On the Calculation of the Virial Coefficients and Low-Pressure Joule-Thomson Effect for Refrigerant Fluids Using Two Equation of State Models

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ABSTRACT: *In this study, we calculate virial coefficients and the Joule-Thomson effect at low pressure for refrigerant fluids in order to evaluate the performance of two models of Equations of State (EOS). The studied refrigerants are R123, R124, R143a, and R152a. The investigated EOSs are van der Waals type consist of van der Waals (vdW), Redlich-Kwong (RK), Soave- Redlich-Kwong (SRK) and Peng-Robinson (PR). In our work, we use Dieterici model of EOS consisting of Dieterici (D) and Dieterici-Carnahan-Starling (DCS). The obtained results show that all EOSs predict the qualitative behavior of the second virial coefficient of refrigerants in wide range temperatures but, cannot provide the qualitative behavior of the third virial coefficient of refrigerants in $T \leq T_c$ in comparing with experimental data. Quantitatively, the EOSs on the basis of vdW model present good results in a wide range of temperatures. Both models of equations of state can also predict the qualitative behavior of changing the low-pressure J-T coefficient with respect to temperature. Our study shows that the EOSs on the basis of vdW model, especially PR, present better results than the other model in a wide range of temperatures.*

KEYWORDS: *Van der Waals equations of state; Dieterici equations of state; Virial coefficients; Refrigerants; Boyle temperature; Joule-Thompson coefficient; Correlation equation.*

INTRODUCTION

Refrigeration is the process of cooling or freezing a substance at a lower temperature than its surroundings and keeping the substance cool. Refrigerant is a material used in refrigeration. It must have suitable thermodynamic properties, including no corrosion of mechanical, non-toxic, and flammable components. In particular, refrigerants should not deplete the ozone layer and change the climate. For these reasons, some refrigerants that were once widely used are now very limited in use, such as chlorofluorocarbons (CFCs).

Due to the importance of refrigerants, a lot of research has been done to determine the physicochemical properties

of them and their effects on the environment [1-6]. In recent years, various studies have been conducted on materials that can be used as refrigerants while not depleting the ozone layer. The results of this research have been the introduction of refrigerants that while maintaining their function as a refrigerant, have much less harmful effects on the environment. They have been considered and used as a replacement for previous refrigerants. Nowadays, a lot of research is in the field of electrocaloric and electrocaloric refrigeration. Many of these technologies show the potential for progress in energy efficiency, compression, noise levels, and environmental impacts [7].

In this paper, we calculate virial coefficients and the Joule-Thomson effect of some refrigerants using two models of EOSs.

Virial coefficients

Thermophysical data of chemical compounds are much needed to design their semi-industrial and industrial production units. It is very useful to predict these characteristics using appropriate models. The use of models is vital when it is difficult or sometimes impossible to determine some thermodynamic data by experimental methods.

One of the important tools for the correlation and prediction of the thermodynamic properties of gases, liquids, and even solids over a wide range of temperatures and pressures is Equations of State (EOSs). Due to their importance, many publications develop or improve them [8-10]. One of the oldest models for calculating the thermophysical properties of fluids is the Virial Equation of State (VEOS), which provides the required information with relatively good accuracy

$$Z = 1 + \sum_{n=2} B_n \rho^{n-1} \tag{1}$$

or

$$Z = \sum B_n^+ P^{n-1} \tag{2}$$

in which

$$B_2^+ = B_2 / RT$$

$$B_3^+ = (B_3 - B_2^2) / R^2 T^2$$

Where Z is the compressibility factor ($Z = pV_m / RT$), ρ is the density ($\rho = 1/V_m$) and B_n is nth-virial coefficient. The virial coefficients are related to intermolecular

interactions by exact statistical-mechanical formulae. In this respect, n th-virial coefficients are related to molecular interactions in clusters of n molecules. Thus, they are the connection bridge between microscopic and macroscopic properties and show the non-ideal behavior of real fluids [11]. By accurately identifying the virial coefficients and how they depend on temperature and also using VEOS, it can be easy to calculate the thermodynamic properties of fluids.

