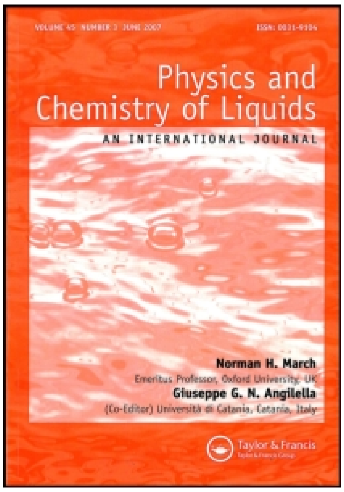

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A HARD-SPHERE SCHEME USED TO MODEL THE VISCOSITY OF TWO TERNARY MIXTURES UP TO 100 MPa

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Recently, an extensive experimental study of two different ternary mixtures has been undertaken which aimed at showing the effect of composition, temperature and pressure on viscosity and density. The ternary mixture heptane + methylcyclohexane + 1-methylnaphtalene has been chosen as it can be, in some ways, a synthetic representation of a C_{5+} distillation fraction of a petroleum crude oil. The ternary mixture water + diacetone-alcohol + 2-propanol has the distinctive feature of involving components which have important interactions. The key issue of this study is the fairly high number of samples within the ternary diagram, as the first mixture is described by 45 compositions and the second one by 66 compositions. The viscosity of both mixtures have then been measured at three temperatures (303.15, 323.15, and 343.15 K) and 6 pressures (0.1, 20, 40, 60, 80 and 100 MPa) using a high pressure falling body viscometer which allowed to collect 1998 experimental data.

Our entire set of data have then been used to test the hard-sphere scheme developed by J. H. Dymond and M. J. Assael. This model has been developed in order to correlate simultaneously thermal conductivity, viscosity and self-diffusion coefficient data for a wide range of temperature and pressure and is derived from the hard-sphere theory.

We have only used the procedure to estimate viscosity and very good results (absolute average deviation equal to 2.3%) are obtained.

Keywords: High pressure; viscosity; density; hard-sphere theory

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INTRODUCTION

Recently [1–7] a scheme has been developed for the simultaneous correlation of self-diffusion, viscosity, and thermal conductivity coefficients data of dense fluids over a wide range of temperatures T and pressures P. The basis of the method is that 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 have been determined empirically for each reduced transport coefficient as a function of reduced density [4]. In this paper, we present the procedure used to estimate the viscosity coefficient. The procedure presented here has been applied to two ternary systems involving 6 different compounds: heptane, methylcyclohexane, and 1-methylnaphtalene on the one hand, and water, 2-propanol, and 4-hydroxy, 4-methyl-2-pentanone (or diacetone alcohol, or DAA) on the other hand. The data used for this study have been presented elsewhere [8–11] and represent a total of 111 different mixtures and 1998 measurement points over the pressure range 0.1–100 MPa and for temperatures 303.15, 323.15, and 343.15 K.

PRESENTATION OF THE CORRELATION SCHEME

It has been shown by Chandler [12] that for rough spherical molecules the transport coefficients can be directly related to the smooth hard-sphere transport coefficients. A corresponding relationship between the experimental transport coefficients of rough nonspherical molecules and the smooth hard-sphere values (subscript shs) can therefore be assumed: $\eta_{\text{exp}} = R_\eta \eta_{\text{shs}}$ where R_η is the roughness factor for viscosity and is assumed to be independent of both temperature and density. Exact smooth hard-sphere transport viscosity are given by the product of the values from Enskog theory [13] and the computed corrections to Enskog theory: $\eta_{\text{shs}} = \eta_E(\eta/\eta_E)_{\text{MD}}$. Reduced coefficient of viscosity η^* is convenient to use [14] and is defined as follows:

$$\eta^*_{\text{exp}} = R_\eta\, \eta^*_{\text{shs}} = R_\eta \left[\frac{\eta_{\text{shs}}}{\eta_0}\right]\left[\frac{V}{V_0}\right]^{2/3}$$

where subscript "0" refers to the low-density hard-sphere coefficient in the first order approximation. Values from the reduced smooth hard-sphere viscosity can be calculated from experiment after substitution of the hard-sphere expressions which give:

$$\eta^*_{\text{shs}} = \frac{\eta^*_{\text{exp}}}{R_\eta} = 6.035 \cdot 10^8 \left[\frac{1}{\text{MRT}}\right]^{1/2} \frac{\eta_{\text{exp}} V^{2/3}}{R_\eta}$$

