J. Chem. Eng. Data **2009,** *54,* 1329–1333

Liquid Viscosities of the Ternary System Benzene + Cyclohexane + *n*-Tetradecane from (313 to 393) K and Pressures up to 60 MPa

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Liquid viscosities of eight mixtures for the ternary system benzene + cyclohexane + *n*-tetradecane were experimentally measured using a rolling-ball viscometer from (313.2 to 393.2) K and at pressures up to 60 MPa. We performed the modeling of the measured mixture viscosity data (256 points) by applying the Grunberg−Nissan (GN) and Katti−Chaudhri (KC) correlations and a liquid viscosity model based on Eyring's theory coupled to a cubic equation of state (ET-EoS) by using a single temperature-independent binary interaction parameter for the benzene + *n*-tetradecane, benzene + cyclohexane, and cyclohexane + *n*-tetradecane systems. Results of the modeling process yielded an average absolute deviation of (4.9, 5.3, and 6.7) % for the GN, KC, and ET-EoS viscosity models, respectively, which show that the GN model is superior to the KC and ET-EoS models in predicting the whole viscosity−temperature−pressure−composition surface of the ternary system studied.

Introduction

Knowledge of the dynamic viscosity of petroleum fluids under reservoir conditions is important for simulating reservoir production systems and designing transport equipments. In this context, a significant number of systematic studies have appeared in the literature that deal with the liquid viscosity of binary mixtures mostly at atmospheric pressure. In an effort to characterize the viscosity behavior of a petroleum fluid or a petroleum cut more realistically, only a few systematic studies have been reported so far for ternary mixtures over wide temperature and pressure ranges. Among these, for example, Iglesias-Silva et al.[1] measured the liquid viscosity of the system pentane + octane + decane using a rolling-ball viscometer at temperatures from (298.15 to 373.15) K and at pressures up to 25 MPa over the entire composition range. The authors compared their experimental results (540 points) to the correlation proposed by Assael et al.[2] and obtained an agreement within ± 6 %.

In another related work, Zeberg-Mikkelsen et al.[3] reported comprehensive viscosity and density measurements for the ternary system 1-methylnaphtalene + *n*-tridecane + 2,2,4,4,6,8,8-heptamethylnonane at high pressures (up to 100 MPa) and within the temperature range of (293.15 to 353.15) K. They used a falling-body viscometer to measure the dynamic viscosity above 0.1 MPa. At atmospheric pressure (0.101 MPa), however, a Ubbelohde viscosimeter was used by the authors. Their viscosity results comprised a total of 882 experimental points and were interpreted in terms of various mixing laws and the Eyring theory as well. Similarly, Zeberg-Mikkelsen et al.[4] measured the dynamic viscosity and density of 13 ternary mixtures containing methylcyclohexane + *cis*-decalin + 2,2,4,4,6,8,8-heptamethylnonane over the temperature range of (293.15 to 353.15) K and up to 100 MPa. Their viscosity measurements yielded a total of 546 experimental points. Two simple mixing laws (Grunberg−Nissan (GN) and Katti−Chaudrhi (KC)) were considered by the authors for correlating purposes.

More recently, Zeberg-Mikkelsen et al.[5] revisited their previous viscosity work for the ternary mixture methylcyclohexane + *cis*-decalin + 2,2,4,4,6,8,8-heptamethylnonane (Zeberg-Mikkelsen et al.[4]) to perform a comparative study of various viscosity models. In doing so, they considered two mixing laws (GN[6] and KC[7]), a self-referencing model,[8] the Lohrenz−Bray−Clark (LBC) correlation,[9] a hard-sphere scheme,[10,11] a free-volume viscosity model,[12,13] and the friction theory.[14,15] As demonstrated by the authors, the performance of all of the models in predicting the viscosity of the aforementioned ternary mixture was quite similar, with the GN and the friction theory approach giving the smallest deviations between calculated and experimental viscosity values.

