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Viscosities of Binary and Ternary Mixtures of Hexadecanoic, Octadecanoic, and *cis*-9-Octadecenoic Acids

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The kinematic viscosity was measured for binary and ternary liquid mixtures for the hexadecanoic, octadecanoic, and *cis*-9-octadecenoic acids, at temperatures ranging from above the solidification point up to 373 K using a Cannon Fenske three-capillary viscosimeter. The measurements were correlated with Andrade's equation for pure components, with McAllister's equation for binary and ternary mixtures, and with Teja and Rice's equation for ternary mixtures. A nearly lineal behavior of the kinematic viscosity with regard to composition was observed in the above-mentioned equations for all the tested mixtures. The equations gave a satisfactory fit to the kinematic viscosity of the liquid mixtures studied. For ternary mixtures, Teja and Rice's equation permitted their estimation over a higher temperature range than McAllister's equation.

Introduction

In the refining of edible oils, gas distillates are obtained that are of great interest to the food and pharmaceutical industries. The majority of the components of the distillates are fatty acids (Ghosh and Bhattacharyya, 1996; Ruiz et al., 1995a). The fatty acids that are present in the raw oils and in the soya, sunflower, and olive refined oils are essentially hexadecanoic, octadecanoic, *cis*-9-octadecenoic, and 9,12-octadecadienoic acids (Ruiz, 1993b).

In Gracianni et al. (1994), a procedure is currently being tested to recover these compounds in tubes and shell exchangers. As heat- and mass-transfer phenomena are involved in the design of these heat exchangers, certain information concerning the kinematic viscosities of each compound and its mixtures is required.

This study presents the results of the measurements of the kinematic viscosity for binary and ternary mixtures of hexadecanoic (palmitic), octadecanoic (stearic), and *cis*-9-octadecenoic (oleic) fatty acids. The measurements were carried out every 5 K between temperatures close to solidification of each mixture and 373 K.

The binary mixtures (palmitic + stearic), (palmitic + oleic), and (oleic + stearic) and the ternary mixture (palmitic + stearic + oleic) were studied. In this last mixture, the presence of oleic acid was increased within the range of compositions so as to reproduce those obtained in raw and deodorized oils, to ascertain the effect of the viscosity as a function of the composition of the mixture.

The experimental values of kinematic viscosity for the pure substances obtained in a previous study (Cedeño et al., 1999) were fitted to Andrade's equation. The experimental data for the binary mixtures were fitted to McAllister's equation, and for the experimental data for the ternary mixtures were fitted to McAllister's and Teja and Rice's equations.

Experimental Section

Measurements of kinematic viscosity were carried out for the binary mixtures (palmitic + stearic), (palmitic + oleic), and (oleic + stearic) and the ternary mixture (palmitic + stearic + oleic). The mixtures were prepared in an analytical balance Mettler AE 240, accurate to 0.1 mg. The mass ratios used for the first and second components for the binary mixtures were respectively (1:1), (1:3), and (3:1). (1:1) means 50% of the mass fraction comes from the first component and 50% of the mass fraction comes from the second, (1:3) means 25% of the mass fraction comes from the first component and 75% of the mass fraction comes from the second, and (3:1) means 75% of the mass fraction comes from the first component and 25% of the mass fraction comes from the second. The first component corresponded to the one with the lower molecular mass. Under the supposition that the compositions of the fatty acids in raw and deodorized oils are practically the same, we employed the same values for the compositions of the ternary mixtures. The mass ratios used for the first, second, and third components were respectively (1:1:1), (3:1:1), (3:1:3), (3:1:6), (3:1:12), (3:1:18), (3:1:24), and (3:1:30). The pure free fatty acids were provided by Merck, their purities for palmitic, stearic, and oleic fatty acids being 99.3 mass %, 97 mass %, and 99.9 mass %, respectively. All the measurements of kinematic viscosity were carried out according to ASTM test methods D 445 and D 2515 using two Canon-Fenske-type Ostwald capillary viscosimeters for transparent liquids, supplied by PROTON (Spain), of sizes 75, 150, and 200. These viscosimeters were used because of the type of fatty acid and the range of working temperature. The kinematic viscosity was determined by the expression

$$\nu = c\tau \qquad (1)$$

where c is the calibration constant in $(\mathrm{mm^2 \cdot s^{-1}}) \cdot \mathrm{s^{-1}}$ for the two viscosimeters used, given by the manufacturer for

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Table 1. Kinematic Viscosity Measurements of Pure Components