The determination of V_0 and R_η at any temperature for the viscosity is accomplished by a well-established curve-fitting procedure. A plot of $\log \eta^*_{\text{exp}}$ *versus* $\log V$ from experiment is superimposed on universal plots of $\log \eta^*_{\text{shs}}$ *versus* $\log (V/V_0)$ from hard-sphere theory by vertical and horizontal adjustment from which the R_η factor and V_0 values are respectively obtained. The universal curves for the viscosity were developed from a consideration of both hard-sphere theory and experimental data for higher *n*-alkanes, and are expressed in terms of reduced volume V_r ($V_r = V/V_0$):

$$\log\left[\frac{\eta^*_{\text{exp}}}{R_\eta}\right] = \sum_{k=0}^{7} a_{\eta_k} (1/V_r)^k \qquad (1)$$

where

$$\begin{array}{ll} a_{\eta_0} = 1.0945 & a_{\eta_4} = 797.6900 \\ a_{\eta_1} = -9.26324 & a_{\eta_5} = -1221.9770 \\ a_{\eta_2} = 71.0385 & a_{\eta_6} = 987.5574 \\ a_{\eta_3} = -301.9012 & a_{\eta_7} = -319.4636 \end{array}$$

The determination of parameters R_{η_i} and V_{0_i} on various pure substances shows that V_{0_i} is a function of T whereas R_{η_i} seems to be temperature independent for pseudo-spherical molecules but shows a temperature dependence for molecules which either depart too much from sphericity or have hydrogen bonds. The first group of molecules includes the linear alkanes [4], the second one includes alcohols [15].

This correlation scheme for the viscosity has been applied separately for *n*-alkanes [4] and for aromatics [7] for which the authors give the following correlations:

Alkanes from C_5H_{12} to $C_{16}H_{34}$:

$$\begin{aligned} R_\eta &= 0.995 - 0.0008944C + 0.005427C^2 \\ 10^6 V_0 &= 117.874 + 0.15(-1)^C + 0.25275T \\ &\quad + 5.48 \cdot 10^{-4}T^2 - 4.2464 \cdot 10^{-7}T^3 \\ &\quad + (C-6)(1.27 - 9 \cdot 10^{-4}T)(13.27 + 0.025C) \end{aligned}$$

Aromatics:

$$\begin{aligned} 10^6 V_0 = {} & -3324.7C^{-2} + 529.47C^{-1} + 12.163C \\ & + T(9.48786C^{-2} - 8.55176 \cdot 10^{-2}C + 6.03463 \cdot 10^{-3}C^2) \\ & + T^2(-1.5797 \cdot 10^{-3} + 3.9901 \cdot 10^{-4}C - 2.2309 \cdot 10^{-5}C^2) \end{aligned}$$

where C is the number of carbon atoms and T the temperature. No definite trend was observed in the R_η values going from benzene to mesitylene.

This model can be adapted in order to be used on mixtures by using the following relationships proposed by Assael *et al.* [5] and Mensah-Brown and Wakeham [16]:

$$V_{0_m} = \sum_{i=1}^{n} x_i V_{0_i} \quad R_{\eta_m} = \sum_{i=1}^{n} x_i R_{\eta_i} \quad M_m = \sum_{i=1}^{n} x_i M_i \tag{2}$$

Let us stress here that in a recent paper Dymond [17] indicates that in order to represent the density dependence at each temperature on a universal curve basis, characteristic volume for self-diffusion must be 5% greater than for dynamic viscosity or thermal conductivity. This seems to be related to the translation-rotation coupling. This phenomenon has already been indicated in the case of particles interacting with a Lennard-Jones type of potential in dilute gases, the cohesion section of hard spheres is lower for diffusion than for viscosity or thermal conductivity. Then, the estimation of V_0 may change from a property to another.

