

An Improved Correlation for Second Virial Coefficients of Pure Fluids

S.A. Razavizadeh

Department of Chemistry, Payame Noor University, P. O. Box: 19395-3697, Tehran, Iran

(Received 2 June 2016, Accepted 13 October 2016)

In the present work, a modified correlation is presented for the second virial coefficients of both polar and nonpolar fluids based on the corresponding states principle. The second virial coefficients of gaseous polar and non-polar compounds were calculated and compared with experimental data and with other correlations. Comparisons with the existing correlations show that the present work is more accurate and reliable for nonpolar and polar compounds. The results also show that the present work is nearly equivalent to the Meng *et al.* correlations for the second virial coefficients of nonpolar fluids. The second virial coefficient of refrigerants, nonhydrogen bonding compounds and alcohols give a satisfactory result with respect to other correlations. The new correlations for the second coefficients need only the critical temperature, critical pressure, acentric factor and reduced dipole moment as parameters. According to the results, the root mean square deviation of second virial coefficients of the nonpolar compound, polar haloalkanes, and nonhydrogen bonding molecules, and alcohols are 10.58, 11.57 and 75.29 respectively.

Keywords: Second virial coefficients, Nonpolar, Polar

INTRODUCTION

The *PVT* behavior of real gases is one of the most important topics that has been studied by physicists and chemists for more than a century. A part of this general interest concerns to the essential role of the investigation of gas imperfections in the clarification of the forces between molecules. From a more practical viewpoint, understanding of *PVT* relationships is fundamental for the resolution of problems in chemical engineering processes where gases are present. The virial equation is one of the most graceful and easy-to-use equations of state because of its outstanding foundation on the statistical-mechanical theory. Actually, the virial equation of state is a foreshortened series expansion of either the grand canonical partition function or the radial distribution function for low-density gasses.

The correlation of Pitzer and Curl [1] for the second virial coefficient, published in 1957, has been used extensively with great success and has been a foundation for several later

correlations. In 1974, Tsonopoulos [2,3] used newer and more extensive data, which improved the polynomials of Pitzer and Curl [1]. Weber [4], Meng *et al.* [5-7] and Di Nicola *et al.* [8, 9] carried out various modifications on Pitzer-Curl correlations, which refitted the coefficients of the Pitzer-Curl correlation and added polar and hydrogen bonding terms and new binary interaction parameter k_{ij} for calculating the second cross virial coefficients of mixtures. The present study, however, presents a new correlation using the selected data for nonpolar gases, polar haloalkanes, other non-hydrogen bonding polar gases, and alcohols.

SECOND VIRIAL COEFFICIENT

The development of a virial equation of state involves knowledge of large numbers of interaction virial coefficients. On the basis of the connection between the virial coefficients and the intermolecular potential, the corresponding state principle (CSP) can be extended to virial coefficients.

Tsonopoulos [2] modified the Pitzer-Curl equation to

*Corresponding author. E-mail: razavizadeh@pnu.ac.ir

give the following widely used expression for the gaseous second virial coefficient:

$$B_r = \frac{BP_c}{RT_c} = f^{(0)}(T_r) + \omega f^{(1)}(T_r) + f^{(2)}(T_r) \quad (1)$$

where

$$f^{(0)} = 0.1445 - \frac{0.330}{T_r} - \frac{0.1385}{T_r^2} - \frac{0.0121}{T_r^3} - \frac{0.000607}{T_r^8} \quad (2)$$

$$f^{(1)} = 0.0637 + \frac{0.331}{T_r^2} - \frac{0.423}{T_r^3} - \frac{0.008}{T_r^8} \quad (3)$$

and B and B_r are the second virial and the reduced second virial, respectively. The contribution of the spherical geometry, non-spherical geometry, and polarity of fluids to the reduced second virial coefficient are represented by $f^{(0)}(T_r)$, $f^{(1)}(T_r)$, and $f^{(2)}(T_r)$, respectively. Recall that P_c and T_c are the critical pressure and critical temperature, R is the universal gas constant, and ω is the acentric factor.

In this study, Eq. (1) is considered as the framework, while Eqs. (2), (3) are redefined by fitting experimental data. The source of these data is originated mainly from the compilation of Dymond *et al.* [10]. Based on the new fitting, the following equations were obtained:

$$f^{(0)} = 0.1333 - \frac{0.3033}{T_r} - \frac{0.1532}{T_r^2} - \frac{0.0105}{T_r^3} + \frac{0.00027}{T_r^8} \quad (4)$$

$$f^{(1)} = 0.1084 - \frac{0.2224}{T_r} + \frac{0.6755}{T_r^2} - \frac{0.6043}{T_r^3} - \frac{0.0041}{T_r^8} \quad (5)$$

The term $f^{(0)}$ is taken from the experimental data of the compounds with almost zero acentric factors such as argon. The PVT data of Gilgen *et al.* [11] and the Estrada-Alexanders and Trusler [12] for argon was used to determine $f^{(0)}$, because they are in superior agreement with each other. The excellent agreement between our calculated second virial coefficients with the values for argon recommended by Dymond [10] gives a more dependable starting point for finding $f^{(1)}$ and $f^{(2)}$.

