

- Godbole, S. P., M. F. Honath, and Y. T. Shah, "Gas Holdup Structure in Viscous Newtonian and Non-Newtonian Liquids," *Chem. Eng. Commun.*, **16**, (1-6), 119 (1982).
- Griffith, R. M., "The Effect of Surfactants on the Terminal Velocity of Drops and Bubbles," *Chem. Eng. Sci.*, **17**, 1057 (1962).
- Hikita, M., S. Asai, K. Tanigawa, K. Segawa, and M. Kitao, "Gas Holdup in Bubble Column," *Chem. Eng. J.*, **20**, 59 (1980).
- Hughmark, G. A., "Holdup and Mass Transfer in Bubble Columns," *Ind. Eng. Chem. Process Des. Dev.*, **6**, 218 (1967).
- Kara, S., B. G. Kelkar, Y. T. Shah, and N. L. Carr, "Hydrodynamics and Axial Mixing in Three Bubble Column," *Ind. Eng. Chem. Process Des. Dev.* (1982).
- Kumar, A., T. T. Dagaleesan, G. S. Laddha, and H. E. Hoelscher, "Bubble Swarm Characteristics in Bubble Columns," *Can. J. Chem. Eng.*, **54**, 503 (1976).
- König, B., R. Buchholz, J. Lücke, and K. Schügerl, "Longitudinal Mixing of the Liquid Phase in Bubble Columns," *Ger. Chem. Eng.*, **1**, 199 (1978).
- Levan, M. D., and J. Newman, "The Effect of Surfactants on the Terminal and Interfacial Velocities of a Bubble or Drop," *AIChE J.*, **22**, 695 (1976).
- Levich, V. G., "Physicochemical Hydrodynamics," Prentice-Hall, Englewood Cliff, NJ (1962).
- Lindland, K. P., and S. G. Terjesen, "The Effect of a Surface-Active Agent on Mass Transfer in Falling Drop Extraction," *Chem. Eng. Sci.*, **5**, 1 (1965).
- Lockett, M. J., and R. D. Kirkpatrick, "Ideal Bubble Flow and Actual Flow in Bubble Columns," *Trans. Inst. Chem. Engrs.*, **53**, 267 (1975).
- Mersmann, A., "Design and Scale-up of Bubble and Spray Columns," *Ger. Chem. Eng.*, **1**, 1 (1978).
- Miller, D. N., "Gas Holdup and Pressure Drop in Bubble Column Reactor," *Ind. Eng. Chem. Process Des. Dev.*, **19**, 371 (1980).
- Oels, U., J. Lücke, R. Buchholz, and K. Schügerl, "Influence of Gas Distributor Type and Composition of Liquid on the Behavior of a Bubble Column Bioreactor," *Ger. Chem. Eng.*, **1**, 115 (1978).
- Panvelker, S. V., J. W. Tierney, Y. T. Shah, and D. F. Rhodes, "Backmixing in a SRC-II Dissolver," *Chem. Eng. Sci.*, **37**, (10), 1582 (1982).
- Raymond, D. R., and S. A. Zieminski, "Mass Transfer and Drag Coefficients of Bubble Rising in Dilute Aqueous Solutions," *AIChE J.*, **17**, 57 (1971).
- Rice, P. G., J. H. I. Tupurainen, and R. M. Hedge, "Dispersion and Holdup in Bubble Columns," ACS Meeting, Las Vegas (Aug., 1980).
- Schügerl, K., J. Lücke, and U. Oels, "Bubble Column Bioreactors," *Adv. Biochem. Eng.*, T. K. Ghose, A. Fiechter, and N. Blakeborough, **7**, 1 (1977).
- Sriram, K., and R. Mann, "Dynamic Gas Disengagement: A New Technique for Assessing the Behavior of Bubble Columns," *Chem. Eng. Sci.*, **32**, 571 (1977).
- Zuber, N., and J. A. Findlay, "Average Volumetric Concentration in Two Phase Systems," *Trans. ASME, J. of Heat Trans.*, **87**, 453 (1965).

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PVT Analysis of a New Cubic-Perturbed, Hard-Sphere Equation of State

The cubic-perturbed, hard-sphere equation of state proposed in 1980 by Ishikawa, Chung and Lu, has been analyzed by PVT criteria, as contrasted to the single component and multicomponent vapor-liquid equilibria to which it was applied. The analysis compares its representation of the PVT behavior of substances with that of a simpler equation.