EXPERIMENTAL DATA

In order to test the representation scheme presented above, we used density and dynamic viscosity data of two ternary mixtures, which involve 6 different compounds. The first mixture is heptane + methylcyclohexane + 1-methylnaphtalene, and is represented by 3 pure components, 21 (3 × 7) compositions for the binary mixtures, and 21 compositions for the ternary mixture (45 compositions in total), as displayed on Figure 1. The second mixture is water + 2-propanol + DAA, and is represented by 3 pure components, 27 (3 × 9) compositions for binary mixtures, 36 compositions for the ternary mixtures (66 compositions in total) as shown on Figure 2. All the measurements of η and ρ have been taken at temperatures $T = 303.15$, 323.15, and 343.15 K and at pressures $P = 0.1$, 20, 40, 60, 80, and

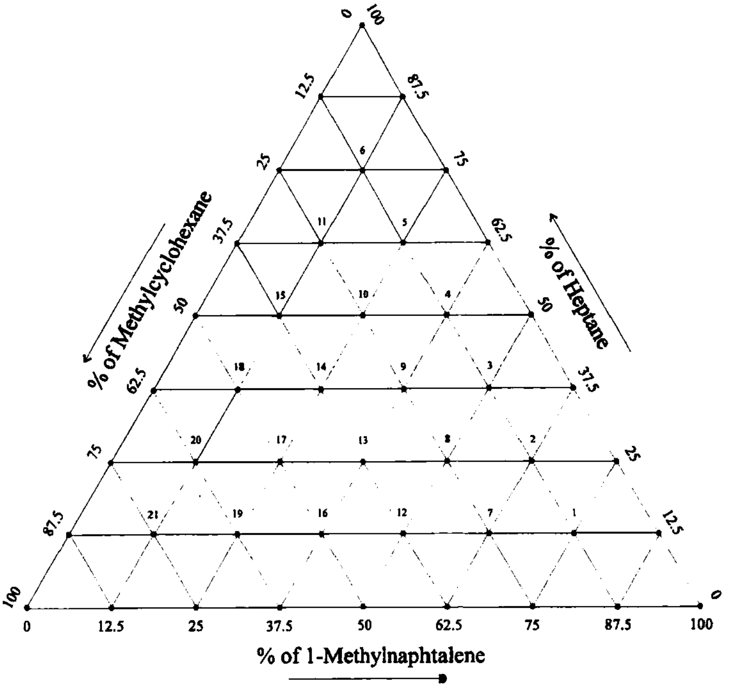

FIGURE 1 Points representative of the three pure substances, the three binaries, and the ternary diagram (as mole percentage) for heptane + methylcyclohexane + 1-methylnaphtalene.

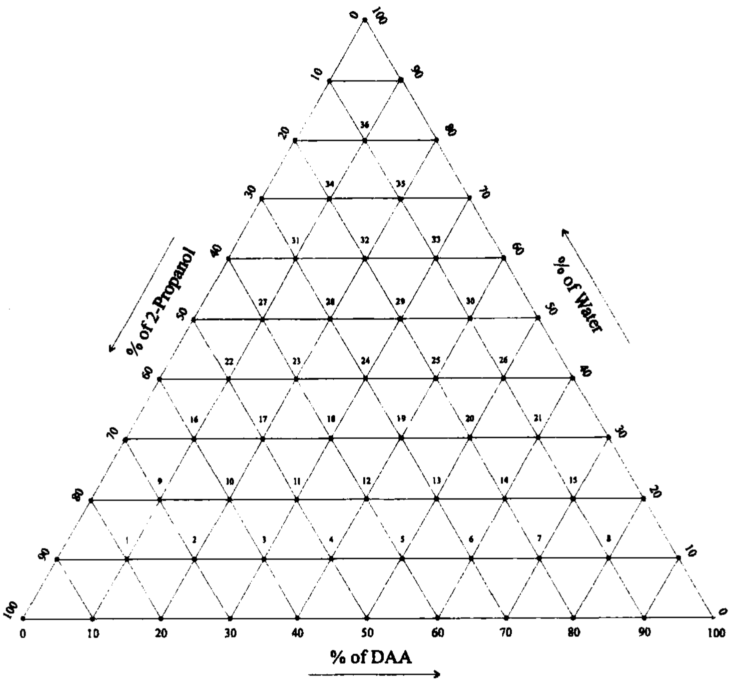

FIGURE 2 Points representative of the three pure substances, the three binaries, and the ternary diagram (as mole percentage) for water + 2-propanol + DAA.

100 MPa for the viscosity [8 – 11]. The entire set of data represent 1998 experimental points ((45 + 66) × 3 × 6 = 1998).