The term $f^{(1)}$ was determined by fitting the data for nonspherical and nonpolar molecules ($\omega \neq 0$), such as normal alkanes, oxygen, nitrogen and carbon dioxide. Also, for

more representation of B over the entire temperature range, the term T_r^{-1} was added to the $f^{(1)}$.

An important fraction of the large number of new B data, available since around 1990, has been gathered in Dymond's 2002 review [10] and now can be examined with the goal of updating the Tsonopoulos correlation for nonpolar and polar fluids. Most of the new B data are on chlorofluoroalkanes and, especially, hydrofluoroalkanes. The ozone-depleting refrigerants containing chlorine are being substituted by the ozone-friendly refrigerants that contain fluorine instead of chlorine. Weber [4] and Meng *et al.* [5] re-examined the correlation of Tsonopoulos. The result of their study differs in the non-polar terms as well as the dependence of the polar contribution of B on μ_r , reduced dipole moment. For polar fluids, also, the data analysis gives almost the same relationship between $f^{(2)}$ and T_r as found earlier by Tsonopoulos [13]:

$$f^{(2)} = \frac{a}{T_r^6} - \frac{b}{T_r^8} \quad (6)$$

The Tsonopoulos investigation of the second virial coefficients of polar haloalkanes demonstrated an apparent difference between polar haloalkanes and non-hydrogen bonding fluids such as ketones and ethers [13]. It is only necessary to use a in Eq. (6) for nonhydrogen-bonding polar compounds such as ketones, that is, setting $b = 0$. Parameter a is determined as a function of μ_r .

Tsonopoulos proposed the following correlation for polar haloalkanes [3]:

$$a = -2.188 \times 10^{-11} \mu_r^4 - 7.831 \times 10^{-21} \mu_r^8 \quad (7)$$

For other nonhydrogen bonding polar compounds, Tsonopoulos also proposed:

$$a = -2.14 \times 10^{-4} \mu_r - 4.308 \times 10^{-21} \mu_r^8 \quad (8)$$

For alcohols, b is a compound-dependent parameter and the term $f^{(2)}$ is expressed by the following equation [2]:

$$f^{(2)} = \frac{0.0878}{T_r^6} - \frac{0.00908 + 0.0006957 \mu_r}{T_r^8} \quad (9)$$

In this study, for polar hydrogen bonding and nonhydrogen bonding compounds, the following relation is considered between $f^{(2)}$ and T_r as suggested earlier by Tsonopoulos only for nonhydrogen-bonding polar compounds:

$$f^{(2)} = a/T_r^6 \quad (10)$$

Parameter a in Eq. (10) is determined as a function of the reduced dipole moment, μ_r , which is always expected to be negative. However, a usually becomes slightly positive for $\mu_r < 100$. Hence, parameter a is found to be related to the reduced dipole moment μ_r as follows:

$$\mu_r = \frac{10^5 \mu^2 P_c}{T_c^2} \quad (11)$$

where μ is the dipole moment in Debye, T_c is critical temperature in Kelvins and P_c is critical pressure in atmosphere. For haloalkanes, a can be represented by the following expression:

$$a = 0.0143 - 1.22 \times 10^{-3} \mu_r + 3.4117 \times 10^{-5} \mu_r^2 - 4.1653 \times 10^{-7} \mu_r^3 + 2.2429 \times 10^{-9} \mu_r^4 - 4.514 \times 10^{-12} \mu_r^5 \quad (12)$$

In addition, for other nonhydrogen bonding polar compounds the following expression is introduced to generate a different dependency on a as a function of μ_r :

$$a = 0.0509 - 4.0371 \times 10^{-3} \mu_r + 8.2884 \times 10^{-5} \mu_r^2 - 7.733 \times 10^{-7} \mu_r^3 + 3.2851 \times 10^{-9} \mu_r^4 - 5.2332 \times 10^{-12} \mu_r^5 \quad (13)$$

Furthermore, for alcohols the following relation is found for the dependency of dipole moment a :

$$a = -0.1856 + 0.004628 \mu_r - 4.3336 \times 10^{-5} \mu_r^2 + 1.2811 \times 10^{-7} \mu_r^3 \quad (14)$$

Recently, Di Nicola *et al.* [8] introduced the following correlation for the second virial coefficient for refrigerants:

$$B = \frac{10^6 RT_c}{P_c} \left(\frac{m \log T_r + n \omega + h \mu_r + k}{T_r^{1.7}} \right) \quad (15)$$

where m , n , h and k are coefficients for each group of refrigerants.

RESULTS AND DISCUSSION

Researchers such as Dymond [10] have found out that many of the older *PVT* data, which have historically been the source of the virial coefficients, are not precise for physical adsorption effects. Therefore, the resulting B is very negative, especially at subcritical temperatures.

The scatter plot of the second virial coefficients versus the reduced temperature for the complete data set is described in Fig. 1. The five groups were expressed by different symbols. The different behavior of the second virial coefficients of the refrigerants is clearly evident in Fig. 1.

Furthermore, the experimental second virial coefficients versus the calculated second virial coefficients are shown in Fig. 2. Small deviations are obvious for all the five groups and for all ranges of the data.

