J. Sci. I. R. Iran **Vol. 11, No. 2, Spring 2000**

COMPARISON OF THE EQUATIONS OF STATE FROM JOULE-THOMSON COEFFICIENT

G. A. Parsafar[1*], E. Noparast and E. Keshavarzi

[1]*Department of Chemistry, Isfahan University of Technology, Isfahan, Islamic Republic of Iran*

Abstract

In the present work, we have examined the ability of some different equations of state in predicting the Joule-Thomson coefficient, $\mu_{J\text{-}T}$, of different fluids. For dense fluids, for which density is greater than the Boyle density, ρ_B, two appropriate equations of state, namely the linear isotherm regularity, LIR, and the dense system equation of state, DSEOS, have been examined. The results show that the DSEOS is in better agreement with the experimental data than the LIR. However, only at very high pressures the LIR gives a better result. For low densities, densities lower than the Boyle density, twelve equations of state namely the van der Waals, Dieterici, Bertholet, Deiters, Virial, Adachi-Lu-Sugie, Kubic-Marthin, Yu-Lu, Twu-Coon-Cunningham, Song-Mason, Ihm-Song-Mason, and the extended linear isotherm regularity, ELIR, have been examined. The results show that the Virial, Song-Mason, Ihm-Song-Mason and ELIR are in a better agreement than the others. Finally we have recommended an appropriate equation of state (ELIR) from which the Joule-Thomson coefficient can be calculated. In this way we found that two harmless refrigerants, R-152a and R-32, have the largest value of $\mu_{J\text{-}T}$, which is in accordance with the experimental observations.

Introduction

The Joule-Thomson expansion is widely used for liquefaction and refrigeration of gases. The determination of the expansion condition is very important in the design of low temperature separation liquefaction plants and in the transport of natural gas processes. The expansion condition indicates whether the system is undergoing a heating or cooling process. It is also important to obtain $\mu_{J\text{-}T}$ by using a theoretical method without involving the experimental measurement. Such an attempt has been done by knowledge of the intermolecular potentials or by using the equations of state. Nain and Aziz [1] predicted $\mu_{J\text{-}T}$ for the noble gases on the basis of numerous intermolecular potentials at zero pressure. Edalate et al.[2] presented a correlation for calculation of the adiabatic Joule-Thomson coefficient of pure gases and their mixtures, by using the Redlich-Kwong, Soav-Redlich-Kwong, Peng-Robinson, and Lee-Kesler equations of state. Maghari and Matin [3] predicted the Joule-Thomson inversion curve from some van der Waals type equations of state.

Keywords: Equation of state; Joule-Thomson coefficient; Refrigerants

---

[*] *E-mail: parsafar@cc.iut.ac.ir*

In the present work, we evaluated different equations of state of predicting the Joule-Thomson coefficient. Owing to the fact that the Joule-Thomson coefficient is very sensitive to small deviations in temperature and pressure, it is a severe test [4] for the accuracy of the equation of state. There is no accurate equation of state, EOS, valid over an entire range of temperature and pressure. Here, the calculation of Joule-Thomson coefficient is divided into two different density ranges, densities greater than and those lower than the Boyle density. Appropriate equations of state for each range have been examined and compared. Finally, we have used more suitable equations of state to predict $\mu_{J\text{-}T}$ for some refrigerants and from such a prediction we have proposed appropriate refrigerants which have a large value of $\mu_{J\text{-}T}$ and a minimal amount of environmental damage.

Appropriate Equations of State for Dense Systems

The Joule-Thomson coefficient in terms of thermodynamic variables is,

$$\mu_{J-T} = \frac{1}{c_p}\left[\frac{-T\left(\frac{\partial P}{\partial T}\right)_v}{\left(\frac{\partial P}{\partial v}\right)_T} - v\right] \tag{1}$$

where, p, v, T, and c_p are pressure, molar volume, absolute temperature and specific heat capacity at constant pressure, respectively. The method of calculation is as follows:

Using a given equation of state, the expression for $\left(\frac{\partial P}{\partial T}\right)_v$ and $\left(\frac{\partial P}{\partial v}\right)_T$ can be obtained and substituted in Equation 1, along with the experimental value of c_p.

For densities greater than the Boyle density we have used the LIR and DSEOS equations of state, because of their simplicity and knowledge of mathematical expressions for the temperature dependencies of their parameters [5,6]. According to the LIR, $(Z\text{-}1)v^2$ is linear versus ρ^2 for each isotherm [5] as,

$$(Z-1)v^2 = A + B\rho^2 \tag{2}$$

where $Z=P/\rho RT$ is the compressibility factor, $\rho=1/v$ is the molar density, A and B are temperature dependent parameters as,

$$A = A_2 - A_1/RT\,, \quad B = B_1/RT \tag{3}$$

and A_1 and B_1 are related to the intermolecular attraction and repulsion respectively, while A_2 is related to the non-ideal thermal pressure. Based on the LIR, $\mu_{J\text{-}T}$ is given as [7],

$$\mu_{J-T} = \frac{1}{c_p}\left[\frac{\rho(3A_1 - 2A_2RT - 5B_1\rho^2)}{RT + 3\rho^2(A_2RT - A_1) + 5B_1\rho^4}\right] \tag{4}$$