It is important to note here that the first ternary system is a simplified representation of a C_{5+} petroleum fraction [9]. The three compounds are weakly associative and the variations of the viscosity, at a given (P, T) set, is a monotonous function of the composition. Figure 3 ($P = 60\,\mathrm{MPa}, T = 323.15\,\mathrm{K}$) shows typical variations of the dynamic viscosity *versus* composition in the ternary diagram. The second ternary system is, on the contrary, very associative. Interactions between water and each alcohol are very strong, and variations of dynamic viscosity *versus* composition, at a given (P, T) set, are not monotonous any longer. Figure 4 ($P = 40\,\mathrm{MPa}, T = 323.15\,\mathrm{K}$) displays this behaviour. The curve shows a very marked maximum for each of the binary mixtures water + DAA and water + 2-propanol,

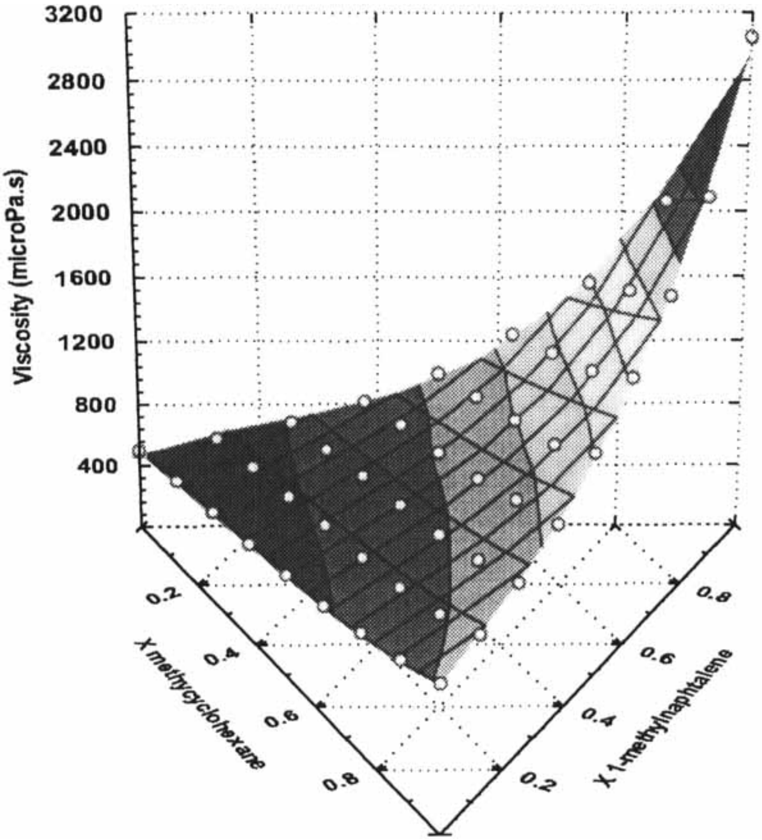

FIGURE 3 Variations of dynamic viscosity ($P = 60\,\text{MPa}, T = 323.15\,\text{K}$) of the ternary heptane + methylcyclohexane + 1-methylnaphtalene.

which are characteristic of the very strong attractive intermolecular forces. The two ternary systems studied here have very different behaviours as regard to dynamic viscosity.

RESULTS

The assessment of the performances of the model are made using the quantities defined as follows:

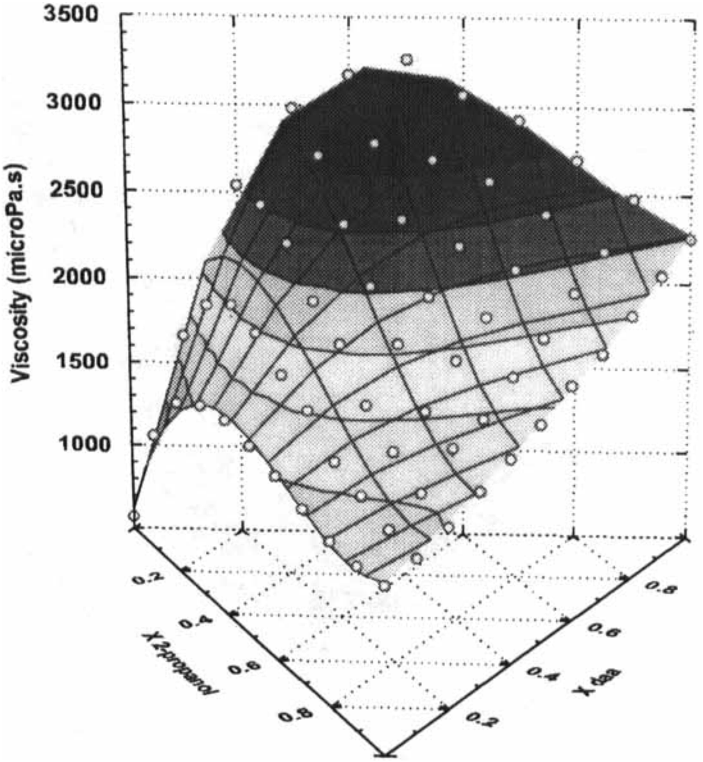

FIGURE 4 Variations of dynamic viscosity ($P = 40\,\text{MPa}, T = 323.15\,\text{K}$) of the ternary water + 2-propanol + DAA.

$$\text{Dev}(i) = 100 \cdot (\eta_{\text{exp}} - \eta_{\text{cal}})/\eta_{\text{exp}}$$

$$\text{Dev abs}(i) = |\text{Dev}(i)| \geq 0$$

$$\text{AAD} = \frac{1}{\text{Nb}} \sum_{i=1}^{\text{Nb}} \text{Dev abs}(i)$$

$$\text{Bias} = \frac{1}{\text{Nb}} \sum_{i=1}^{\text{Nb}} \text{Dev}(i)$$

$$\text{DM} = \text{MAX}(\text{Dev abs}(i))$$

in which Nb is the number of experimental points, η_{exp} the measured viscosity and η_{cal} the value calculated using the 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. Finally DM characterises the maximum error that can be generated using the representation.

Ternary System: Heptane + Methylcyclohexane + 1-Methylnaphtalene

Firstly, we applied (for heptane and methylcyclohexane) the model using the coefficients adjusted on linear alkanes. For heptane we obtained AAD = 2.55%, DM = 5.05% and Bias = 2.16% which is a very good result emphasising the good evaluation of the coefficients by Assael *et al.* [4]. However, the estimation of the viscosity for methylcyclohexane using the same coefficients leads to AAD = 117%, DM = 185% and Bias = −117%. These results can be explained by the fact that this substance is a cyclo-alkane and not a linear alkane. In the case of 1-methylnaphtalene, no result has been obtained: the correlation of dense fluid transport coefficients has been applied [7] to seven simple aromatic hydrocarbons: benzene, toluene, o-xylene, m-xylene, p-xylene, mesitylene and ethylbenzene, and give a correlation for the coefficients V_0 but none has been given for R_η so that it has been impossible to estimate a value for R_η for 1-methylnaphalene.

Secondly, we have fitted the coefficients a_{η_k}, V_{0_i} and R_{η_i} on the 54 experimental values of viscosity for the pure substances. We then obtained better results (Tab. I) for the pure substances (for heptane, AAD = 0.5%) as AAD are below the experimental uncertainty (2%). 1-methylnaphtalene is the substance which is less good represented with AAD = 2.0% even though. The parameters have been used to generate the viscosity of the binary and ternary mixtures (using Eqs. (2)) for which we obtained AAD = 8.49%, DM = 29.6%, Bias = −8.46% (for the binaries) and AAD = 13.6%, DM = 35.0%, Bias = −13.6% (for the ternary).

The parameters a_{η_k}, R_{η_i} and V_{0_i} have been fitted on the 810 experimental values of viscosity of pure substances as well as of mixtures. Table II gives the detailed results. For the overall results, AAD = 3.5% and DM = 18.7% which are very satisfying results, and it can be noticed that 1-methylnaphtalene gives the less good results (AAD = 13.9%). The reason might be the complex chemical structure

TABLE I Results obtained from the adjustment of a_{η_k}, R_{η_i} and V_{0_i} on pure substances

$a_{\eta 0}$	$a_{\eta 1}$	$a_{\eta 2}$	$a_{\eta 3}$	$a_{\eta 4}$	$a_{\eta 5}$	$a_{\eta 6}$	$a_{\eta 7}$
1.192929	−8.104825	70.617766	−343.986946	910.564048	−1260.1597	858.064815	−219.453339

