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Dynamic Viscosity and Density of Heptylbenzene and Heptylcyclohexane up to 100 MPa

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The dynamic viscosity η and the density ρ of heptylbenzene (1-phenylheptane) and heptylcyclohexane (1-cyclohexylheptane) have been studied as a function of temperature (293.15 K to 353.15 K in 10 K steps) and pressure (up to 60 MPa for density and up to 100 MPa for viscosity). The dynamic viscosity was measured with a falling-body viscometer, except at atmospheric pressure, where a classical capillary viscometer was used. The uncertainty for the viscosity is less than 2%. The uncertainty in the density measurements is less than 1 kg·m^{-3}. Using a Tait-type relation, the density has been extrapolated to 80 and 100 MPa. The dynamic viscosities of these two compounds have been modeled with three recently proposed models which have a physical and theoretical background but represent three different approaches. The models considered are the hard-sphere viscosity scheme, the free-volume viscosity model, and the friction theory. For these three models, the absolute average deviation is of the order of the experimental error.

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1. Introduction

Despite the large number of works already completed on hydrocarbons, there is still a lack of experimental data concerning some of these compounds as a function of pressure, especially for heavy hydrocarbons with high normal boiling points belonging to the naphthenic and aromatic families. The viscosity and the density are important properties required within many chemical engineering disciplines, such as simulation of processes or the design of chemical equipments. This work presents the dynamic viscosities and the densities of heptylbenzene (1-phenylheptane) and heptylcyclohexane (1-cyclohexylheptane) in the temperature range 293.15 K to 353.15 K, at pressures up to 100 MPa.

The viscosity values have been modeled by three recently proposed viscosity models applicable to hydrocarbon fluids over wide ranges of temperature and pressure. The three models are based on the hard-sphere scheme, the free-volume, and the friction theory. These models have a physical and theoretical background, and they represent three different approaches for modeling the viscosity.

2. Experimental Techniques

The dynamic viscosity η was measured up to 100 MPa using a falling-body viscometer of the type designed by Ducoulombier et al.[1] In this apparatus, a stainless steel cylinder falls through a fluid of unknown viscosity under given conditions. The viscosity is a function of the falling time, of the density difference between the cylinder and the fluid, and of one constant obtained by calibrating the viscometer with a substance of known viscosity and density. The technical details for this viscometer are described by Et-Tahir et al.[2] We have used toluene[3] as a calibrating fluid, and *n*-decane[4,5] has been used to verify the calibration. At 0.1 MPa the dynamic viscosity was obtained by measuring the kinematic viscosity, $\nu = \eta/\rho$, with a classical capillary viscometer (Ubbelohde). For this purpose several tubes, connected to an automatic Schott Geräte analyzer, were used. The falling-body viscometer and the Ubbelohde viscometer belong to the types of well-characterized viscometer for which fully developed working equations exist. The interested reader will find a presentation and a discussion of several viscometers in ref 6. For the types of viscometer used in this work, the flow must be laminar. A complete discussion of this aspect has been developed,[7] and for all the experiments carried out in this work, the flow is in the laminar region. The density ρ was measured with an Anton-Paar DMA60 unit combined with an additional 512P high-pressure cell for use up to 60 MPa. The density measurements were extrapolated up to 100 MPa using the following Tait-type equation[2] for the variation of the density versus pressure

$$\frac{1}{\rho(P,T)} = \frac{1}{\rho(0.1\ \mathrm{MPa},\ T)} + A \ln\left(1 + \frac{P/\mathrm{MPa} - 0.1}{B}\right) \quad (1)$$

where A and B are two adjustable constants.

For the viscosity measurements performed with the falling-body viscometer and the density measurements, the uncertainty in the temperature was estimated to be ±0.5 K and ±0.05 K, respectively. The uncertainty in the pressure was estimated to be ±0.1 MPa for the viscosity measurements and ±0.05 MPa for the density measurements (except at 0.1 MPa). The overall uncertainty in the reported density values is lower than 1 kg·m^{-3}, while the uncertainty in the viscosity is of the order of 2% at high pressure. As it has been discussed previously,[5,8] this uncertainty is comparable to the uncertainties obtained by other authors for similar experimental devices. For example, comparative curves for heptane and methylcyclohexane,[9] and for water and 2-propanol[10] plot the experimental values obtained in our laboratory and those obtained by other authors with other techniques. For the measurements of the kinematic viscosity performed with the classical capillary viscometer at atmospheric pressure, the uncertainty in the temperature was ±0.05 K. After mul-