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SCOPE

There have been many different cubic equations of state proposed since van der Waals over a century ago. However, a form was suggested by Ishikawa, Chung and Lu (1980) that had never appeared before in the literature. Because of its novelty, it seemed worthwhile to study it in some detail from the point of view of its PVT behavior which had been examined only on a limited basis when it was presented. In this work it is compared with other cubic equations on seven basic counts: (1) prediction of the compressibility factor at the critical point; (2)

comparison with the general volume-cubic equation of state of Martin (1979); (3) its Z -chart sum; (4) its critical isotherm and prediction of the critical pressure from the critical temperature and volume; (5) its prediction of a collection of PVT data for argon for reduced temperatures from 0.6 to 1.5 and up to twice the critical density; (6) its prediction of pressures from saturated liquid and vapor densities of argon; and (7) the same comparisons for isopentane as for argon where the former has a Z_c of only 0.270 compared to 0.291 for the latter.

CONCLUSIONS AND SIGNIFICANCE

The new equation is shown to predict a low critical pressure and an impossible critical isotherm. When compared to a simpler form of cubic equation of state, it is less satisfactory in representing PVT data up to twice the critical density or in predicting vapor pressure from saturated liquid density for two

quite different fluids, argon and isopentane. Because it was originally utilized largely for its use in equilibrium comparisons, its PVT behavior was not examined. Only in the three-root region is its prediction of a more constant pressure isotherm closer than a simpler equation, but this results from its greater deviation of pressure at smaller volumes. The analysis is useful because it can be applied to evaluate the PVT performance of any equation of state.

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TABLE 1. CRITICAL COMPRESSIBILITY FACTORS FOR SEVERAL CUBIC EQUATIONS

Equation	Z_c
van der Waals	0.3750
Redlich-Kwong	0.3333
Ishikawa-Chung-Lu	0.3152
Peng-Robinson	0.3074
Martin (Set by Volume Translation)	0.25-0.375

Recently, a new equation of state was suggested by Ishikawa, Chung and Lu (1980), which they termed a cubic perturbed, hard sphere equation of state. The proposed equation has the form,

$$P = \frac{RT(2V + b)}{V(2V - b)} - \frac{a}{T^{0.5}V(V + b)} \quad (1)$$

where the parameters, a and b , are both taken to be temperature-dependent. In the development of van der Waals (1873) equation, $P = RT/V(V - b) - a/V^2$, with hard-spherical molecules subjected to an intermolecular attraction force, b may be thought of as a measure of the volume of the hard spheres. If b is allowed to vary with temperature, one can speak of a perturbed hard-sphere equation. In Eq. 1, b can be considered similar to van der Waals b (even though it is in the attraction as well as the repulsion term), so that if it is allowed to vary with temperature, the equation may be termed a perturbed hard sphere equation.

Other two-term cubic equations containing a hard-sphere parameter b have been subjected to perturbation of b by previous writers, including the two senior authors, Chung and Lu (1977), of Eq. 1. The important thing here is that Eq. 1 is a form of cubic equation that has never appeared before, so it is worthwhile to examine it carefully to determine if it has any characteristics that make it better than any of the other cubic equations proposed during the century after van der Waals. Is the perturbation of the b term in Eq. 1 significantly different from perturbation of other two-term cubic equations so that it is "superior to all the other (12) variants of the van der Waals and the Redlich-Kwong equations," studied by the authors?

In the usual way at the critical point the derivatives, $(dP/dV)_T$ and $(d^2P/dV^2)_T$, may be set to 0 to solve for the constants in Eq. 1,

$$a = 0.467123 R^2 T_c^{2.5} / P_c \text{ at } T_c \quad (2)$$

and

$$b = 0.108762 RT_c / P_c \text{ at } T_c \quad (3)$$

Substituting these values at the critical point gives the critical compressibility factor,

$$Z_c = P_c V_c / RT_c = 0.315206 \quad (4)$$

Unfortunately, the authors did not give the values in Eqs. 2, 3 and 4 even though they implied using the critical point derivatives. The last value, Z_c , may be compared with several other two-term cubics in Table 1. Since the experimental values of Z_c for most substances normally encountered are less than 0.3, the proposed equation is better than the Redlich-Kwong (1949), but not as good as the Peng-Robinson (1976), or the Martin (1967, 1979), when a suitable volume translation is selected.

