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# NOTE ON THE APPLICABILITY OF THE LEE–KESLER EQUATION OF STATE FOR $P$–$V$–$T$ CALCULATIONS

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## ABSTRACT

The predictive applicability of the Lee–Kesler equation of state was studied over a wide range of $P-V-T$ values for a series of pure compounds by means of the available experimental compressibility data. A comparison of calculated and experimental compressibility values indicates that the Lee–Kesler equation should be used with caution in the vicinity of the vapour–liquid critical point and in the low pressure region ($Z < 0.1$). The compressibility root mean square (RMS) percent errors increase with increasing dipole moment values, and relate non-linearly to Pitzer's acentric factor, $\omega$.

## INTRODUCTION

The demand for accurate volumetric properties of various fluids is increasing in process industries. One of the equations of state which has proved to be successful in process industries in predicting the phase equilibria is the Lee–Kesler equation [1].

This study has analyzed the predictive applicability of the Lee–Kesler equation of state through the use of the available compressibility data including those found in the low pressure range, near the critical point and in the liquid state. In this work the calculated compressibility values were compared with the experimental compressibility data. The comparison was done for each experimental data point in terms of the root mean square (RMS) percent error defined as

$$\mathrm{RMS} = \left[ \frac{\sum_{i=1}^{n} (\%\ \mathrm{error})^2}{n} \right]^{1/2} \qquad (1)$$

in which

$$\%\ \mathrm{error} = \left[ \frac{Z_{\mathrm{calc}} - Z_{\mathrm{exp}}}{Z_{\mathrm{exp}}} \right] \times 100 \qquad (2)$$

Table 1 presents the summary of data used and contains the results

TABLE 1

Input data and results

| Compound | Critical properties [a] |  | Physical properties [a] |  |  | Input data |  |
|---|---|---|---|---|---|---|---|
|  | $T_c$ (K) | $P_c$ (atm) | Acentric factor $\omega$ | Dipole moment (D) | Molecular weight $M$ | Number of data points $N$ | Refs. |
| Helium | 5.19 [c] | 2.24 [c] | −0.387 [c] | – | 4.003 | 34 | 14,15 |
| Hydrogen | 33.2 [c] | 12.8 [c] | −0.220 [c] | – | 2.016 | 52 | 14 |
| Argon | 150.8 | 48.1 | −0.004 | – | 39.944 | 31 | 14 |
| Krypton | 209.4 | 54.3 | −0.002 | – | 83.3 | 55 | 15 |
| Neon | 44.25 | 27.2 | 0.000 | – | 20.179 | 81 | 16 |
| Xenon | 298.58 | 57.6 | 0.002 | – | 131.3 | 87 | 14 |
| Methane | 190.6 | 45.4 | 0.008 | – | 16.043 | 24 | 14 |
| Oxygen | 154.6 | 49.8 | 0.021 | – | 31.999 | 14 | 17 |
| Nitrogen | 126.2 | 33.5 | 0.040 | – | 28.013 | 37 | 14 |
| Ethane | 305.4 | 48.2 | 0.098 | – | 30.07 | 29 | 14 |
| Hydrogen sulfide | 373.2 | 88.2 | 0.100 | 0.97 | 34.08 | 27 | 17 |
| *n*-Propane | 369.8 | 41.9 | 0.152 | 0.05 | 44.097 | 77 | 18 |
| Nitrous oxide | 309.6 | 71.5 | 0.160 | 0.167 | 44.013 | 84 | 19 |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| Isobutane | 408.1 | 36.0 | 0.176 | 0.100 | 58.124 | 90 | 20 |
| Chlorotrifluoromethane | 302.0 | 38.7 | 0.180 | 0.50 | 104.459 | 36 | 21 |
| 1-Butene | 419.56 | 39.7 | 0.187 | 0.34 | 56.108 | 52 | 22 |
| Isobutylene | 417.9 | 39.5 | 0.190 | 0.48 | 45.108 | 61 | 23,24 |
| *n*-Butane | 425.2 | 37.5 | 0.193 | 0.084 | 58.124 | 69 | 14,25 |
| Neopentane | 433.75 | 31.545 | 0.197 | – | 72.00 | 70 | 26 |
| Ethylene oxide | 469.0 | 71.0 | 0.200 | 1.89 | 44.054 | 12 | 27 |
| *cis*-2-Butene | 435.6 | 41.5 | 0.202 | 0.30 | 56.108 | 57 | 28 |
| Carbon dioxide | 304.2 | 72.8 | 0.225 | – | 44.01 | 39 | 14 |
| Isopentane | 460.39 | 33.37 | 0.227 | 0.13 | 72.0 | 76 | 29 |
| Ammonia | 405.6 | 111.3 | 0.250 | 1.47 | 17.0 | 22 | 13 |
| *n*-Pentane | 496.65 | 33.25 | 0.251 | 0.10 | 72.0 | 83 | 30 |
| Sulfur dioxide | 430.8 | 77.8 | 0.251 | 1.63 | 64.063 | 67 | 31 |
| 1,1,1,2,2-Pentafluoropropane | 380.11 | 30.962 | 0.302 | – | 134.0 | 28 | 32 |
| Isooctane | 544.25 | 25.49 | 0.303 | – | 114.224 | 63 | 14 |
| Acetone | 508.10 | 46.4 | 0.309 | 2.88 | 58.08 | 54 | 33 |
| Water | 647.3 | 217.6 | 0.344 | 1.85 | 18.015 | 43 | 34 |
| *n*-Heptane | 540.2 | 27.00 | 0.351 | 0.10 | 100.205 | 46 | 35 |
| *n*-Decane | 617.6 | 20.8 | 0.490 | – | 142.286 | 36 | 36 |
| tert-Butyl alcohol | 506.20 | 39.2 | 0.618 | 1.76 | 74.123 | 54 | 37 |