Virial coefficients can be obtained using both experimental and theoretical methods. Experimental methods consist of PVT measurements, speed of sound measurements, Joule–Thomson measurements, refractive index, relative permittivity measurements, and vapor pressure and enthalpy of vaporization measurements. Theoretical approaches usually consist of using equations of state and interaction potential functions [11-22].

On the other side, based on the corresponding state principle, all fluids when compared at the same reduced temperature and reduced pressure, have approximately the same compressibility factor and all deviate from ideal gas behavior to about the same degree. In this regard, using experimental data of virial coefficients, they can be formulated as correlation functions based on the corresponding state principle.

In this study, we calculated the second and third virial coefficients for some refrigerants using several equations of state and compared the obtained results with correlation functions which are based on experimental data of the virial coefficients.

In general, the nth-virial coefficient (B_n) can be obtained from any equation of state *via* the following relationship:

$$B_n = \frac{1}{(n-1)!} \left(\frac{\partial^{n-1} Z}{\partial \rho^{n-1}} \right)_{\rho=0} \tag{3}$$

and so the second and third virial coefficients are

$$B_2 = \left(\frac{\partial Z}{\partial \rho} \right)_{\rho=0} \quad , \quad B_2 = B \tag{4}$$

$$B_3 = \frac{1}{2} \left(\frac{\partial^2 Z}{\partial \rho^2} \right)_{\rho=0} \quad , \quad B_3 = C$$

Joule-Thomson effect

The Joule-Thomson (J-T) effect is of considerable importance in refrigeration and gas liquefaction

processes. In this regard, the J-T coefficient (μ_{JT}) is defined as follows

$$\mu_{JT} = \left(\frac{\partial T}{\partial P} \right)_H \tag{5}$$

This coefficient can be positive, negative, or zero. For refrigeration to occur, the thermodynamic state of the fluid must be in the area bounded by the inversion curve or the location of points where the J-T coefficient is zero ($\mu_{JT} = 0$). The inversion curve is plotted at *T-P* coordinates and is specific to each refrigerant. The range of this curve is from the minimum temperature (T_{min}) for the saturated state on the vapor pressure line to the maximum temperature (T_{max}) for the ideal gas limit at zero density and pressure. Based on thermodynamic relations, we have

$$\mu_{JT} = -\frac{1}{C_{p,m}} \left(\frac{\partial H}{\partial P} \right)_T \tag{6}$$

in which, $C_{p,m}$ is heat capacity. The thermodynamic properties of fluids can be easily calculated from a knowledge of the virial coefficients. In this sense, it can be shown that

$$\left(\frac{\partial H}{\partial P} \right)_T = -RT^2 \left[\frac{dB^+}{dT} + \frac{dC^+}{dT}P + \frac{dD^+}{dT}P^2 + ... \right] \tag{7}$$

Given equation (6), we have

$$\mu_{JT} = \frac{RT^2}{C_{P,m}} \left(\frac{dB^+}{dT} + \frac{dC^+}{dT}P + \frac{dD^+}{dT}P^2 + ... \right) \tag{8}$$

and finally, it can be shown as the expansion by pressure in which expansion coefficients depend on virial coefficients as follows

$$\mu_{JT} = \frac{1}{C_{P,m}} \sum_{n=0} A_n P^n = \frac{1}{C_{P,m}} \left(A_0 + A_1 P + ... \right) \tag{9}$$

$$A_0 = T\frac{dB}{dT} - B$$

$$A_1 = \frac{1}{R}\left(\frac{dC}{dT} - 2B\frac{dB}{dT} \right) + \frac{2}{RT}\left(B^2 - C \right)$$

At zero pressure, we have

$$\mu_{JT}^0(T) = \frac{A_0(T)}{C_{P,m}^0} = \frac{1}{C_{P,m}^0}\left(T\frac{dB(T)}{dT} - B(T) \right) \tag{10}$$

$$A_0(T) = \mu_{JT}^0(T)\, C_{P,m}^0(T) = -\left(\frac{\partial H}{\partial P} \right)_T^0$$

in where, μ_{JT}^0 and $C_{P,m}^0$ are Joule-Thomson coefficient and heat capacity at zero pressure respectively. Also, $(\partial H / \partial P)_T^0$ the enthalpy changes relative to the pressure at zero pressure or ideal conditions.