It is desirable first to show that Eq. 1 is just a special case of the all-inclusive volume-cubic equation of state presented in 1979,

$$P = \frac{RT}{V} - \frac{\alpha}{(V + \beta)(V + \gamma)} + \frac{\delta}{V(V + \beta)(V + \gamma)} \quad (5)$$

With temperature included in a , Eq. 1 may be rearranged to

$$P = \frac{RT(V + b/2) - RTb/2 + RTb/2}{V(V - b/2)} - \frac{a}{V(V + b)}$$

or

$$P = \frac{RT}{V} + \frac{RTb(V + b) - a(V - b/2)}{V(V - b/2)(V + b)} \quad (6)$$

TABLE 2. Z-CHART SUMS FOR SEVERAL CUBIC EQUATIONS

Equation	Σ , Z-Chart Sum
van der Waals	0.6719
Redlich-Kwong	0.6742
Ishikawa-Chung-Lu	0.6736
Peng-Robinson	0.6868
Martin (For All Translations)	0.6719

The last term may be split so that

$$P = \frac{RT}{V} + \frac{RTb - a}{(V - b/2)(V + b)} + \frac{RTb^2 + ab/2}{V(V - b/2)(V + b)} \quad (7)$$

Now in Eq. 5 set $\beta = -b/2$, $\gamma = b$, $\alpha = a - RTb$, and $\delta = RTb^2 + ab/2$. With these values Eqs. 5 and 7 are quickly seen to be identical, so that Eq. 1 is simply a form of Eq. 5 with suitable specialization of constants.

Returning to Eq. 1 another test of its behavior is its Z-chart sum, $\Sigma = Z_c - (dZ/dP_R)_{T_R} = 1, P_R = 0$, where $(dZ/dP_R)_{T_R} = 1, P_R = 0$ is the generalized second virial coefficient. Using the constants a and b , and carrying out the necessary differentiation to obtain the second virial coefficient gives

$$\Sigma = 0.673567 \quad (8)$$

This may be compared with the Z-chart sum for several other equations in Table 2. The differences of Σ are small, but significant. It is to be recalled from Martin (1979), that Σ is about 0.62 for practically all substances, so the lowest value in Table 2 is preferable, other characteristics being the same. This makes the Ishikawa-Chung-Lu equation second in the listing and slightly better than the Redlich-Kwong.

The actual PVT behavior of Eq. 1 will now be examined for two substances out of 22 presented in the article. These are argon and isopentane which have been chosen because of their range of Z_c values from 0.291 for argon to 0.270 for isopentane. It is convenient to put Eq. 1 into generalized form by multiplying through by the critical constants,

$$P_R = \frac{RTV_c T_c (Z_c / V_c) (2V + b)}{V P_c V_c T_c (Z_c / V_c) (2V - b)} - \frac{a T_c^{0.5} V_c R T_c P_c}{T^{0.5} T_c^{0.5} P_c V V_c R^2 T_c^2 (Z_c V_c) (V + b)}$$

or

$$P_R = \frac{T_R (2Z_c V_R + b P_c / RT_c)}{Z_c V_R (2Z_c V_R - b P_c / RT_c)} - \frac{a P_c / R^2 T_c^{2.5}}{T_R^{0.5} Z_c V_R (Z_c V_R + b P_c / RT_c)} \quad (9)$$

Now the generalized a and b for temperatures other than T_c have been presented as moderately long power series expansions of T_R , as follows:

$$a P_c / R^2 T_c^{2.5} = \sum_{i=0}^3 a_i T_R^i \quad (10)$$

and

$$b P_c / RT_c = \sum_{i=0}^3 b_i T_R^i \quad (11)$$

Values of a_i and b_i have been given for the 22 substances, which include argon and isopentane.

The behavior along the critical isotherm will be studied first for argon. Two possibilities exist, for the numbers in Eqs. 2 and 3 may be used or the values that come from the correlating Eqs. 10 and 11 at $T_R = 1$ may also be used. The latter are

$$a = 0.45345 R^2 T_c^{2.5} / P_c \text{ at } T_c \quad (12)$$

From Eqs. 10
and 11

$$b = 0.10425 RT_c / P_c \text{ at } T_c \quad (13)$$

The required constants to be employed for argon are $T_c = 150.86$ K, $P_c = 48.34$ atm, $V_c = 1/13.41$ L/gmol, $R = 0.0820535$ L·atm/gmol·K, and $Z_c = 0.29121$. Comparison with the data of the National Bureau of Standards (1969) shows pressure predictions are from 10 to 50% low for reduced densities from 1.6 to 2.0 for either possible equation. (All comparisons referred to in this paper have been filed with the National Auxiliary Publication Service, 214-13 Jamaica Ave., Queens Village, NY 11428.)

