Comparison of bulk modulus as Benzene dense fluid using the LIR equation of state with the extended coefficients and comparison with Peng-Robinson equation of state

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ARTICLE INFO

ABSTRACT

New parameters of the linear isotherm regularity, the so-called LIR equation of state, are used to calculate the bulk modulus of dense fluids. In this work, we drive an expression for the bulk modulus of dense fluids (CO, C_6H_6, $C_6H_5CH_3$) using the linear isotherm regularity (LIR). In later stages, bulk modulus calculated by Peng - Robinson (PR) equation of state as a test of the other equation of state. Comparison of the calculated values of bulk modulus with the extended coefficients of the linear isotherm regularity with the values obtained experimentally shows the accuracy of this method to be is general, quite good.

1. Introduction

The bulk modulus is a scalar quantity relating an isotropic pressure to an average change in volume. It is the average of the three inverse linear compressibilities (change of length induced by pressure). The bulk modulus is defined as:

$$B = -\frac{1}{\rho}\left(\frac{\partial p}{\partial V}\right)_T = \rho\left(\frac{\partial p}{\partial \rho}\right)_T \quad (1)$$

Huang and O'Connell [1] discovered a regularity in which all isotherms of the reduced bulk modulus of a compressed liquid as a function of density intersect at a common point called _common bulk modulus point'. The reduced bulk modulus is defined as:

$$B_r = \left(\frac{\partial p}{\partial \rho}\right)_T \Big/ KT \quad (2)$$

where Br is the reduced bulk modulus and R is the gas constant. Huang and O'Connell checked the regularity for more than 250 fluids and used it as the basis of a correlation scheme for the volumetric properties of compressed liquids and liquid mixtures. Boushehri et al. [2] presented a theoretical basis for this regularity in terms of a statistical–mechanical equation of state [3].

The linearity of bulk modulus as a function of pressure: More than 100 years ago, a regularity has been discovered by Tait [4] in which isotherms of bulk modulus (reciprocal isothermal compressibility), $1/\beta = \rho(\partial p/\partial \rho)_T$, of a liquid as a function of pressure vary linearly [5-8].

A new scale for measuring the overall elastic stiffness of these compounds is introduced and its correlation with the calculated bulk modulus and lattice constants is analyzed. The overall elastic stiffness is calculated and found to be directly proportional to bulk modulus and inversely proportional to lattice constants. bulk modulus has been found to correlate well with strength and hardness in many materials and those with largest the bulk moduli are usually expected to be the hardest materials [9]. Therefore, one of the important parameters that characterize the physical property of a material system is the material stiffness and its corresponding bulk modulus which measures the degree of stiffness or the energy required to produce a given volume deformation. The bulk modulus reflects important bonding characters in the material and, for many applications, is used as an indicator for material strength and hardness. Early

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experimental and theoretical investigations for bulk modulus were reported in [10,11]. Cohen [10] obtained an empirical expression for the bulk modulus based on the nearest-neighbor distance. His theoretical and experimental results were in agreement. Lam et al. [11] obtained an expression for bulk modulus using the total energy method with acceptable results. The bulk modulus for the semiconductor compounds was found to be inverse proportionally correlated to the lattice constants [11,12]. The overall elastic stiffness of II-IV semiconductor compounds CdS, CdSe, and CdTe is calculated and found to be directly proportional to bulk modulus and inversely proportional to lattice constants. Among these compounds, CdS has the largest overall elastic stiffness and bulk modulus and the smallest lattice constant. Meanwhile, CdTe has the smallest overall elastic stiffness and bulk modulus and the largest lattice constant [13].

