MASTER

An Extension of the Law of Corresponding States
to Rare-Gas Solids*

by Gerald L. Pollack

Department of Physics, Michigan State University, East Lansing, Michigan

ABSTRACT

It is shown that the law of corresponding states can be applied to sublimation pressure curves of rare-gas solids. The vapor pressure (P) and temperature (T) below the triple point are written in terms of the usual reduced variables $P^* = P\sigma^3/\epsilon$ and $T^* = kT/\epsilon$, where ϵ and σ are Lennard-Jones potential parameters. It is then demonstrated that a single curve fits the P^* , T^* data for Ar, Kr, and Xe over the 5 decade range in P^* for which measurements have been made, $2 \times 10^{-8} \leq P^* \leq 2 \times 10^{-3}$ and $0.36 \leq T^* \leq 0.72$.

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The equation of the curve for Ar, Kr, and Xe is $\ln P^* = 5.302 - 8.206/T^*$. The sublimation pressure curve for Ne is different from that of the heavier rare gases because of zero-point energy effects. The equation of the Ne curve is $\ln P^* = 4.287 - 7.077/T^*$, over about 3 decades in P^* . The historical background of the law of corresponding states, and its derivation by De Boer et al., from quantum-mechanical first principles are presented in a way natural to this problem. It is shown that extension of the law of corresponding states to the solid-vapor phase transition is natural and proper. As a corollary, the sublimation pressure for Radon is predicted as $\log_{10} P(\text{torr}) = 7.868 - 1034/T$.

INTRODUCTION

The law of corresponding states may be expressed this way:
The equations of state of certain simple substances are identical when expressed in terms of suitable reduced variables.

This idea, that the thermodynamic properties of a large class of substances can be described in common if the right variables are used, was suggested by van der Waals in 1873.^{1/} The van der Waals equation of state may be conveniently expressed for this purpose in terms of the reduced variables $P_r = P/P_{cr}$, $V_r = V/V_{cr}$, and $T_r = T/T_{cr}$ where P_{cr} , V_{cr} , and T_{cr} are the coordinates of the critical point.^{2/} In this form the law of corresponding states (l.o.c.s.) is used to describe the properties of gases in the neighborhood of and above the critical point. Actually, as was first pointed out by Byk in 1921,^{3/} these three reduced variables must be supplemented by a fourth one when quantum mechanical effects become significant.

More modern forms of the l.o.c.s. usually use reduced variables which depend explicitly on the characteristics of the intermolecular potential. A typical intermolecular potential for molecular solids, the substances of interest to us here, is the Lennard-Jones potential:

$$\phi(r) = 4\epsilon[(\sigma/r)^{12} - (\sigma/r)^6], \quad (1)$$

where r is the intermolecular separation. The maximum well depth is ϵ ; it occurs at an intermolecular separation of

$r_0 = 2^{1/6}\sigma$, and $\phi(\sigma) = 0$. The modern reduced variables are $P^* = P\sigma^3/\epsilon$, $V^* = V/N\sigma^3$, $T^* = kT/\epsilon$, and the quantum parameter $\Lambda^* = h/\sigma(m\epsilon)^{1/2}$. In these definitions N is Avogadro's number,

k and h are, respectively, Boltzmann's constant and Planck's constant, and m is the molecular mass. These dimensionless variables were first introduced by De Boer, Michels, et al.,^{4,5,6} in an application of the l.o.c.s. first to gaseous helium and then to condensed permanent gases.

For the rare-gas solids the reduced variables P^* , V^* , T^* , and Λ^* may be expressed in another interesting way that is approximately equivalent. The well depths ϵ for Ar, Kr, and Xe are approximately proportional to the heats of sublimation of the solids at 0° K, L_0 .⁷ The ratios of L_0 per atom to ϵ are about: 7.8 for Ar, 8.2 for Kr, and 8.3 for Xe. The corresponding ratio for Ne is 6.2. Thus, we could write the l.o.c.s. approximately in terms of $T^{\prime*} = kT/L_0$ instead of the usual T^* . In the same way the values of σ for Ne, Ar, Kr, and Xe are closely proportional to the atomic radii.⁸ The ratios of atomic radius to σ are: 0.57 for Ne, 0.56 for Ar, 0.55 for Kr, and 0.56 for Xe. Thus, we could continue the l.o.c.s. for Ar, Kr, and Xe in terms of $P^{\prime*} = P(\text{atomic radius})^3/L_0$, $V^{\prime*} = V/N(\text{atomic radius})^3$, and $\Lambda^{\prime*} = h/(\text{atomic radius})(mL_0)^{1/2}$. The same formulation may be applied to Ne, but it holds not as well.