The other suitable EOS for such a density range is the DSEOS. It predicts that ρv^2 is quadratic versus ρ for each isotherm as [6],

$$\rho v^2 = A_0 + A_1\rho + A_2\rho^2 \tag{5}$$

where A_0, A_1, and A_2 are temperature dependent parameters defined as,

$$A_i(T) = a_i + b_iT + c_iT^2 - d_iT\ln T \quad i = 0,1,2 \tag{6}$$

The values of constants a_i, b_i, c_i, and d_i may be obtained from a least square fit of the experimental ρ-v-T data in Equation 5, then the results obtained for A_is may be fitted into Equation 6. This EOS is valid for densities greater than the Boyle density and dose not have any temperature limitation [6]. According to the DSEOS, $\mu_{J\text{-}T}$ is given as,

$$\mu_{J-T}\frac{1}{c_p} \times\left[\frac{T(A_0' + A_1'\rho + A_2'\rho^2) - (2A_0 + 3A_1\rho + 4A_2\rho^2}{(2A_0\rho + 3A_1\rho^2 + 4A_2\rho^3)}\right] \tag{7}$$

where $A_i' = \frac{dA_i}{dT}$.

These two equations of state (LIR and DSEOS) are compared with the experimental data through their ability for predicting $\mu_{J\text{-}T}$. Because the experimental values of $\mu_{J\text{-}T}$ are not reported for such a density range, we have used Goodwin's $\mu_{J\text{-}T}$ reported data for toluene which is calculated by an accurate EOS [8]. The values of $\mu_{J\text{-}T}$ for toluene at 1.01325, 70, 250, 700, and 1000 bar are calculated. The results for 1.01325 bar are given in Table 1 and for other isobars are summarized in Figure 1 in which percent deviation is plotted versus temperature for LIR, Figure 1a and DSEOS Figure 1b. The agreement between the DSEOS and Goodwin's reported values is quite well. We may conclude that the DSEOS can predict $\mu_{J\text{-}T}$ in this density range better than the LIR. However, the LIR predictions are better than those of the DSEOS at high pressure such as 1000 bar, especially at low temperatures.

Appropriate Equations of State for Density Range Lower than the Boyle Density

We have also examined the accuracy of some different equations of state in low density range for predicting $\mu_{J\text{-}T}$ such as ELIR, five van der Waals type equations of state, Song-Mason, and Ihm-Song-Mason. In this section, we briefly introduce these equations and

Table 1. The Joule-Thomson coefficient of toulene at 1.01325 bar for given temperature, predicted by the LIR, DSEOS, along with the experimental values

T, K	$(\mu_{J\text{-}T})_{\text{exp}}$ (K bar^{-1})	$(\mu_{J\text{-}T})_{\text{LIR}}$ (K bar^{-1})	$(\mu_{J\text{-}T})_{\text{exp}}$-$(\mu_{J\text{-}T})_{\text{LIR}}$ (K bar^{-1})	$(\mu_{J\text{-}T})_{\text{DSEOS}}$ (K bar^{-1})	$(\mu_{J\text{-}T})_{\text{exp}}$-$(\mu_{J\text{-}T})_{\text{DSEOS}}$ (K bar^{-1})
210	-0.0583	-0.0601	0.0018	-0.0600	0.0017
220	-0.0573	-0.0595	0.0022	-0.0591	0.0018
230	-0.0563	-0.0586	0.0023	-0.0577	0.0014
240	-0.0551	-0.0576	0.0025	-0.0563	0.0012
250	-0.0539	-0.0567	0.0028	-0.0550	0.0011
260	-0.0526	-0.0558	0.0032	-0.0536	0.0010
270	-0.0512	-0.0549	0.0037	-0.0523	0.0011
280	-0.0498	-0.0536	0.0038	-0.0506	0.0008
300	-0.0467	-0.0516	0.0049	-0.0475	0.0008
320	-0.0434	-0.0493	0.0059	-0.0441	0.0007
340	-0.0398	-0.0469	0.0071	-0.0402	0.0004
360	-0.0360	-0.0446	0.0086	-0.0360	-0.0000
380	-0.0319	-0.0423	0.0104	-0.0311	-0.0008

