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Predicting the Viscosity of Natural Gas[1]

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The viscosity of natural gas has been evaluated by four methods: the Lohrentz–Bray–Clark (LBC), Pedersen et al. (PFCT), SUPERTRAPP, and Vesovic–Wakeham (VW) methods. The predictions have been compared with available experimental data that cover the temperature range from 240 to 444 K and pressures up to 55 MPa. The PFCT and VW methods showed the smallest rms deviations, while the predictions of SUPERTRAPP were only marginally worse. The results indicate that these three methods are capable of predicting the viscosity of natural gas with an rms deviation of 3% and maximum deviations of 5 to 6%. The LBC method proved less reliable with maximum deviations of 8 to 9%.

KEY WORDS: high pressure; natural gas; petroleum fluids; prediction; viscosity.

1. INTRODUCTION

The increasing demand for natural gas has led to the need to develop a more reliable reservoir characterization and simulation. The upstream gas industry, through the gas suppliers, is also being faced with increasing demand for precision in the monitoring of gas supplies. For the exploitation and usage to be optimal, an accurate and reliable knowledge of the viscosity, along with other thermophysical properties, of natural gas is a prerequisite. The wide range of possible natural gas mixtures and of conditions of interest precludes obtaining the relevant data by experimental means alone, thus requiring the development of prediction methods.

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In principle, the viscosity of a fluid can be related to molecular motion and intermolecular forces acting among molecules. In practice, there is no rigorous theory that allows the complete evaluation of viscosity as a function of temperature and pressure in terms of a realistic intermolecular potential. Thus, any prediction method has to rely to a significant extent on a number of assumptions. Nevertheless, the need for greater accuracy and internal consistency of predicted viscosity data favors the prediction methods that are based on a theoretical framework and a stringent choice of primary data rather than those that are merely empirical correlations [1]. Modern developments in the field of prediction of viscosity of fluid mixtures have tended to follow two distinct routes [1]. Namely, these are methods that are based on the corresponding-states principle or methods that have a basis in kinetic theory. Embedded in each class of method is a mixing rule that allows the binary interaction to be estimated. It is at this level that most methods resort to assumptions that are difficult to verify a priori. In general, modern methods do not require any experimental information on mixture viscosities and can be used to predict mixture viscosities from limited information on pure component properties. Their versatility, thermodynamic consistency, and usually good accuracy makes them preferable to the empirical correlations of viscosity data that abound in the petroleum industry.

The existence of a number of prediction methodologies [2–10] led us to perform a series of tests to find out what is the most suitable method for predicting the viscosity of natural gas mixtures. Although comparisons have been reported previously in the literature [11, 12], recent publications of new measurements on the viscosity of natural gas and the developments in the predictive methodologies allow for more comprehensive testing than has previously been possible. The choice of the prediction methods was limited to four, which encompass the methods employed in the petroleum industry and those that are at the forefront of recent developments in the field. Traditionally, practitioners within the petroleum industry employ the Lohrentz–Bray–Clarke methodology [2] for estimating the viscosity of natural gas. This method is essentially empirical in nature, and its present form is based on experimental data available in the 1960s. Recently, methods that take advantage of the theoretical advances made in the thermophysical properties field have begun to make in-roads. The Pedersen et al. [3, 4] method is the most popular choice, since it was specifically developed for hydrocarbon mixtures. Two further methods were chosen for the purposes of this work, one [5, 6] based on the corresponding-states principle and one based on the kinetic theory of fluid mixtures [7–9]. This paper reports on comparisons of predictions of each method with experimental viscosity data for natural gas mixtures.

2. THEORY

To put the subsequent discussion of the results in a proper context, it is useful to highlight and contrast the four methods chosen in this study. For brevity only, essential elements of the four methodologies are presented here, and the reader is referred, in each case, to the original publications for more details.