The l.o.c.s. may be applied not only to gases, but also to liquids and solids. This was made clear in early work of Pitzer¹ in which he derived the classical l.o.c.s. from first principles under certain assumptions about the interacting molecules. The key assumptions are: that the molecules be effectively spherical so that the two-body intermolecular potential depends only on separation, and that the potential be expressible in the form $\phi = \epsilon\phi(r/\sigma)$, where ϵ and σ are general

*--- Gred/phi
--- Gred/sigma*

energy and length scaling parameters, and $\phi(r/\sigma)$ is a universal potential function. The prototype elements for correspondence in the condensed states are: Ar, Kr, Xe, and to a lesser extent Ne, CH₄, O₂, N₂, CO, H₂, etc. This work was extended and applied by Guggenheim.⁹ These two workers^{6,9} demonstrated correspondence in terms of P_r , V_r , and T_r in many properties of the liquid and gaseous states, e.g., triple point properties, virial coefficients, entropy of vaporization, heat capacities, surface tension, and thermal expansivity.

A demonstration of correspondence proceeds in this way: The experimental data of some quantity for several substances is reduced, i.e. made nondimensional, by multiplying and dividing the data by appropriate combinations of the respective P_{cr} , V_{cr} , and T_{cr} . This reduced data is then plotted as a function of the reduced variables, P_r , V_r , or T_r . Correspondence generally consists in demonstrating that all the reduced data fall on a universal curve. If the more modern version of the l.o.c.s. is used, then the data is reduced by use of ϵ , σ , and m . The results are then plotted as a function of P^* , V^* , T^* , and, in the quantum case, Λ^* .

Tests of correspondence for specific properties must be derived individually from first principles. Thus, a test of correspondence in triple-point properties consists in showing that P_{tr}^* , the reduced triple-point pressure, T_{tr}^* , the reduced triple-point temperature, $V_{tr,liq}^*$ and $V_{tr,sol}^*$ the reduced triple-point liquid and solid specific volumes, are the same for

several substances. Modern values of these quantities are shown in Table I. A test of correspondence in vapor pressure or other phase equilibrium pressure consists in examining P^* as a universal function of T^* . A test of correspondence of the reduced isothermal bulk modulus $K_T^* = -V^* (\partial P^* / \partial V^*)_{T^*}$ a quantity derived directly from the equation of state, consists in showing that K^* is a universal function of Λ^* , at constant P^* and T^* . ^{7,10}

In this paper we should like to demonstrate that the l.o.c.s. applies accurately to sublimation curves of the rare-gas solids, Ne, Ar, Kr, and Xe from the triple points down to very low pressures. We shall be interested then in the experimental sublimation and vapor pressure data when plotted as P^* versus T^* . Modern values of ϵ , σ , m , and Λ^* for the rare gases Ne, Ar, Kr, Xe, and Rn are shown in Table I.

THEORETICAL BASIS

An excellent derivation of the law of corresponding states in application to condensed gases was given by De Boer and Blaisse.^{5,6} Recently, a discussion of this work and other applications of the l.o.c.s. has been presented as part of a review of rare-gas solids.⁷ The essence of the argument is to consider the eigenvalues E_n of the Schrödinger equation for N_A interacting molecules. If the intermolecular forces are two-body forces which depend only on the separation, the equation is⁵:

$$\left[-(\hbar^2/2m) \sum_i \nabla_i^2 + \sum_{i>k} \phi(r_{ik}) - E_n \right] \psi_n(r_1, \dots, r_{N_A}) = 0. \quad (2)$$

Summation sign Greek psi

under summation sign

The sums are over all particles and $\phi(r_{ik})$ is, say, the same potential function as in Eq. (1). In Eq. (2), ϵ , σ , and m are different for different simple substances and, therefore, the

Please
Note $\hbar = \hbar$
 ∇ is "del" symbol

Please
Note
unlabeled

eigenvalues and eigenfunctions are different for different substances.

If, however, Eq. (2) is rewritten, in terms of reduced variables, in the nondimensional form:

$$[-\Lambda^2 \sum_i (1/8\pi^2) \nabla_i^2 + \sum_{i>k} \phi^*(r_{ik}^*) - N E_n^*] \psi_n(x_1^*, \dots, x_{N_A}^*) = 0, \quad (3)$$

Note $\sum_i$ is summation symbol

then the same equation holds for all substances which have a common form for the intermolecular potential. In Eq. (3),

$\nabla_i^* = \sigma \nabla_i$, $\phi^* = \phi/\epsilon$, $E_n^* = E_n/N\epsilon$, and $r^* = r/\sigma$. For our purposes, the simple substances to be discussed are solid and liquid Ar, Kr, Xe, and Ne. The intermolecular potential is taken to be Eq. (1).