	Heptane	*Methylcyclo.*	1-*Methylnaph.*	*Binaries*	*Ternary*	*Total*
R_η	1.1526970	1.0593564	0.8293858			
V_0 (303.15 K)	$9.517 \cdot 10^{-5}$	$8.902 \cdot 10^{-5}$	$10.898 \cdot 10^{-5}$			
V_0 (323.15 K)	$9.445 \cdot 10^{-5}$	$8.859 \cdot 10^{-5}$	$10.8 \cdot 10^{-5}$			
V_0 (343.15 K)	$9.382 \cdot 10^{-5}$	$8.844 \cdot 10^{-5}$	$10.71 \cdot 10^{-5}$			
AAD (%)	0.50	1.37	2.00	8.49	13.6	10.4
DM (%)	1.53	5.87	7.75	29.6	35.0	35.0
Bias (%)	0.03	−0.04	0.82	−8.46	−13.6	−10.3

TABLE II Results obtained from the adjustment of a_{η_k}, R_{η_i} and V_{0_i} on the 810 experimental values

a_{η_0}	a_{η_1}	a_{η_2}	a_{η_3}	a_{η_4}	a_{η_5}	a_{η_6}	a_{η_7}
1.19998917	−7.93991139	61.6482962	−332.83701	964.00417	−1371.82892	903.146976	−206.904386

	Heptane	*Methylcyclo.*	1-*Methylnaph.*	*Binaries*	*Ternary*	*Total*
R_η	1.17092351	1.12421949	0.76885643	–	–	–
V_0 (303.15 K)	$9.5499 \cdot 10^{-5}$	$8.9109 \cdot 10^{-5}$	$11.022 \cdot 10^{-5}$	–	–	–
V_0 (323.15 K)	$9.4278 \cdot 10^{-5}$	$8.8296 \cdot 10^{-5}$	$10.937 \cdot 10^{-5}$	–	–	–
V_0 (343.15 K)	$9.3354 \cdot 10^{-5}$	$8.7644 \cdot 10^{-5}$	$10.852 \cdot 10^{-5}$	–	–	–
AAD (%)	4.37	9.01	13.9	3.38	2.93	3.55
DM (%)	7.46	14.8	18.7	12.6	13.2	18.7
Bias (%)	4.18	9.63	13.9	2.23	−1.86	0.77

of this compound. It must also be noticed that the values for a_{η_k} parameters presented in Tables I and II are very close to those given by Assael *et al.* [3].

We finally used a_{η_k} parameters given by Assael *et al.* [3] and fitted only R_{η_i}, V_{0_i} on the 810 experimental values of viscosity. The results are displayed in Table III, and it can be noticed that those obtained for the mixtures are better than those obtained on the same mixtures with the a_{η_k}, R_{η_i} and V_{0_i} parameters adjusted only on pure substances (Tab. I). This shows the universality of the a_{η_k} parameters and that the R_{η_i} and V_{0_i} parameters are characteristic of the system studied.

Ternary System: 2-Propanol + DAA + Water

First, the model has been applied to pure substances re-adjusting parameters V_0 and R_η *versus* experimental data of dynamic viscosity keeping the a_{η_k} values already determined by Assael *et al.* [3] because, as explained in the previous paragraph, the a_{η_k} coefficients seem to be universal. No problem has been encountered for 2-propanol or DAA, but for water V_0 and R_η can not be determined simultaneously as dynamic viscosity has a weak pressure dependence. One parameter has to be fixed first. Assael *et al.* [18] give, for water, the following linear relationship $V_0 = b_0 + b_1 T$ with T expressed in K ($b_0 = 6.228 \cdot 10^{-6}$ m$^3 \cdot$ mol^{-1}, $b_1 = 6.238 \cdot 10^{-9}$ m$^3 \cdot$ mol$^{-1} \cdot$ K^{-1}), estimated from thermal conductivity experiments. According to the work of Dymond [17], these values can be used for the estimation of dynamic viscosity (see discussion above). The results then obtained minimising AAD on the three pure substances are presented in Table IV.