Joule-Thomson coefficient of an ideal gas is zero because there is no interaction between their molecules. However, equation (10) shows that J-T coefficient of a real gas is not zero in the limit of zero pressure. In this regard, the zero pressure J-T coefficient can be used as a measure of the deviation from the ideal state due to the interactions related to the pair of molecules. These deviations can provide useful information about the nature of intermolecular forces. As told before, in the inversion curve, maximum temperature (T_{max}) correspond to the ideal gas limit at zero density and pressure, then

$$\mu_{JT}^0(T_{max}) = 0 \quad \text{or}$$

$$A_0(T_{max}) = 0 \tag{11}$$

In this paper, we calculate the Joule-Thomson coefficient at zero pressure limit and T_{max} for some refrigerants using several equations of state.

Equations of state

In our work, we used four EOSs of the van der Waals model and two EOSs of the Dieterici model for calculating and predicting the second and third virial coefficients and zero pressure J-T coefficient for refrigerants.

The van der Waals EOS, proposed in 1873 in his doctoral thesis, was the first equation to predict vapor-liquid coexistence [23]

$$p = \frac{RT}{V_m - b} - \frac{a}{V_m^2} \tag{12}$$

The parameter *a* is a measure of attractive forces between the molecules and the parameter *b* is the covolume occupied by the molecules. A and *b* can be obtained from the critical properties of the fluid. In the spirit of vdW equation, the equation of state in the literature almost invariably follows the "repulsive +attractive" formula, *i.e.*

$$p = p_{rep} + p_{att} \quad \text{or} \quad Z = Z_{rep} + Z_{att} \tag{13}$$

in which the subscripts identify the contributions to pressure or compressibility factor of intermolecular

Table 1: The models used in this work.

Model	EOS	Formula	a	b
vd Waals	Van der Waals (vdW)	$Z = \dfrac{V_m}{V_m - b} - \dfrac{a}{R\,T\,V_m}$	$27\,R^2T^2_c/64\,P_c$	$RT_c/8P_c$
	Redlich-kwong (RK)	$Z = \dfrac{V_m}{V_m - b} - \dfrac{a}{R\,T^{0.5}\,(V_m + b)}$	$0.4278\,R^2T^{2.5}_c/P_c$	$0.0867\,RT_c/P_c$
	Peng-Robinson (PR)	$Z = \dfrac{V_m}{V_m - b} - \dfrac{a(T)\,V_m}{R\,T\{V_m\,(V_m + b) + b(V_m - b)\}}$	$0.45724(R^2T^2_c/P_c)\{1+k[1-(T/T_c)^{0.5}]\}^2$	$0.07780\,RT_c/P_c$
	Soava- Redlich-kwong (SRK)	$Z = \dfrac{V_m}{V_m - b} - \dfrac{a}{R\,T\,(V_m + b)}$	$0.4274(R^2T^2_c/P_c)\{1+m[1-(T/T_c)^{0.5}]\}^2$	$0.5732\,RT_c/P_c$
Dieterici	Dieterici(D)	$Z = \dfrac{V_m}{V_m - b}\,e^{-a/RTV_m}$	$0.5411\,R^2T^2_c/P_c$	RT_c/e^2P_c
	Dieterici-Carnahan-starling(DCS)	$Z = \dfrac{(1 + \eta + \eta^2 - \eta^3)}{(1 - \eta)^3}\,e^{-a/RTV_m}$, $\eta = \dfrac{b}{4V_m}$	$0.8143\,R^2T^2_c/P_c$	$1.5285\,Vc$

repulsion and attraction. Many of the equations of state that have been introduced later, with different degrees of success for non-ideal liquids and gases, are based on this model [8-10]. Many of them can be categorized in terms of modifications to the repulsive and attractive vdW model.