2.1. Lohrenz–Bray–Clark (LBC) Method

The LBC method [3] is still the most common method employed in the petroleum industry for estimating the viscosity of petroleum fluids. It is based on the observation that the excess viscosity of a fluid is a function primarily of density and that, to first approximation, it can be assumed to be independent of temperature. The LBC method incorporates this behavior into a general two-parameter corresponding-states methodology for estimating the viscosity of petroleum fluids from critical parameters of its constituent species. The viscosity of a fluid mixture is thus given by

$$[(\eta-\eta^0)\,\xi+10^{-4}]^{-0.25}=\sum_{j=0}^{4} a_j\left(\frac{\rho}{\rho_{\mathrm{c}}}\right)^j \tag{1}$$

$$\xi^6=\left[\sum x_i T_{\mathrm{c}i}\right]\left[\sum x_i M_i\right]^{-3}\left[\sum x_i P_{\mathrm{c}i}\right]^{-2} \tag{2}$$

where x_i is the mole fraction of each pure species i, while $T_{\mathrm{c}i}$, $P_{\mathrm{c}i}$, and M_i are the critical temperature, critical pressure, and molecular weight, respectively. The coefficients a_j are temperature independent and are those already suggested by Jossi et al. [13]. The zero-density mixture viscosity is given by a simple mixing rule,

$$\eta^0=\frac{\sum_{i=1}^{N} x_i\eta_i^0 M_i^{1/2}}{\sum_{i=1}^{N} x_i M_i^{1/2}} \tag{3}$$

while the critical density is calculated from the critical volume obtained from a mole fraction average mixing rule [3]. In principle, the zero-density viscosity, η_i^0, of each pure species can be obtained from a number of reliable sources. In this work the original expressions employed by Lohrentz et al. [3] are used, where the viscosity is calculated by means of a simple corresponding states expression in terms of reduced temperature.

2.2. SUPERTRAPP Method

The method behind the SUPERTRAPP computer program [6] was originally developed by Ely and Hanley [5] and is based on the extended corresponding-states principle. The viscosity of a given fluid mixture is calculated from the viscosity of the predefined reference fluid, by

$$\eta(T,\rho)=\eta_{\mathrm{ref}}(T/g,\rho h)\left[\frac{M}{M_{\mathrm{ref}}}\right]^{1/2} g^{1/2}h^{-2/3}X_\eta \tag{4}$$

The quantity X_η is the correction factor for noncorrespondence, which, for mixtures, tends to be different from unity. The parameters g and h and the molecular weight of the mixture M are given by the following empirical mixing rules:

$$g=\frac{\sum_i\sum_j x_i x_j\sqrt{g_i g_j}\,h_{ij}}{h} \tag{5}$$

$$h=\sum_i\sum_j x_i x_j h_{ij}=\frac{1}{8}\sum_i\sum_j x_i x_j(h_i^{1/3}+h_j^{1/3})^3 \tag{6}$$

$$M=\frac{[\sum_i\sum_j x_i x_j g_{ij}^{1/2}h_{ij}^{4/3}M_{ij}^{1/2}]^2}{gh^{8/3}} \tag{7}$$

For each pure species the parameters g_i and h_i can be calculated from the expressions that involve reduced temperature, reduced density, and the Pitzer acentric factor [5].

In the original implementation [5] the reference fluid was methane, but in recent versions of the SUPERTRAPP program, propane was chosen as the more appropriate reference. For the purposes of this work, SUPERTRAPP Version 3.0 was used for all the calculations of the viscosity of natural gas.

2.3. PFCT Method

Pedersen, Fredenslund, Christensen, and Thomassen (PFCT) [3, 4] modified the method of Ely and Hanley, applying it specifically to the petroleum fluids. This method is nowadays gaining acceptance within the petroleum industry as the method of choice for viscosity predictions. The viscosity of the fluid mixture of interest is still calculated from the viscosity of the predefined reference fluid, but the reduced pressure rather than density is used as one of the corresponding-states parameters. This approach avoids the need for estimation of mixture density from the equation of state, but the choice of pressure, rather than density, leads to difficulties in dealing with the discontinuity in the viscosity at the saturation line.

In the PFCT method [3, 4] the viscosity is calculated from the following equation:

$$\eta(T,P)=\eta_{\mathrm{ref}}\left(\frac{TT_{\mathrm{cref}}\alpha_{\mathrm{ref}}}{T_{\mathrm{cmix}}\alpha_{\mathrm{mix}}},\frac{PP_{\mathrm{cref}}\alpha_{\mathrm{ref}}}{P_{\mathrm{cmix}}\alpha_{\mathrm{mix}}}\right)\left[\frac{T_{\mathrm{cmix}}}{T_{\mathrm{cref}}}\right]^{-1/6}\left[\frac{P_{\mathrm{cmix}}}{P_{\mathrm{cref}}}\right]^{2/3}\left[\frac{M_{\mathrm{mix}}}{M_{\mathrm{ref}}}\right]^{2/3}\left[\frac{\alpha_{\mathrm{mix}}}{\alpha_{\mathrm{ref}}}\right] \tag{8}$$