A more fundamental defect occurs where the correlating Eqs. 10 and 11 leading to Eqs. 12 and 13 give an impossible critical isotherm with the pressure exceeding the critical pressure for volumes greater than the critical volume and being less than the critical pressure for volumes less than the critical volume, meaning pressure decreases for increasing density which is not possible and violates equilibrium state conditions. This defect extends to temperatures a little above the critical temperature and was not detected by Ishikawa, Chung and Lu in focusing their attention on developing correlating equations for a and b to yield low-temperature ($T_R < 1$) predictions of vapor pressures. Using the usual derivatives of pressure with respect to volume at the critical giving Eqs. 2 and 3, the expected horizontal inflection results with no violation of equilibrium.

It is instructive to look at the behavior of the Martin equation with two different values of the translation, the first as given in the paper and the second with the translation reduced by 0.022 and the temperature exponent increased by 0.25; thus,

$$P_R = \frac{T_R}{Z_c V_R - 0.082} - \frac{27/64 T_R^{0.55}}{(Z_c V_R + 0.043)^2} \quad (14)$$

and

$$P_R = \frac{T_R}{Z_c V_R - 0.06} - \frac{27/64 T_R^{0.8}}{(Z_c V_R + 0.065)^2} \quad (15)$$

The predictions along the critical isotherm above the critical density can be made similar to those of the ICL equation by proper selection of the translation and this has been done in Eq. 15; however, as with the ICL equation, the fit of the NBS data is good at $V_R \approx 1/1.5$, but very poor at $1/2$ while the reverse is true for Eq. 14, a dilemma which is true for all cubics. The main reason that Eq. 15 is similar to the ICL is because its calculated Z_c is $0.25 + 0.06$ or 0.31 which is close to that given in Eq. 4. Both Eqs. 14 and 15 give the proper inflection to the critical isotherm.

The next comparison is over the whole plateau of NBS argon data up to twice the critical density and for reduced temperatures from 0.5 to 1.5. Below the critical density the differences between Eq. 9 with constants from Eqs. 10 and 11, and Eqs. 14 and 15 are small. Above the critical density Eq. 14 or 15 is generally better except for one region which is, as expected, at temperatures below the critical temperature. This is a near-liquid region where perturbation has its powerful beneficial effect, and it will be seen shortly how perturbation can be applied to Eq. 15, or Eq. 14 for that matter, to make it just as good as the ICL for saturated liquid.

To show how the ICL equation has been fitted to the saturated liquid, a series of temperatures have been selected down to a reduced temperature of about 0.65 for comparison. As anticipated from the low-temperature high-density results, the ICL equation does a fair job of predicting pressures from saturated liquid densities, while Eq. 15 is extremely poor, giving large negative pressures as the temperature decreases. To make a fair comparison, however, one must allow the b term of Eq. 15 to be perturbed in the same manner as the ICL equation by letting it vary with temperature. The generalized form of Eq. 15 is

$$P_R = \frac{T_R}{Z_c V_R - bP_c/RT_c} - \frac{27/64 T_R^{0.8}}{(Z_c V_R + 1/8 - bP_c/RT_c)^2} \quad (16)$$

For reduced temperatures between 1 and 1.5 good results are obtained if

$$bP_c/RT_c = 0.06 \text{ for } T_R \geq 1.0 \quad (17)$$

which is precisely Eq. 15; however, pressure predictions of Eq. 15 at $T_R < 1$ are on the low side. To raise pressures it is necessary that bP_c/RT_c be increased (as much as 50% or so over the 0.06 of Eq.

17). A number of temperature functions were examined and after a little trial a simple expression was developed that gives results generally better than the ICL equation. The expression is

$$bP_c/RT_c = 0.09406 - 0.03406 T_R^{0.4} \text{ for } T_R \leq 1.0 \quad (18)$$

where at $T_R = 1$, it merges identically into Eq. 17. The only place where the ICL equation is better is for a saturated liquid density point just below the critical. This is largely due to the fact that the ICL equation was designed to misfit at the critical (thereby giving the impossible critical isotherm), whereas Eq. 16 with its b perturbed as in Eq. 18 fits the critical point almost perfectly and yields a logical critical isotherm. The two equations do not differ a lot for prediction of pressures using saturated vapor densities.