In an attempt to expand the availability of viscosity measurements for pure hydrocarbon and their mixtures over a wide range of temperature and pressure, we previously initiated a systematic study of liquid viscosities of mixtures containing paraffin, aromatic, and naphthenic compounds. Accordingly, we first presented experimental liquid viscosities for *n*-tetradecane, benzene, and cyclohexane, and its corresponding binary systems at the temperature range of (313.2 to 393.2) K and at pressures up to 60 MPa (Hernández-Galván et al.[16,17]). As a continuation of previous work, we report here liquid viscosity measurements for the ternary system benzene + cyclohexane + *n*-tetradecane from (313.2 to 393.2) K and at pressures up to 60 MPa over the entire composition range. Dynamic viscosity data for this system were determined in a high-pressure rolling-ball viscometer using a density Tait-type correlation reported by Assael et al.[18] for *n*-tetradecane and by Cibulka and Takagi[19,20] for benzene and cyclohexane.

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10.1021/je8009122 CCC: $40.75

Experimental Section

Materials. Cyclohexane, normal tetradecane, and benzene were acquired from Aldrich with a minimum mole fraction purity of 99.9 %, 99 %, and 99.9 %, respectively. All of these chemicals were used without further purification.

Apparatus and Procedure. The apparatus and procedures employed in its operation have been previously described[16] and will not be repeated in detail in this report. Briefly, the apparatus mainly consists of a commercial rolling-ball viscometer (Ruska, model 1602-828) that can be maintained within ± 0.1 K for temperatures between (303 and 423) K. Pressure within the viscometer can be maintained within 0.03 % of gauge reading up to 69 MPa.

The experimental procedure first consists of a comprehensive cleaning of the whole system to remove any trace of dust or grease that prevents the stainless steel ball from freely rolling through the liquid inside the barrel of the viscometer. Subsequently, the system is evacuated using a vacuum pump, and the fluid is introduced to the viscometer through a high-pressure cell by means of an ISCO syringe pump (model 100 DM) with a pressure rating of 69 MPa. Finally, the experimental temperature and pressure are fixed. Once the equilibrium conditions are reached and stabilized, measurements of the fall times of the steel ball are performed.

In the rolling-ball viscometer, a stainless steel sphere rolls through a fluid of unknown viscosity under given conditions of temperature and pressure for a precise distance. The viscosity is then a function of the rolling time and the density difference between the sphere and fluid, which can be expressed as

$$\eta = K(T,p,t)(\rho_{\text{ball}} - \rho_{\text{fluid}}) \tag{1}$$

where η is the absolute viscosity, t is the roll time, ρ_{ball} is the density of the sphere, ρ_{fluid} is the density of the fluid, and $K(T,p,t)$ is a function of temperature, T, pressure, p, and the rolling time of the ball, which is obtained by calibrating the viscometer with different substances of known viscosity and density. Further details involving the calibration of the viscometer are given in a previous work.[16] Once the calibration of the viscometer is completed and after applying eq 1, viscosities are determined with a overall uncertainty of about ± 2 % over the whole temperature and pressure ranges studied, which is comparable to that obtained, for instance, by Iglesias-Silva et al.[1] using the same experimental technique.

Results

Experimental viscosity data at high pressures for the ternary mixture benzene + cyclohexane + n-tetradecane have not been previously reported. Liquid viscosities for this system were therefore experimentally measured here over the temperature range from (313.2 to 393.2) K and pressures up to 60 MPa. All of the measurements were performed in a rolling-ball viscometer that had a sphere diameter of 6.22 mm ($\rho_{\text{ball}} = 7.6185\ \text{g}\cdot\text{cm}^{-3}$) and an inclination angle of 23°. For each viscosity value, an average value of 10 measurements of all of the roll time was used. The viscometer was rotated during 15 min to obtain a uniform temperature and mixing within the viscometer. Thermal and mechanical equilibrium was reached in the viscometer prior to the time recording.

Fluid densities of n-tetradecane are determined using the following Tait-like equation suggested by Assael et al.[18] for normal alkane compounds (from ethane to n-hexadecane)

$$\rho_{\text{fluid}}(T,p) = \rho_0(T,p_0)\left[1 - 0.2\log\left(\frac{B(T)+p}{B(T)+p_0}\right)\right]^{-1} \tag{2}$$

where ρ_{fluid} and ρ_0 are the densities of the fluid at pressures p and p_0, respectively, and $B(T)$ is a temperature-dependent parameter. The density of the liquid at $p_0 = 0.101325$ MPa is determined from the correlation proposed by Cibulka and Hnědkovský[21]

$$\rho_0 = \rho_c[1 + \sum_{i=1}^{5} a_i(1 - T_r)^{1/3}] \tag{3}$$

where ρ_c is the critical density of the fluid, T_r is the reduced temperature ($T_r = T/T_c$), and coefficient a_i ($i = 1,...,5$) is a parameter that depends on the type of fluid under study.