In 1899, Dieterici proposed an alternative theory based EOS involving an exponential term [24]

$$p = \frac{R\,T}{V_m - b}\,e^{-a/RTV_m} \qquad (14)$$

This model can be generalized as follows

$$Z = Z_{rep}\,e^{Z_{att}} \qquad (15)$$

The Diererici equation provides an alternative framework for the vdW equation for developing new equations of state. The same as van der Waals EOS, attractive or repulsive term of Dieterici EOS can be replaced with the other terms. In this respect, Sadus replaced the repulsive term of this model with the Carnahan-Starling expression and observed the generally good results in predicting the phase behavior of some fluids and binary mixtures [25-26]. Therefore, the generalized Dieterici formula can be used as the basis for the equation of state development.

EOSs on the basis of Dieterici and van der Waals models have been used in our work for calculating and predicting the virial coefficients for some refrigerants and their details are shown in Table 1.

RESULTS AND DISCUSSION

In this paper, we chose refrigerants R123, R124, R143*a,* and R152*a* in order to determine their viral and Joule-Thomson coefficients. So far, a lot of research has been done on virial coefficients of them [27-40]. In our work, four EOSs based on vdW model and two EOSs on the basis of the Dieterici model are used to calculate and predict virial coefficients of these refrigerant fluids. Critical properties and other parameters of studied refrigerants have been shown in Table 2.

Fig. 1 shows the calculated second virial coefficient of refrigerants as a function of temperature using the above-mentioned EOSs. The obtained results have been compared with the correlated equation on the basis of Meng's work [41]. Meng developed the well-known Tsonopoulos correlation [27-28] for second virial coefficients based on the corresponding-states principle. This correlation has been obtained using new, high-quality experimental data and were determined by fitting them and enhancing the prediction accuracy and reliability based on the corresponding-states form. Therefore, this equation has a very good correlation with the experimental results and so, we have compared our results with it. The correlated and developed equation is as follows:

Table 2: Critical properties and other parameters of Refrigerants.

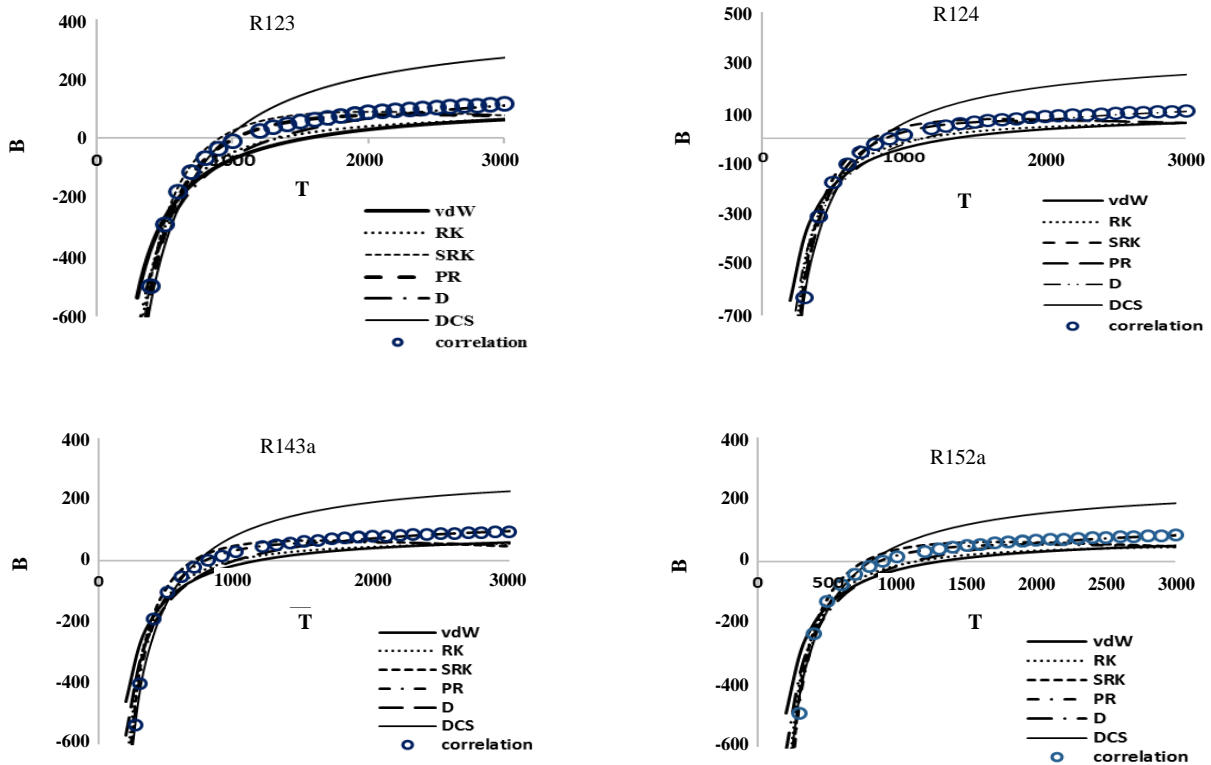| Refrigerant	formula	T_c (K)	P_c (atm)	ρ_c (mol/L)	ω	μ_r	a
R123	$CHCl_2CF_3$	456.831	36.139	3.596418	0.28192	31.84	-0.00091
R124	$CHClFCF_3$	395.425	35.76901	4.103316	0.28810	49.36	-0.00069
R143a	CF_3CH_3	345.857	37.118	5.12845	0.2615	169.91	-0.01703
R152a	CHF_2CH_3	386.411	44.5769	5.571450	0.27521	152.76	-0.01661