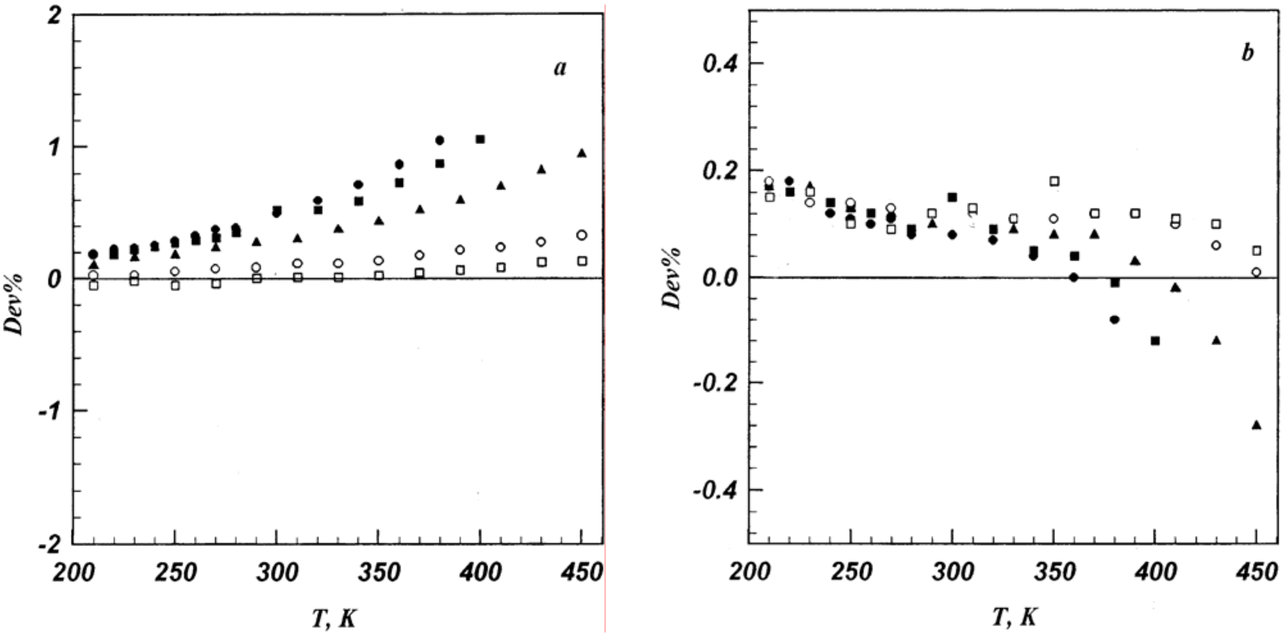

Figure 1. Deviation plot for Joule-Thomson coefficient predicted by (a) the LIR and (b) the DSEOS versus temperature at 1.01325 bar (●), 70 bar (■), 250 bar (▲), 700 bar (○), and 1000 bar (□).

then compare their validity for predicting $\mu_{J\text{-}T}$. The method of calculation is the same as that which was explained earlier.

ELIR Equation of State

Recently, the LIR has been extended to lower density range (lower than Boyle density). This new equation of state [9] is called "extended linear isotherm regularity", or simply "ELIR". According to which

$$\frac{(Z-1)}{\rho} = B_2 + C\rho + a_2\rho^2 + a_3\rho^3 + a_4\rho^4 \qquad (8)$$

with

$$a_2 = \frac{3}{\rho_B}(A-C) - \frac{6B_2}{\rho_B^2}$$

$$a_3 = \frac{-3}{\rho_B^2}(A-C) + \frac{8B_2}{\rho_B^3} + B$$

and

$$a_4 = \frac{1}{\rho_B^3}(A-C) + \frac{3B_2}{\rho_B^4} \qquad (9)$$

where B_2 and C are the second and third Virial coefficients, respectively, A and B are LIR parameters.

Five van der Waals Type Cubic Equations of State

The general reduced form of van der Waals type cubic equations of state [3] can be expressed as:

$$p_r = \frac{1}{Z_c}\left[\frac{T_r}{v_r - U} - \frac{W(T)}{v_r^2 + Y_1 v_r + Y_2}\right] \qquad (10)$$

where Z_c is the critical compressibility factor. The four parameters U, W, Y_1, and Y_2 for the cubic equations of state are given as follows,

$$U \equiv B/Z_c, \quad W(T) \equiv \frac{A\alpha}{Z_c}, \quad Y_1 \equiv \frac{\lambda_1}{Z_c}, \quad Y_2 \equiv \frac{\lambda_2}{Z_c^2}$$

where A, B, and λ_2 are given below for different cubic equations of state.

Adachi-Lu-Sugie, ALS, equation of state

A=0.44869+0.04024ω+0.01111ω^2-0.00579ω^3

B=0.08974-0.03452ω+0.00330ω^2

C=0.03686+0.00405ω-0.01073ω^2+0.00157ω^3

D=0.15400+0.141222ω-0.00272ω^2-0.00484ω^3

α=0.4070+1.3787ω-0.2933ω^2

$$\lambda_1 = C - D, \quad \lambda_2 = -CD \qquad (11)$$

where ω is the acentric factor.

