at any temperature for which the saturation pressure of the real gas is known. The following relationships may be derived by the method outlined by Hicks (28).

$$\ln p_i = \ln p_s - \ln \frac{p_s \bar{V}}{RT} + \frac{2B}{\bar{V}} + \frac{3C}{2\bar{V}^2} + \frac{4D}{3\bar{V}^3} + \frac{5E}{4\bar{V}^4} \quad (1)$$

$$\ln p_i = \frac{M_1}{M_a} \ln p_s - \frac{x_2}{2} \ln k_2 - \frac{x_4}{4} \ln k_4 + \frac{M_1}{M_a} [N_1 \ln N_1 + N_2 \ln N_2 + N_4 \ln N_4] \quad (2)$$

Imperfection terms in Equation 1 are based on the virial equation of state in the volume expansion form. A term in this equation for the compressibility of the liquid is small and was ignored. The imperfection terms in Equation 2 are based on a specific type of quasichemical equation of state as reported by Ewing *et al.* (16), in which the metal vapor is treated as an ideal mixture of monomeric, dimeric, and tetrameric species. The second equation may be reduced, as would be expected from Dalton's law, to the simple form,  $p_i = N_1 p_s$ .

The equations of state also permit a third-law calculation of the enthalpy of vaporization of the ideal monomeric gas at any observed saturation pressure. This quantity, which was used as a criterion for consistency in this analysis, was calculated from the following relationship,

$$(\Delta H \bar{v})_c = - \left[ \left( \frac{f^\circ - h \bar{v}^\circ}{T} \right)^6 - \left( \frac{f^\circ - h \bar{v}^\circ}{T} \right)^4 \right] T - \frac{RT}{M_1} \left( \frac{2B}{\bar{V}} + \frac{3C}{2\bar{V}^2} + \frac{4D}{3\bar{V}^3} + \frac{5E}{4\bar{V}^4} + \ln p_s - \ln \frac{p_s \bar{V}}{RT} \right) \quad (3)$$

The equation is in the virial form and can likewise be derived by the method of Hicks (28). The free-energy functions of the monomeric gas and liquid required for the computations with this equation were those selected in previous studies (13, 14, 15, 50).

If the calculated pressures ( $p_i$ ) of the ideal gas are to be useful in relating vapor-pressure results, a simple equation which accurately represents the quantity over a wide temperature range is needed. For an ideal gas at temperatures well below the critical point, there are two well-known forms of the vapor pressure equation which have been generated from phase equilibrium considerations. The first form (22)

$$\ln p_i = I_c - (\Delta H \bar{v})_c / RT + \frac{(C_p^\circ - C_p)}{R} \ln T \quad (4)$$

was obtained by assuming that  $(C_p^\circ - C_p)$  is independent of temperature. The alternate form (57, 58)

$$\ln p_i = I_c - (\Delta H \bar{v})_c / RT + \frac{G_c}{R} \ln T + \frac{H_c}{R} T + \frac{J_c T^2}{2R} + \frac{M_c T^3}{3R} \quad (5)$$

was obtained by using a polynomial equation with the coefficients  $G_c$ ,  $H_c$ ,  $J_c$ , and  $M_c$  to represent the molar specific-heat difference.

Equation 4 was selected in this study to relate the vapor pressure of the perfect gas with temperature. One could equally well have used Equation 5 by assigning values to the heat-capacity coefficients. This was done for one of the metal vapors using heat-capacity coefficients selected in a previous study (13). Each equation in turn was used for potassium to correlate vapor pressures of the ideal gas above the normal boiling point and to extend this information to lower temperatures. Corresponding values of  $p_i$  extrapolated to lower temperatures with the two equations were compared. A maximum difference of 4% occurred at 400° F. with differences of less than 1% from 500° F. to the normal boiling point at 1394° F. Since the extrapolated pressures with the two equations exhibited only small differences and since the liquid heat-capacity information at higher temperatures is incomplete, one is not justified in using the additional heat-capacity coefficients. The quantity  $(C_p^g - C_p^l)/R$  was taken to be independent of temperature and the equation form

$$\log p_i = A_c - B_c/T + C_c \log T \quad (6)$$

was used for the correlation of  $p_i$  in this study.