The effects of bulk modulus nonlinearity on the performance of a hydrostatic transmission control system have been analyzed through system modeling and simulation. This study has demonstrated that omitting the bulk modulus dynamics in hydrostatic transmission control systems may lead to major errors in system response and have implications on the safety of operation. Therefore, bulk modulus should be considered as a variable parameter to obtain a more realistic overall model and to determine more accurate control parameters in PID controller. Analysis including bulk modulus dynamics in an HST-control system model with this control design feature has not been described in the literature to date. Therefore, it may be useful for the early design of an HST system used for PID control application. In addition, it is clearly seen that a fuzzy controller has the capability of eliminating the adverse effects of variable bulk modulus. This will also benefit the control design process in terms of developing a robust controller. For future research, model development will be expanded to include swashplate dynamics, valve dynamics and more complex flow and torque models of the pump and the motor. Furthermore, an adaptive control method will be applied for changeable velocity reference and load moment [14]. A general regularity was reported for pure dense fluids, namely testing literature results for *p*–*v*–*T* for pure dense fluids, according to which $(Z-1)V^2$ is linear with respect to ρ^2 for each isotherm, where $Z \equiv pV/RT$ is the compression factor [15]. This equation of state works very well for all types of dense fluids, for densities greater than the Boyle density but for temperatures below twice the Boyle temperature. The regularity was originally suggested on the basis of a simple lattice-type model applied to a Lennard-Jones (12, 6) fluid. We shall refer to this equation of state as the 'linear isotherm regularity', or simply LIR from now on. The LIR is used to investigate some empirically known regularities [16, 17].

In the present work, LIR has been used to calculate the bulk modulus. The purpose of this paper is to point out an expression for the bulk modulus of dense fluids using LIR. In this paper, in section 1, we present a simple method that keeps the first-order temperature dependence of parameters in LIR versus inverse temperature. Then, the bulk modulus is calculated from LIR. In section 2, the temperature dependence of the parameters in LIR has been developed to second order. In section 3, the temperature dependence of parameters in LIR has been developed to third order and then the bulk modulus is calculated by LIR in each state.

2. Theory

Liquids and dense fluids are usually considered to be complicated on a molecular scale; they show a number of simple regularities [18]. The first is the Tait–Murnaghan relation, in which the bulk modulus (reciprocal compressibility) of a liquid (or solid) is linear in pressure [7]. The second is the linear relation between the temperature and density at unit compression factor [19], which was discovered empirically in 1906. The third is the common bulk modulus point, in which all liquid isotherms of the reduced bulk modulus as a function of molar volume intersect at essentially a single point. The fourth is the linear isotherm for dense fluids [20], for which we attempt to calculate the internal pressure by modeling the average configurational potential energy and then take its derivative with respect to volume. This assumes that any kinetic energy contribution to the internal energy will vanish on taking the derivative, since the temperature is held constant. It also approximates the average potential energy by summing the contribution from nearest neighbors only, and assuming that the average number of nearest neighbors is proportional to the density, as is the case for liquid argon, rubidium, and cesium [21]. Combining the foregoing results, a general regularity that was reported for pure dense fluids, according to which $(Z-1)V^2$ is linear with respect to ρ^2, each isotherm is,

$$(Z-1)V^2 = A + B\rho^2 \qquad (3)$$

where $Z \equiv pV/RT$ is the compression factor, $\rho = 1/V$ is the molar density, and *A* and *B* are the temperature-dependent parameters. It is shown that this regularity is compatible with equations of state based on statistical–mechanical theory [22]. The model thus not only mimics the linearity of $(Z-1)V^2$ versus ρ^2 but also predicts the temperature dependence intercept and slope. This result immediately shows why *A* has much similarities to the second virial coefficient. It has the same temperature dependence as that of a van der Waals gas, which usually gives a fair representation of the temperature dependence for real gases in the vicinity of the Boyle temperature [21]. The compression factor of the system can be given by using the LIR:

$$\frac{p}{\rho RT} = 1 + A\rho^2 + B\rho^4 \qquad (4)$$

Therefore, the compression factor $Z \equiv pV/RT$ versus ρ^2 becomes quadratic for each isotherm. Using the experimental data, the temperature dependence of the parameters will be tested in the following sections.

2.1. First-order temperature dependence of parameters

We first derive the pressure by applying LIR, and then use the first-order temperature dependence of the parameters to get the final $\left(\frac{\partial p}{\partial \rho}\right)_T$ and bulk modulus for the dense fluid, where

$$A = A_2 - \frac{A_1}{RT} \qquad (5)$$

$$B = \frac{B_1}{RT} \qquad (6)$$

Here A_1 and B_1 are related to the intermolecular attractive and repulsive forces, respectively, while A_2 is related to the nonideal thermal pressure and RT has its usual meaning. In the present work, the starting point in the derivation is equation (4). By substitution of equations (5) and (6) in equation (4), the pressure can be given by using the LIR:

$$p = \rho RT + A_2\rho^3 RT - A_1\rho^3 + B_1\rho^5 \qquad (7)$$

We first derive an expression for $\left(\frac{\partial p}{\partial \rho}\right)_T$ using the first-order temperature dependence of the parameters

$$\left(\frac{\partial p}{\partial \rho}\right)_T = RT + 3A_2\rho^2 RT - 3A_1\rho^2 + 5B_1\rho^4 \qquad (8)$$

and then by substitution of equations (8) in equation (1) we obtain the bulk modulus for a dense fluid. The final result is to form $B^{(1)}$.