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Table 1. Dynamic Viscosities η and Densities ρ of Heptylbenzene and Heptylcyclohexane versus Temperature T and Pressure P

		heptylbenzene		heptylcyclohexane	
T/K	P/MPa	η/mPa·s	ρ/kg·m^{-3}	η/mPa·s	ρ/kg·m^{-3}
293.15	0.1	2.100	857.5	2.810	810.6
293.15	20	2.570	869.5	3.530	822.8
293.15	40	3.060	880.2	4.410	833.6
293.15	60	3.670	889.9	5.520	843.2
293.15	80	4.510	898.7	7.020	852.0
293.15	100	5.610	906.8	8.980	860.0
303.15	0.1	1.740	850.1	2.230	803.6
303.15	20	2.150	862.7	2.950	816.4
303.15	40	2.580	873.8	3.630	827.6
303.15	60	3.050	883.7	4.490	837.6
303.15	80	3.620	892.8	5.570	846.5
303.15	100	4.320	901.1	6.920	854.7
313.15	0.1	1.480	842.5	1.830	796.5
313.15	20	1.840	855.7	2.390	809.9
313.15	40	2.150	867.4	2.990	821.6
313.15	60	2.530	877.8	3.670	831.9
313.15	80	2.990	887.1	4.450	841.3
313.15	100	3.530	895.7	5.320	849.8
323.15	0.1	1.260	835.1	1.540	789.4
323.15	20	1.590	849.0	2.030	803.5
323.15	40	1.870	861.1	2.500	815.6
323.15	60	2.180	871.7	3.040	826.3
323.15	80	2.530	881.3	3.680	835.9
323.15	100	2.920	889.9	4.410	844.6
333.15	0.1	1.090	827.6	1.320	782.3
333.15	20	1.370	842.1	1.730	797.0
333.15	40	1.610	854.7	2.110	809.7
333.15	60	1.880	865.8	2.570	820.8
333.15	80	2.180	875.8	3.120	830.7
333.15	100	2.500	884.9	3.750	839.7
343.15	0.1	0.944	820.3	1.140	775.3
343.15	20	1.200	835.5	1.480	790.8
343.15	40	1.400	848.5	1.800	804.0
343.15	60	1.630	860.0	2.180	815.4
343.15	80	1.860	870.2	2.610	825.5
343.15	100	2.170	879.4	3.130	834.7
353.15	0.1	0.834	812.7	0.987	768.2
353.15	20	1.060	828.7	1.290	784.4
353.15	40	1.240	842.2	1.600	798.1
353.15	60	1.450	854.1	1.860	809.9
353.15	80	1.670	864.6	2.240	820.3
353.15	100	1.890	874.2	2.720	829.7

tiplying the kinematic viscosity by the density, the dynamic viscosity is obtained with an uncertainty of less than 1%.

The two compounds studied are commercially available chemicals with the following purity levels: heptylbenzene ($C_{13}H_{20}$: Fluka, purity > 99%, M = 176.30 g·mol^{-1}); heptylcyclohexane ($C_{13}H_{26}$: Tokyo Kasei, purity > 99%, M = 182.35 g·mol^{-1}).

3. Results

Measurements of the dynamic viscosity η were made at seven temperatures, (293.15, 303.15, 313.15, 323.15, 333.15, 343.15, and 353.15) K, and at six pressures, (0.1, 20, 40, 60, 80, and 100) MPa, for heptylbenzene and heptylcyclohexane. A total of 84 values were obtained for the dynamic viscosity η. The density measurements were carried out at the same temperatures at pressures from 0.1 MPa to 60 MPa, corresponding to 56 experimental values for the density ρ, which were extrapolated with the aid of the Tait-type relationship, eq 1, to obtain the densities at 80 and 100 MPa (28 values).