The experimental data from the *PVT* measurements of Gilgen *et al.* [11] and from the speed-of-sound measurements of Estrada-Alexanders and Trusler [12] were used to investigate the second virial coefficients for argon. The comparison of the deviations of predicted second virial coefficients in Fig. 3 shows that the present correlation clearly improves the Tsonopoulos correlations with its too negative values at low temperatures and positive values at high temperatures. The comparison in Fig. 3 for methane shows that the present correlation gives the satisfactory results over the entire range, while the Tsonopoulos correlation is too negative below a reduced temperature of 1.5.

The second virial coefficients for n-butane were taken from the experimental data of the isochoric Burnett measurements of Gupta and Eubank [14] at 264.9-450 K. According to Fig. 3, our correlation is in excellent compliance with the experimental data.

According to Table 1, the root mean square deviations (RMSD) for the second virial coefficient of 15 nonpolar fluids are presented. These results show that new correlation is similar to Meng *et al.* correlation, though it is much closer to the experimental data than the one obtained by Tsonopoulos and Weber.

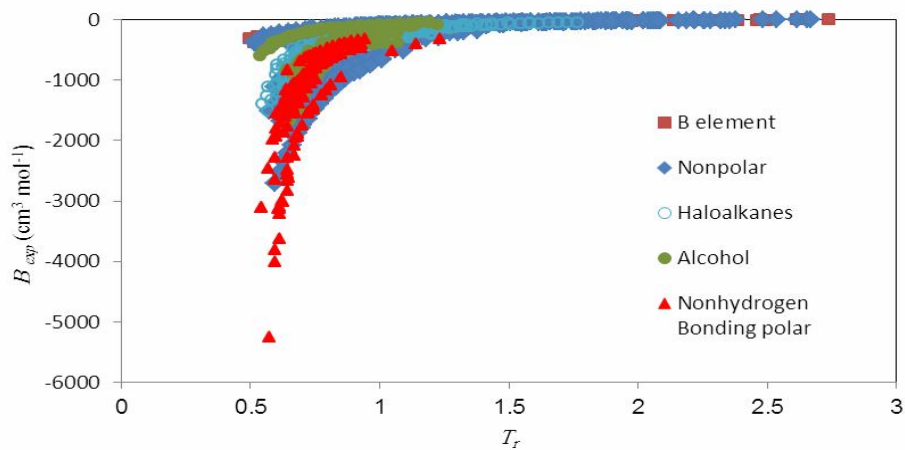


Fig. 1. Experimental second virial coefficients versus the reduced temperature.

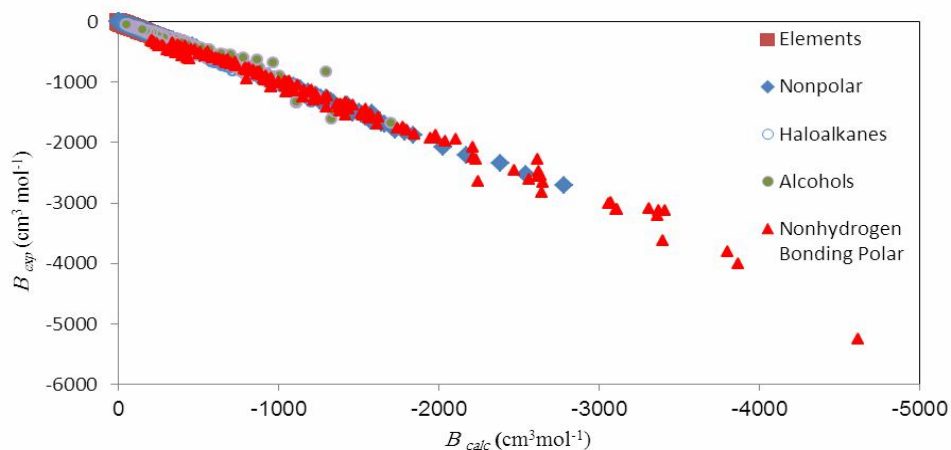


Fig. 2. Experimental second virial coefficients versus the calculated second virial coefficients.

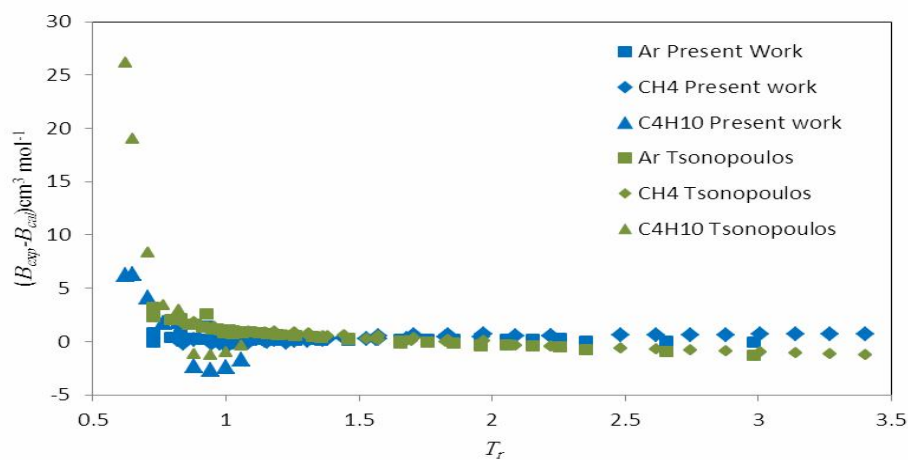


Fig. 3. Deviations of the second virial coefficients of argon, methane, and butane from the experimental data [11-19].