According to equation (8), the experimental value of density and the value of A_1, A_2 and B_1 by applying LIR can be used to calculate the value of $\left(\frac{\partial p}{\partial \rho}\right)_T$. According to the LIR, A is linear versus $1/T$; the intercept gives the value of A_2 and the slope gives the value $-A_1/R$. Also, B is linear versus $1/T$; the slope gives the value B_1/R. The values of A_1, A_2 and B_1 for six fluids (Ar, N_2, CO, CO_2, C_6H_6 and C_6H_5–CH_3) are listed in Table 1, together with the temperature range of the experimental data and the coefficient of determination. We begin with C_6H_6 in its liquid range for calculating $\left(\frac{\partial p}{\partial \rho}\right)_T$ and bulk modulus.

Figure 1 shows the experimental values of the bulk modulus versus density for C_6H_6 fluid compared with the and bulk modulus using the $B^{(1)}$ at 280 K. The bulk modulus using the $B^{(1)}$ model yields inaccurate results for the liquid phase. Also, this deviation is significant for the supercritical phase. Moreover, we predict that this deviation leads to inaccurate values of A_1 and A_2; for this purpose, we plot A and B versus inverse temperature ($1/T$) for carbon monoxide [23] and plots show in Figure 2. It is clear that A and B versus inverse temperature are not first order.

Table 1. The calculated values of A_1, A_2 and B_1 for different fluids using equations (1) and (2) and the coefficient of determination (R^2).

Fluid	A_1	A_2	R^2	B_1	R^2	T_{min} to T_{max}
Ar	1.717	0.729	1.00	0.162	1.00	85 to 250
N_2	1.539	0.810	0.99	0.143	0.99	75 to 200
[a] CO	1.542	0.770	1.00	0.132	1.00	70 to 210
CO_2	3.318	0.705	1.00	0.218	0.99	250 to 400
[b] C_6H_6	9.443	1.301	1.00	0.762	1.00	280 to 680
[c] C_6H_5-CH_3	16.861	3.179	0.97	1.398	0.97	200 to 660

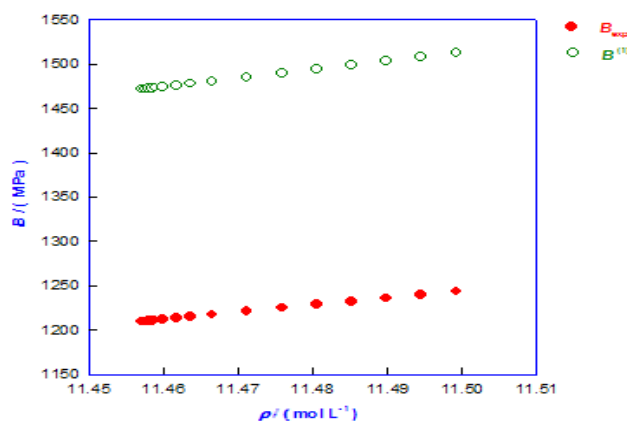[a] Reference [23], [b] Reference [24], [c] Reference [25]