Table 1 presents the measured dynamic viscosity and the density values as functions of temperature T and pressure P for heptylbenzene and heptylcyclohexane, respectively. In Figures 1 and 2 the variations of the density versus pressure for various temperatures are shown for respectively heptylbenzene and heptylcyclohexane. Figures 3 and 4 show the dynamic viscosity versus density for respectively heptylbenzene and heptylcyclohexane. The dynamic viscosities and the densities of the two substances are very different for a given P and T. The dynamic viscosity is higher for the naphthenic compound (heptylcyclohexane) than for the aromatic compound (heptylbenzene), whereas the density is lower for heptylcyclohexane compared with the density of heptylbenzene. For instance at P = 100 MPa and T = 293.15 K, η = 5.61 mPa·s and ρ = 906.8 kg·m^{-3} for heptylbenzene, whereas for heptylcyclohexane η = 8.98 mPa·s and ρ = 860.0 kg·m^{-3}.

An examination of Table 1 reveals a general pattern consistent with previous observations made by other

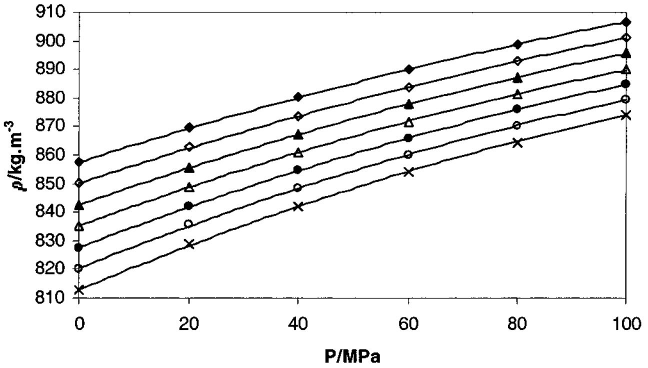

Figure 1. Density ρ of heptylbenzene as a function of pressure for T = 293.15 K (◆), 303.15 K (◇), 313.15 K (▲), 323.15 K (△), 333.15 K (●), 343.15 K (○), 353.15 K (×); (—) polynomial fitting for guiding the eye.

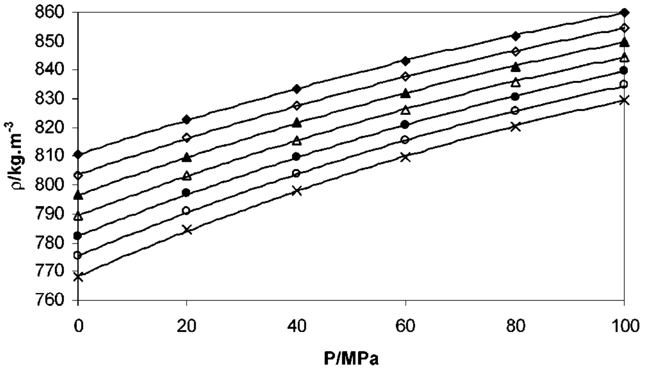

Figure 2. Density ρ of heptylcyclohexane as a function of pressure for T = 293.15 K (◆), 303.15 K (◇), 313.15 K (▲), 323.15 K (△), 333.15 K (●), 343.15 K (○), 353.15 K (×); (—) polynomial fitting for guiding the eye.

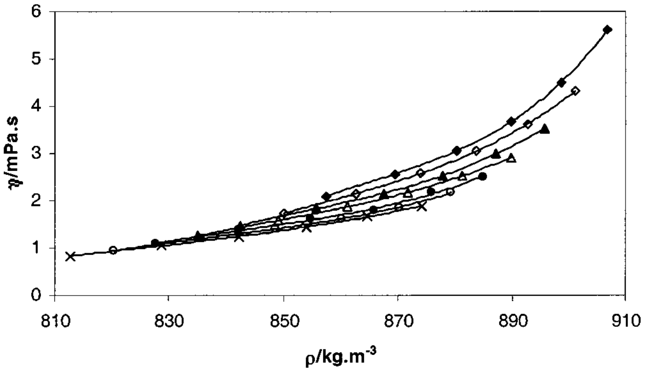

Figure 3. Viscosity η of heptylbenzene as a function of density ρ for T = 293.15 K (◆), 303.15 K (◇), 313.15 K (▲), 323.15 K (△), 333.15 K (●), 343.15 K (○), 353.15 K (×); (—) polynomial fitting for guiding the eye.