Table 1. Comparison of the Calculated Results from the Present Work and other Correlation for Nonpolar Molecules

| Substance | T-range | No. point | RMSD (cm ³ mol ⁻¹) ^a | | | | Ref. |
|--------------------------------|---------|-----------|---|--------------------|-------------|--------|------------|
| | | | Present work | Meng <i>et al.</i> | Tsonopoulos | Weber | |
| Ar | 110-450 | 44 | 0.36 | 0.35 | 1.20 | 0.89 | [11,12] |
| Kr | 107-873 | 77 | 8.22 | 8.41 | 4.25 | 11.13 | [10] |
| Xe | 160-973 | 68 | 3.62 | 3.75 | 2.81 | 4.43 | [10] |
| CH ₄ | 160-673 | 49 | 0.40 | 0.13 | 0.90 | 0.73 | [15-19] |
| C ₂ H ₆ | 200-623 | 24 | 3.02 | 2.62 | 4.07 | 2.32 | [10,18,19] |
| C ₃ H ₈ | 270-573 | 46 | 32.49 | 31.88 | 33.87 | 30.39 | [10] |
| C ₄ H ₁₀ | 265-450 | 9 | 3.69 | 1.80 | 11.27 | 28.78 | [14] |
| C ₅ H ₁₂ | 270-648 | 20 | 23.85 | 17.59 | 42.72 | 44.36 | [10] |
| C ₆ H ₁₄ | 313-663 | 40 | 16.05 | 18.97 | 14.86 | 75.84 | [10] |
| C ₇ H ₁₆ | 350-598 | 19 | 14.23 | 20.35 | 10.85 | 86.27 | [10] |
| C ₈ H ₁₈ | 337-623 | 25 | 38.60 | 42.65 | 53.02 | 150.63 | [10] |
| CO ₂ | 217-773 | 45 | 1.28 | 1.31 | 1.43 | 2.24 | [10] |
| N ₂ | 98-748 | 46 | 1.31 | 0.85 | 1.52 | 1.22 | [10,19] |
| O ₂ | 85-476 | 53 | 0.99 | 1.02 | 2.49 | 2.01 | [10] |
| Average | | | 10.58 | 10.83 | 13.23 | 31.52 | |

^aRoot mean square deviation.

The data analysis of polar molecules leads to optimum values of parameter a for 21 haloalkanes, 7 other non-associating fluids and 5 alcohols listed in Tables 2 and 3. These values exhibit a more definitive relationship between a and μ_r for polar compounds.

The optimum values of a for the 21 haloalkanes and 7 other non-associating fluids are plotted versus μ_r in Fig. 4. Both equations (12) and (13) for nonpolar fluids are plotted in Fig. 4.

Figure 5 shows the deviation of the second virial for R13 (CCl₃), R32 (CH₂F₂), R141b (C₂H₃Cl₂F), R125 (C₂HF₅), R134a (C₂H₂F₄), R123 (C₂HCl₂F₃) and

nitromethane from experimental data. The results show how the present correlation is much closer to experimental data than that obtained by Tsonopoulos and Meng. The comparison in Fig. 5, for R32, shows that the present correlation gives satisfactory results over the entire range, while the Tsonopoulos correlation is too negative below a reduced temperature of 0.7. We present deviation between the calculated values of the second virial coefficients of R141b and experimental data of Goodwin and Moldover [36] in Fig. 5. According to this Figure, the new correlation has a good prediction of the second virial coefficient for R125 although the deviation increase at $T_r < 0.8$.

Table 2. Comparison of the Calculated Results from the Present Work and other Correlations for Polar Haloalkanes Molecules