where the parameters T_{cmix} and P_{cmix} are obtained from empirical mixing rules [3, 4] that involve the critical temperature and pressure of each species in the mixture. The parameters α and M_{mix} account for the molecular size of different species. Both parameters have been treated as adjustable in the present method [3, 4] and are given in terms of empirical relationships. Originally, methane was used as the reference fluid [3, 4], but a more advanced version of the PFCT method exists [14] that is based on the two reference fluids (methane and decane). For natural gas, which is primarily methane, both versions are expected to give similar predictions and, therefore, a one-reference fluid version of the method was employed in this work.

2.4. Vesovic–Wakeham (VW) Method

The VW methodology is based on rigid-sphere theory which is adequately modified to take into account the behavior of real fluids in a self-consistent manner. The viscosity of a fluid mixture, η, is written in the form [7–9]

$$\eta=-\begin{vmatrix} H_{11} & \cdots & H_{1N} & Y_1 \\ \vdots & & \vdots & \vdots \\ H_{N1} & \cdots & H_{NN} & Y_N \\ Y_1 & \cdots & Y_N & 0 \end{vmatrix} \Bigg/ \begin{vmatrix} H_{11} & \cdots & H_{1N} \\ \vdots & & \vdots \\ H_{N1} & \cdots & H_{NN} \end{vmatrix} + \kappa_{\mathrm{mix}} \tag{9}$$

$$Y_i = x_i\left[1+\sum_{j=1}^{N}\frac{m_j}{m_i+m_j}x_j\alpha_{ij}\bar{\chi}_{ij}\rho\right] \tag{10}$$

$$H_{ii}=\frac{x_i^2\bar{\chi}_{ii}}{\eta_i^0}+\sum_{j\neq i}\frac{x_ix_j\bar{\chi}_{ij}}{2A_{ij}^*\eta_{ij}^0}\frac{m_im_j}{(m_i+m_j)^2}\left[\frac{20}{3}+\frac{4m_j}{m_i}A_{ij}^*\right] \tag{11}$$

$$H_{ij}=-\frac{x_ix_j\bar{\chi}_{ij}}{2A_{ij}^*\eta_{ij}^0}\frac{m_im_j}{(m_i+m_j)^2}\left[\frac{20}{3}-4A_{ij}^*\right] \tag{12}$$

$$\kappa_{\mathrm{mix}}=\frac{15}{5\pi}\rho^2\sum_{i=1}^{N}\sum_{j=1}^{N}x_ix_j\bar{\chi}_{ij}\alpha_{ij}^2\eta_{ij}^0 \tag{13}$$

where η_{ij}^{0} is the zero-density interaction viscosity, while A_{ij}^{*} is a weakly temperature-dependent function for the i–j pair interaction. The parameter α_{ij} accounts for the mean free path shortening for an i–j collision in the dense fluid, whereas $\bar{\chi}_{ij}$ is the pseudo-radial distribution function for the species i and j in the presence of all other species in the mixture. The pseudo-radial distribution function, $\bar{\chi}_{ij}$, is constructed by means of the following mixing rule, which has no adjustable parameters,

$$\bar{\chi}_{ij}(\rho, T)=1+\frac{2}{5}\sum_{k=1}^{N} x_k(\bar{\chi}_k-1)+\frac{6(\bar{\chi}_i-1)^{1/3}(\bar{\chi}_j-1)^{1/3}}{5[(\bar{\chi}_i-1)^{1/3}+(\bar{\chi}_j-1)^{1/3}]}\sum_{k=1}^{N} x_k(\bar{\chi}_k-1)^{2/3} \tag{14}$$

while α_{ij} is obtained, for each isotherm, from

$$\alpha_{ij}(T)=\tfrac{1}{8}(\alpha_{ii}^{1/3}+\alpha_{jj}^{1/3})^{3} \tag{15}$$

The pseudo-radial distribution function for a pure component i is calculated from the viscosity of the pure species. To construct a realistic pseudo-radial distribution function that increases monotonically with increasing density, one is constrained in the choice of parameter α_{ii}. In fact, at each temperature, the requirement that the pseudo-radial distribution function is a continuous function of the molar density uniquely determines the value of parameter α_{ii}. This, in turn, allows for a unique determination of the pseudo-radial distribution function $\bar{\chi}_i$, as a function of the molar density at each isotherm for each pure component [7–9].