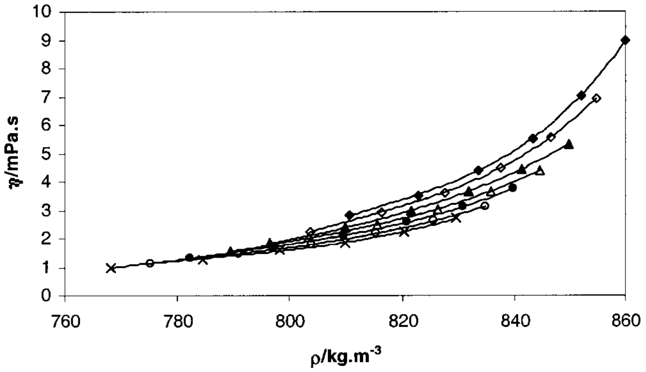

Figure 4. Viscosity η of heptylcyclohexane as a function of density ρ for T = 293.15 K (◆), 303.15 K (◇), 313.15 K (▲), 323.15 K (△), 333.15 K (●), 343.15 K (○), 353.15 K (×); (—) polynomial fitting for guiding the eye.

authors and by us on different hydrocarbon systems. For the studied compounds, the variation of the viscosity as a function of pressure, at constant temperature, shows a sharp increase, and its variation as a function of temperature, at constant pressure, shows a decreasing behavior. The shape of $\rho(P)$ at constant temperature is compatible with the logarithmic form proposed by Tait to model the influence of pressure on $1/\rho$, which is the form used for the extrapolation of the measured densities up to 100 MPa. Further, the variations of ρ versus T are practically linear because of the narrow temperature interval (60 K) considered in this investigation.

4. Discussion

The viscosity data of these two pure hydrocarbons were modeled using three recently proposed viscosity models, which have a physical and theoretical background. The models are based on the hard-sphere viscosity scheme, the free-volume, and the friction theory. To assess and compare the performances of the various models, the following quantities are defined:

$$\mathrm{Dev}_i = 100(1 - \eta_{\mathrm{calc},i}/\eta_{\mathrm{exp},i})$$

$$\mathrm{AAD} = \frac{1}{\mathrm{Nb}}\sum_{i=1}^{\mathrm{Nb}}|\mathrm{Dev}_i|$$

$$\mathrm{Bias} = \frac{1}{\mathrm{Nb}}\sum_{i=1}^{\mathrm{Nb}}\mathrm{Dev}_i$$

$$\mathrm{MD} = \mathrm{Max}|\mathrm{Dev}_i|$$

in which Nb is the number of experimental points, η_{exp} the measured viscosity, and η_{calc} the value calculated using a given model. The quantity AAD (average absolute deviation) indicates how close the calculated curves are to the experimental curves, and the quantity Bias indicates how well the experimental points are distributed to either side of the calculated curves. If Bias = AAD, then all the experimental points are above the calculated curves. Finally, MD characterizes the maximum absolute deviation that can be generated using a given representation.

4.1. Hard-Sphere Viscosity Scheme. A scheme has been developed for the simultaneous correlation of self-diffusions, viscosities, and thermal conductivities of dense fluids.[11,12] The transport coefficients of real dense fluids expressed in terms of $V_r = V/V_0$, with V_0 the close-packed volume and V the molar volume, are assumed to be directly proportional to values given by the exact hard-sphere theory. The proportionality factor, described as a roughness factor R_x (for the property x), accounts for molecular roughness and departure from molecular sphericity. Universal curves for the viscosity have been developed and are expressed as

Table 2. Results Obtained by Adjustment of R_η and V_0 in the Hard-Sphere Viscosity Scheme

	V_0/m^3·mol^{-1}	
T/K	heptylbenzene R_η = 1.669 345 1	heptylcyclohexane R_η = 1.470 922 5
293.15	0.000 150 0	0.000 169 4
303.15	0.000 149 2	0.000 168 4
313.15	0.000 148 1	0.000 167 3
323.15	0.000 147 3	0.000 166 4
333.15	0.000 146 4	0.000 165 6
343.15	0.000 145 6	0.000 164 7
353.15	0.000 144 9	0.000 163 9
AAD/%	1.21	1.25
MD/%	8.14	5.85
Bias/%	0.29	0.37

$$\ln\left(\frac{\eta^*_{\mathrm{exp}}}{R_\eta}\right) = \sum_{i=0}^{7} a_{\eta,i}\left(\frac{1}{V_r}\right)^i \qquad (2)$$

with

$$\eta^*_{\mathrm{exp}} = (6.035 \times 10^8)\left(\frac{1}{MRT}\right)^{1/2}\eta_{\mathrm{exp}}V^{2/3} \qquad (3)$$

where M is the molecular weight and R the gas constant. η_{exp} is the experimental viscosity of the fluid. The dimensionless coefficients $a_{\eta,i}$ are universal, independent of the chemical nature of the compound. This has been verified by Baylaucq et al.,[13,14] who used the hard-sphere scheme to model the viscosities of two ternary systems composed of heptane + methylcyclohexane + 1-methylnaphthalene and water + 2-propanol + 4-hydroxy-4-methyl-2-pentanone.