| Substance | Formula | μ_r | Optimum a | RMSD(cm ³ mol ⁻¹) ^a | | | | | | Ref. |
|-----------|---|---------|-------------|---|----------------------|--------------------|-------------|--------|-------------------------|---------|
| | | | | Present work (optimum a) | Present work Eq. (9) | Meng <i>et al.</i> | Tsonopoulos | Weber | Di Nicola <i>et al.</i> | |
| R-11 | CCl ₃ F | 3.97 | 0.01419 | 11.27 | 14.05 | 32.43 | 34.24 | 27.35 | 26.65 | [20] |
| R-12 | CCl ₂ F ₂ | 7.16 | 0.00180 | 5.17 | 27.01 | 10.02 | 16.62 | 8.95 | 14.28 | [20] |
| R-13 | CCIF ₃ | 10.92 | 0.00470 | 2.10 | 2.11 | 5.06 | 6.44 | 4.01 | 13.19 | [20] |
| R-22 | CHClF ₂ | 76.76 | -0.00440 | 4.86 | 13.98 | 5.25 | 8.55 | 31.47 | 14.42 | [10] |
| R-23 | CHF ₃ | 144.77 | -0.01350 | 5.31 | 5.33 | 4.66 | 3.26 | 11.24 | 4.10 | [10] |
| R-32 | CH ₂ F ₂ | 180.95 | -0.02920 | 11.74 | 12.58 | 10.48 | 43.38 | 125.55 | 75.6 | [21-25] |
| R40 | CH ₃ Cl | 118.41 | -0.00890 | 8.40 | 9.95 | 8.92 | 9.78 | 26.45 | 10.39 | [10] |
| R-41 | CH ₃ F | 198.08 | -0.04900 | 5.82 | 5.82 | 5.15 | 5.28 | 25.88 | 9.60 | [10] |
| R-114 | C ₂ Cl ₂ F ₄ | 7.93 | 0.00240 | 12.35 | 32.25 | 22.98 | 34.98 | 18.50 | 21.23 | [10] |
| R-115 | C ₂ ClF ₅ | 6.68 | 0.01100 | 3.59 | 4.09 | 10.21 | 12.48 | 11.25 | 7.19 | [10] |
| R-123 | C ₂ HCl ₂ F ₃ | 31.84 | -0.00154 | 1.58 | 2.88 | 6.22 | 21.83 | 84.58 | 47.89 | [26] |
| R-124 | C ₂ HClF ₄ | 49.36 | -0.00050 | 2.64 | 2.98 | 13.06 | 7.21 | 41.22 | 31.53 | [27] |
| R-125 | C ₂ HF ₅ | 75.82 | 0.00120 | 0.98 | 3.25 | 9.90 | 5.21 | 7.83 | 10.06 | [27-29] |
| R-134a | C ₂ H ₂ F ₄ | 121.17 | -0.00750 | 2.72 | 3.86 | 3.10 | 7.42 | 48.94 | 19.71 | [30-34] |
| R-141b | C ₂ H ₃ Cl ₂ F | 77.5 | 0.00025 | 16.87 | 22.73 | 78.78 | 57.12 | 49.62 | 41.45 | [35] |
| R-142b | C ₂ H ₃ ClF ₂ | 109.29 | -0.00400 | 11.46 | 20.17 | 26.14 | 13.37 | 66.16 | 26.34 | [27] |
| R-143a | C ₂ H ₃ F ₃ | 169.91 | -0.01670 | 6.39 | 7.06 | 5.69 | 10.49 | 29.70 | 8.27 | [27,33] |
| R-152a | C ₂ H ₄ F ₂ | 152.76 | -0.01600 | 8.11 | 10.64 | 20.05 | 10.60 | 73.15 | 35.16 | [33] |
| R-227ea | C ₃ HF ₇ | 43.58 | -0.00220 | 8.11 | 8.83 | 8.87 | 12.56 | 26.56 | 9.37 | [10] |
| R-236ea | C ₃ H ₂ F ₆ | 25.9 | 0.00230 | 10.12 | 21.80 | 20.33 | 24.36 | 30.63 | 4.56 | [27] |
| Average | | | | 6.98 | 11.57 | 15.37 | 17.26 | 37.45 | 21.55 | |

^aRoot mean square deviation.

Table 3. Comparison of the Calculated Results from the Present Work and other Correlations for Nonhydrogen Bonding Molecules and Alcohols

| Substance | μ_r | Optimum a | RMSD ($\text{cm}^3 \text{mol}^{-1}$) ^a | | | | Ref. |
|---------------|---------|-------------|--|----------------------------------|-----------------------|-------------|------------|
| | | | Present work (optimum a) | Present Work Eq. (9), (10) | Meng <i>et al.</i> | Tsonopoulos | |
| Nitromethane | 215.483 | -0.05627 | 13.81 | 13.82 | 150.74 | 359.89 | [36,37] |
| Diethyl ether | 21.812 | -0.00503 | 47.45 | 47.45 | 69.07 | 39.16 | [38,39] |
| Acetonitrile | 246.157 | -0.1229 | 168.62 | 168.63 | 209.08 | 203.25 | [10,39] |
| Acetone | 151.078 | -0.0332 | 26.21 | 28.88 | 28.80 | 30.04 | [38,40,41] |
| 2-Butanone | 111.29 | -0.023 | 34.06 | 35.41 | 72.52 | 84.82 | [10] |
| 2-Pentanone | 84.42 | -0.0215 | 156.37 | 174.11 | 203.71 | 157.82 | [10] |
| 3-Pentanone | 92.84 | -0.0179 | 61.11 | 70.83 | 62.74 | 73.25 | [10] |
| Methanol | 87.878 | -0.0272 | 46.09 | 46.12 | 19.89 | 85.20 | [38-40] |
| Ethanol | 66.398 | -0.0292 | 81.14 | 82.04 | 63.37 | 55.63 | [10,38] |
| 1-Propanol | 51.178 | -0.0412 | 82.20 | 83.95 | 128.45 | 175.66 | [10] |
| 2-Propanol | 52.569 | -0.0505 | 75.82 | 83.78 | 191.65 | 249.00 | [10] |
| 1-Butanol | 43.902 | -0.05385 | 68.01 | 68.42 | 243.10 | 357.43 | 10 |
| Average | | | 71.74 | 75.29 | 120.26 | 155.93 | |

^aRoot mean square deviation.