Kubic-Marthin, KM, equation of state

A=0.421875

B=0.081946-0.06487ω-0.01157ω^2-0.01037ω^3

C=0.043$\gamma^{(0)}$+0.0713$\gamma^{(1)}$[0.000756+0.90984ω+0.1622ω^2+ 0.14549ω^3]

$\alpha(T_{r,}\ \omega)=\alpha^{(0)}+\alpha^{(1)}$[0.000756+0.90984ω+0.16226ω^2+ 0.14549ω^3]

$\gamma^{(0)}=4.275051-8.87889T_r^{-1}+37.433095T_r^{-2}-18.05842\ T_r^{-3}+3.514050T_r^{-4}$

$\alpha^{(0)}=-0.1514T_r+0.7895+0.3314T_r^{-1}+0.029T_r^{-2}+0.0015T_r^{-7}$

$\alpha^{(1)}=0.237T_r-0.786T_r^{-1}+1.0019T_r^{-7}$

$$\lambda_1 = 2C, \quad \lambda_2 = C^2 \qquad (12)$$

Yu-Lu, YL, equation of state

A=0.468630-0.0378304ω+0.00751969ω^2

B=0.0892828-0.0340903ω-0.00518289ω^2

C=-1.29917+0.648463ω+0.895926ω^2

$\log\alpha = M(\omega)(A_0 + A_1 T_r + A_2 T_r^2)(1 - T_r)$

for ω≤0.49

$M(\omega)$=0.406849+1.87907ω-0.792636ω^2+0.737519ω^3

A_0=0.536843, A_1=-0.39244, A_2=0.26507

for 1≥ω≥0.49

$M(\omega)$=0.581981-0.171414ω+1.84441ω^2-1.19074ω^3

A_0=0.76355, A_1=-0.53409, A_2=0.37273

$$\lambda_1 = B(1 + C/C_0), \quad \lambda_2 = B^2(C/C_0), \quad C_0 = 1m^3 \qquad (13)$$

Twu-Coon-Cunningham, TCC, equation of state

$A=3Z_c+B+(1-3Z_c)+4B^2$

$B^3-(3Z_c+1)B^2+(3Z_c^{\ 2}-6Z_c+2)B-Z_c^{\ 3}=0$

$C=1-3(Z_c+B)$

$\alpha(T_{r,}\ \omega)=\alpha^{(0)}+\omega[\alpha^{(1)}+\alpha^{(0)}]$

$\alpha^{(0)}=T_r^{\ 0.076554}\mathrm{e}^{1.04734}[1-\ T_r^{\ 0.304777}]$

$$\alpha^{(1)}=T_r^{\ -0.629327}\mathrm{e}^{0.482355}[1-\ T_r^{\ 2.38492}] \qquad (14)$$

Deiters Equation of state

Another van der Waals type equation of state was derived by Deiters for a set of spheres interacting through a square well potential as [10],

$$p = \frac{\rho RT}{b}\left[1 + Cc_0\frac{4\xi - 2\xi^2}{(1-\xi)^3} - \frac{R\alpha}{b}\rho^2\frac{T+\lambda_p}{y}[\exp(\frac{y}{T+\lambda_p}) - 1]I_1\right] \qquad (15)$$

The parameters of this equation are given in [10].

Song-Mason, SM, EOS

Song and Mason derived an EOS based on a statistical-mechanical perturbation theory for both spherical [11], and molecular fluids [12]. Their equation for non-polar spherical molecules is derived as,

$$Z = 1 + B_2\rho + \alpha(T)\rho[g(\sigma^+) - 1] \qquad (16)$$

where $a(T)$ is a temperature dependent parameter that scales for the softness of repulsive forces and $g(\sigma^+)$ is the pair correlation function at contact. The extension of Equation 16 to molecular fluid becomes as [12],

$$Z = 1 + B_2\rho + \alpha(T)\rho[G(b(T)\rho) - 1] \qquad (17)$$

where $b(T)$ is the temperature-dependent parameter and is analogous to the hard sphere diameter and $G(b\rho)$ is the effective pair correlation function for molecular fluids at contact.

Ihm-Song-Mason, ISM, EOS

Ihm, Song, and Mason derived an accurate EOS [13, 14] based on statistical mechanical perturbation theory as:

Table 2. The Joule-Thomson coefficient for argon at 40 atm predicted from different equations of state.

($\mu_{J\text{-}T}$)	T, K						
(K atm^{-1})	383.15	353.15	313.15	273.15	233.15	193.15	153.15
EXP	0.2043	0.243	0.308	0.392	0.513	0.698	0.970
EOS							
ELIR	0.1942	0.233	0.297	0.379	0.494	0.668	0.789
Virial	0.1941	0.233	0.298	0.381	0.499	0.684	0.953
ISM	0.1950	0.234	0.299	0.386	0.509	0.693	0.993
SM	0.1932	0.232	0.297	0.386	0.510	0.700	1.037
Deiters	0.3225	0.354	0.409	0.489	0.622	0.924	-1.400
vdW	0.2395	0.273	0.327	0.398	0.493	0.626	0.764
Dieterici	0.3477	0.390	0.459	0.544	0.653	0.780	0.711
Bertholet	0.0684	0.106	0.173	0.269	0.420	0.664	1.166
ALS	0.0528	0.057	0.064	0.072	0.082	0.90	0.070
TCC	-0.0436	-0.035	-0.020	-0.001	0.024	0.051	0.050
YL	-0.0028	-0.003	0.020	0.054	0.104	0.166	0.173
KM	0.0776	0.086	0.102	0.121	0.145	0.172	0.130

$$Z = 1 + \frac{(B_2 - \alpha)\rho}{1 + 0.22\lambda b\rho} + \frac{\alpha\rho}{1 - \lambda b\rho}$$

where λ is an adjustable parameter.