For each metal, at temperatures above the normal boiling point there were several sets of saturation data in good agreement. The initial step in this analysis was to select for each metal one representative set of results. For three metals—sodium, potassium, and cesium—the results of Stone *et al.* (52) covered a wide temperature range and were representative of the bulk of the determinations. For rubidium, the measurements of Tepper (54) covered the widest temperature range and were most representative of existing data. These four sets of data were, therefore, selected as representative of all results above normal boiling points.

For each selected set of saturation pressures, values of  $p_i$  were computed with Equation 1 at all temperatures for which saturation pressures were reported, and the least squares method was used to derive the three coefficients in Equation 6. For rubidium the precision of Tepper's data was not sufficient to define a value for the coefficient  $C_c$ . Therefore, the similarity principle was used to select an arbitrary but consistent value of 1.2.

The coefficients from the least squares equations for  $p_i$  are compared with their theoretical values (Equation 4) in Table I. Although there is an interrelationship through vapor pressure between the theoretical value,  $(\Delta H_0^g)_v/R$ , of the coefficient  $B_c$  for each metal and its corresponding empirical value, the excellent agreement between the two provides a good check on internal consistency. To allow a comparison of coefficient  $C_c$  with its theoretical counterpart, the values of  $(C_p^g - C_p^l)/R$  at the normal boiling points of the metals are used in place of the integrated quantities. The theoretical values are, therefore, only approximations, but they show that the coefficients are of the proper sign and magnitude.

The next step in the analysis involved a graphical plot for each metal of  $(\log p_i - C_c \log T)$  vs.  $1/T$  using values computed for all existing real gas data above and below the normal boiling point. A consistent value of coefficient  $C_c$  derived from the representative measurements above 1 atm. was used for all data. Experimental, rather than smoothed, values were used in computing the required  $p_i$ , except in the few cases where investigators reported their results only in smoothed form. Some of the measurements at low temperatures were obtained by effusion or transpiration methods in which the authors assumed a perfect gas. Before any data of this type were used, corrections were made for the effect of the molecular weight of the vapor

Table I. Comparison of Empirical Values of Coefficients in  $p_i$  Equations with Their Theoretical Values

Metal	Coefficient $A_c$ , Empirical Value from Eq. 6	Coefficient $B_c$		Coefficient $C_c$	
		Empirical value from Eq. 6	$(\Delta H_0^g)_v/R$	Empirical value from Eq. 6	$(C_p^g - C_p^l)/R$
Sodium	8.8126	10,232	10,080 (14)	-1.1859	-1.0 (21)
Potassium	8.6853	8,505	8,513 (13)	-1.2642	-1.2 (10)
Cesium	8.1636	7,305	7,329 (15)	-1.2036	-1.5 (36)
Rubidium	8.2817	7,680	7,706 (50)	-1.200 <sup>a</sup>	-1.0 (1)

<sup>a</sup> Value estimated from those for other metals.

using the virial equation of state and the method outlined by Evans *et al.* (12). These corrections were generally small.

The simplest procedure with each graphical plot would have been to fit the most consistent linear curve to the existing measurements for the full temperature range, using the third law as a guide. However, inconsistencies in the observed data at lower temperatures would not permit this. It appeared likely that more reliable and more consistent saturation pressures at lower temperatures could be obtained for all the metals by a linear extrapolation of the representative higher temperature data. It was necessary to justify this procedure by third-law calculations and by comparing pressures obtained in this manner with those directly observed.

Saturation pressures for each metal were generated for the full temperature range from the representative higher temperature data by computing values for the ideal gas from Equation 6 (numerical coefficients from Table I) and converting these with Equation 1 to corresponding values for the real gas. The selected and extended saturation pressures so obtained were analyzed and compared to existing experimental values.

**Sodium.** The selected saturation pressures for sodium are compared graphically to existing experimental measurements in Figure 1. The selected values are taken as a standard, and the per cent deviation of the vapor pressure reported from that selected is plotted as a function of temperature. The deviation curve for each set of results was obtained by drawing the best curve through the deviations of the individual experimental values. No comparison has been made for those experimental results (20, 27, 40, 46) which exhibit large positive and negative deviations. Close inspection of Figure 1 reveals that many of the more recent measurements, even though they are mostly at higher temperatures, effectively overlap the temperature range of two groups of measurements at lower temperatures. It is