Fluid densities for benzene and cyclohexane are determined using the following Tait-like equation proposed by Cibulka and Takagi[19,20] for aromatic and nonhydrocarbon compounds

$$\rho_{\text{fluid}}(T,p) = \rho_0(T,p_0)\left[1 - C(T)\ln\left(\frac{B(T)+p}{B(T)+p_0}\right)\right]^{-1} \tag{4}$$

where $B(T)$ and $C(T)$ are temperature-dependent parameters, and the reference density $\rho_0(T, p_0)$ is evaluated at $p_0 = 0.101325$ MPa by using the correlations also reported by these authors.

For the ternary mixtures, fluid densities are calculated from the expression

$$\rho_{\text{mix}} = \left(\sum_{i=1}^{3} \frac{w_i}{\rho_i}\right)^{-1} \tag{5}$$

where w_i and ρ_i are the mass fraction and density of component i, respectively. Et-Tahir et al.[22] showed that although eq 5 assumes that the mixing is ideal, it can be used for nonideal mixtures by taking into account that an error of $10\ \text{kg}\cdot\text{cm}^{-3}$ in liquid density generates a relative error of 0.001 for viscosity.

Table 1 presents the measured liquid viscosity values of the ternary system benzene + cyclohexane + n-tetradecane at different pressures, temperatures, and compositions. As can be seen in Table 1, eight different compositions in terms of mole fraction of benzene and cyclohexane were chosen in this study. Dynamic viscosity was determined at four temperatures (313.2, 333.2, 353.2, and 393.2 K) and eight pressures (1, 5, 10, 20, 30, 40, 50, and 60 MPa) for the eight compositions, thus yielding a total of 256 viscosity values.

To illustrate how the experimental viscosity of the ternary system benzene + cyclohexane + n-tetradecane under study varies with pressure and temperature at a fixed composition, Figure 1 presents the viscosity behavior as a function of pressure at different temperatures for a specific composition of 0.291 benzene mole fraction and 0.373 cyclohexane mole fraction, whereas Figure 2 shows the behavior of the viscosity as a function of temperature at different pressures for the same composition.

Figure 1 shows that viscosity increases almost linearly as pressure is increased for all isotherms investigated, but it decreases as temperature increases for a given pressure. This effect can be seen in more detail in Figure 2. As depicted in this Figure, viscosity decreases as temperature increases for all isobars considered, following a regular trend. This Figure also

Table 1. Experimental Dynamic Viscosities, η/mPa·s, as a Function of Temperature, Pressure, and Composition for the Benzene (1) + Cyclohexane (2) + *n*-Tetradecane (3) System