TABLE 1 (*continued*)

| Compound | Reduced property range | | $N > 2\%$ | Error analysis | | | |
|---|---|---|---|---|---|---|---|
| | $\Delta T_r$ | $\Delta P_r$ | | RMS in[b] $Z$-values | Absolute maximum error | | |
| | | | | | % Error | Reduced coordinates | |
| | | | | | | $T_r$ | $P_r$ |
| Helium | 3.85–129.07 | 0.44–88.12 | 5 | 3.76 | 13.05 | 3.85 | 44.07 |
| Hydrogen | 1.05–15.06 | 0.77–38.56 | 17 | 2.60 | 11.22 | 0.54 | 0.77 |
| Argon | 0.66–3.32 | 0.04–4.10 | 2 | 0.79 | 2.40 | 1.06 | 1.23 |
| Krypton | 1.07–1.54 | 0.02–3.64 | 2 | 0.56 | 2.27 | 1.07 | 1.23 |
| Neon | 1.58–2.71 | 0.25–11.21 | 6 | 1.15 | 5.49 | 1.58 | 11.12 |
| Xenon | 0.99–1.14 | 0.02–3.43 | 5 | 1.23 | 7.79 | 1.00 | 1.03 |
| Methane | 0.68–2.62 | 0.04–1.52 | 2 | 1.24 | 4.37 | 1.05 | 1.09 |
| Oxygen | 0.02–1.00 | 0.58–1.00 | 10 | 8.26 | 24.39 | 1.00 | 1.00 |
| Nitrogen | 0.79–3.17 | 0.06–2.95 | 3 | 1.07 | 3.10 | 0.99 | 1.03 |
| Ethane | 0.65–1.64 | 0.04–2.05 | 2 | 1.15 | 3.74 | 1.02 | 2.05 |
| Hydrogen sulfide | 0.57–0.97 | 0.01–0.83 | 17 | 5.56 | 22.77 | 0.97 | 0.83 |
| *n*-Propane | 0.84–1.30 | 0–1.19 | 7 | 4.81 | 64.99 | 0.89 | 0.24 |
| Nitrous oxide | 0.78–1.12 | 0.08–1.82 | 4 | 0.82 | 3.76 | 1.04 | 1.40 |
| Isobutane | 1.00–1.47 | 0.03–11.11 | 0 | 0.66 | 1.76 | 1.47 | 9.72 |
| Chlorotrifluoromethane | 0.90–1.32 | 0.50–2.49 | 2 | 1.74 | 8.60 | 1.00 | 0.98 |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| 1-Butene | 0.95–1.43 | 0.03–1.00 | 15 | 2.55 | 9.69 | 1.02 | 1.00 |
| Isobutylene | 0.96–1.08 | 0.03–2.53 | 17 | 4.84 | 29.97 | 0.98 | 0.63 |
| *n*-Butane | 0.69–1.20 | 0.04–10.53 | 18 | 1.57 | 2.60 | 0.77 | 5.44 |
| Neopentane | 0.67–1.38 | 0.03–12.68 | 8 | 1.11 | 3.17 | 0.99 | 0.92 |
| Ethylene oxide | 0.63–1.00 | 0.02–1.00 | 11 | 10.78 | 35.23 | 0.98 | 0.90 |
| *cis*-2-Butene | 0.92–1.38 | 0.02–1.20 | 19 | 5.50 | 22.04 | 1.03 | 1.20 |
| Carbon dioxide | 0.95–1.87 | 0.03–1.90 | 5 | 1.18 | 5.84 | 0.99 | 0.95 |
| Isopentane | 0.67–1.30 | 0.03–8.99 | 13 | 2.74 | 19.63 | 1.02 | 1.20 |
| Ammonia | 0.55–0.83 | 0.40–3.31 | 22 | 18.47 | 19.07 | 0.76 | 2.31 |
| *n*-Pentane | 0.62–1.27 | 0.03–21.05 | 60 | 7.79 | 80.93 | 0.91 | 0.72 |
| Sulfur dioxide | 0.66–1.00 | 0.01–1.54 | 2 | 0.72 | 2.68 | 1.00 | 0.13 |
| 1,1,1,2,2-Pentafluoropropane | 0.61–1.00 | 0.01–1.00 | 28 | 0.88 | 3.34 | 0.99 | 0.95 |
| Isooctane | 0.53–0.81 | 0.19–4.65 | 25 | 1.67 | 2834.0 | 0.81 | 0.19 |
| Acetone | 1.00–1.06 | 1.99–3.84 | 54 | 15.29 | 16.12 | 1.06 | 6.64 |
| Water | 0.43–1.00 | 0–1.00 | 22 | 7.25 | 36.68 | 1.00 | 0.97 |
| *n*-Heptane | 0.56–0.97 | 0.26–13.01 | 21 | 1.78 | 1702.19 | 0.78 | 0.27 |
| *n*-Decane | 0.48–0.72 | 0.65–9.81 | 0 | 0.74 | 1.50 | 0.72 | 5.72 |
| tert-Butyl alcohol | 0.03–0.71 | 0.94–1.05 | 27 | 2.63 | 6.15 | 0.97 | 0.61 |