These equations of state along with the known van der Waals, Dieterici and Bertholet are examined for predicting $\mu_{J\text{-}T}$.

Experimental Test

We have calculated $\mu_{J\text{-}T}$ of argon for the temperature range of 153. 15-383.15 K at 40 atm, the experimental values of c_p are taken from [15]. The results are shown in Table 2 and are compared with the experimental data [16]. This table shows that four equations of state, namely the Virial, SM, ISM, ELIR give a better result than the others. The van der Waals type equations of state predict $\mu_{J\text{-}T}$ with a very large deviation from the experiment. The Dieterici, Deiters, Bertholet and van der Waals equations of state have also deviations, which are more significant for Deiters and Bertholet at high temperatures. However, these equations of state give better results than van der Waals type equations of state. In this way, we select the more accurate equations of state, namely the Virial, SM, ISM, and ELIR and examine their validity for different isobars.

Figures 2 and 3 show the results of $\mu_{J\text{-}T}$ for Ar at 160 and 200 atm obtained using four selected equations of state. Even though these equations of state are in agreement with the experimental data at high temperatures, the Virial EOS shows a large deviation at low temperatures, at which it diverges. However, we have used the Virial, ELIR, SM, and ISM, using Boushehri and Mason Correlation [17], to calculate $\mu_{J\text{-}T}$ for different compounds such as C_2H_6 [18], C_3H_8 [19], CO_2 [20], and C_7H_8 [8], for which the results are shown in Figures 4-7. Figure 4 shows the comparison between the experimental data of $\mu_{J\text{-}T}$ for CO_2 with those calculated from four selected equations of state in terms of temperature at 15 bar. As it is clear all four equations of state agree with the experimental data. Figure 5 shows prediction of the ISM, Virial, and ELIR for toluene for the temperature range of (530-600 K) at 20 bar. As shown the result of ISM is more accurate at least at high temperatures. Figure 6 shows the value of $\mu_{J\text{-}T}$ versus pressure for 373.15 K isotherm of ethane (C_2H_6). The results show that the Virial EOS is more accurate, except at high pressures. Figure 7 is prediction value of $\mu_{J\text{-}T}$ for propylene at 398.15 K. The results show that all three equations of state give good agreement with the experiment at low pressures.

Therefore, we may consider these four equations of state namely, ELIR, SM, ISM, and Virial to calculate $\mu_{J\text{-}T}$ for refrigerants, because of the fact that they give the most accurate value for $\mu_{J\text{-}T}$ at low densities. Since almost all refrigerants are generally polar and the SM and ISM equations of state are proposed for the non-polar and slightly polar compounds [21], we didn't use them to calculate $\mu_{J\text{-}T}$ for such compounds. Therefore, we have calculated $\mu_{J\text{-}T}$ for several refrigerants only by using the Virial and ELIR equations of state. These two equations of state have been tested for some different refrigerants such as R-22 [22], R-23 [23] and R-32 [24].

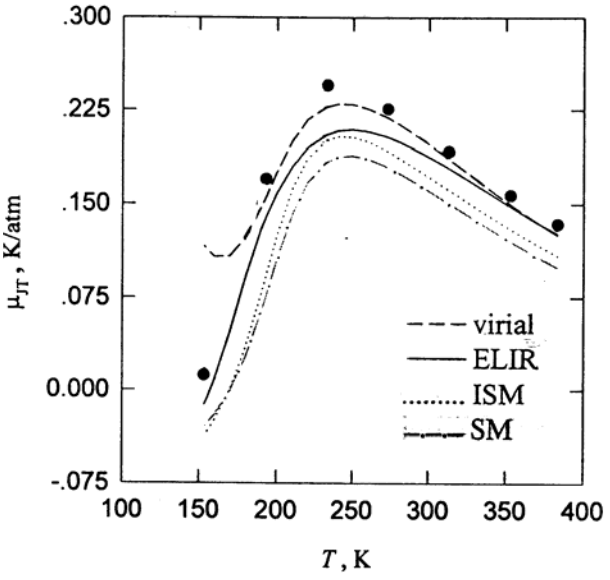

Figure 2. $\mu_{J\text{-}T}$ versus temperature for Ar obtained by the ISM, SM, Virial and ELIR at 160 atm.