In total, the results give AAD = 13.4%, DM = 49.2%, and Bias = 4.42%, which shows a deterioration from those obtained on pure substances for which AAD is lower than the experimental uncertainty. Let us remind here that there are very strong attractive interactions between water and each alcohol. We then considered the case where R_{η_i} is temperature independent. The overall results give AAD = 13.5%, DM = 46.3%, and Bias = 1.14%. With 12 parameters to fit, the representation is as satisfactory as in the case of 18 parameters previously presented, with better DM and Bias even though and only a small deterioration on AAD (13.5% instead of 13.4%).

TABLE III Results obtained from the adjustment of R_{η_i} and V_{0_i} on the 810 experimental values (a_{η_k}: original coefficients)

	Heptane	*Methylcyclo.*	1-*Methylnaph.*	*Binaries*	*Ternary*	*Total*
R_η	1.34756612	1.29609442	0.91217406	–	–	–
V_0 (303.15 K)	$9.1943 \cdot 10^{-5}$	$8.5719 \cdot 10^{-5}$	$10.631 \cdot 10^{-5}$	–	–	–
V_0 (323.15 K)	$9.0758 \cdot 10^{-5}$	$8.5099 \cdot 10^{-5}$	$10.511 \cdot 10^{-5}$	–	–	–
V_0 (343.15 K)	$8.9937 \cdot 10^{-5}$	$8.4385 \cdot 10^{-5}$	$10.429 \cdot 10^{-5}$	–	–	–
AAD (%)	4.50	9.12	18.9	3.55	3.20	3.87
DM (%)	9.02	14.6	33.3	15.7	14.6	33.3
Bias (%)	4.15	8.90	18.9	2.39	−2.26	0.77

TABLE IV Results obtained from the adjustment of R_{η_i} and V_{0_i} (except V_{0_i} for water) on the 54 experimental values of pure substances (18 parameters)

	Water	2-*Propanol*	*DAA*	*Binaries*	*Ternary*	*Total*
R_η (303.15 K)	5.31243466	3.56289665	1.46827431	–	–	–
R_η (323.15 K)	3.70813287	2.4971206	1.62174013			
R_η (343.15 K)	2.64531402	1.96552638	1.50271262			
V_0 (303.15 K)	$0.8119 \cdot 10^{-5}$	$5.0779 \cdot 10^{-5}$	$9.2023 \cdot 10^{-5}$	–	–	–
V_0 (323.15 K)	$0.8244 \cdot 10^{-5}$	$5.0634 \cdot 10^{-5}$	$8.9763 \cdot 10^{-5}$	–	–	–
V_0 (343.15 K)	$0.8369 \cdot 10^{-5}$	$5.0161 \cdot 10^{-5}$	$8.8745 \cdot 10^{-5}$	–	–	–
AAD (%)	1.83	0.72	0.89	16.3	12.3	13.4
DM (%)	6.48	2.29	3.41	49.2	47.6	49.2
Bias (%)	0.35	0.17	0.00	6.86	2.95	4.42

In order to improve the representation for mixtures, characteristic volumes can be developed as follows:

$$V_{0_m}(x,T) = \sum x_i x_j V_{0_{ij}} \quad \text{with} \quad V_{0_{ij}} = \frac{1}{2}(V_{0_i} + V_{0_j})(1 - k_{ij})$$

where the parameters k_{ij} are introduced in order to take into account the interactions between components (i) and (j), as usual. If $k_{ij} = 0$, the representation is the same as the one already presented.

The results of this new development of V_{0_m} are displayed in Table V. If we consider k_{ij} temperature dependent, an improvement can be observed compared to the results presented with $k_{ij} = 0$, but this is done with the addition of 9 interaction parameters (27 in total). It is possible to reduce the number of parameters down to 15 considering R_{η_i} and k_{ij} temperature independent. The deterioration is small (AAD = 8.96% instead of 7.30%) if we take into account the number of parameters (15 instead of 27).

TABLE V Comparison of the results obtained with the development of $V_{0_m}(T)$ using k_{ij} parameters as a function of T and T independent (adjustment on the entire set of data)

	2-Propanol + *DAA*	*Water* + *DAA*	*Water* + *2-Propanol*	*Binaries*	*Ternary*	*Total*
			$k_{ij}(T)$ and $R_\eta(T)$ (27 parameters)			
AAD (%)	4.30	12.4	13.4	10.0	5.77	7.30
DM (%)	9.90	39.0	39.5	39.5	38.6	39.5
Bias (%)	4.30	4.93	5.09	4.77	1.42	2.73
			k_{ij} and R_η, T independent (15 parameters)			
AAD (%)	4.31	14.6	14.5	11.1	7.93	8.96
DM (%)	8.82	39.4	40.6	40.6	38.4	40.6
Bias (%)	3.83	3.20	5.03	4.02	−0.42	1.42

It must be noticed that the results are very sensitive to the precision on V_0, as an error of 2% on V_0 can induce 10% error on dynamic viscosity.