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Figure 1. The experimental values of the bulk modulus versus density for C_6H_6 fluid are compared with the bulk modulus using the $B^{(1)}$ at 280 K.
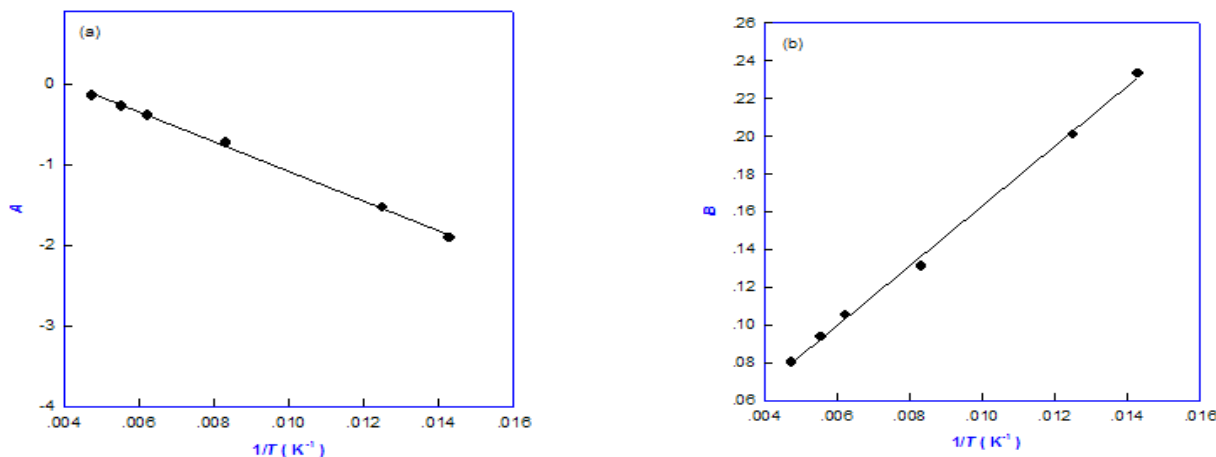
Figure 2. (a) Plot of A versus inverse temperature. The solid line is the linear fit to the A data points, for CO. (b) Plot of B versus inverse temperature. The solid line is the linear fit, for CO.

2.2. Second-order temperature dependence of parameters

In order to solve this problem, the LIR equation of state in the form of truncated temperature series of A and B parameters has been developed to second order for dense fluids. Figures 3(a) and (b) show plots of A and B parameters versus inverse temperature for carbon monoxide fluid. It is clear that A and B versus inverse temperature are second order. Thus, extending parameters A and B, we obtain the second-order equation as:

$$A = A_1 + \frac{A_2}{T} + \frac{A_3}{T^2} \tag{9}$$

$$B = B_1 + \frac{B_2}{T} + \frac{B_3}{T^2} \tag{10}$$

The starting point in the derivation is equation (4) again. By substitution of equations (9) and (10) into equation (4), we obtain the pressure for a dense fluid:

$$p = \rho RT + A_1 RT\rho^3 + A_2 R\rho^3 + \frac{A_3 R\rho^3}{T} + B_1 RT\rho^5 +$$

$$B_2 R\rho^5 + \frac{B_3 R\rho^5}{T} \tag{11}$$

The A and B parameters and their density derivatives were calculated from this model, and the final result is, for $\left(\frac{\partial p}{\partial \rho}\right)_T$ to form,

$$\left(\frac{\partial p}{\partial \rho}\right)_T = RT + 3A_1 RT\rho^2 + 3A_2 R\rho^2 + \frac{3A_3 R\rho^2}{T} + 5B_1 RT\rho^4 +$$

$$5B_2 R\rho^4 + \frac{5B_3 R\rho^4}{T} \tag{12}$$

Finally, by substitution of equations (12) into equation (1), we obtain the bulk modulus for a dense fluid to form $B^{(2)}$.

Therefore, it is possible to calculate the bulk modulus at each density and temperature by knowing A_1, A_2, A_3, B_1, B_2 and B_3. For this purpose we have plotted extended parameters of A and B versus $1/T$ that intercept, and the coefficients show the values of A_1, A_2, A_3, B_1, B_2 and B_3 that are given in Table 2. For comparison, Figure 4 shows the experimental values of the bulk modulus versus density for C_6H_6 fluid, which are compared with the bulk modulus using $B^{(1)}$ and LIR $B^{(2)}$ at 280 K.

Table 2. The calculated values of A_1, A_2 and A_3 using equation (9) and B_1, B_2 and B_3 using equation (10) for different fluids, and the coefficient of determination (R^2).