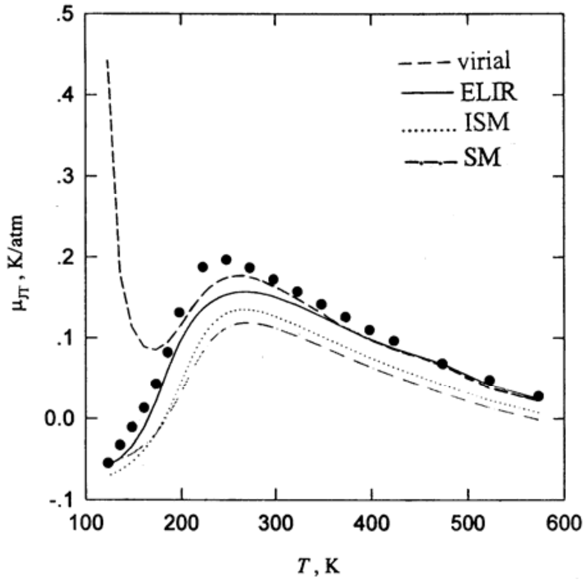

Figure 3. Same as Figure 2 for 200 atm.

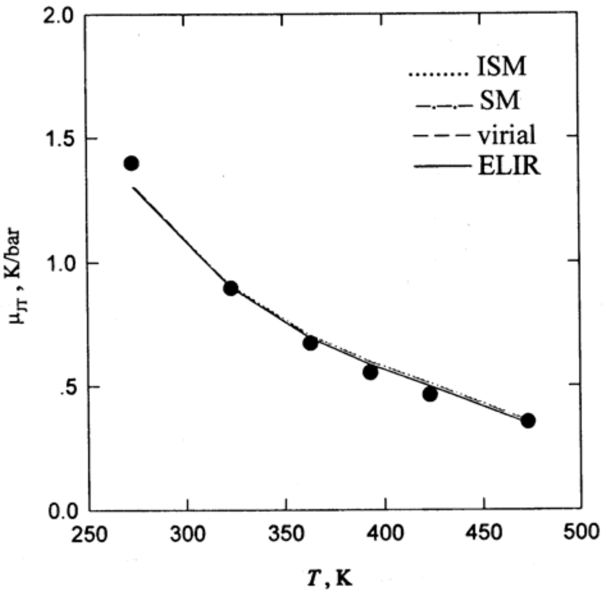

Figure 4. Comparison between the experimantal Joule-Thamson coefficient with those calculated from ISM, SM, Virial and ELIR for CO_2 at 15 bar.

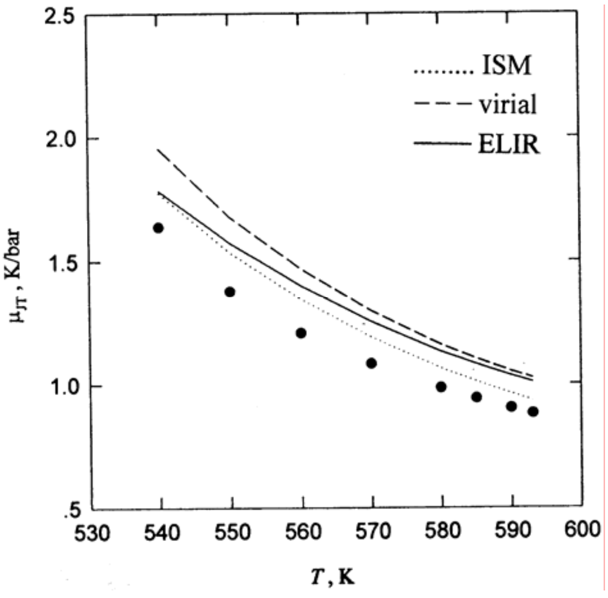

Figure 5. Comparison of the ISM, Virial and ELIR for predicting the Joule-Thamson coefficient for toluene with the experimental data (●) at 20 bar.

The results are shown in Figure 8 that reveals the Virial and ELIR can predict $\mu_{J\text{-}T}$ with a good agreement with the experimental data. Similar calculations have been done for these refrigerants for different pressure and temperature ranges, and also for other refrigerants. The results are almost similar to those shown in Figure 8. But, when the deviation from the experimental data for Virial equation of state at high densities becomes significant, the ability of the ELIR becomes remarkable. Therefore, we have used only this equation of state to predict the Joule-Thomson coefficient for the ozone friendly refrigerants such as R-23, R-32, R-152a, R-134a [25]. Figure 9 shows the calculated value of $\mu_{J\text{-}T}$ from the ELIR versus temperature at 0.5 MPa for the mentioned refrigerants. As it is obvious two ozone safe refrigerants, R-152a and R-32 have the largest value of