	T/K			
p/MPa	313.2	333.2	353.2	393.2
$x_1 = 0.101$, $x_2 = 0.151$				
1	1.379	1.030	0.812	0.456
5	1.435	1.083	0.854	0.481
10	1.501	1.117	0.902	0.514
20	1.662	1.240	1.995	0.575
30	1.827	1.354	1.080	0.642
40	2.019	1.462	1.169	0.711
50	2.242	1.581	1.264	0.780
60	2.530	1.710	1.337	0.842
$x_1 = 0.100$, $x_2 = 0.451$				
1	1.058	0.802	0.618	0.377
5	1.104	0.842	0.659	0.396
10	1.154	0.895	0.701	0.423
20	1.261	0.995	0.780	0.479
30	1.384	1.083	0.862	0.538
40	1.514	1.177	0.939	0.594
50	1.648	1.268	1.016	0.653
60	1.793	1.356	1.093	0.711
$x_1 = 0.105$, $x_2 = 0.770$				
1	0.780	0.650	0.485	0.325
5	0.820	0.660	0.526	0.338
10	0.868	0.698	0.555	0.359
20	0.963	0.775	0.619	0.405
30	1.057	0.850	0.682	0.446
40	1.149	0.925	0.748	0.490
50	1.245	1.001	0.816	0.537
60	1.345	1.075	0.878	0.583
$x_1 = 0.298$, $x_2 = 0.151$				
1	1.054	0.790	0.633	0.392
5	1.101	0.829	0.669	0.410
10	1.154	0.875	0.706	0.435
20	1.261	0.965	0.791	0.487
30	1.374	1.056	0.863	0.550
40	1.494	1.137	0.940	0.597
50	1.630	1.223	1.015	0.654
60	1.766	1.311	1.078	0.709
$x_1 = 0.291$, $x_2 = 0.373$				
1	0.942	0.742	0.588	0.384
5	0.984	0.774	0.622	0.398
10	1.044	0.820	0.651	0.418
20	1.141	0.904	0.727	0.474
30	1.241	0.987	0.802	0.530
40	1.347	1.072	0.873	0.576
50	1.456	1.144	0.936	0.626
60	1.572	1.224	1.005	0.679
$x_1 = 0.504$, $x_2 = 0.143$				
1	0.820	0.641	0.513	0.326
5	0.861	0.661	0.534	0.346
10	0.901	0.698	0.558	0.365
20	0.994	0.777	0.624	0.405
30	1.078	0.853	0.685	0.445
40	1.167	0.921	0.744	0.481
50	1.260	0.993	0.808	0.530
60	1.344	1.065	0.869	0.571
$x_1 = 0.510$, $x_2 = 0.344$				
1	0.628	0.495	0.407	0.279
5	0.653	0.517	0.429	0.288
10	0.688	0.548	0.451	0.305
20	0.764	0.605	0.497	0.336
30	0.842	0.664	0.546	0.368
40	0.916	0.727	0.596	0.399
50	0.987	0.787	0.639	0.435
60	1.057	0.849	0.688	0.466
$x_1 = 0.702$, $x_2 = 0.149$				
1	0.648	0.513	0.420	0.288
5	0.669	0.531	0.436	0.299
10	0.705	0.554	0.456	0.318
20	0.771	0.609	0.496	0.349
30	0.833	0.660	0.545	0.375
40	0.900	0.719	0.588	0.405
50	0.977	0.771	0.634	0.431
60	1.042	0.824	0.679	0.462

shows that the viscosity is larger at high pressures and low temperatures than at low pressures and high temperatures. A similar viscosity behavior as a function of either pressure or temperature was observed for the other seven mixtures varying in composition.

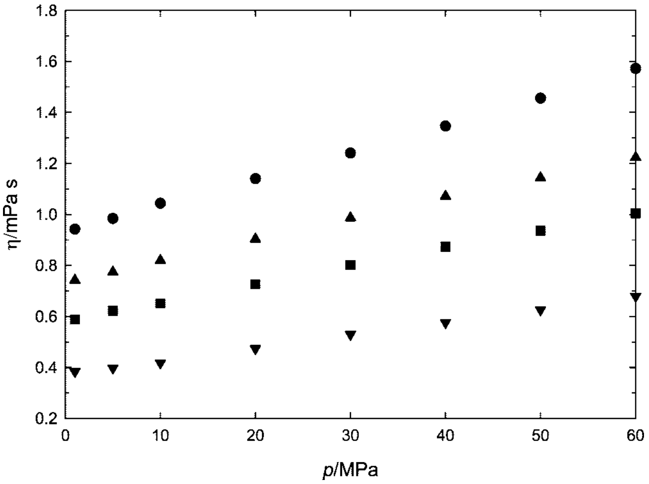

Figure 1. Experimental dynamic viscosity as a function of pressure for the system benzene (1) + cyclohexane (2) + *n*-tetradecane (3) at $x_1 = 0.291$ and $x_2 = 0.373$: ●, 313.2 K; ▲, 333.2 K; ■, 353.2 K; ▼, 393.2 K.