Solution of Eq. (3) gives then a universal set of reduced eigenvalues, valid for all condensed rare gases. From this set of E_n^* a reduced partition function may be calculated and this, in turn, gives a universal equation of state of the form⁵⁻⁷:

all on one line

$$p^* = p^*(V^*, T^*, \Lambda^*). \quad (4)$$

These arguments are most useful for the liquid and solid states of the rare gases. Their application to these states becomes limited, for example, as the two-body potential law becomes inaccurate due to three-body or electron exchange effects.

As corollaries of this work, one can show many interesting correspondences among the rare-gas liquids and solids. Some examples of these are: (a) There is correspondence among triple-point coordinates and among critical-point coordinates (Table I). (b) There is correspondence among reduced isotherms, p^* versus V^* at constant T^* , if $\Lambda^* \rightarrow 0$.⁶ This follows from Eq. (4). (c) The transition lines between different phases show correspondence in an especially interesting and simple

way. The condition describing such a transition is:

$$G_1^*(V^*, T^*, \Lambda^*) = G_2^*(V^*, T^*, \Lambda^*). \quad (5)$$

In Eq. (5), $G_{1,2}^*$ are the reduced Gibbs free energies for phases 1 and 2. This means that the phase transition may be described by universal functions of V^* and T^* (and of Λ^* if quantum effects are important).⁵

Another reason phase changes are important for study of the l.o.c.s. is that different ones give information about correspondence in different parts of the intermolecular potential. Thus, correspondence in melting curves at high P^* and low V^* depends partly on the repulsive part of $\phi(r)$ near the hard core $r \approx \sigma$.¹¹ Correspondence in vapor pressure and sublimation pressure curves at comparatively low P^* and low T^* depends on the properties of $\phi(r)$ from the neighborhood of the minimum on out in the attractive direction $r > r_0$.

There have been many interesting and useful tests of the l.o.c.s. on rare-gas liquids. A usual form of the test, as already remarked above, is to examine curves of P^* versus T^* from the respective triple-point coordinates (P_{tr}^*, T_{tr}^*) to the critical point coordinates (P_{cr}^*, T_{cr}^*) .^{1,9,12-14} In general the curves for Ar, Kr, and Xe fall closely together, with Ne slightly displaced from this universal curve due to its larger Λ^* .¹² These curves span from $P_{tr}^* \approx 1.5 \times 10^{-3}$, $T_{tr}^* \approx 0.7$ to $P_{cr}^* \approx 0.1$, $T_{cr}^* \approx 1.3$.

Sublimation pressures of rare-gas solids have until now not been tested for correspondence in this same way, partly because of lack of sufficient data. The advantage of examining the l.o.c.s.

below P_{tr}^* , T_{tr}^* , i.e. for sublimation, is that in principle a much wider decade range of P^* and T^* is available. From the development described in connection with Eqs. (2) - (5)^{5,6} we expect that correspondence in sublimation pressure curves might obtain for a good distance below the triple point and maybe down to the P^* , $T^* \rightarrow 0$ limit. This is the problem we examine in the next section.

RESULTS

Figure 1 displays good, representative, sublimation pressure data on Ne, Ar, Kr, and Xe over the range in which data have been reported. The P versus T curves have all been terminated at the triple points (marked x). Respective coordinates of the triple points are those in Table I. On the four curves of Figure 1, the marked dots are either actual data,¹⁵⁻¹⁹ a short theoretical extrapolation of data,¹⁹ averages over a small number of data points that are close together,¹⁵ or calculated from curves which fit actual data points.^{20,21} Smooth curves to connect these points are also shown.

Two sources have been used for the Ne sublimation pressure. From 16.4° K to the triple point data of Bigeleisen and Roth¹⁵ have been used, and from 15°-18° K rather old but very good data of Keesom and Haantjes¹⁶ have been used. The Ar sublimation pressure has been taken as alternate points of the data of Flubacher et al.,¹⁷ who measured the Ar sublimation pressure between 66° K and the triple point.

Two sources have also been used for the Kr sublimation pressure. Between 83° K and the triple point data of Beaumont et al.¹⁸ have been used, and from 63°-80° K results of Fisher and McMillan¹⁹ have been used. In the range 63°-69° K, Fisher

and McMillan¹⁹ reported theoretical sublimation pressures since their experimental data showed effects of what are probably trace nitrogen impurities. The data points for Kr on Figure 1 in this region are, therefore, taken from their theoretical results.