Fig. 1: The calculated second virial coefficient of refrigerants vs temperature.

$$B_r = \frac{B P_c}{R T_c} = f^{(0)}(T_r) + \omega f^{(1)}(T_r) + f^{(2)}(T_r) \qquad (16)$$

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

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

$$f^{(2)} = \frac{a}{T_r^6}$$

in which, T_r ($=T/T_c$) is the reduced temperature, P_c and T_c are the critical pressure and critical temperature,

ω is the acentric factor and a is proposed to be a function of the reduced dipole moment μ_r. The acentric factor ω is said to be a measure of the non-sphericity of molecules. It is defined as $\omega = -\log\left(P_r^{sat}\right) - 1$ at $T_r = 0.7$.

The obtained results show that all EOSs predict the qualitative behavior of the second virial coefficient of fluids with respect to temperature, but quantitatively, they present different results. As shown, from among the EOSs based on van der Waals model, PR, and then SRK predict well second virial coefficient of the fluids and provide a good consistent correlation equation over a wide range of temperatures.

Table 3: Temperature range and %AAD in second virial coefficient of studied refrigerants.

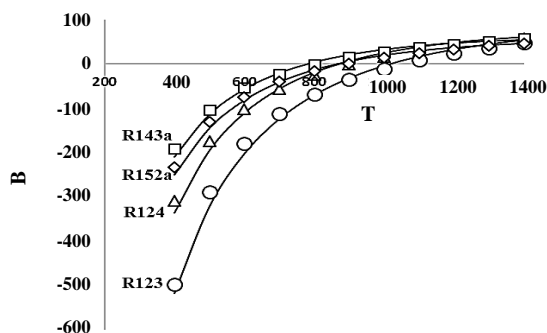| Refrigerant	formula	Temperature range (K)	%AAD
R123	$CHCl_2CF_3$	300-2000	11.13
R124	$CHClFCF_3$	300-1800	10.21
R143a	CF_3CH_3	300-1500	10.9
R152a	CHF_2CH_3	300-1700	12.18

Fig. 2: Calculated second virial coefficient of studied refrigerants vs temperature, line: PR, symbols: correlation equation [41]

For better consideration, Figure 2 shows the results which only consist of PR. As is clear, obtained results from PR EOS are in very good agreement with the correlation data. In this regard, Table 3 shows %AAD of this EOS for studied refrigerants.