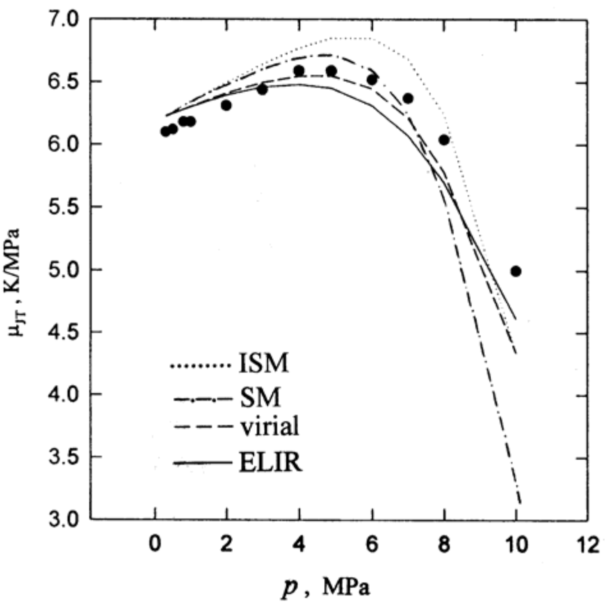

Figure 6. $\mu_{J\text{-}T}$ versus pressure at 373.15 K for C_2H_6 obtained by four selected equations of state and experimental values(●).

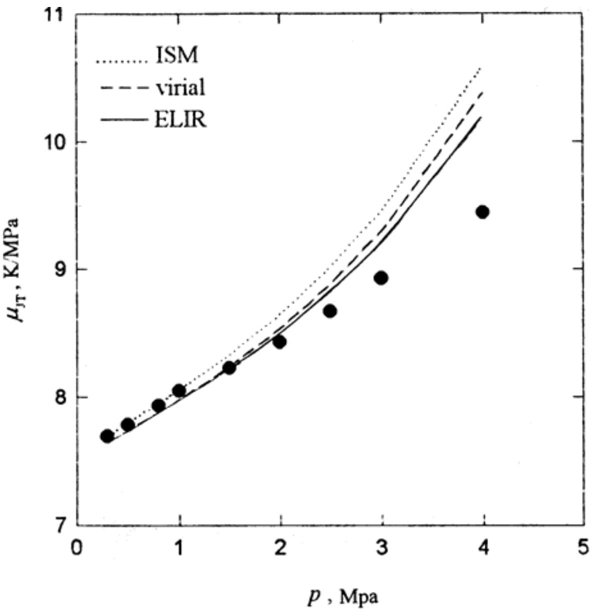

Figure 7. Same as Figure 4 for propylene at 398.15 K.

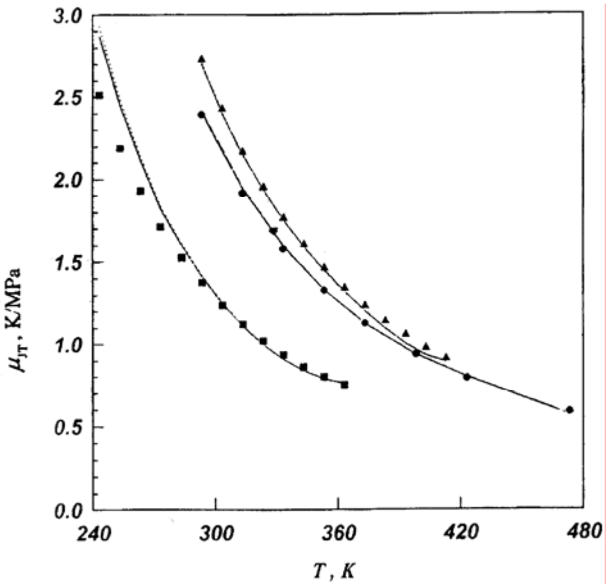

Figure 8. The Joule-Thamson coefficient for R-22 at 5 bar(●), R-23 at 10 bar (■), and R-32 at 5 bar (▲) and the calculated values predicted by the ELIR (—) and Virial (……).

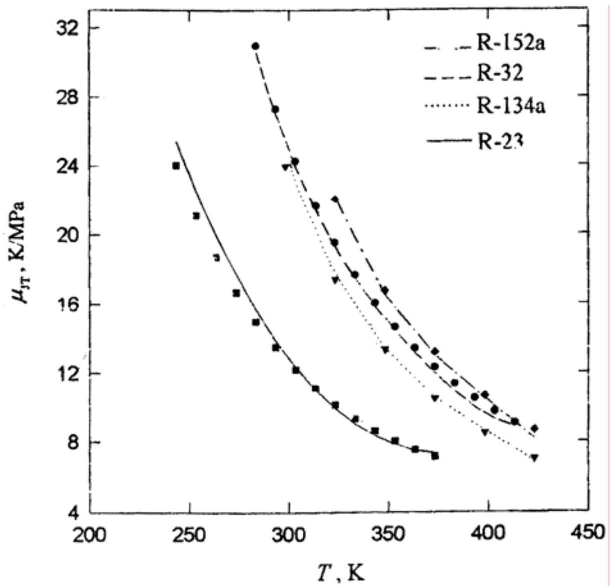

Figure 9. Joule-Thamson coefficient predicted by ELIR for R-32, R-23, R-152a and R-134a at 0.5 MPa (points are the experimental data).