For alkanes and aromatics, correlations are given for V_0 and R_η.[11,15] It has been observed for various compounds that R_η is temperature independent, whereas V_0 depends on the temperature. The experimental viscosity and density data have been used to fit V_0 (dependent on T) and R_η (independent of T) for the two pure compounds by keeping constant the universal coefficients $a_{\eta,i}$. The results are given in Table 2. As usual, V_0 decreases with temperature. The resultant AAD of the modeling for each compound is within the experimental uncertainty.

4.2. Free-Volume Viscosity Model. Very recently,[16] a new approach has been proposed for the modeling of the viscosity of Newtonian fluids in the dense phase (density ρ > 200 kg·m^{-3}) with small molecules. This approach connects the dynamic viscosity η to the molecular structure via a representation of the free-volume fraction. The dynamic viscosity is expressed as

$$\eta = \frac{\rho l\left(E_0 + \frac{PM}{\rho}\right)}{\sqrt{3RTM}}\exp\left(B\left(\frac{E_0 + \frac{PM}{\rho}}{2RT}\right)^{3/2}\right) \qquad (4)$$

where l is analogous with a length and E_0 is the energy within the molecule. This equation involves three physical parameters l, E_0, and B which are characteristic of the molecule. This model is only valid for dense fluids, but it has been extended to the dilute gas limit.[17] The free-volume

Table 3. Results Obtained by Adjusting the Coefficients E_0, B, and l, in the Free-Volume Viscosity Model

	E_0/J·mol^{-1}	B	l/Å	AAD/%	MD/%	Bias/%
heptylbenzene	127 164.19	0.022 355	0.354 089	1.66	6.68	−0.47
heptylcyclohexane	125 875.28	0.026 534	0.307 960	1.53	5.06	−0.48

Table 4. Results Obtained with the Friction Theory in Conjunction with the PR EOS by Adjusting the Characteristic Critical Viscosity η_c and the Third-Order Friction Constant d_2 (for Heptylcyclohexane)

	η_c/mPa·s	d_2/μP·MPa^{-3}	AAD/%	MD/%	Bias/%
heptylbenzene	0.025 102	0	2.11	6.50	0.09
heptylcyclohexane	0.030 100	$3.501\ 98 \times 10^{-6}$	1.57	6.57	0.38