3. RESULTS

To make a sensible comparison of the predictions of the four methods, it is important that the experimental data used are obtained in a well-characterized apparatus with well-defined uncertainty limits. A literature search produced only three sets [15–17] of data that fulfill the above characteristics, highlighting the general issue of the scarcity of reliable measurements of the viscosity of fluid mixtures. Although a plethora of viscosity measurements must have been made in routine industrial analysis, such data are usually not available in the open literature and are not, in general, suitable for comparison purposes due to lack of adequate characterization.

It is customary to measure and report viscosity as a function of pressure at a given temperature. Three of the prediction methods used in this study, work on the basis of predicting the viscosity at a given temperature and density. Thus, it is necessary to convert temperature, pressure pairs into the appropriate temperature, density pairs. The density of the natural gas mixture was calculated by means of the AGA8-DX92 correlation of Jaeschke and Schley [18], which is based on an extended virial-type equation. The uncertainty of this correlation is ±0.1% for the temperature range from 265 to 335 K and pressures up to 12 MPa and ±0.3% for the other temperature and pressure ranges.

In the 1960s Lee and his collaborators [15] performed extensive measurements of the viscosity of natural gas mixtures. Four natural gas mixtures were examined, and the measurements of viscosity were reported for temperatures from 311 to 444 K and pressures from 1.3 to 55 MPa. The measurements were performed by means of the capillary viscometer with an estimated accuracy of ±2.0% or better. The authors have also performed the density measurements and, in this case, their quoted values are used as inputs in the three prediction methods that require temperature, density pairs.

Table I summarizes the rms deviations obtained by comparing the predictions of the four methods with 111 available experimental data points. The overall agreement is very good. The PFCT, SUPERTRAPP, and VW methods predict the experimental data equally well, with rms deviations of approximately 2.8%. The LBC method produces a larger rms deviation, and closer examination of the deviation plots indicates that for a number of isotherms the predicted density dependence of viscosity is not correct.

It is illustrative to examine in more detail the predictions of the four methods for each mixture. Mixture 1 is a nine-component mixture, with the major components being methane ($x_{CH_4} = 0.863$), ethane ($x_{C_2H_6} = 0.068$), carbon dioxide ($x_{CO_2} = 0.032$), and propane ($x_{C_3H_8} = 0.024$). As illustrated in Table I, the PFCT, SUPERTRAPP, and VW methods predict the viscosity of this mixture equally well with maximum deviations of 2.0, 3.2, and −3.2%, respectively. The LBC method, in general, underpredicts the viscosity, with a maximum deviation of −5.7%. Closer examination of the deviation plots indicates that the only systematic trends are observed at

Table I. The rms Deviations of the Experimental Data from Ref. 15

	LBC (%)	PFCT (%)	SUPERTRAPP (%)	VW (%)
Overall	**4.9**	**2.6**	**3.0**	**2.8**
Mixture 1	4.2	1.0	1.4	1.9
Mixture 2	4.8	3.7	4.8	1.9
Mixture 3	5.4	1.9	1.7	3.1
Mixture 4	5.5	3.0	2.8	4.0

very low densities, up to 3000 mol · m^{-3}, where all the methods predict a faster increase in viscosity with density than indicated by the experimental data.

Mixture 2 is an 11-component mixture, with the major components being methane ($x_{CH_4} = 0.717$), ethane ($x_{C_2H_6} = 0.14$), and propane ($x_{C_3H_8} = 0.083$). Figure 1 illustrates the deviations of the viscosity predicted by the four methods from the experimental data. The VW method predicts these data with the same rms deviation as for mixture 1, while the PFCT and SUPERTRAPP methods tend to overpredict the data, which results in an increase in rms deviations to 3.7 and 4.8 %, respectively. The largest deviations, for PFCT and SUPERTRAPP, of the order of 6 to 10 %, are observed at the highest densities. The LBC method predicts a faster increase in viscosity, as a function of density, than is observed experimentally.