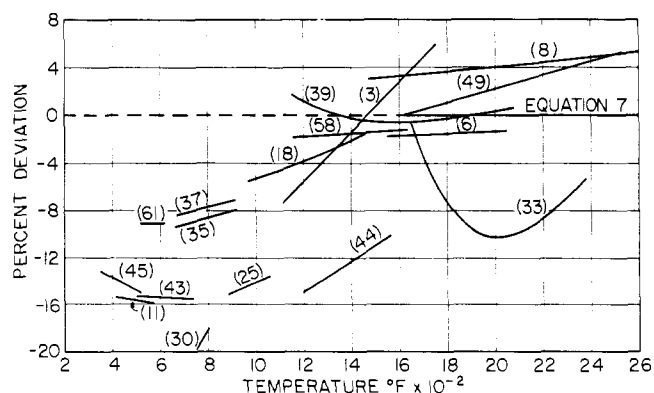


Figure 1. Deviations of experimental vapor pressure of sodium from recommended Equation 7

difficult to explain why most of the low-temperature measurements made before 1936 are 8 to 16% lower than the recent measurements. However, it is known that alkali metals with significantly higher purity have been available in recent years, and one cannot help but suspect that the purity of the metal must be an important factor in the large discrepancy.

It is extremely unlikely that the large group of recent measurements could be in error by as much as 8%. Therefore, the more reliable saturation pressures at lower temperature would appear to be those based on an extrapolation of the higher temperature results. This selection is further justified by a third-law analysis of the extrapolated and measured results at lower temperatures. Using the selected values, the computed vaporization quantity  $(\Delta H_8)_v$  exhibits only a slight change (1%) over the temperature range from 2550° to 400° F., whereas the corresponding change is 2% or more for a smooth vapor-pressure curve drawn through the experimental results at high and low temperatures.

**Potassium.** A similar comparison of all existing experimental work for potassium is presented as Figure 2. Selected results based on the higher temperature work and derived from Equations 6 and 1 are again used as the basis of comparison. There is even less consistency in the data at lower temperatures for this metal than for sodium. Some of the early measurements are as much as 8 to 12% below those defined by the higher temperature work, while others are significantly higher than the base curve. Again, the saturation pressures selected at lower temperatures appear to be more reliable than the measured quantities. A third-law analysis of the selected vapor pressures shows a uniform increase in the value of  $(\Delta H_8)_v$  of less than 0.4% from 2400° to 400° F.

**Cesium and Rubidium.** A graphical analysis, as described above, was made of the existing data for both cesium and rubidium (Figures 3 and 4, respectively). For each metal there are several consistent measurements at higher temperatures but only a few scattered ones at lower temperatures, and it is believed that the low-temperature values obtained by the extrapolation procedure represent the most reliable values now available. Again this extrapolation procedure is justified to an extent by third-law calculations; the value of  $(\Delta H_8)_v$  changes by only 0.3% for cesium for the temperature range from 2350° to 500° F. and 0.6% for rubidium from 2200° to 500° F.

The saturation pressures for each metal as selected by the arguments above are represented for the full tem-

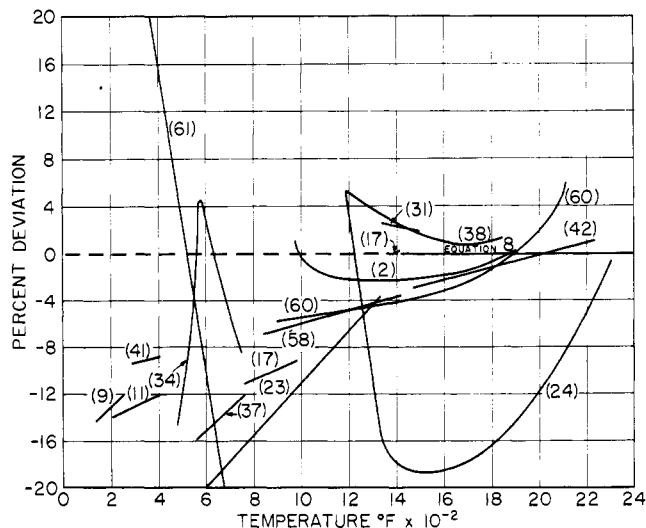


Figure 2. Deviations of experimental vapor pressure of potassium from recommended Equation 8