$\mu_{J\text{-}T}$ in the entire temperature range.

Conclusion

In the present work, we have compared the validity of some different equations of state in predicting the Joule-Thomson coefficient. Our results for densities greater than the Boyle density show that the DSEOS gives better results in comparison with the LIR except at very high pressures. Such a result may be expected due to the fact that in the derivation of the DSEOS, the thermal pressure is treated in exact, while it is assumed to be constant in the LIR derivation [5]. Therefore, we may expect that all properties which are related to the temperature derivatives of pressure such as $\mu_{J\text{-}T}$ given by the DSEOS becomes more accurate than that given by the LIR.

In the low density range, four equations of state, namely the Virial, ELIR, SM, ISM give more accurate results, therefore we recommend them for the calculation of μ_{J-T}. Since the Joule-Thomson coefficient calculation is mainly important for refrigerants, which are mostly polar, the ELIR and Virial equations of state are appropriate for such a calculation. We have also examined these two equations for some refrigerants. The results show that the ELIR is in better agreement with the experimental data than the Virial, especially at high densities. Owing to the fact that the ELIR works up to the Boyle density (ρ_B is about twice of the critical density), at which the Virial equation diverges, such a result is expected. Therefore, we may conclude that by using the ELIR EOS we may obtain the most accurate value of μ_{J-T} for refrigerants. We have also shown the predictions of the ELIR for R-134a, R-23, R-132, and R-152a, (Figure 9) which are in a good agreement with the experimental data. It is obvious from this figure that the value of μ_{J-T} for R-152a and R-132 are the largest, in the entire range of temperature, which is in accordance with the ELIR prediction. Therefore, this EOS can be used to predict the thermodynamic state at which μ_{J-T} has an appropriate value, without getting involved in experimental measurements.

Acknowledgements

We acknowledge the Iranian National Research Council for its financial support.

References

1. Nain, V. P. S. and Aziz, R. A. *Can. J. Chem.* **54,** 2617, (1976).
2. Edalat, M., Bozargnehri, R. and Basiri-Parsa, J. *Iran. J. Chem. & Chem. Eng.* **11,** 43, (1992).
3. Maghari, A. and Matin, N. S. *J. Chem. Eng. Jpn.* **30,** 520, (1997).
4. Gine, R. D. and Pravsnitze, J. M. *Cryogenic,* **6,** 324, (1966).
5. Parsafar, G. A. and Mason, E. A. *J. Phys. Chem.* **97,** 9084, (1993).
6. Parsafar, G. A., Farzi, N., and Najafi, B. *Int. J. Thermophys.* **18,** 1197, (1997).
7. Alavi, S., Parsafar, G. A. and Najafi, B. *Int. J. Thermophys.* **16,** 1421, (1995).
8. Goodwin, R. D. *J. Phys. Chem. Ref. Data,* **18,** 1565, (1989).
9. Najafi, B. and Parsafar, G. A. *J. Sci. I. R. Iran,* **8,** 236, (1997).
10. Deiters, U. *Chem. Eng. Sci.* **36,** 1139, (1981).
11. Song, Y. and Mason, E. A. *J. Chem. Phys.* **91,** 7840, (1989).
12. Song, Y. and Mason, E. A. *Fluid Phase Equilib.* **75,** 105, (1992).
13. Ihm, G., Song, Y. and Mason, E. A. *J. Chem. Phys.* **94,** 3839, (1991).
14. Ihm, G., Song, Y. and Mason, E. A. *Fluid Phase Equilib.* **75,** 117, (1992).
15. Stewrt, R. B. and Jacobsen, R. T., *J. Phys. Chem. Ref. Data,* **18,** 639, (1989).
16. Strackey, J. P. and Bennett, C. O. *AICHE. Journal,* **20,** 803, (1979).
17. Boushehri, A. and Mason, E. A. *Int. J. Thermophys.,* **14,** 685, (1993).
18. Bier, K., Kunze, J. and Maurer, G. *J. Chem. Thermodyn.* **8,** 857, (1979).
19. Bier, K., Ernst, G. Kunze, J. and Maurer, G. *J. Chem. Thermodyn.* **6,** 1039, (1974).
20. Bender, R., Bier, K. and Maurer, G. *Ber. Bunsenger, Phys. Chem.* **85,** 778, (1981).
21. Ihm, G., Song, Y., and Mason, E. A. *Mol. Phys.,* **75,** 197, (1992).
22. Bier, K., Maurer, G. and Sand, H. *Ber. Bunsenger. Phys. Chem.* **84,** 430, (1980).
23. Rasskazov, D. S., Kryvkarl, A. *Tr. Mosk. Energ. Inst.* **179,** 108, (1974).
24. Tillner-Roth, R. and Yokozeki, A. *J. Phys. Chem. Ref. Data,* **26,** 1237, (1997).
25. Turk, M., Grone, M. and Bier, K. *J. Chem. Thermodyn.* **28,** 1179, (1996).