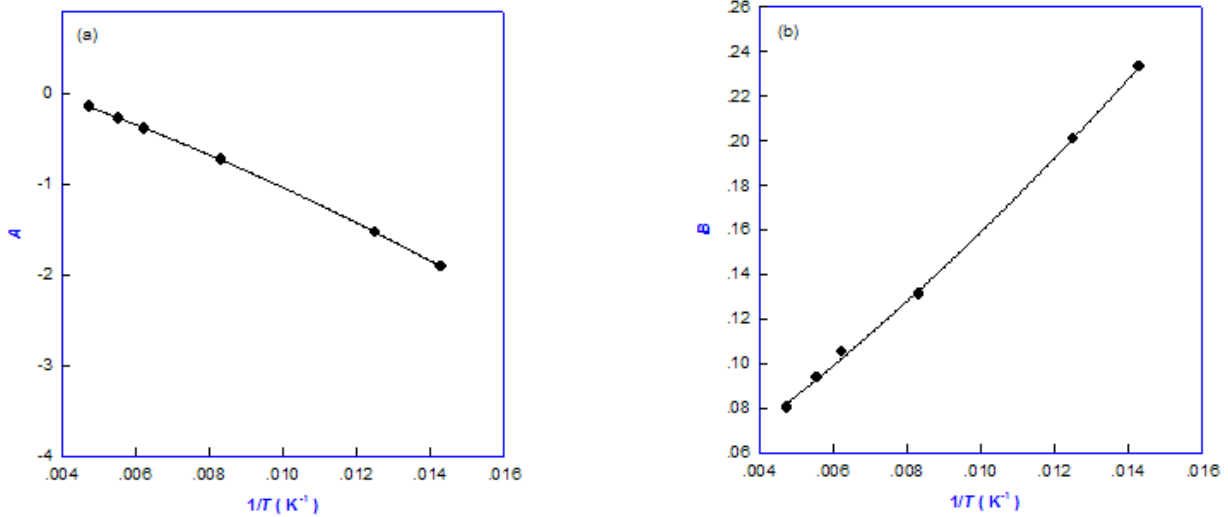| Fluid	A_1	A_2	A_3	R^2	B_1	B_2	B_3	R^2
Ar	0.402	-108.077	-6349.597	1.000	0.028	10.502	577.487	0.999
N_2	0.368	-75.165	-6058.263	1.000	0.040	6.200	602.767	0.999
CO	0.489	-116.150	-3637.325	1.000	0.025	10.665	273.036	0.999
CO_2	0.592	-328.327	-10866.137	0.999	0.036	25.980	42.039	0.991
C_6H_6	0.819	-701.175	-87611.297	1.000	-0.026	95.381	-757.645	1.000
$C_6H_5\text{-}CH_3$	-0.142	279.692	353569.439	1.000	0.084	-16.791	28340.004	0.999

Figure 3. (a) Plot of *A* versus inverse temperature. The solid line is the linear fit to the *A* data points, for CO. (b) Plot of *B* versus inverse temperature. The solid line is the linear fit, for CO.
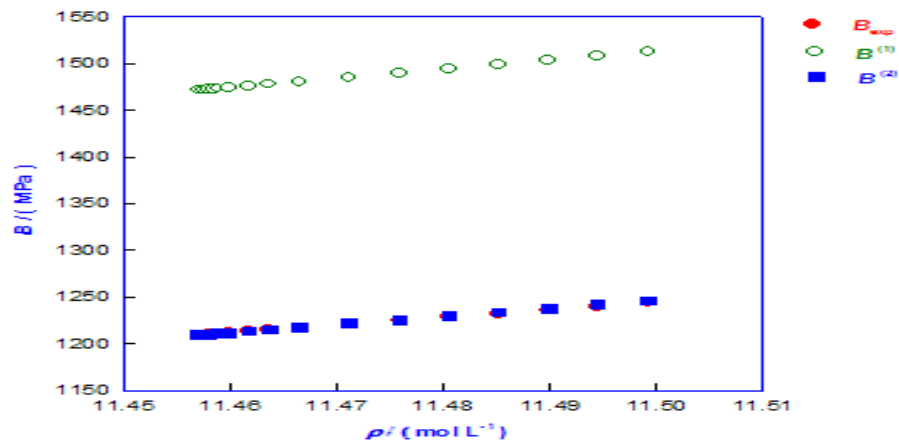

Figure 4. Experimental values of the Bulk modulus versus density for C_6H_6 fluid are compared with the Bulk modulus using $B^{(1)}$ and $B^{(2)}$ at 280 K.