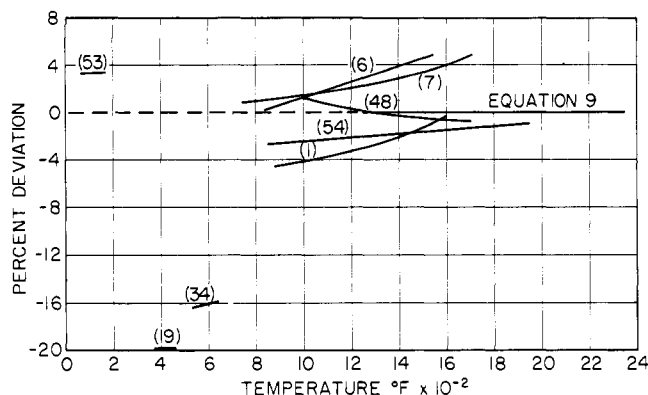


Figure 3. Deviations of experimental vapor pressure of cesium from recommended Equation 9

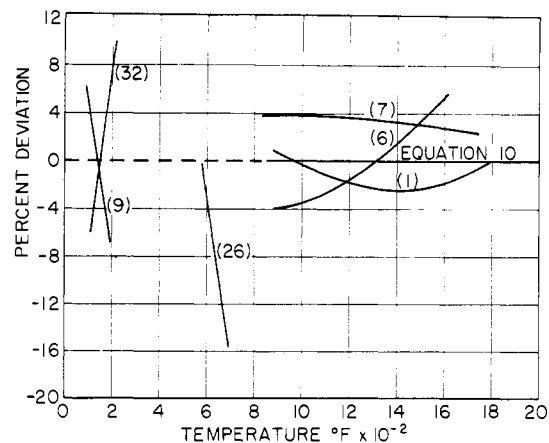


Figure 4. Deviations of experimental vapor pressure of rubidium from recommended Equation 10

perature range by two equations. One is the simple  $p_i$  relationship (Equation 6); but the other, relating  $p_s$  to  $p_i$  (Equation 1), is more complex and requires an equation of state for solution. This mathematical complexity made it desirable to generate simpler relationships for the four metals.

From the work of Vinogradov *et al.* (58) and Thorn and Winslow (57), it appeared that one or more exponential terms would be needed to reflect the influence of association in the vapor-pressure equation, and that saturation pressures over the full temperature range could be fitted by an equation of the type,

$$\log p_s = \log p_i + (D_s/T) (e^{E_s/T})$$

Saturation pressures at intervals of 25° F. were computed for each metal from the more complex relationships. It was found that the values for each metal could be precisely fitted with the appropriate  $p_i$  equation by adding exponential terms to represent the association influence. The following simple equations were obtained by least-squares treatments of the four sets of selected values.

$$\log p_s \text{ (sodium)} = 8.81256 - \frac{10,232.4}{T} - 1.18589 \log T + \frac{3872.7}{T} e^{-7716/T} + 191.9 e^{-1.0431 \times 10^{+8}/T^2} \quad (7)$$

$$\log p_s \text{ (potassium)} = 8.68528 - \frac{8504.7}{T} - 1.26422 \log T +$$

$$\frac{4627.7}{T} e^{-7936.7/T}$$

$$\log p_s (\text{cesium}) = 8.16361 - \frac{7305.1}{T} - 1.20357 \log T + \frac{4942.4}{T} e^{-7608.8/T} \quad (9)$$

$$\log p_s (\text{rubidium}) = 8.28170 - \frac{7680.3}{T} - 1.20000 \log T + \frac{4502.66}{T} e^{-7182.5/T} \quad (10)$$

The degree of fit obtained with each of the simplified vapor-pressure equations is compared to that obtained with two other equational forms in Table II. Although we have tested and recommended each simplified equation for use below its experimental range, meaningful precision indices can be generated only for the experimental range.

For each metal, the table compares the standard deviation of the experimental data from the vapor-pressure equation reported by the original investigators with those from both the complex and the simplified equations generated in this article. It is significant that the standard deviations for a particular metal are almost identical for all three equational forms. This demonstrates clearly that the simplified equation for each metal, as generated in this article and recommended for an extended temperature range, is equivalent over the experimental range to that reported by the original investigators.