[a] Taken from Reid et al. [13] unless otherwise indicated.

[b] The calculated errors obtained with the Lee–Kesler equation fall within the experimental range.

[c] Noting that the Lee–Kesler equation works reasonably well for $H_2$ at all temperatures and for He at temperatures > 50 K [12], we did not introduce quantum corrections [12] but used experimental $T_c$, $P_c$ and $\omega$ values.

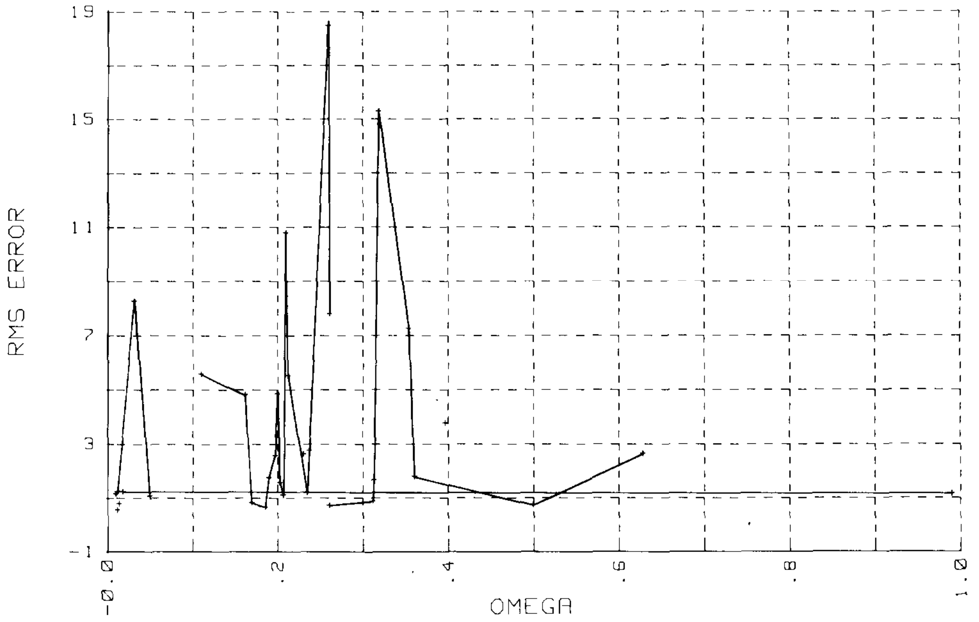

Fig. 1. RMS percent error plot vs. absolute value of Pitzer's acentric factor, $\omega$.