Mixture 3 is also an 11-component mixture, with the major components being methane ($x_{CH_4} = 0.807$), ethane ($x_{C_2H_6} = 0.087$), nitrogen ($x_{N_2} = 0.048$), and propane ($x_{C_3H_8} = 0.029$). The PFCT and SUPERTRAPP methods predict these data with similar rms deviations as for mixture 1, while the VW method tends to underpredict the data which results in an increase in rms deviations to 3.1 %. The maximum deviations observed for the three methods are 3.4, 3.4, and −5.2 %, respectively. No systematic trends with either density or temperature are observed, and, for brevity, no deviation plot is included.

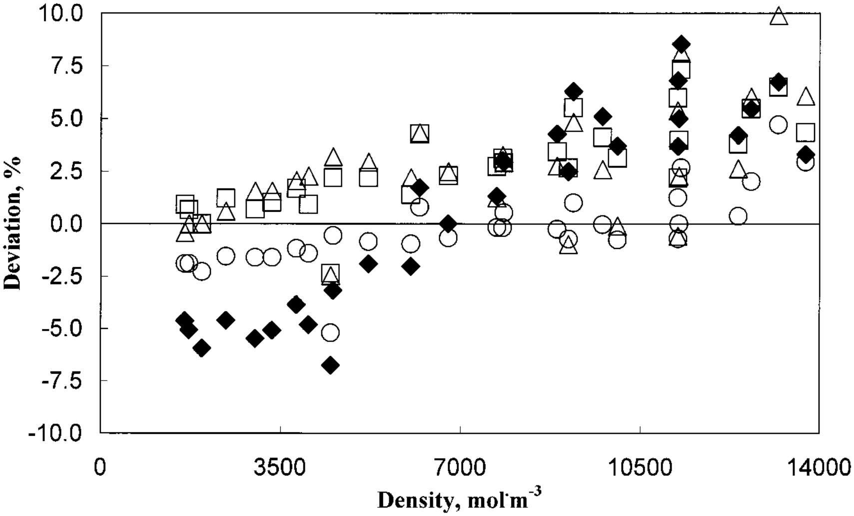

Fig. 1. Deviations, $[(\eta_p - \eta_{exp})/\eta_{exp}] \times 100\,\%$, of the predicted values, η_p, of the viscosity of natural gas from the experimental values, η_{exp}, of mixture 2 [15] as a function of molar density, ρ. (◆) LBC, (□) PFCT, (△) SUPERTRAPP, and (○) VW methods.

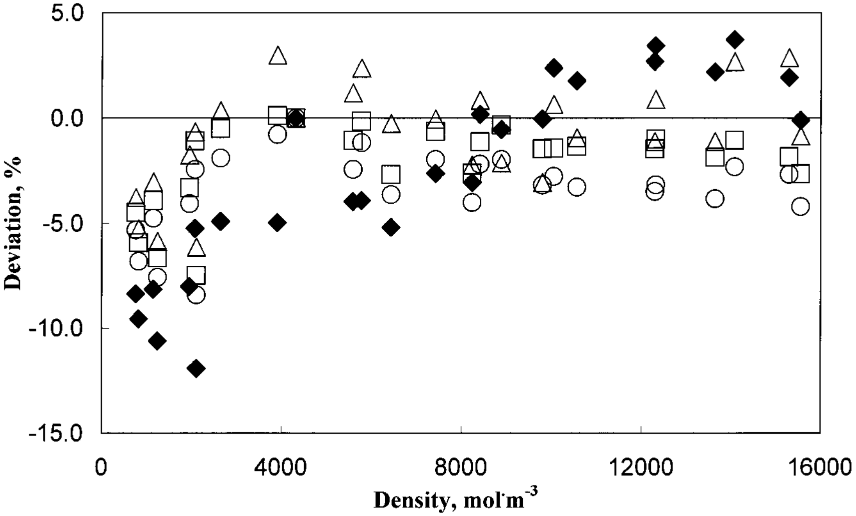

Fig. 2. Deviations, $[(\eta_p - \eta_{exp})/\eta_{exp}] \times 100\,\%$, of the predicted values, η_p, of the viscosity of natural gas from the experimental values, η_{exp}, of mixture 4 [15] as a function of molar density, ρ. (◆) LBC, (□) PFCT, (△) SUPERTRAPP, and (○) VW methods.