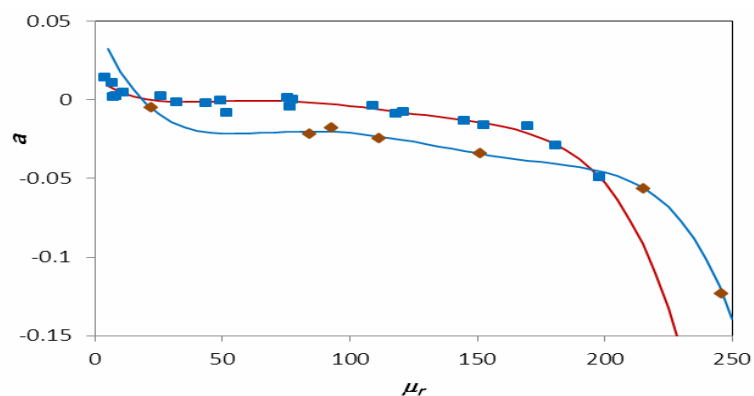


Fig. 4. Dependence of the polar parameter a on the reduced dipole moment. (■) Optimum values for haloalkanes; (◆) optimum values for other nonhydrogen bonding polar fluids; (—) Eq. (12); (—) Eq. (13).

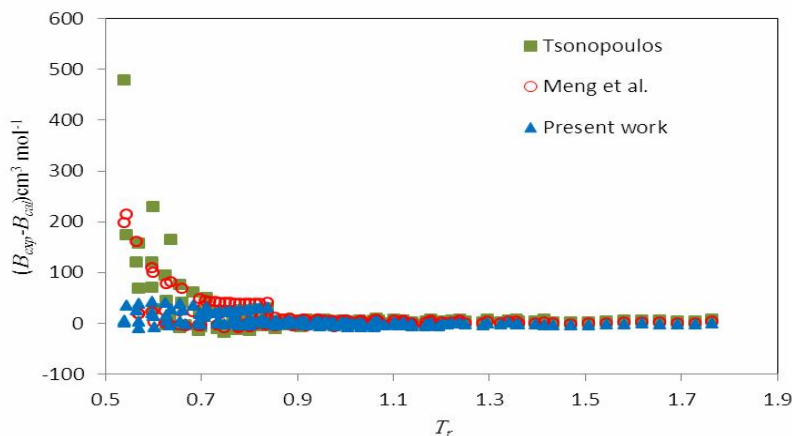


Fig. 5. Deviations of second virial coefficients of R13, R32, R141b, R125, R134a, R123 and nitromethane from experimental data [20-38].

CONCLUSIONS

In the foregoing sections, the well-known Pitzer-Curl and Tsonopoulos correlations have been modified for the second virial coefficients for nonpolar and polar fluids based on the corresponding states principle. Comparing the results of the present correlation with the new, high-quality experimental data and also existing models shows that the present correlation is more accurate, dependable and satisfactory for nonpolar and polar compounds.

The comparison of the present and other correlations for the nonpolar gases illustrate that, in terms of precision, the present correlation is equivalent to the correlation proposed by Meng *et al.* and better than Tsonopoulos' correlation. However, for polar haloalkanes, polar non-hydrogen bonding fluids, and alcohols, the present correlation is often more accurate than other correlations.

New correlation presents a highly satisfactory scheme for the prediction of the second virial coefficients for the nonpolar fluids, but is less satisfactory for polar fluids. Therefore, a more accurate approximation to polar contribution is needed to improve the predictive capability of the model.

Although Meng *et al.* correlation modified nonpolar term; it cannot provide significantly improved predictions for the second virial coefficients of the polar fluids.

Finally, the overall comparison of the correlations

shows that the present work offers quite satisfactory results. Although there is a slight decrease in the overall RMSD for the second virial coefficients of polar haloalkanes, there occurs an important improvement in the overall RMSD for B of other non-hydrogen bonding and alcohols.

List of Symbols

- a parameter of polar contribution to B , $f^{(2)}$
- B second virial coefficients
- f dimensionless functions of T_r in B correlations
- P pressure
- R universal gas constant
- T temperature

Greek Letters

- μ dipole moment, in Debye
- ω acentric factor

Subscripts

- c critical property
- cal calculated result
- exp experimental result
- r reduced property

ACKNOWLEDGMENTS

This work was supported by the Payame Noor

University.