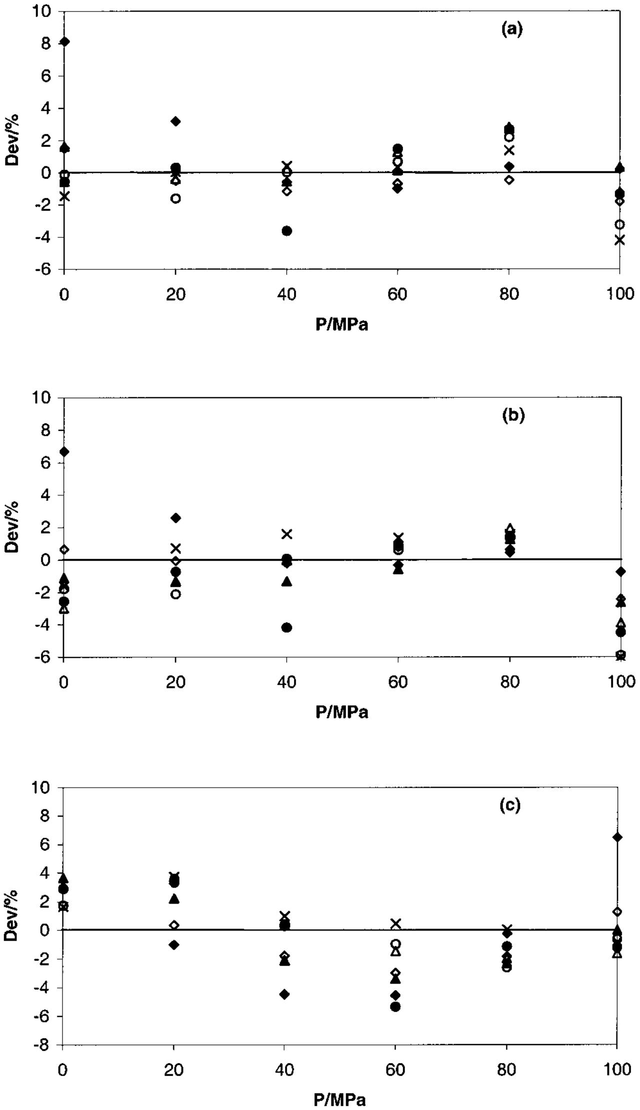

Figure 5. Deviation of the viscosity of heptylbenzene modeled with the hard-sphere scheme (a), the free-volume model (b), and the friction theory (c), as a function of pressure P for T = 293.15 K (◆), 303.15 K (◇), 313.15 K (▲), 323.15 K (△), 333.15 K (●), 343.15 K (○), and 353.15 K (×).

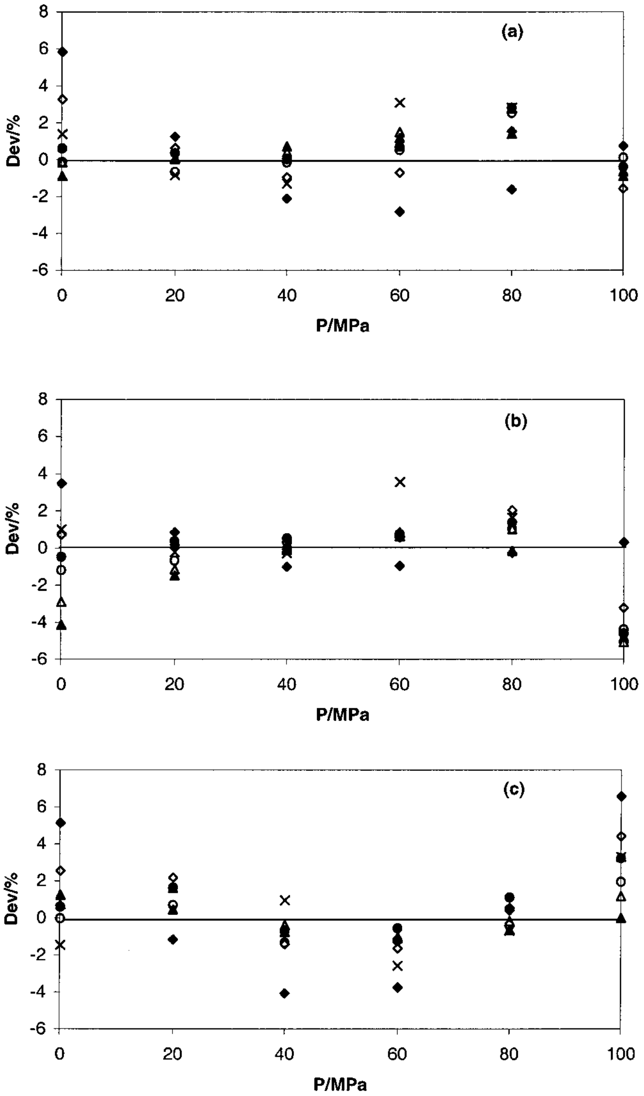

Figure 6. Deviation of the viscosity of heptylcyclohexane modeled with the hard-sphere scheme (a), the free-volume model (b), and the friction theory (c), as a function of pressure P for T = 293.15 K (◆), 303.15 K (◇), 313.15 K (▲), 323.15 K (△), 333.15 K (●), 343.15 K (○), and 353.15 K (×).

model presented in eq 4 has been applied[16] using a database containing 41 compounds of very different chemical species: alkanes (linear and ramified, light and heavy), alkylbenzenes, cycloalkanes, alcohols, fluoroalkanes (refrigerants), carbon dioxide, and water. For the pressure range $P < 110$ MPa and the density range $\rho > 200$ kg·m^{-3} (dense fluids) there are 3012 points in the database,[13] for which an AAD = 2.8% is obtained. Thus, since the compounds studied in this work are in the dense state, the free-volume model[16] presented in eq 4 has been used for the modeling of the viscosity. For each of the studied fluids, the three parameters in eq 4 have been adjusted and are reported in Table 3 along with the AAD, the Bias, and the MD. The obtained AADs are within the experimental uncertainty. Since this viscosity approach has already been applied to very different chemical species, it seems consequently very general.