Mixture 4 is a 10-component mixture, with the major components being methane ($x_{CH_4} = 0.915$) and ethane ($x_{C_2H_6} = 0.031$). Figure 2 illustrates the deviations of the viscosity predicted by the four methods from the experimental data. With such a large mole fraction of methane, one would expect that the four prediction methods would have minimal problems predicting the viscosity of this mixture. In fact, nearly the opposite is true, and the rms deviations observed are larger than average, as illustrated in Table I. Surprisingly, the maximum deviations are observed at low densities and, for the 411 K isotherm, are exceptionally large, ranging from 5 to 6% for SUPERTRAPP to 10 to 12% for the LBC method. This might indicate that for this particular mixture the uncertainty in the experimental viscosity might be higher than anticipated.

Recently Nabizadeh and Mayinger measured the viscosity of a natural gas mixture as a part of a more general program of measuring the viscosity of hythane [16]. The measurements were carried out in an oscillating-disk viscometer with a claimed uncertainty of ±1%. Only one synthetic natural gas mixture ($x_{CH_4} = 0.9467$; $x_{C_2H_6} = 0.035$; $x_{N_2} = 0.0183$) was examined and the viscosity measurements were reported in the temperature range from 298 to 400 K at pressures from atmospheric up to 7.1 MPa.

Figure 3 illustrates the deviations of the viscosity predicted by the four methods from the experimental data. The overall agreement is excellent.

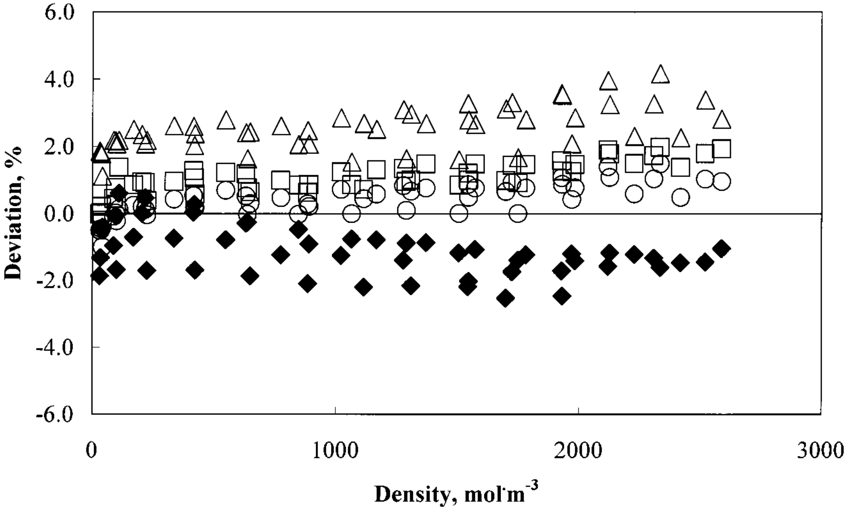

Fig. 3. Deviations, $[(\eta_p - \eta_{exp})/\eta_{exp}] \times 100\%$, of the predicted values, η_p, of the viscosity of natural gas from the experimental values, η_{exp} [16], as a function of molar density, ρ. (◆) LBC, (□) PFCT, (△) SUPERTRAPP, and (○) VW methods.

The LBC method tends to slightly underpredict the data, while the other three methods tend to overpredict the data. The rms deviations of the LBC, PFCT, SUPERTRAPP, and VW methods are 1.4, 1.2, 2.6, and 0.7%, respectively, while the maximum deviations are −2.5, 1.8, 4.2, and 1.5%, respectively. Thus, all the observed deviations are well within the combined uncertainty of the experimental data and the uncertainty of the methods. No systematic trends with temperature were observed, and all four methods seem to predict the correct density dependence.

Assael and co-workers [17] also reported measurements of the viscosity of a natural gas mixture. The measurements were carried out in a vibrating-wire viscometer with a claimed uncertainty of ±1%. Only one synthetic natural gas mixture ($x_{CH_4} = 0.8484$, $x_{C_2H_6} = 0.084$, $x_{C_3H_8} = 0.005$, $x_{CO_2} = 0.0066$, $x_{N_2} = 0.056$) was examined, and the viscosity measurements were reported in the temperature range from 240 to 353 K at pressures from atmospheric up to 15 MPa.

Figure 4 illustrates the deviations of the viscosity predicted by the four methods from the experimental data. The overall agreement is very good, and the trends are similar to those displayed in Fig. 3. The rms deviations of the LBC, PFCT, SUPERTRAPP and VW methods are 0.9, 1.9, 3.4, and 1.5%, respectively, while the maximum deviations are −2.4, 3.1, 5.9, and −3.8%, respectively. No systematic trends with temperature were observed, and all four methods seem to predict the correct density dependence.