REFERENCES

- [1] Pitzer, K. S.; Curl Jr., R. F., The volumetric and thermodynamic properties of fluids. III. Empirical equation for the second virial coefficient. *J. Am. Chem. Soc.* **1957**, *79*, 2369-2370, DOI: 10.1021/ja01567a007.
- [2] Tsonopoulos, C., An empirical correlation of second virial coefficients. *AIChE J.* **1974**, *20*, 263-272, DOI: 10.1002/aic.690200209.
- [3] Tsonopoulos, C., Second virial coefficients of polar haloalkanes. *AIChE J.* **1975**, *21*, 827-829, DOI: 10.1002/aic.690210435.
- [4] Weber, L. A., Estimating the virial coefficients of small polar molecules. *Int. J. Thermophys.* **1994**, *15*, 461-482.
- [5] Meng, L.; Duan, Y. Y.; Li, L., Correlations for second and third virial coefficients of pure fluids. *Fluid Phase Equilib.* **2004**, *226*, 109-120, DOI: 10.1016/j.fluid.2004.09.023.
- [6] Meng, L.; Duan, Y. Y.; Wang, X. D., Binary interaction parameter *kij* for calculating the second cross-virial coefficients of mixtures. *Fluid Phase Equilib.* **2007**, *260*, 354-358, DOI: 10.1016/j.fluid.2007.07.044.
- [7] Meng, L.; Duan, Y. Y., Prediction of the second cross virial coefficients of nonpolar binary mixtures. *Fluid Phase Equilib.* **2007**, *238*, 229-238, DOI: DOI: 10.1016/j.fluid.2005.10.007.
- [8] Di Nicola, G.; Coccia, G.; Pierantozzi, M.; Falone, M., A semi-empirical correlation for the estimation of the second virial coefficients of refrigerants. *Int. J. Ref.* **2016**, *68*, 242-251.
- [9] Di Nicola, G.; Falone, M.; Pierantozzi, M.; Stryjek, R., An improved group contribution method for the prediction of second virial coefficients. *Ind. Eng. Chem. Res.* **2014**, *53*, 13804-13809.
- [10] Dymond, J. H.; Marsh, K. N.; Wilhoit, R. C.; Wong, K. C., *Virial coefficients of pure gases and mixtures, subvol. A, Virial Coefficients of Pure Gases*, Landolt-Bornstein, **2002**.
- [11] Gilgen, R.; Kleinrahm, R.; Wagner, W., Measurement and correlation of the (pressure, density, temperature) relation of argon I. The homogeneous gas and liquid regions in the temperature range from 90 K to 340 K at pressures up to 12 MPa. *J. Chem. Thermodyn.* **1994**, *26*, 383-398, DOI: 10.1006/jcht.1994.1048.
- [12] Estrada-Alexanders, A. F.; Trusler, J. P. M., Thermodynamic properties of gaseous argon at temperatures between 110 and 450 K and densities up to 6.8 mol dm⁻³ determined from the speed of sound. *Int. J. Thermophys.* **1996**, *17*, 1325-1347.
- [13] Tsonopoulos, C.; Dymond, J. H., Second virial coefficients of normal alkanes, linear 1-alkanols (and water), alkyl ethers, and their mixtures. *Fluid Phase Equilibria* **1997**, *133*, 11-34, DOI: 10.1016/S0378-3812(97)00058-7.
- [14] Gupta, D.; Eubank, P. T., Density and virial coefficients of gaseous butane from 265 to 450 K at Pressures to 3.3 MPa. *J. Chem. Eng. Data* **1997**, *42*, 961-970, DOI: 10.1021/je970034v
- [15] Holleran, E. M., Accurate virial coefficients from P, V, T data. *J. Chem. Thermodyn.* **1970**, *2*, 779-786, DOI: 10.1016/0021-9614(70)90019-4
- [16] Roe, D. R., Ph.D. Thesis, Univ. London, London, England, **1972**.
- [17] Haendel, G.; Kleinrahm, R.; Wagner, W., Measurements of the (pressure, density, temperature) relation of methane in the homogeneous gas and liquid regions in the temperature range from 100-260 K and at pressures up to 8 MPa. *J. Chem. Thermodyn.* **1992**, *24*, 685-695, DOI: 10.1016/S0021-9614(05)80076-X.
- [18] Trusler, J. P. M., The speed of sound in (0.8CH₄ + 0.2C₂H₆)(g) at temperatures between 200 K and 375 K and amount-of-substance densities up to 5 mol dm⁻³. *J. Chem. Thermodyn.* **1994**, *26*, 751-763, DOI: 10.1006/jcht.1994.1089.
- [19] Hou, H. J.; Holste, C.; Hall, K. R.; Marsh, K. N.; Gammon, B. E., Second and Third Virial Coefficients for Methane + Ethane and Methane + Ethane + Carbon Dioxide at (300 and 320) K. *J. Chem. Eng. Data* **1996**, *41*, 344-353. DOI: 10.1021/je9502307.
- [20] Kunz, R. G.; Kapner, R. S., Second virial coefficients from tabulated P-V-T data. Results for chlorotrifluoromethane, dichlorodifluoromethane,