The conclusion to be reached for argon is that the ICL Eqs. 9, 10 and 11 with its eight constants is no better in predicting PVT data than the much simpler set of Eqs. 16, 17 and 18 with just three constants. Around the critical point and along the critical isotherm the ICL equation is inferior as it also is for vapor pressures from saturated liquid densities. Since the predictions for the gas at low pressure are about alike for the two equations, the second virial coefficients will be practically the same.

It is desirable next to look at isopentane where Z_c is much less than for argon. The ICL equation is again the combination of Eqs. 9, 10 and 11, with the a_i and b_i for isopentane. By trying several values for the constants, Eqs. 16, 17, and 18 become for isopentane

$$P_R \frac{T_R}{Z_c V_R - bP_c/RT_c} - \frac{27/64 T_R^{1.2}}{(Z_c V_R + 1/8 - bP_c/RT_c)^2}, \quad (19)$$

$$bP_c/RT_c = 0.045 \text{ for } T_R \geq 1.0 \quad (20)$$

and

$$bP_c/RT_c = 0.0852 - 0.0402 T_R^{1.0} \text{ for } T_R \leq 1.0 \quad (21)$$

The required constants for isopentane are $T_c = 460.39$ K, $P_c = 33.37$ atm, $V_c = 306$ cm³/gmol, $R = 82.05606$ cm³·atm/gmol·K, $Z_c = 0.270297$. Comparison with the data of Das, Reed, and Eubank (1977) shows that the ICL equation is generally not as good up to twice the critical density and reduced temperatures from 0.67 to 1.3. Since the predictions at low pressure are similar, the second virial coefficients will be about alike, as with argon. For pressures predicted from saturated liquid densities the ICL equation is inferior, as it predicts negative pressures at the lowest temperatures. Also, the ICL equation misses the critical point, as expected from its development. Along the critical isotherm the ICL equation has the same defect for isopentane as it does for argon, in that it predicts an impossible maximum and minimum.

Since vapor pressures for both argon and isopentane have been computed for true saturated liquid and vapor volumes directly from the equations of state, the conditions of equilibrium have been accounted for only approximately. If fugacities are calculated at these points, neither the ICL nor Martin equations will give the identical values expected for equilibrium between the saturated liquid and saturated vapor. This is simply because the usual three-root oscillation of a cubic isotherm on pressure-volume coordinates under the saturation dome does not in general give $\int V dP = 0$, as would occur if a horizontal line at the vapor pressure joined the true saturated liquid and vapor volumes. It is, of course, easy to find a pressure for a cubic isotherm under the dome such that the $\int V dP$ does vanish, but this will not occur at the true saturated liquid and vapor volumes.

However, this pressure and the saturated liquid and vapor volumes determined by its outer intersections with the isotherm will be closer to the true values for the ICL equation than for the Martin equation, which is its only advantageous representation. The reason for this is because the ICL equation does not give as deep a dip below this pressure as the Martin equation as shown in Figure 1. This smaller dip under the dome, however, leads to problems outside the dome because it causes the isotherm to rise too slowly for volumes less than the saturated liquid volume, which results in calculated pressures considerably less than those measured ex-

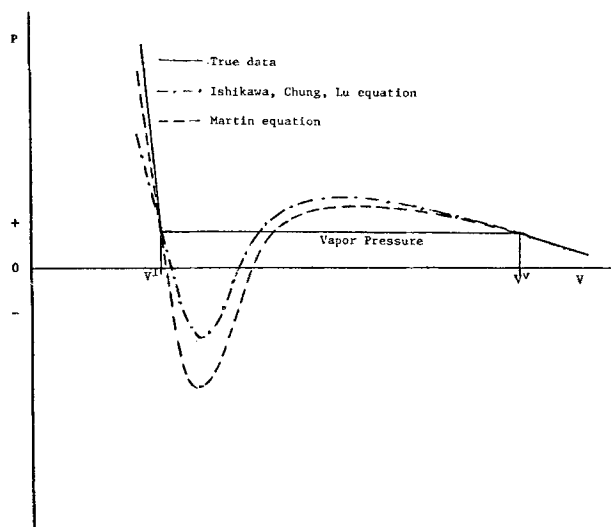


Figure 1. Isotherms of cubic equations of state in the three-root region when correct pressure given at the saturated liquid and vapor volumes or densities.

perimentally. The Martin equation with its greater dip under the dome rises more rapidly outside the dome and predicts pressures much closer to the experimental values.