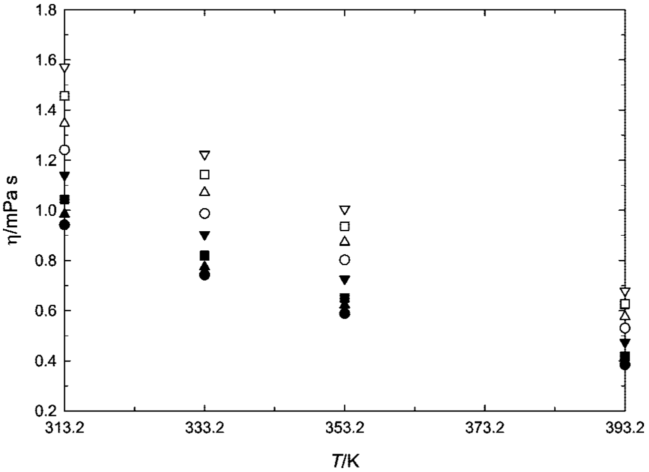

Figure 2. Experimental dynamic viscosity as a function of temperature for the system benzene (1) + cyclohexane (2) + *n*-tetradecane (3) at $x_1 = 0.291$ and $x_2 = 0.373$: ●, 1 MPa; ▲, 5 MPa; ■, 10 MPa; ▼, 20 MPa; ○, 30 MPa; △, 40 MPa; □, 50 MPa; ▽, 60 MPa.

Modeling

The measured viscosity data of the present ternary system (256 experimental points) have been used to perform a comparative study and evaluation of the performance of various representative viscosity models as a function of temperature, pressure, and composition. The considered models are the classical mixing laws by Grunberg and Nissan[6] and Katti and Chaudhri[7] and a recent approach with a more physical and theoretical background, the Eyring-theory-based model coupled to a cubic equation of state (ET-EoS).[16] These viscosity models were chosen because they are thought to give a satisfactory representation of the $\eta-T-p-x$ surface from low to elevated pressures. An important feature of these three viscosity models is that for each binary system, they use a single temperature-independent binary interaction parameter to describe the whole $\eta-T-p-x$ surface of interest.

For a ternary mixture, the GN equation can be expressed as

$$\ln \eta = \sum_{i=1}^{3} x_i \ln \eta_i + x_1 x_2 g_{12}^{\mathrm{GN}} + x_1 x_3 g_{13}^{\mathrm{GN}} + x_2 x_3 g_{23}^{\mathrm{GN}} \quad (6)$$

whereas the KC equation is given by

$$\ln(\eta V) = \sum_{i=1}^{3} x_i \ln(\eta_i V_i) + x_1 x_2 g_{12}^{\mathrm{KC}} + x_1 x_3 g_{13}^{\mathrm{KC}} + x_2 x_3 g_{23}^{\mathrm{KC}} \quad (7)$$

where η_i is the viscosity of component i, η is the viscosity of the mixture, x_i is the mole fraction of component i, and g_{ij}^{GN} and g_{ij}^{KC} are the interaction parameters that characterize the binary formed by components i and j for the GN and KC models, respectively.

The form of the viscosity model ET-EoS for a ternary mixture can be written as

$$\eta = \frac{(\eta V)^{\mathrm{id}}}{V_{\mathrm{m}}} \exp[\sum_{i=1}^{3} x_i (\ln \phi_i - \ln \phi_i^0) + x_1 x_2 g_{12}^{\mathrm{EE}} + x_1 x_3 g_{13}^{\mathrm{EE}} + x_2 x_3 g_{23}^{\mathrm{EE}}] \quad (8)$$

where $\phi_i(T, p, x)$ is the fugacity coefficient of component i in the mixture, $V_{\mathrm{m}}(T, p, x)$ is the molar volume of the liquid mixture, $\phi_i^0(T, p)$ is the fugacity coefficient of the pure fluid, g_{ij}^{EE} is the binary interaction parameter for this viscosity model, and $(\eta V)^{\mathrm{id}}$ is the kinematic viscosity of an ideal solution given by

$$(\eta V)^{\mathrm{id}} = \exp[\sum_{i=1}^{2} x_i \ln(\eta_i^0 V_i^0)] \quad (9)$$

where $V_i^0(T, p)$ is the molar volume of the pure liquid.

In this work, the calculation of the fugacity coefficients $\phi_i(T, p, x)$ and $\phi_i^0(T, p)$ and molar volumes $V_m(T, p, x)$ and $V_i^0(T, p)$ was performed using the Peng−Robinson equation of state;[23] however, the calculation of the kinematic viscosity of an ideal solution $(\eta V)^{\mathrm{id}}$ in eq 9 requires the viscosity values of the pure components, η_i^0, at the temperature and pressure of interest. For tetradecane and benzene, the experimental viscosities are given by Hernández-Galván et al.,[16] whereas for cyclohexane, the experimental viscosities are reported elsewhere.[17]