obtained thus providing an idea of the magnitude and nature of errors found in terms of the compound involved. Figures 1 and 2 strengthen this analysis of errors through a histogram of $|\omega|$ values, and a corresponding scattergram

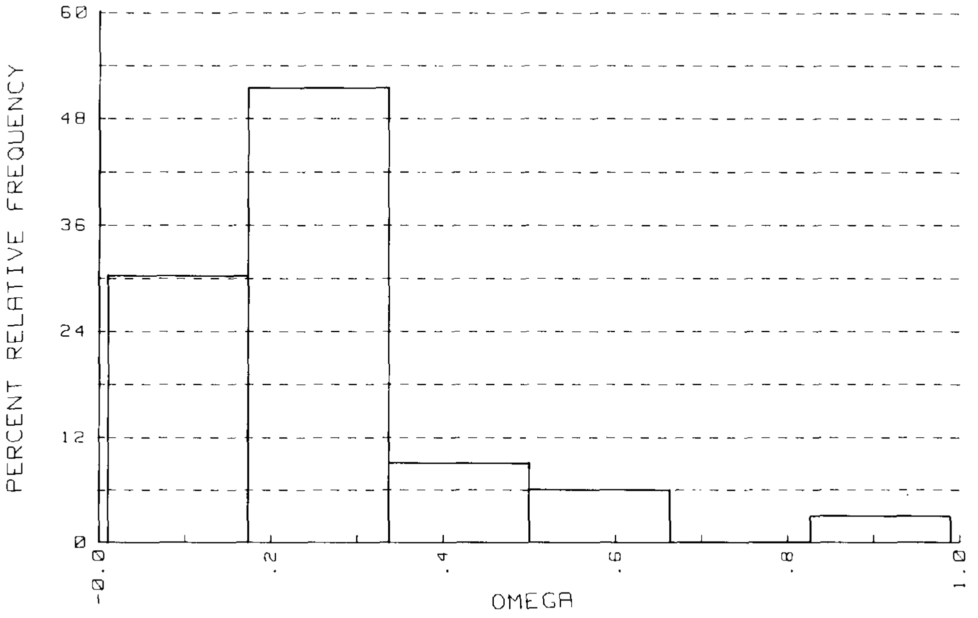

Fig. 2. Histogram of the absolute value of Pitzer's acentric factor, $\omega$.

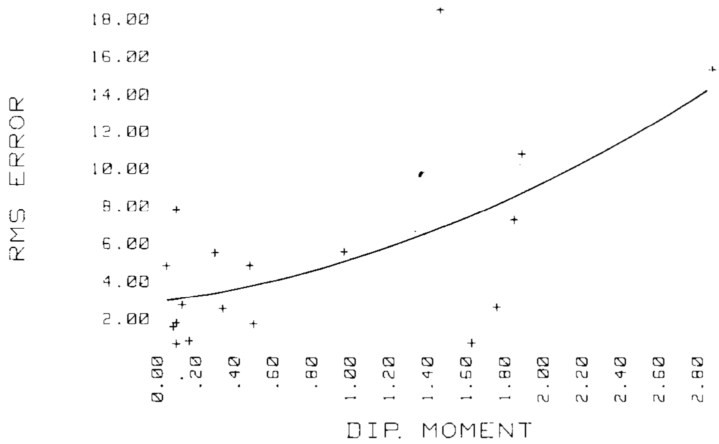

Fig. 3. RMS percent error plot vs. dipole moment, $\mu$, values.

of RMS percent error and the acentric factor $|\omega|$ distribution. Figure 3 on the other hand implies that there may be a connecting relation between the magnitude of the obtained RMS percent error and the isothermal dipole moment, $\mu$, value in terms of compounds considered. Finally, Table 2 provides a list of Lee–Kesler constants.