## DISCUSSION

The virial equations published by this laboratory were used for all imperfection calculations in this article. These equations were derived (or estimated in the case of rubidium) from compressibility data extending from the normal boiling point to about 2500° F., so an extrapolation of each state equation was required to compute the ideal pressure,  $p_i$ , at all temperatures below the boiling point. It was recognized that other equations of state for potassium, cesium, and rubidium (4, 5, 55, 56, 59) had been published and that many of these were obtained from compressibility information extending to temperatures below the normal boiling point. Even so, the state equations by the present authors were preferred since they were directly based on compressibility data observed at higher temperatures where the deviations from ideality are large. Compressibility information at lower temperatures is gen-

erally subject to larger uncertainties (51), and it was our opinion that imperfection calculations based on an extrapolation of the higher temperature equations of state would be more consistent and more reliable than those based on existing equations of state at lower temperatures. In any event, the degree of association and the corresponding corrections to ideality are small at temperatures below the boiling point.

For sodium and potassium, the authors developed two equations of state—the virial and the quasichemical—from the same compressibility information. The two equations for each metal were used to test the reliability of the extrapolation to lower temperatures. The pressure,  $p_i$ , of the ideal gas was calculated for each metal from about 400° to 2500° F., using experimental saturation pressures in Equations 1 and 2, and the two values at each saturation pressure generally showed an agreement of better than 1%. Therefore, the choice of one state equation over the other is arbitrary, and the more accepted virial form was generally used for the calculations in this article.

One might expect from the standard deviations in Table II that the vapor-pressure equations generated in this article would be exactly equivalent to those published by Stone *et al.* (52) and Tepper (54) over the experimental range of each particular metal. This is true to a point, but the method of correlating  $p_i$  and adding exponential terms to account for the nonideality leads in each case to a slightly different fitting of the saturation data, even though the standard deviations are essentially identical. In comparing equations for a particular metal, one would expect, and find, a slightly differing vapor pressure at a given temperature and a slightly differing enthalpy of vaporization as derived from the Clapeyron equation.

The authors believe that the arguments and procedures outlined have generated the most reliable saturation pressures now available for four of the alkali metals over the temperature range from 400° to about 2500° F. The equations should be most useful in generating consistent thermodynamic quantities for these metals.

## NOMENCLATURE

- $B$  = second virial coefficient, cu. ft./mole
- $C$  = third virial coefficient, (cu. ft.)<sup>2</sup>/(mole)<sup>2</sup>
- $D$  = fourth virial coefficient, (cu. ft.)<sup>3</sup>/(mole)<sup>3</sup>
- $E$  = fifth virial coefficient, (cu. ft.)<sup>4</sup>/(mole)<sup>4</sup>
- $C_p$  = specific heat at constant pressure, B.t.u./lb.-mole° F.
- $f$  = free energy, B.t.u./lb.
- $h$  = enthalpy per unit mass, B.t.u./lb.
- $\Delta h_v$  = enthalpy change upon vaporization of a unit mass at equilibrium, B.t.u./lb.
- $\Delta H_v$  = enthalpy change upon vaporization of a mole of equilibrium vapor, B.t.u./lb.-mole
- $k$  = equilibrium constant
- $M$  = molecular weight
- $N$  = mole fraction
- $p_s$  = saturation pressure, absolute atm.
- $p_i$  = saturation pressure of hypothetical ideal gas, absolute atm.
- $R$  = gas constant
- $T$  = absolute temperature, °R.
- $\bar{V}$  = molal volume per formula weight of monomer, cu. ft./lb.-mole
- $x$  = weight fraction

## Subscripts

- $a$  = quantity for equilibrium molecular mixture
- $c$  = empirical constant
- $i$  = quantity for vapor in any specific state
- $0$  = quantity at 0° R.
- $1$  = quantity for monatomic species
- $2$  = quantity for diatomic species
- $4$  = quantity for tetratomic species

Table II. Standard Deviations of Equations

Metal	Standard Deviations ( $\log p_s$ ) of Experimental Data		
	From published equation by original investigators	From complex equations combining Eq. 6 (coefficients from Table I) with Eq. 1	From simplified equation generated for $p_s$ (Eq. 7 through 10)
Sodium	0.0023 (52)	0.0023	0.0023
Potassium	0.0019 (52)	0.0019	0.0018
Cesium	0.0023 (52)	0.0023	0.0022
Rubidium	0.0053* (54)	0.0054	0.0054

\* Figure obtained from experimental points of Tepper *et al.*, with their equation, is comparable to standard per cent deviation of 1.22 which they reported.

## Superscripts

- g = quantity in gas state  
 l = quantity in liquid state  
 ° = standard state, 1 atm. for gas

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RECEIVED for review April 18, 1968. Accepted January 10, 1969.