Equations 6, 7, and 8 were applied to the modeling of the experimental liquid viscosities obtained in this work. The binary interaction parameters used in the viscosity prediction of the ternary system benzene (1) + cyclohexane (2) + *n*-tetradecane (3) were previously obtained from the viscosity correlation of the binary systems benzene + cyclohexane, benzene + *n*-tetradecane, and cyclohexane + *n*-tetradecane; the corresponding g_{ij} values for the three viscosity models were[16,17] $g_{12}^{\mathrm{GN}} = 0.509$, $g_{13}^{\mathrm{GN}} = -0.365$, $g_{23}^{\mathrm{GN}} = 0.441$ and $g_{12}^{\mathrm{KC}} = 1.035$, $g_{13}^{\mathrm{KC}} = -0.293$, $g_{23}^{\mathrm{KC}} = 0.803$ for the GN and KC models, respectively, and $g_{12}^{\mathrm{EE}} = 0.879$, $g_{13}^{\mathrm{EE}} = -0.336$, $g_{23}^{\mathrm{EE}} = 0.897$ for the ET-EoS viscosity model. Results of the representation for the binary systems with the GN, KC, and ET-EoS viscosity modeling yielded, respectively, average absolute deviations of (3.1, 3.8, and 4.0) % for the benzene + cyclohexane system,[17] (2.4, 3.5, and 4.0) % for the benzene + *n*-tetradecane system,[16] and (3.4, 3.8, and 3.5) % for the cyclohexane + *n*-tetradecane system,[17] whereas for pure components, a comparison between our measured viscosities and the correlation given by Assael et al.[2] showed average absolute deviations of 1.5 % for benzene,[16] 2.3 % for cyclohexane,[17] and 2.7 % for *n*-tetradecane.[16]

For the ternary system benzene + cyclohexane + *n*-tetradecane, the agreement between experimental viscosity values (η_i^{exptl}) and those calculated by each model considered here was established through the average absolute deviation (AAD)

$$\mathrm{AAD}\ \% = \frac{100}{N_{\mathrm{p}}} \sum_{i=1}^{N_{\mathrm{p}}} \left| \frac{\eta_i^{\mathrm{exptl}} - \eta_i^{\mathrm{calcd}}}{\eta_i^{\mathrm{exptl}}} \right| \quad (10)$$

the point at which the maximum deviation (MD) occurs

$$\mathrm{MD}\ \% = \mathrm{Max}\left(100 \left| \frac{\eta_i^{\mathrm{exptl}} - \eta_i^{\mathrm{calcd}}}{\eta_i^{\mathrm{exptl}}} \right| \right) \quad (11)$$

and the bias (how well the calculated values are distributed around the experimental ones)

$$\mathrm{bias} = \frac{100}{N_p} \sum_{i=1}^{N_p} \frac{\eta_i^{\mathrm{exptl}} - \eta_i^{\mathrm{calcd}}}{\eta_i^{\mathrm{exptl}}} \quad (12)$$

where subscript i in this case means the ith experimental point in the mixture.

For each viscosity model considered here, the corresponding AAD, MD, and bias values based on a total of 256 experimental points in all cases are (4.9, 32.1 and 0.6) % for the GN correlation, (5.3, 32.4, and 1.0) % for the KC correlation, and (6.7, 18.4, and −0.3) % for the ET-EoS viscosity model. An inspection of these results shows that the correlations of GN and KC give better prediction results with AAD values of (4.9 and 5.3) %, respectively; however, the lowest MD and bias values obtained from the ET-EoS model clearly indicate that the deviations between experimental viscosity data and those estimated from this model exhibit a better distribution around zero. In this context, the prediction performance of the ET-EoS model should be regarded to be quite acceptable.

To illustrate the performance of the GN and ET-EoS models in predicting viscosities for the ternary system benzene + cyclohexane + *n*-tetradecane over the pressure and temperature range experimentally studied at a fixed composition, Figure 3 presents the viscosity behavior as a function of pressure at different temperatures for a specific composition of 0.291 benzene mole fraction and 0.373 cyclohexane mole fraction, whereas Figure 4 shows the behavior of the viscosity as a function of temperature at different pressures for the same composition. In these Figures, the symbols are the experimental pressure and temperature data, whereas the solid and dashed lines are those calculated with the ET-EoS and GN models, respectively.