The results of the second virial coefficient of studied refrigerants can be used to evaluate the strength of their intermolecular interaction and compare them with each other from this viewpoint. As we know, the second virial coefficient depends on intermolecular interaction potential energy, so with increasing intermolecular attractive forces, the second virial coefficient decreases, and the Boyle temperature increases. As can be seen in Fig. 2, at a given temperature, the value of the second virial coefficient of studied refrigerants in comparison with each other is as $B_{R123} \langle B_{R124} \langle B_{R152a} \langle B_{R143a}$ and Table 4 also shows that the order of Boyle temperature increase is as $T_B(R143a) < T_B(R152a) < T_B(R124) < T_B(R123)$. Therefore according to the above, R123 has the strongest interaction and R143a has the lowest. In this regard, as shown in Fig. 2, PR EOS correctly predicts these results qualitatively and quantitatively.

Given the good results obtained, it can be calculated the second virial coefficient for binary mixtures using PR EOS. To calculate the second virial coefficient of the binary mixture and to compare with the correlation, has been used the classical van der Waals mixing rules for determining parameters a and b of EOS and calculating the correlated second virial coefficient as follows [44]

$$a = a_1 x_1^2 + a_2 x_2^2 + 2(a_1 a_2)^{1/2} x_1 x_2 \qquad (17)$$

$$b = b_1 x_1 + b_2 x_2$$

in which *a* and *b* are the parameters of the mixture with mole fractions of x_1 and x_2. Also correlated second virial coefficient of the mixture is [44]

$$B = x_1^2 B_1 + 2 x_1 x_2 B_{12} + x_2^2 B_2 \qquad (18)$$

in which B_{12} can be estimated as $B_{12} = 1/2 (B_1+B_2)$ [44]. Figure 3 shows the calculated results for binary mixture R143a+R152a. There is a very good agreement between computational data and the correlation with %AAD = 4.36

In Dieterici model, those cannot provide the second virial coefficient of refrigerants in wide range temperatures. DCS shows a large deviation and D provides almost good results in $T > 4T_c$.

Also, since the Boyle temperature (T_B) was found to be sensitive to the equation of state [42], therefore we determined this temperature using different EOSs for evaluating them. The Boyle temperature (T_B) is the temperature in which $B(T_B) = 0$. The results has been shown in Table 4. The same as before, PR EOS presents fairly good results.

Fig. 4 shows the calculated third virial coefficient of refrigerants as a function of temperature using the above-mentioned EOSs and the results have been compared with the correlated equation on the basis of Meng's work [41]. *Meng* improved the Weber correlation [29-32] for third

Table 4: Comparison of predicted Boyle Temperature of EOSs for Refrigerants.

EOS	R123	R124	R143*a*	R152*a*
vdW	1541	1335	1167	1304
RK	1322	1154	1002	1119
SRK	901	774	687	765
PR	1013	875	777	861
D	1318	1141	998	1115
DCS	959	831	726	812
correlation	1053	910	807	892
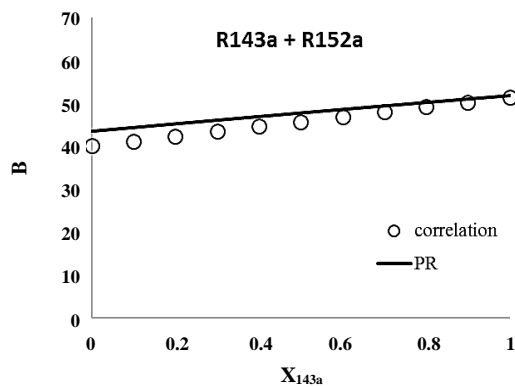
Fig. 3: Second virial coefficient of binary mixture vs mole fraction of x_{143a}.

virial coefficients. Just like the second virial coefficient, this correlation was also determined by fitting experimental data. Therefore, this equation has a very good correlation with the experimental results and so, we have compared our results with it. The improved equation is as follows

$$C_r = \frac{C P_c^2}{R^2 T_c^2} = c_0 + (B_r - c_1)^2 \left[f^{(0)}(T_r) + \mu_r^4 f^{(1)}(T_r) \right] \quad (19)$$

$$f^{(0)} = 1094.051 - \frac{3334.145}{T_r^{0.1}} + \frac{3389.848}{T_r^{0.2}} - \frac{1149.580}{T_r^{0.3}}$$