2.3. Third-order temperature dependence of parameters

In another step, we performed a test to form the truncated temperature series of *A* and *B* parameters to third order:

$$A = A_1 + \frac{A_2}{T} + \frac{A_3}{T^2} + \frac{A_4}{T^3} \qquad (13)$$

$$B = B_1 + \frac{B_2}{T} + \frac{B_3}{T^2} + \frac{B_4}{T^3} \qquad (14)$$

The starting point in the derivation is equation (4) again. By substitution of equations (13) and (14) into equation (4), we obtain the pressure for a dense fluid:

$$p = \rho RT + A_1\rho^3 RT + A_2\rho^3 R + \frac{A_3\rho^3 R}{T} + \frac{A_4\rho^3 R}{T^2} +$$

$$B_1\rho^5 RT + B_2\rho^5 R + \frac{B_3\rho^5 R}{T} + \frac{B_4\rho^5 R}{T^2}$$

$$(15)$$

The final result is for $\left(\dfrac{\partial p}{\partial \rho}\right)_T$ to form:

$$\left(\frac{\partial p}{\partial \rho}\right)_T = RT + 3A_1\rho^2 RT + 3A_2\rho^2 R + \frac{3A_3\rho^2 R}{T} + \frac{3A_4\rho^2 R}{T^2} +$$

$$5B_1\rho^4 RT + 5B_2\rho^4 R + \frac{5B_3\rho^4 R}{T} + \frac{5B_4\rho^4 R}{T^2}$$

$$(16)$$

Finally, by substitution of equations (16) into equation (1), we obtain the bulk modulus for a dense fluid to form $B^{(3)}$.

On the basis of equation (16), to obtain $\left(\dfrac{\partial p}{\partial \rho}\right)_T$ it is necessary to determine the values of A_1, A_2, A_3, A_4, B_1, B_2, B_3 and B_4; these values are given in Table 3. In contrast, Figures 5 and 6 show the experimental values of the bulk modulus versus density for benzene as a liquid and as a supercritical fluid, which are compared with the bulk modulus using $B^{(1)}$, $B^{(2)}$, $B^{(3)}$ and PR at 280 and 680 K, respectively

Table 3. The calculated values of A_1, A_2, A_3 and A_4 using equation (13) and B_1, B_2, B_3 and B_4 using equation (14) for different fluids, and the coefficient of determination (R^2).

| Fluid	A_1	A_2	A_3	A_4	R^2	B_1	B_2	B_3	B_4	R^2
Ar	0.509	-154.639	-204.726	-252605.473	1.000	0.004	21.106	-821.958	57528.871	0.999
N_2	0.890	-269.399	16473.524	-818810.762	1.000	-0.053	40.973	-3431.035	146589.395	1.000
CO	0.584	-151.286	393.909	-143448.013	1.000	0.001	19.588	-750.712	36429.199	0.999
CO_2	2.146	-1807.800	451655.351	-47486485.896	1.000	-0.286	332.220	-95697.637	9829469.221	0.996
C_6H_6	-0.029	409.238	-539867.462	58435819.360	1.000	0.071	-31.658	50983.732	-6685480.436	1.000
C_6H_5-CH_3	-0.383	541.788	441148.442	9018987.070	1.000	0.142	-79.596	49357.824	2164439.453	0.999
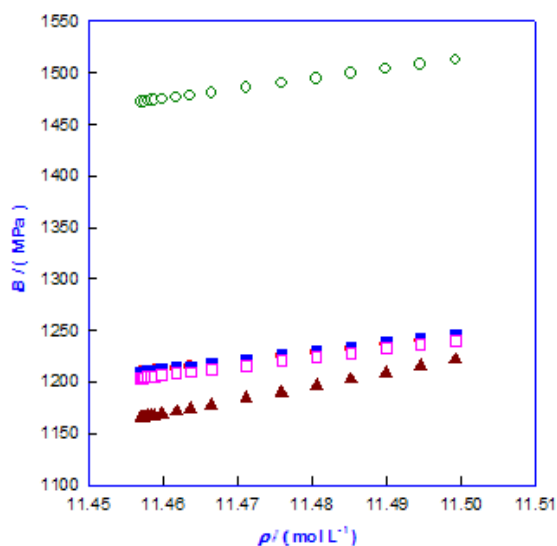
Figure 5. Experimental values of the Bulk modulus versus density for C_6H_6 fluid are compared with the Bulk modulus using $B^{(1)}$, $B^{(2)}$, $B^{(3)}$ and B_{PR} at 280 K.
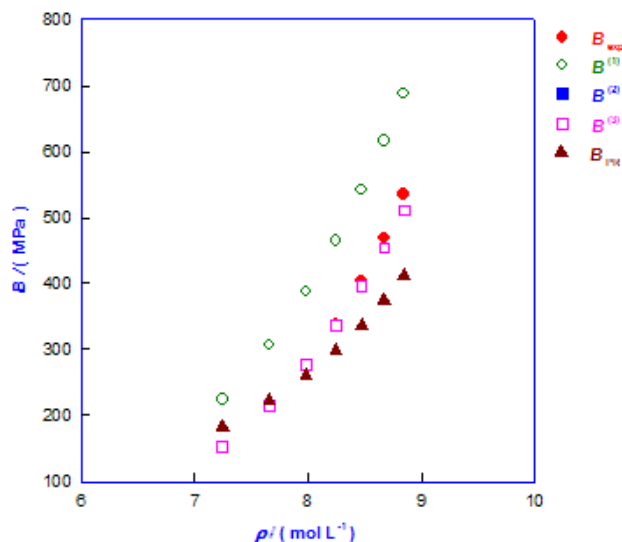