- fluorotrichloromethane, and trimethyl borate. *J. Chem. Eng. Data* **1969**, *14*, 190-191, DOI: 10.1021/je60041a003.
- [21] Utenkov, V. F., Ph.D. Thesis, MEI, Moscow, **1976**.
- [22] Defibaugh, D. R.; Morrison, G.; Weber, L. A., Thermodynamic properties of difluoromethane. *J. Chem. Eng. Data* **1994**, *39*, 333-340, DOI: 10.1021/je00014a032.
- [23] Hozumi, T.; Sato, H.; Watanabe, K., Speed of sound in gaseous difluoromethane. *J. Chem. Eng. Data* **1994**, *39*, 493-495, DOI: 10.1021/je00015a020.
- [24] Sato, T.; Sato, H.; Watanabe, K., PVT property measurements for difluoromethane. *J. Chem. Eng. Data* **1994**, *39*, 851-854, DOI: 10.1021/je00016a048.
- [25] Sun, L. Q.; Duan, Y. -Y.; Shi, L.; Zhu, M. S.; Han, L. Z., Speed of sound and ideal-gas heat capacity at constant pressure of gaseous difluoromethane. *J. Chem. Eng. Data* **1997**, *42*, 795-799, DOI: 10.1021/je970035n.
- [26] Goodwin, A. R. H.; Moldover, M. R., Thermophysical properties of gaseous refrigerants from speed-of-sound measurements. III. Results for 1,1-dichloro-2,2,2-trifluoroethane (CHCl₂-CF₃) and 1,2-dichloro-1,2,2-trifluoroethane (CHClF-CClF₂). *J. Chem. Phys.* **1991**, *95*, 5236-5242, <http://dx.doi.org/10.1063/1.461831>.
- [27] Gillis, K. A., Thermodynamic properties of seven gaseous halogenated hydrocarbons from acoustic measurements: CHClFCF₃, CHF₂CF₃, CF₃CH₃, CHF₂CH₃, CF₃CHFCHF₂, CF₃CH₂CF₃, and CHF₂CF₂CH₂F. *Int. J. Thermophys.* **1997**, *18*, 73-135.
- [28] Boyes, S. J.; Weber, L. A., Vapour pressures and gas-phase (p , ρ_n , T) values for CF₃CHF₂(g) (R125). *J. Chem. Thermodyn.* **1995**, *27*, 163-174, DOI: 10.1006/jcht.1995.0013.
- [29] Duarte-Garza, H. A.; Stouffer, D. E.; Hall, K. R.; Holste, J. C.; Marsh, K. N.; Gammon, B. E., Experimental critical constants, vapor pressures, and vapor and liquid densities for pentafluoroethane (R-125). *J. Chem. Eng. Data* **1997**, *42*, 745-753, DOI: 10.1021/je960362f.
- [30] Goodwin, A. R. H.; Moldover, M. R., Thermophysical properties of gaseous refrigerants from speed of sound measurements. I. Apparatus, model, and results for 1,1,1,2-tetrafluoroethane R134a. *J. Chem. Phys.* **1990**, *93*, 2741-2753, <http://dx.doi.org/10.1063/1.458913>.
- [31] Qian, Z. Y.; Sato, H.; Watanabe, K., Compressibility factors and virial coefficients of HFC-134a by the Burnett method. *Fluid Phase Equilib.* **1992**, *78*, 323-329, DOI: 10.1016/0378-3812(92)87044-N.
- [32] Bignell, C. M.; Dunlop, P. J., Second virial coefficients for seven fluoroethanes and interaction second virial coefficients for their binary mixtures with helium and argon. *J. Chem. Phys.* **1993**, *98*, 4889-4891, <http://dx.doi.org/10.1063/1.464943>.
- [33] Beckermann, W.; Kohler, F., Acoustic determination of ideal-gas heat capacity and second virial coefficients of some refrigerants between 250 and 420 K. *Int. J. Thermophys.* **1995**, *16*, 455-464.
- [34] F. Weber, Measurement of temperature-dependence of the 2nd virial-coefficients of the coolants R134a and R124 in the range of 290-360 K at pressures below 0.1 MPa. *PTB-Mitt.* **1996**, *106*, 17-26.
- [35] Goodwin, A. R. H.; Moldover, M. R., Thermophysical properties of gaseous refrigerants from speed-of-sound measurements. II. Results for 1,1-dichloro-1-fluoroethane (CCl₂FCH₃). *J. Chem. Phys.* **1991**, *95*, 5230-5235, <http://dx.doi.org/10.1063/1.461694>.
- [36] McCullough, J. P.; Scott, D. W.; Pennington, R. E.; Hossenlopp, I. A.; Waddington, G., Nitromethane: The vapor heat capacity, heat of vaporization, vapor pressure and gas imperfection; the chemical thermodynamic properties from 0 to 1500 °K. *J. Am. Chem. Soc.* **1954**, *76*, 4791-4796, DOI: 10.1021/ja01648a008.
- [37] Brown, I.; Smith, F., Liquid-vapour equilibria. X. The systems acetone + Nitromethane and acetone + acetonitrile at 45 °C. *Aust. J. Chem.* **1960**, *13*, 30-37, DOI: 10.1071/CH9600030.
- [38] Knoebel, D. H.; Edmister, W. C., Second virial coefficients of binary mixtures of benzene with methanol, ethanol, acetone, and diethyl ether. *J. Chem. Eng. Data* **1968**, *13*, 312-317, DOI: 10.1021/je60038a005.
- [39] Olf, G.; Schnitzler, A.; Gaube, J., Virial coefficients of binary mixtures composed of polar substances. *Fluid Phase Equilib.* **1989**, *49*, 49-65, DOI: 10.1016/0378-3812(89)80005-6.

- [40] Bottomley, G. A.; Spurling, T. H., Measurement of the temperature variation of virial coefficients. III. The second virial coefficient of Carbon Disulphide, Methyl Chloride, Acetone, and Methanol, and certain of their binary mixtures. *Aust. J. Chem.* **1967**, *20*, 1789-1796. DOI: 10.1071/CH9671789.
- [41] Hajjar, R. F.; Kay, W. B.; Leverett, G. F., Second virial coefficients of eight compounds in the range 40.deg. to 200 deg. *J. Chem. Eng. Data.* **1969**, *14*, 377-380. DOI: 10.1021/je60042a039.