For example, for isopentane at 398.15 K the saturated liquid volume is $147 \text{ cm}^3/\text{gmol}$ and the vapor pressure is 11.8 atm. If the pressure is computed for a volume of $142.27 \text{ cm}^3/\text{gmol}$, the ICL equation gives 17.865 atm and the Martin equation gives 30.612 atm while the measured pressure of Isaac, Li and Canjar (1954) is 35.563 atm. This difference between the two equations is damped out as the critical temperature is approached, but is more pronounced at lower temperatures. In general ICL isotherms under the dome (three-root regime) are better because they do not rise and fall so severely, but they are not as good outside the dome because they do not rise steeply enough for volumes less than the saturated liquid volume.

The conclusion to be reached in this study is that for PVT behavior of two quite different substances, argon and isopentane, except in the three-root region, the ICL equation is not as good as another. A simpler equation has been shown to be equal to or better when considering the critical point prediction, the critical isotherm, the Z-chart sum, the super-heated or saturated vapor phase behavior, and the vapor pressure for true saturated liquid volumes. Equation 1 is an interesting form of cubic equation, but the PVT analysis shows it has little to recommend it by comparison with Eq. 16 or 19 with their associated perturbed equations. It is probable that the ICL equation is better than the Redlich-Kwong because of its smaller value of Z_c , but on that score the Peng-Robinson would be better. If an equation such as Eq. 1 is to be useful, in the calculation of PVT behavior it should outperform a much simpler equation such as Eq. 16 or 19, and this is not demonstrated here.

NOTATION

a	= constant or temperature function in equations of state
b	= constant or temperature function in equations of state
P	= pressure
R	= gas constant
t	= translation in volume
T	= absolute temperature
V	= specific volume
Z	= compressibility factor
α	= constant in equation of state
β	= constant in equation of state
γ	= constant in equation of state
δ	= constant in equation of state
Σ	= Z-chart sum

Subscripts

c	= at critical point
i	= sequence of constants in series expansions
R	= reduced property meaning that property divided by its value at the critical point

LITERATURE CITED

- Das, T. R., C. O. Reed, Jr., and P. T. Eubank, "PVT Surface and Thermodynamic Properties of Isopentane," *J. Chem. Eng. Data*, **22**, 9 (1977).
- Isaac, R., K. Li, and L. N. Canjar, "Volumetric Behavior of Isopentane," *Ind. Eng. Chem.*, **46**, No. 1, 199 (1954).
- Ishikawa, T., W. K. Chung, and B. C. Y. Lu, "A Cubic Perturbed, Hard Sphere Equation of State for Thermodynamic Properties and Vapor-Liquid Equilibrium Calculations," *AIChE J.*, **26**, 372 (1980).
- Martin, J. J., "Equations of State," *Ind. Eng. Chem.*, **59** (12), 34 (1967).
- Martin, J. J., "Cubic Equations of State-Which?" *Ind. Eng. Chem. Fund.*, **18**, 81 (1979).
- National Bureau of Standards, "Thermodynamic Properties of Argon from the Triple Point to 300 K at Pressures to 1000 Atmospheres," NSRDA-NBS 27 (1969).
- Peng, D. Y., and D. B. Robinson, "A New Two-Constant Equation of State," *Ind. Eng. Chem. Fund.*, **15**, 59 (1976).
- Redlich, O., and J. N. S. Kwong, "On the Thermodynamics of Solutions V," *Chem. Rev.*, **44**, 233 (1949).
- Salah, E. M. H., W. K. Chung, I. M. Elshayal, and B. C. Y. Lu, "Generalized Temperature-Dependent Parameters of the Redlich-Kwong Equation of State for Vapor-Liquid Equilibrium Calculations," *Ind. Eng. Chem. Proc. Des. Dev.*, **16**, 51 (1977).
- Soave, G., "Equilibrium Constant from a Modified Redlich-Kwong Equation of State," *Chem. Eng. Sci.*, **37**, 1197 (1972).
- Van der Waals, Jr., J. D., "Over de continuïteit von den gasen Vloeistof-toestand," Doctoral Dissertation, Leiden, Holland, (1873).

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