The last step of this work was to minimise the AAD of the entire set of points for this ternary (1188 data points) in order to estimate $V_{0_i}(T)$ and R_{η_i} for the pure components and binary interaction parameters k_{ij}. We used the following development for $R_\eta: R_\eta = G(1/V_0)^2 + F(1/V_0) + E$. In the case of mixtures $V_{0_m}(T)$ are calculated according to the equations involving k_{ij}.

The results displayed in Table VI have been obtained following the three different configurations of this method:

1. $k_{ij}(T)$ and $E(T)$, $F(T)$, and $G(T)$ (27 parameters).
2. k_{ij} temperature independent, $E(T)$, $F(T)$, and $G(T)$ (21 parameters).
3. k_{ij} and E, F, and G temperature independent (15 parameters).

The improvement obtained with method 1 is very important as AAD = 2.1% is of the magnitude of the experimental uncertainty, and DM = 16.6% shows very good performances of the model due to the fact that the system studied here is very associative. Method 2 has the advantage to reduce the number of parameters from 27 to 21 with almost no deterioration as AAD = 2.16% and DM = 16%. Method 3 gives less good results than the first two (AAD = 6.41%), but it involves only 15 parameters. These final results are at least better than

TABLE VI Comparison of the results obtained with the development of $V_{0_m}(T)$ using k_{ij} parameters, and $R_\eta(V_0)$. (Bin 1: 2-propanol + DAA, Bin 2: Water + DAA, Bin 3: Water + 2-propanol)

	Water	*2-Propa.*	*DAA*	*Bin* 1	*Bin* 2	*Bin* 3	*Binaries*	*Ternary*	*Total*
				Method 1 (27 parameters)					
AAD (%)	2.57	3.49	0.94	1.33	2.91	2.64	2.29	1.94	2.10
DM (%)	9.33	9.69	4.01	5.45	16.6	7.06	16.6	7.72	16.6
Bias (%)	−0.63	3.45	0.38	1.20	−2.44	−0.97	−0.74	−0.19	−0.36
				Method 2 (21 parameters)					
AAD (%)	2.57	3.00	1.20	1.41	2.91	2.93	2.42	1.95	2.16
DM (%)	9.34	8.93	4.90	4.76	16.0	7.37	16.0	8.13	16.0
Bias (%)	−0.69	2.77	0.51	1.27	−2.37	−1.24	−0.78	−0.17	−0.37
				Method 3 (15 parameters)					
AAD (%)	1.70	3.22	1.08	2.49	9.33	10.5	7.43	6.01	6.41
DM (%)	7.89	10.8	3.66	8.13	28.8	34.7	34.7	32.8	34.7
Bias (%)	−0.61	0.57	−0.03	1.87	−0.41	2.22	1.23	0.18	0.60

those presented in Table V for which AAD = 8.96% (27 parameters) and AAD = 7.30% (15 parameters).

CONCLUSION

A scheme based on a consideration of the hard sphere theory of transport properties (viscosity, self-diffusion and thermal conductivity) on a wide range of temperature and pressure has been used to correlate dynamic viscosity of two ternary mixtures. One with weak intermolecular interactions, and one with strong attractive intermolecular interactions between the components involved. The results obtained show a very good agreement between the calculated and experimental data. It has been shown that the ternary mixtures heptane + methylcyclohexane + 1-methylnaphtalene (weakly interactive), and water + 2-propanol + DAA (strongly associative) can be correlated with AAD = 3.87% (810 experimental data, 12 parameters) and AAD = 2.16% (1188 experimental data, 21 parameters) respectively. This work shows that the scheme which has been used on pure substances and on some binary mixtures can be extended to much more complex systems such as the one studied here. In addition, it is important to emphasise the fact that the coefficients a_{η_k} of Eq. 1 seem to be universal (the only parameters characteristic of the components being V_{0_i} and R_{η_i}).

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