4.3. Friction Theory. Recently, starting from basic principles of mechanics and thermodynamics, the friction theory (*f-theory*) for viscosity modeling[18] has been developed. In the *f-theory*, the total viscosity η can be written as

$$\eta = \eta_0 + \eta_f \tag{5}$$

where η_0 is the dilute gas viscosity term and η_f the residual friction contribution. The friction contribution is correlated with the van der Waals repulsive pressure term p_r and the attractive pressure term p_a of a cubic equation of state (EOS), such as the Peng and Robinson (PR) EOS.[19] Following these concepts, a general one-parameter *f-theory* model[18] has been proposed. In the case of hydrocarbons with a simple molecular structure, it has been shown that the *f-theory* models[18,20] consisting of a linear correlation on p_a and a quadratic correlation on p_r suffice to accurately represent the viscosity over wide ranges of temperature and pressure. However, in some cases, such as 2,2,4,4,6,8,8-heptamethylnonane,[21] the molecular structure of the compound may induce an interlinking effect that results in an important reduction of the fluid mobility (high viscosity) when brought under pressure. For many systems, such dragging effects can be taken into account by a simple extension of the *f-theory* models' repulsive pressure dependency from quadratic to third order. A third-order *f-theory* model can be written as

$$\eta_f = \kappa_a p_a + \kappa_r p_r + \kappa_{rr} {p_r}^2 + \kappa_{rrr} {p_r}^3 \tag{6}$$

where the κ parameters are the temperature-dependent friction coefficients.

According to the foregoing, to apply the general one-parameter *f-theory* model[20] to fluids with high dragging effects, it is necessary to introduce a third-order corrective term to the general model. Thus, eq 6 can be written as

$$\eta_f = \eta^{GM} + \eta^{III} \tag{7}$$

where η^{GM} is the friction viscosity contribution of the general one-parameter *f-theory* model[20] and $\eta^{III} = \kappa_{rrr} {p_r}^3$ is the third-order correction to the general *f-theory* model. The following expression for the third-order friction coefficient has been used[21]

$$\kappa_{rrr} = d_2(\exp(2\Gamma) - 1)(\Gamma - 1)^3 \tag{8}$$

with

$$\Gamma = \frac{T_c}{T} \tag{9}$$

where d_2 is a component-related parameter.

In this work, the general one-parameter *f-theory* model[20] with a third-order repulsive pressure correction and in conjunction with the PR EOS has been used to model the viscosities of the two pure compounds (Figures 5 and 6). The dilute gas viscosity term has been obtained by the model of Chung et al.[22] The modeling of the viscosity of a pure fluid requires the fitting of two parameters per compound: the characteristic critical viscosity η_c, used in the general one-parameter *f-theory* model, and the d_2 constant. These parameters are given in Table 4 with the obtained AAD and MD. Thus, for heptylbenzene, the pressure effect on the viscosity is not so pronounced that a third-order correction is required. The obtained AADs are also for this model within the experimental uncertainty. These results are interesting because all the parameters involved are universal, except the characteristic critical viscosity (compound related) and the parameter d_2. The used critical properties are for heptylbenzene,[23] $T_c = 713.5$ K, $P_c = 2.2$ MPa, and $\omega = 0.530$, whereas, for heptylcyclohexane,[23] $T_c = 708.6$ K, $P_c = 1.96$ MPa, and $\omega = 0.498$.

5. Conclusion

The viscosities and the densities of heptylbenzene and heptylcyclohexane have been measured up to 100 MPa in the temperature range 293.15 to 353.15 K. The experimental uncertainty for the viscosity measurements is of the order of 2%, except at 0.1 MPa, where the uncertainty is 1%. For the densities, the uncertainty is 1 kg·m^{-3}. It follows from the discussion that some simple viscosity approaches with a physical and theoretical background (the hard-sphere scheme, the free-volume model, and the friction theory) are able to model the viscosities of these two compounds. The experimental data obtained (84 points for the dynamic viscosity) could be included in databases and used to carry out further tests of other more sophisticated models, such as, for instance, models based on molecular dynamic simulation.

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