The Xe sublimation pressure has been taken from results of Freeman and Halsey²⁰ between 110° K and the triple point, and from results of Podgurski and Davis²¹ between 83°-90° K. Both of these investigations gave results in the form:

$$\log_{10} P(\text{torr}) = A - B/T. \quad (6)$$

The Xe curve on Figure 1 has been calculated from this equation using the two sets of A and B given by the original investigators.^{20,21} Equation (6) is frequently used to fit sublimation pressure curves.⁷ We shall use it below to fit reduced sublimation pressure curves.

The reduced sublimation pressure curves, $P^* = P\sigma^3/\epsilon$ versus $T^* = kT/\epsilon$, are plotted on Figure 2. Values of the potential parameters σ and ϵ which were used to reduce the data are those given on Table I. The data on Figure 2 terminate in the respective reduced triple point coordinates, also given on Table I. All the data plotted on Figure 1 for each of the four elements have been included on Figure 2.

Reduced vapor pressure curves for liquid Ne, Ar, Kr, and Xe from the triple points to the critical points have been studied by Hamrin and Thodos,¹² by Boato and Casanova,¹³ and by Malyshev.¹⁴ A plot analogous to our Figure 2 but for the liquid range, $1.2 < 1/T^* < 1.4$, for Ne, Ar, Kr, Xe, and other liquids, appears in Figure 1 of Ref. 13. Reduced curves of melting pressure versus temperature have been studied by De Boer

and Blaisse⁶ and by Danon and Pitzer.²²

It is clear from Figure 2 that the P^* versus T^* curve for Ne is significantly higher than that for the other elements. Behavior of this kind is found also in other properties of Ne, for example, in thermal expansivity.⁷ This anomalously high sublimation pressure is fundamentally due to the larger contribution of zero-point energy in the properties of solid Ne than in the properties of the other solids. In terms of our parameters, Ne has a significantly larger Λ^* than the heavier rare gas elements (Table I). This means that in equating $G_{\text{solid}}^*(V^*, T^*, \Lambda^*) = G_{\text{vapor}}^*(V^*, T^*, \Lambda^*)$, as in Eq. (5), we cannot in the case of Ne neglect the contribution of Λ^* . In the same manner in the equation of state, Eq. (4), Λ^* cannot be neglected for Ne. Thus we do expect a universal sublimation pressure curve P^* versus T^* in the limit $\Lambda^* \rightarrow 0$, as is approximated by Ar, Kr, and Xe, but as Λ^* gets larger, as for Ne, then the corresponding P^* versus T^* curves will be different.

Departures from the reduced curves obtained in the $\Lambda^* = 0$ limit are of course even more pronounced for such substances as deuterium and hydrogen, whose Λ^* 's are still higher than for Ne.^{12,13} A quantitative test of the functional dependence of P^* versus T^* curves on Λ^* requires a study of these data for substances with several different values of Λ^* . This is beyond our aim here. However, quantitative tests of the dependence on Λ^* of reduced solid state properties in the limit $P^* = 0$ and $T^* = 0$, i.e. $T = 0^\circ \text{K}$, have been carried out.^{10,23}

The Ne curve on Figure 2 can be fit by an equation of the form:

$$\ln P^* = a^* - \frac{b^*}{T^*} \quad (7)$$

The coefficients which give a least squares fit are $a^*(\text{Ne}) = 4.287$ and $b^*(\text{Ne}) = 7.077$. This smooth curve has been included on Figure 2.

Finally, Figure 2 shows that the P^* versus T^* reduced sublimation pressure data for the heavier rare gases, Ar, Kr, and Xe, all fall close to a universal curve. This is what we have deduced above and in the theoretical section of this paper. We expect also that if sublimation pressure data for Rn were available, they would also fit along this universal curve, since Λ^* is smallest for Rn of all the rare-gas elements. The best fitting, least squares, curve of the form of Eq. (7), to these data has coefficients $a^*(\text{Ar, Kr, Xe}) = 5.302$ and $b^*(\text{Ar, Kr, Xe}) = 8.206$. This smooth curve has also been included on Figure 2. The average absolute deviation of the combined Ar, Kr, and Xe data from this universal curve is 9.1%.

It is interesting to note on Figure 2 that the P^* , T^* data for Ar are closer to the data of Xe than they are to those of Kr. This is some kind of unexpected slight rearrangement in the order of these reduced properties of Ar, Kr, and Xe. The expected order is Ne, Ar, Kr, Xe. This is the order of the unreduced triple-point coordinates, critical-point coordinates, and potential parameters in the first eight columns of Table I. The rearranged order Ar, Xe, Kr is apparent in some of the next six columns, e.g., in T_{tr}^* and P_{tr}^* .