Figure 6. Experimental values of the Bulk modulus versus density for C_6H_6 fluid are compared with the Bulk modulus using $B^{(1)}$, $B^{(2)}$, $B^{(3)}$ and B_{PR} 680 K.

2.4. Peng-Robinson EOS

The Peng-Robinson (PR) EOS [26] may be used to compute the solubility parameter. This equation is given as:

$$p = \frac{RT}{V-b} - \frac{a(T)}{V(V+b)+b(V-b)} \quad (17)$$

Where

$$a(T) = a(T_c) \cdot \alpha(T_r, \omega) \quad (18)$$

$$a(T_c) = 0.45724 \frac{R^2 T_c^2}{P_c} \quad (19)$$

$$b(T_c) = 0.07780 \frac{RT_c}{P_c} \quad (20)$$

$$\alpha = \left(1 + \kappa\left(1 - T_r^{1/2}\right)\right)^2 \quad (21)$$

$$\kappa = 0.37464 + 1.5422\omega - 0.26992\omega^2 \quad (22)$$

$$T_r = T/T_C \quad (23)$$

where T_C and P_C are the critical temperature and pressure, respectively, and ω is acentric factor.

In later stages, bulk modulus calculated by Peng-Robinson (PR) equation of state as a testing the other equation of state.

$$\left(\frac{\partial p}{\partial V}\right)_T = -\frac{RT}{(V-b)^2} + \frac{(2V+2b)a(T)}{\left(V^2+2bV-b^2\right)^2} \quad (24)$$

3. Experimental tests and discussion

The bulk modulus is computed for dense liquid and supercritical fluids using three different models. To investigate the first-order temperature dependence of the parameter A, the six fluids serve as our primary test, because of the abundance of available p–v–T data. The results are summarized in Table 1. In examining the ability of the LIR theory to calculate the bulk modulus of dense fluids, benzene and toluene serve because of the abundance of available bulk modulus data. Such data are more limited for the other fluids examined. When we restricted the temperature series of the parameters A and B to first order, it was seen that the points from the low densities for $B^{(1)}$ deviate significantly from the experimental data.

To decrease adequately the deviation in the bulk modulus from the experimental data, it was necessary to extend the temperature series of the parameters A and B to second order. Nevertheless, it is only for some monatomic fluid similar to Ar that the temperature

dependences of the parameters A and B themselves are satisfactory to first order. Therefore, the present approach for obtaining the bulk modulus from p–v–T data contrasts with the experimental data by extension of the temperature series of the parameters A and B to second order and its derivatives. So, the bulk modulus gives the form of $B^{(2)}$.

We also considered an even more accurate estimate, namely extension of the temperature series of the parameters A and B to third order. Then we introduce the explicit parameters and temperature dependences resulting from the p–v–T data. The final result is for bulk modulus to form $B^{(3)}$. In contrast, Figures 7 and 8 show the experimental values of the bulk modulus versus density for liquid and supercritical fluid toluene, compared with the thermal pressure coefficient using $B^{(1)}$, $B^{(2)}$, $B^{(3)}$ and B_{PR} at 200 and 660 K, respectively.