## LEE–KESLER EQUATION OF STATE

The Lee–Kesler equation is a classical corresponding states correlation [1,2–6]. To predict the compressibility factors, Lee and Kesler [1] took the

TABLE 2

Lee-Kesler constants (cf. footnote on p. 214; eqns. 4–7)

| | Simple fluid $Z^{(0)}$ calculation | Reference fluid $Z^{(r)}$ calculation |
|---|---|---|
| $b_1$ | 0.1181193 | 0.2026579 |
| $b_2$ | 0.265728 | 0.331511 |
| $b_3$ | 0.154790 | 0.027655 |
| $b_4$ | 0.030323 | 0.203488 |
| $c_1$ | 0.0236744 | 0.0313385 |
| $c_2$ | 0.0186984 | 0.0503618 |
| $c_3$ | 0.0 | 0.016901 |
| $c_4$ | 0.042724 | 0.041577 |
| $d_1 \times 10^4$ | 0.155488 | 0.48736 |
| $d_2 \times 10^4$ | 0.623689 | 0.0740336 |
| $\beta$ | 0.65392 | 1.226 |
| $\gamma$ | 0.060167 | 0.03754 |

macroscopic corresponding states correlation of Pitzer et al. [2–6]

$$Z = Z^{(0)} + \omega Z^{(1)} \tag{3}$$

and used a modified Benedict–Webb–Rubin equation given in reduced coordinates as

$$Z = \frac{P_r V_r}{T_r} = 1 + \frac{B}{V_r} + \frac{C}{V_r^2} + \frac{D}{V_r^5} + \frac{c_4}{T_r^3 V_r^2}\left(\beta + \frac{\gamma}{V_r^2}\right)\exp\left(-\frac{\gamma}{V_r^2}\right) \tag{4}$$

to predict the values of $Z^{(0)}$ and $Z^{(1)}$ (eqn. 3). Constants $B$, $C$ and $D$ in eqn. (4) are defined as *

$$B = b_1 - (b_2/T_r) - (b_3/T_r) - (b_4/T_r^3) \tag{5}$$

$$C = c_1 - (c_2/T_r) + (c_3/T_r^3) \tag{6}$$

$$D = d_1 + (d_2/T_r) \tag{7}$$

The pseudo-reduced volume, $V_r$, is given as

$$V_r = P_c V/RT_c \tag{8}$$

The theoretical basis of the predictive applicability of the Lee–Kesler equation is found in the inherent characteristics of Pitzer's use of the macroscopic theorem of corresponding states [2–5] and the range of applicability of the modified BWR relation [7] (eqn. 4). It is well known that Pitzer's macroscopic theorem of corresponding states being a first order perturbation about simple fluid (reference fluid, acentric factor $\omega = 0$) corresponding states, effectively can be applied only to moderately large nonspherical molecules. As such the work of Lee and Kesler is based on experimental data for hydrocarbons ranging from methane to *n*-octane as the heavy reference fluid with a subsequent adjustment to other substances. It is questionable whether the linear form of the $Z - \omega$ relation (eqn. 3) may be extended through the inclusion of higher order Taylor expansion terms [2–5,8]. Attempts have been made [8–12] to extend the applicability of the linear $Z - \omega$ relation (eqn. 3) at regular intervals of $T_r$ and $P_r$ in the region of $0.2 < T_r < 5.0$ and $0.0 < P_r < 12.0$. These extensions are valid strictly only for simpler molecules and certain groups of compounds, therefore, quantum gases and highly polar molecules may not be included in this type of correlation [8,12].

## EXPERIMENTAL DATA USED

The summary of experimental data used is presented in Table 1. The experimental gas and liquid state compressibility values for the given set of

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* Constants used (eqns. 5–7) are found in Table 1. Note that $\omega$ in the Pitzer relation (eqn. 3) is replaced by $\omega/0.3978$ for the Lee–Kesler equation. A value of 0.3978 represents the $\omega$ value for the reference fluid used (*n*-octane).

compounds (Table 1) were taken from various sources. These, from the literature data obtained, were thought to be sufficiently reliable, therefore, no further study to reevaluate their accuracy was made. Indeed, the data set used * for this study was regarded to be only a representative one. Following the prototype of McFee et al. [12], our program reads-in the experimentally determined compressibility factor $Z$ at the $T$ and $P$ of interest, and compares these data with the compressibility factor calculated by means of the Lee–Kesler equation of state at the same thermodynamic conditions. The RMS percent error (eqn. 1) is used as the basis for a comparison yielding a measure of accuracy of fit for the given set of state data points.