CONCLUSIONS

We have shown that the law of corresponding states can be extended to sublimation pressures of the rare-gas solids. When sublimation pressure curves are plotted in terms of the reduced quantities P^* versus T^* , as on Figure 2, then a single curve

of the form Eq. (7) fits the data for the heavier rare gases, Ar, Kr, and Xe, to better than 10%. The data for Ne fall on a separate curve, close to but higher than that for Ar, Kr, and Xe, because of the larger contribution of zero-point energy to the thermodynamic properties of Ne. The theoretical basis of the l.o.c.s. has been presented in a form natural to the extension we have carried out.

The sublimation pressures show correspondence over the entire 5 decades in P^* for which they have been measured below the triple point. It is expected that this same correspondence will continue down to the limit $P^* = 0$, $T^* = 0$.

As a corollary to this work the sublimation pressure for Rn can be predicted. Since Λ^* for Rn is small, the reduced sublimation pressure curves for Rn should follow Eq. (7) with the same coefficients as those for heavy rare gases, i.e. $\ln P^* = 5.302 - 8.206/T^*$. Using the values for the potential parameters for Rn of Table I, the resulting expression in the form of Eq. (6) is:

$$\log_{10} P(\text{torr}) = 7.868 - 1034/T \text{ (for Rn).}$$

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Table I: Potential parameters and triple-point and critical-point properties of rare gases.

- (a) G. K. Horton, Am. J. Phys. 36, 93(1968). Values for ϵ/k and σ were calculated from AN values on Table II.
- (b) G. A. Miller, J. Phys. Chem. 64, 163 (1960), also ref. 7.
- (c) Compiled by J. S. Brown as given in Table I of G. K. Horton, op. cit.
- (d) Ref. 8.

The data in the first six columns all are taken from ref.7. Definitions of the reduced quantities are $T^* = kT/\epsilon$, $P^* = P_0^{1/3}/\epsilon$, $V^* = V/No^{1/3}$, and $\Lambda^* = h/\sigma\sqrt{m\epsilon}$. The quantities in the last seven columns are calculated from these definitions and the data.

Figure 1: Sublimation pressure curves for Ne, Ar, Kr, and Xe. The data points shown for Ne are taken from refs.(15) and (16), those for Ar are from ref.(17), those for Kr are from refs.(18) and (19), and those for Xe are from refs.(20) and (21). The curves have been terminated at the triple points (marked x).

Figure 2: Reduced sublimation pressure curves for Ne, Ar, Kr, and Xe. The data points shown on this figure are taken from those ^{data on the} on Figure 1. The curves have been terminated, at the high P^* , T^* end, at the triple points.

This is the left half of Table I.

Table I

Element	$T_{tr} (^{\circ}K)$	$P_{tr} (torr)$	$V_{tr, liq}$ ($cm^3/mole$)	$V_{tr, sol}$ ($cm^3/mole$)	$T_{cr} (^{\circ}K)$	$P_{cr} (atm)$	$\epsilon/k (^{\circ}K)$	$\sigma (\text{\AA})$ ^{symbol for Angstroms}
Ne	24.56	323.5	16.18	13.98	44.5	25.9	36.7 ^a	2.789 ^a
Ar	83.810	516.86	28.321	24.60	150.9	48.3	120 ^a	3.403 ^a
Kr	115.78	548.7	34.18	29.64	209.4	54.3	164 ^a	3.638 ^a
Xe	161.37	612.2	42.68	38.62	298.8	57.64	231 ^a	3.961 ^a
Rn	202	(500)	(39)		377.5	62.4	290 ^b	4.34 ^b

Caption for Table I is on a separate page

This is the right half of Table I

Table I (cont'd)

m (10^{-24} g/atom)	T_{tr}^*	P_{tr}^*	$V_{tr,liq}^*$	$V_{tr,sol}^*$	T_{cr}^*	P_{cr}^*	Λ^*
33.510 [✓]	0.669	0.00185	1.238	1.070	1.21	0.113	0.577
66.280 [✓]	0.698	0.00165	1.193	1.036	1.26	0.117	0.186
139.05 [✓]	0.706	0.00155	1.178	1.022	1.28	0.117	0.102
217.9 [✓]	0.699	0.00159	1.140	1.031	1.25	0.114	0.0635
(368) [✓]	0.697	(0.0014)			1.30	0.129	(0.040)