Figures 3 and 4 show that the ET-EoS model predicts low and moderate pressures for all isotherms studied well, but it is less satisfactory when pressure increases. The overall agreement between experimental and predicted viscosity values with the GN model is quite satisfactory, especially at the highest pressures. Similar plots were prepared for the other seven compositions reported in Table 1and yielded similar results.

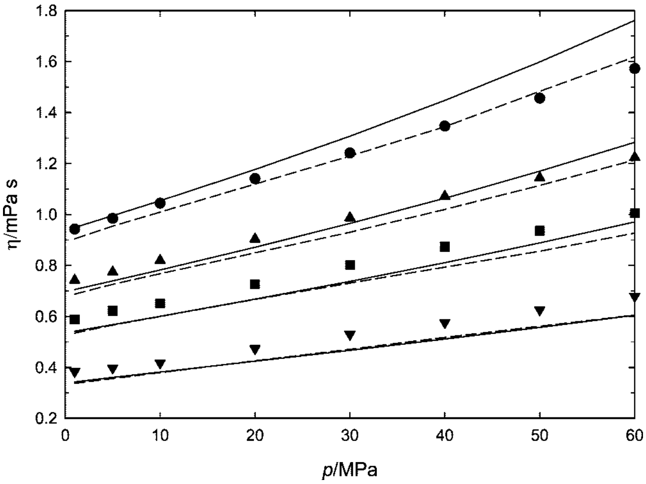

Figure 3. Experimental and calculated dynamic viscosity as a function of pressure for the system benzene (1) + cyclohexane (2) + *n*-tetradecane (3) at $x_1 = 0.291$ and $x_2 = 0.373$: ●, 313.2 K; ▲, 333.2 K; ■, 353.2 K; ▼, 393.2 K; —, ET-EoS viscosity model; ---, GN correlation.

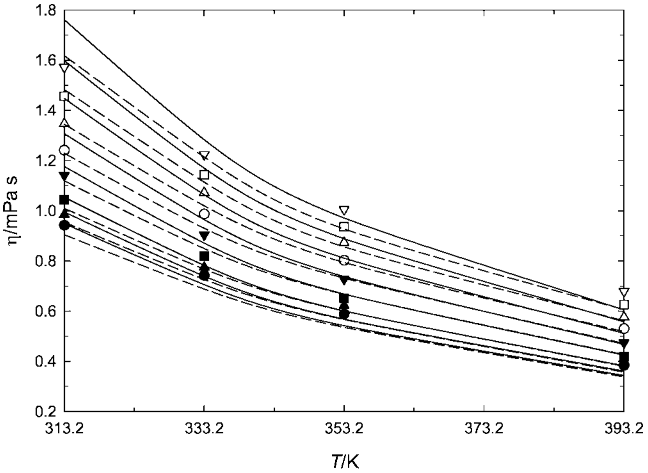

Figure 4. Experimental and calculated dynamic viscosity as a function of temperature for the system benzene (1) + cyclohexane (2) + *n*-tetradecane (3) at $x_1 = 0.291$ and $x_2 = 0.373$: ●, 1 MPa; ▲, 5 MPa; ■, 10 MPa; ▼, 20 MPa; ○, 30 MPa; △, 40 MPa; □, 50 MPa; ▽, 60 MPa; —, ET-EoS viscosity model; ---, GN correlation.

Conclusions

A rolling-ball viscometer has been used to experimentally determine liquid viscosities of the ternary system benzene + cyclohexane + *n*-tetradecane over the temperature range from (313.2 to 393.2) K and pressures up to 60 MPa at eight different mixture compositions with an estimated experimental uncertainty in the measured viscosity data of ± 2 %.

The modeling of this ternary system with three different viscosity models (i.e., the GN and KC correlations and the ET-EoS model) by using a single temperature-independent binary interaction parameter per each binary subsystem showed that despite its simplicity, the use of the GN approach gave the best prediction of the dynamic viscosity of the ternary system studied. Nevertheless, the use of the Eyring-based modeling approach (ET-EoS) also yielded quite acceptable prediction results for the present ternary system with an AAD value of 6.7 %.

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Received for review November 26, 2008. Accepted February 16, 2009. This work was supported by the Molecular Engineering Research Program of the Mexican Petroleum Institute under projects D.00406 and I.00432. M.A.H.-G. gratefully acknowledges the National Council for Science and Technology (CONACyT) of Mexico for their financial support through a Ph.D. fellowship.

JE8009122