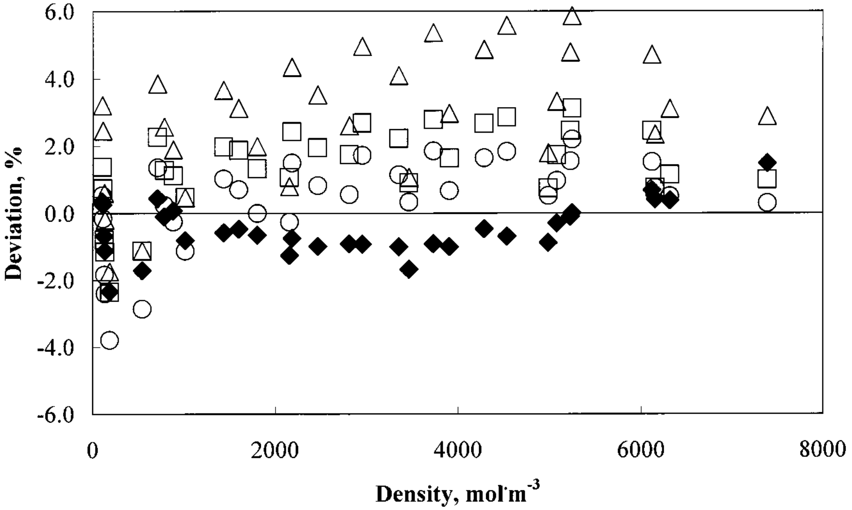

Fig. 4. Deviations, $[(\eta_p - \eta_{exp})/\eta_{exp}] \times 100\%$, of the predicted values, η_p, of the viscosity of natural gas from the experimental values, η_{exp} [17], as a function of molar density, ρ. (◆) LBC, (□) PFCT, (△) SUPERTRAPP, and (○) VW methods.

Finally, a comparison was made with the viscosity data pertaining to a real reservoir fluid, as reported by Ali [11] in Table 5 of his work. The viscosity measurements are reported at one temperature, $T = 306$ K, and at pressures from 1.35 to 13.5 MPa. No details of the experimental setup and uncertainty are available, and the only justification for the inclusion of this data set is to extend the comparison to real reservoir fluids. In fact, the presence of heavier components ($x_{C_{6+}} = 0.005$) in this mixture causes some retrograde condensation on lowering the pressure, thus making this fluid strictly speaking a condensate rather than a natural gas. Nevertheless, the amount of liquid formed during the retrograde condensation is sufficiently small that it hardly affects the composition and the viscosity of the vapor phase.

All four prediction methods reproduce the nine experimental points very well. The rms deviations of the LBC, PFCT, SUPERTRAPP and VW methods are 2.0, 2.4, 2.9, and 1.4%, respectively, while the maximum deviations are −2.9, 2.4, 2.9, and −2.6%, respectively. No systematic trends are observed, and, for brevity, no deviation plot is included.

4. CONCLUSIONS

The four methods for predicting the viscosity of fluid mixtures, namely, the Lohrentz–Bray–Clark (LBC) method, the Pedersen et al.

(PFCT) method, the SUPERTRAPP method, and the Vesovic–Wakeham (VW) method, have been examined and used to predict the viscosity of natural gas mixtures. The predictions have been tested against available experimental data. The comprehensive data of Lee et al. [15], containing 111 data points, were reproduced by the PFCT, SUPERTRAPP, and VW methods with an rms deviation of approximately 2.8%. This is only marginally worse than the claimed accuracy of the data, and adequate for most industrial requirements. The more accurate, but limited, data of Nabizadeh and Mayinger [16] and Assael et al. [17] were reproduced by the PFCT and VW methods with rms deviations of less than 2%, while the SUPERTRAPP exhibited a larger rms deviation, of the order of 3%. The LBC method performed relatively poorly, compared with the other three, reproducing the Lee et al. [15] data with an rms deviation of 4.9%, while it did surprisingly well reproducing the more accurate data [16, 17], with the deviations only marginally worse than the claimed experimental uncertainty.

The viscosity of real reservoir condensate [11] was reproduced equally well by all the methods. The smallest deviations were exhibited by the VW method, while the largest deviations were produced by SUPERTRAPP method.

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