The experimental and calculated values of $\left(\dfrac{\partial p}{\partial \rho}\right)_T$

using $LIR^{(1)}$, $LIR^{(2)}$ and $LIR^{(3)}$ and the bulk modulus using $B^{(1)}$, $B^{(2)}$, $B^{(3)}$ and B_{PR} are also compared in Tables S1–S4 for benzene and toluene fluids. Although all three models capture the qualitative features for dense fluids, only the calculated values of the bulk modulus using the $B^{(2)}$ model produce quantitative agreement. Tables S1-S4 present a greater test of these models, because only the $B^{(2)}$ model is able to predict accurately

Figure 8. Experimental values of the Bulk modulus versus density for $C_6H_5CH_3$ fluid are compared with the Bulk modulus using $B^{(1)}$, $B^{(2)}$, $B^{(3)}$ and B_{PR} at 660 K.

4. Conclusion

In this paper, we have derived an expression for the bulk modulus of dense fluids (Ar, N_2, CO, CO_2, C_6H_6 and C_6H_5–CH_3) using the linear isotherm regularity. Unlike previous models, it has been shown in this work that the bulk modulus can be obtained without employing any reduced Helmholtz energy. Only p–v–T experimental data have been used for calculating the bulk modulus. Comparison of the calculated values of the bulk modulus using the linear isotherm regularity

both the bulk modulus of the liquid and supercritical fluids. The $B^{(3)}$ model yields good results for toluene, but deviates significantly for benzene, in contrast to the experimental values of the bulk modulus coefficient, whereas the $B^{(1)}$ model is rather inaccurate for both benzene and toluene fluids.
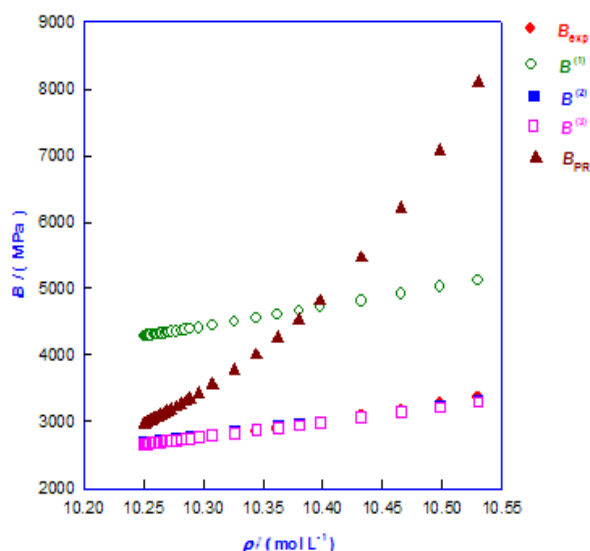

Figure 7. Experimental values of the Bulk modulus versus density for $C_6H_5CH_3$ fluid are compared with the Bulk modulus using $B^{(1)}$, $B^{(2)}$, $B^{(3)}$ and B_{PR} at 200 K.

with the values obtained experimentally show that validity of the use of the linear isotherm regularity for studying the bulk modulus of monatomic dense fluids is doubtful. The validity of the use of the linear isotherm regularity equation state for calculating the bulk modulus of polyatomic dense fluids is also imprecise. In this work, it has been shown that the temperature dependences of the intercept and the slope using the linear isotherm regularity are nonlinear. This problem has led us to try to obtain the expression for the bulk modulus using the extended intercept and slope of the linearity parameters versus inverse temperature to second order. The bulk modulus predicted from this simple model are in good agreement with experimental data. The results show that the accuracy of this method is generally quite good.
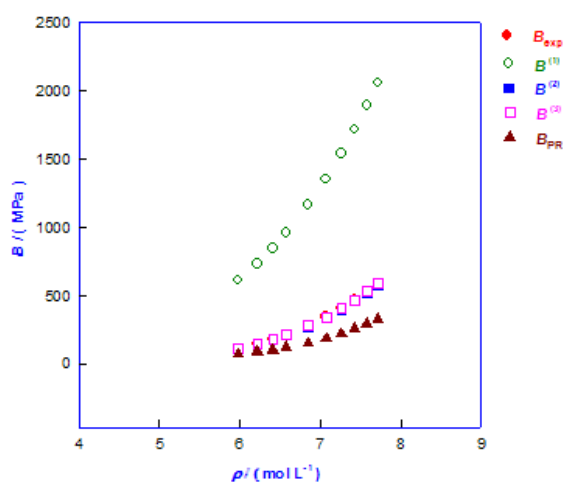
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