$$f^{(1)} = \left(2.0243 - \frac{0.85902}{T_r} \right) \times 10^{-10}$$

$$c_0 = 5.476 \times 10^{-3} \quad , \quad c_1 = 0.0936$$

It is clear that the third virial coefficient of fluids has the maximum value near the critical temperature. As shown, none of the EOSs can predict the qualitative behavior of third virial coefficients of fluids in $T<T_c$. However, in the temperature region beyond the critical temperature, all EOSs, except DCS, behave in a good manner compared to the experimental data qualitatively. In this respect, just like before, PR and then SRK present good results in comparison with correlated equation quantitatively. In relation to EOSs based on Dieterici model, those cannot provide the third virial coefficient of refrigerants in wide range temperatures and show a large deviation.

Fig. 5 shows enthalpy changes with respect to pressure $(A_0 = (-(\partial H / \partial P)_T^0)$ of refrigerants as a function of temperature and results have been compared with experimental data from NIST [43].

As seen, with decreasing temperature, especially temperatures below the critical temperature, $-(\partial H/\partial P)_T^0$ increases sharply and with increasing temperature, this coefficient goes to zero. This behavior of fluid is qualitatively well predicted by all EOSs. But quantitatively, except at high temperatures, there is a large deviation. These deviations are due to the high interactions of the molecules at low temperatures, and the EOS listed above cannot represent them correctly.

Under zero pressure and high-temperature conditions, the maximum temperature (T_{max}) can be calculated by equations of state. Table 5 presents T_{max} of studied refrigerants.

CONCLUSIONS

By reviewing the figures and results, we obtain the following conclusions:

1- Two models of EOSs predict the qualitative behavior of the second virial coefficient of refrigerants in wide range temperatures but, cannot provide the qualitative behavior of the third virial coefficient of refrigerants in $T \leq T_c$.
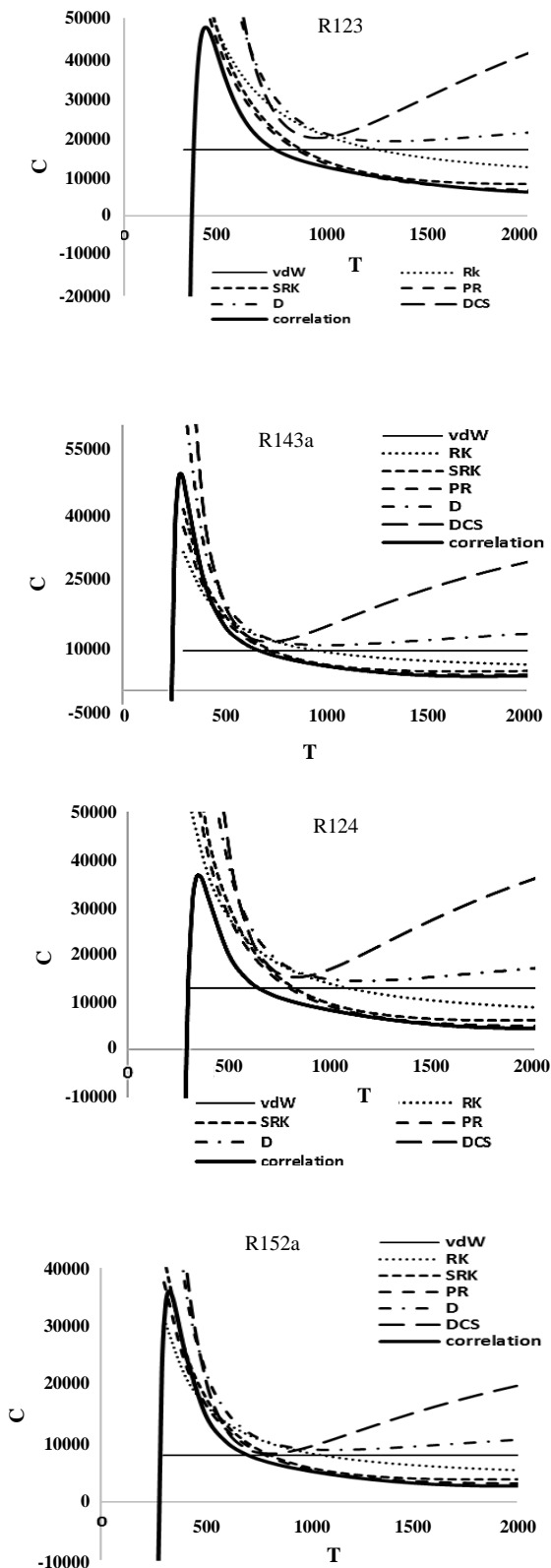
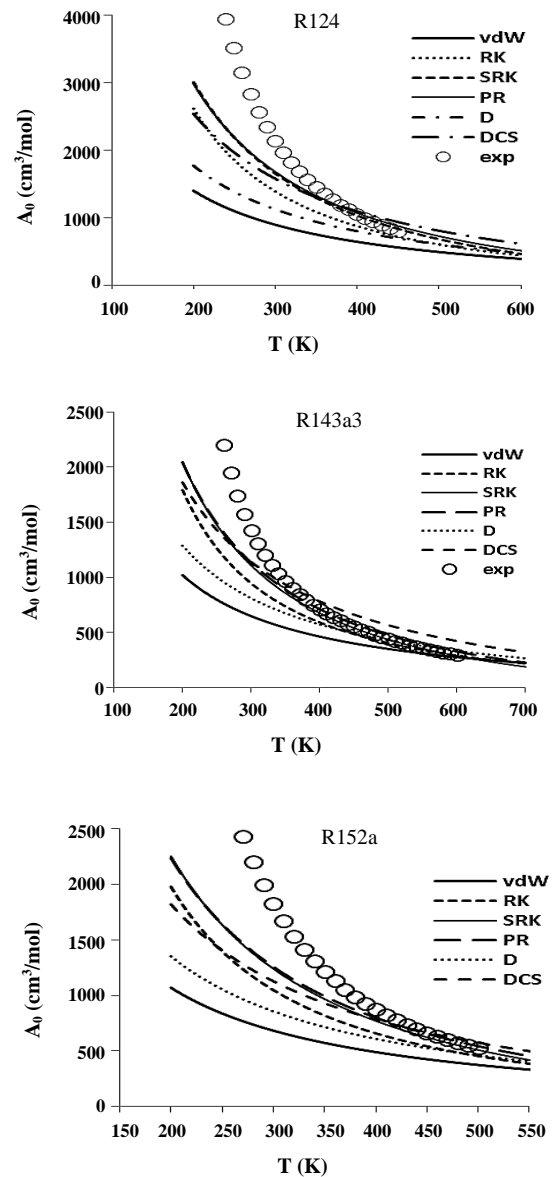*Fig. 4: The calculated third virial coefficient of refrigerants vs temperature.*

Fig. 5: The calculated $A_0 = -(\partial H / \partial P)_T^0$ of refrigerants vs temperature.

2- The EOSs on the basis of vdW model present accurate results and these are consistent well with experimental data based on correlation equations in a wide range of temperatures.

3- Our study show PR and then SRK equations of state predict good results for the second and third virial coefficients of studied refrigerants and their Boyle temperatures. In Dieterici model, D provides fairly good results for the second virial coefficient in high temperature.

Table 5: T_{max} of studied refrigerants.

EOS	R123	R124	R143a	R152a
vdW	3082	2669	2330	2806
RK	2437	2110	1845	2062
SRK	1446	1245	1116	1231
PR	1641	1412	1271	1399
D	2635	2282	1996	2230
DCS	1916	1659	1451	1621

4- Given that these models show two different physical attitudes about the contribution of interaction between molecules to thermodynamic functions, it seems that the vdW model exhibits a better attitude than the other. In fact, the "attractive + repulsive" formula for the pressure or compressibility factor seems to be more accurate in line with the system physics.

5- Both models of equations of state, qualitatively, have a good prediction of changing the zero pressure J-T coefficient with respect to temperature. However, among different EOSs, PR shows slightly more appropriate behavior.

Acknowledgments

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