## RESULTS AND DISCUSSION

The fully generalized, classical corresponding states method of Lee and Kesler was studied with physical constant values (critical temperature and pressure, acentric factor, $\omega$, dipole moment, $\mu$) taken from Reid et al. [13]. The results show a varying degree of reliability for all the compounds in all regions studied. For instance, of the sample of 33 compounds considered (Table 1), 14 compounds have an RMS percent error of less than 1.5%, while 15 compounds have an RMS percent error in excess of 2%. Table 1 indicates that compounds which follow the corresponding states principle for simple and normal fluids (eqn. 3) and for which the intermolecular forces are conformal, show a high degree of compatibility with the Lee–Kesler equation of state in all regions considered as expected **.

Compounds which do not follow the corresponding states principle accurately and whose intermolecular forces are not conformal, do show deviations from the experimental results where compressibility calculations are performed using the Lee–Kesler method of $P$–$V$–$T$ data prediction. Table 1 shows that for non-conformal fluids these deviations appear to be associated first with various forms of compounds and natures of intermolecular forces (i.e., quantum fluids such as He; hydrogen bonded fluids such as $NH_3$ or $H_2O$; olefinic type fluids such as *cis*-2-butene), and secondly, with thermodynamic state condition (i.e., pressure and temperature range of the experimental study).

Since the predictive ability of the Lee–Kesler equation of state involves the use of basic assumptions of Pitzer's corresponding states principle (cf.

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* This also includes the listed (Table 1) acentric factor, $\omega$, and dipole moment, $\mu$, values.

** Note that the Lee–Kesler analytical method expresses Pitzer's macroscopic corresponding states correlation (eqn. 3) through a reduced form of a modified Benedict–Webb–Rubin equation of state (eqn. 4) in which a set of constants (eqns. 5–7) for the simple fluid are determined from the data of argon, krypton and methane and that of the reference fluid are from the data of *n*-octane [1]. See also footnote on p. 214.

eqn. 3; footnotes on pp. 214, 215) it seems feasible to expect that the RMS percent error found in compressibilities (Table 1) may relate to physical parameters expressing intermolecular force interactions.

Two parameters were considered in this study: the acentric factor, $\omega$ (eqn. 3) which measures the overall deviation of the intermolecular potential function from that of the simple spherical molecules; and the dipole moment, $\mu$, which account specifically for the polarity effects of the given compound. The scattering (Fig. 1) shows that RMS percent error values increase with increasing absolute value of the acentric factor, reaching a maximum at around an $|\omega|$ of 0.26. The $|\omega|$ histogram (Fig. 2) shows that most of $|\omega|$ values fall within the same $|\omega|$ range as that obtained for the RMS percent error (Fig. 1). However Fig. 3 indicates that the Lee–Kesler equation following the simple corresponding states principle (eqn. 3) may not completely account for polarity effects since RMS percent errors are shown to correlate distinctly with dipole moment, $\mu$, values. Whether this indicates that the derivation of the Lee–Kesler equation should be modified through the introduction of an extended theorem of a simple corresponding states [12] in preference to the presently used Pitzer's simple corresponding states principle (eqn. 3), is an open question. Yet it should be recalled that there are many non-conformal substances whose interactions cannot be described through an intermolecular potential function and which therefore cannot be subject to a corresponding states principle.

Table 1 shows that in general the non-conformal fluids possess large absolute maximum error values within the critical region and at high pressures. Yet some of the simpler paraffins, such as *n*-propane, iso-pentane, *n*-pentane, iso-octane and *n*-heptane, show considerable irregularities at low pressures. Although recently Hsiao and Lu [8] extended the applicability range of the linear Pitzer correlation (eqn. 3) in the $T_r - P_r$ region of $0.2 < T_r < 5.0$ and $0 < P_r < 12.0$ and the experimental compressibility values were erratic at given low pressures, it is still felt that at these low pressures, the use of the Lee–Kesler equation should be tempered with caution.

## LIST OF SYMBOLS

$B$, $C$, $D$, $b_1$, $b_2$, $b_3$, $b_4$, $c_1$,
$c_2$, $c_3$, $c_4$, $d_1$, $d_2$, $\beta$, $\gamma$ Lee–Kesler equation constants
$P$ pressure
$R$ universal gas constant
$T$ temperature
$Z$ compressibility factor
$\rho$ density
$\omega$ acentric factor

*Superscript*

0 reference, ideal

*Subscripts*

c critical state
r reduced state with respect to the vapour–liquid critical state

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