	$10^6\nu/\mathrm{m^2 \cdot s^{-1}}$		
T/K	palmitic	stearic	oleic
298			32.948
303			27.343
308			22.990
313			19.588
318			16.778
323			14.455
328			12.537
333			11.017
338	9.956		9.735
343	8.867		8.598
348	7.856	10.127	7.560
353	7.039	8.989	6.774
358	6.322	8.022	5.998
363	5.703	7.188	5.419
368	5.158	6.485	4.926
373	4.694	5.868	4.499

Table 2. Kinematic Viscosity Measurements of Binary Mixtures

	$10^6\nu_m/\mathrm{m^2 \cdot s^{-1}}$					
T/K	x_1[a]	palmitic + stearic	x_1	oleic + stearic	x_1	palmitic + oleic
333					0.2686	11.514
338	0.2700	12.302	0.2513	12.358	0.2686	10.101
343	0.2700	10.751	0.2513	10.845	0.2686	8.817
348	0.2700	9.538	0.2513	9.617	0.2686	7.834
353	0.2700	8.470	0.2513	8.424	0.2686	7.000
358	0.2700	7.558	0.2513	7.541	0.2686	6.286
363	0.2700	6.860	0.2513	6.767	0.2686	5.655
368	0.2700	6.118	0.2513	6.091	0.2686	5.123
373	0.2700	5.608	0.2513	5.528	0.2686	4.645
323					0.5241	15.294
328					0.5241	13.253
333	0.5259	12.884	0.5018	13.416	0.5241	11.554
338	0.5259	11.269	0.5018	11.709	0.5241	10.139
343	0.5259	9.922	0.5018	10.287	0.5241	8.949
348	0.5259	8.801	0.5018	9.105	0.5241	7.923
353	0.5259	7.774	0.5018	8.040	0.5241	7.077
358	0.5259	7.006	0.5018	7.188	0.5241	6.356
363	0.5259	6.259	0.5018	6.440	0.5241	5.714
368	0.5259	5.703	0.5018	5.818	0.5241	5.179
373	0.5259	5.180	0.5018	5.263	0.5241	4.702
333	0.7690	12.303				
338	0.7690	10.767	0.7255	10.464	0.7430	10.927
343	0.7690	9.528	0.7255	9.247	0.7430	8.793
348	0.7690	8.475	0.7255	8.166	0.7430	7.848
353	0.7690	7.576	0.7255	7.301	0.7430	7.011
358	0.7690	6.780	0.7255	6.540	0.7430	6.308
363	0.7690	6.071	0.7255	5.901	0.7430	5.695
368	0.7690	5.509	0.7255	5.341	0.7430	5.089

[a] x_1: mole fraction of the component with lower molecular mass of the binary mixtures (first component).

310.93 K, 323.15 K, and 372.04 K, and τ is the flow average time of the three times measured, with an accuracy of 0.01 s.

A thermostat model Summer & Runge D 1000 (Germany), with a temperature range of 283 to 373 K and a temperature accuracy of ±0.1 K, was also employed, the thermometers used having subdivisions of 0.1 K.

Results and Discussion

Applications for Andrade's equation for fatty acids appear in Noureddini and Clements (1992b) and Valeri and Meirelles (1997). The equation is as follows:

$$\ln \nu = A + \frac{B}{T} \qquad (2)$$

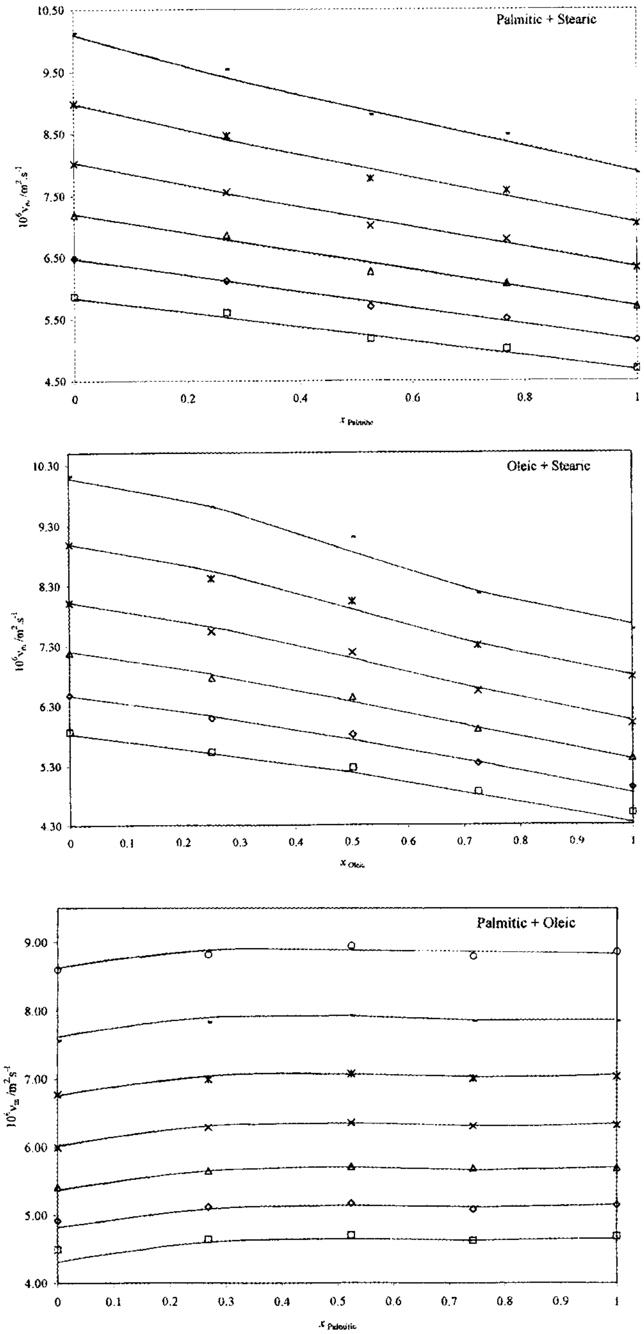

Figure 1. Estimated kinematic viscosity and the measurements for the binary mixtures as a function of the mole fraction, from McAllister's equation. Solid lines are correlations, and the points are experimental data. (□) 373 K; (◇) 368 K; (△) 363 K; (×) 358 K; (∗) 353 K; (-) 348 K; (○) 343 K.

where ν is the kinematic viscosity, A and B are the constants obtained by fitting to the experimental data, and T is the thermodynamic temperature.

McAllister's method for the estimation of the kinematic viscosities of binary mixtures of fatty acids is reported in Valeri and Meirelles (1997), Irving (1977), and Barata and Serrano (1994). The resulting equation has the following form:

$$\ln \nu_m = x_1^3 \ln \nu_1 + (1 - x_1)^3 \ln \nu_2 + 3x_1^2(1 - x_1) \ln \nu_{12} + 3x_1(1 - x_1)^2 \ln \nu_{21} + r^\circ \quad (3)$$

with

Table 3. Kinematic Viscosity Measurements of Ternary Mixtures

			$10^6\nu_m$/m^2·s^{-1}														
mixture[a]	x_1[b]	x_2[b]	303 K	308 K	313 K	318 K	323 K	328 K	333 K	338 K	343 K	348 K	353 K	358 K	363 K	368 K	373 K
1:1:1	0.3560	0.3209					16.669	14.411	12.552	11.002	9.740	8.604	7.659	6.888	6.181	5.583	5.096
3:1:1	0.6238	0.1874						13.771	12.063	10.502	9.288	8.232	7.348	6.590	5.948	5.373	4.881
3:1:3	0.4528	0.1361					15.459	13.340	11.631	10.209	9.018	8.013	7.162	6.424	5.782	5.242	4.756
3:1:6	0.3209	0.0964				17.917	15.419	13.358	11.675	10.297	9.082	8.026	7.161	6.433	5.835	5.258	4.796
3:1:12	0.2027	0.0609		25.023	21.018	17.887	15.326	13.379	11.642	10.277	9.180	8.213	7.126	6.374	5.753	5.251	4.709
3:1:18	0.1482	0.0445		24.052	20.286	17.434	15.015	13.028	11.440	10.042	8.866	7.946	7.101	6.377	5.735	5.196	4.734
3:1:24	0.1168	0.0351	28.035	23.511	19.830	17.049	14.686	12.780	11.161	9.844	8.721	7.765	6.992	6.248	5.637	5.136	4.661
3:1:30	0.0963	0.0290	28.135	23.630	20.021	17.142	14.812	12.899	11.269	9.946	8.815	7.860	7.019	6.313	5.709	5.181	4.715

[a] Mixture: mass ratios used in the ternary mixtures. [b] x_1, x_2: mole fraction for the palmitic and stearic fatty acids in the ternary mixtures.

Table 4. Constants *A* and *B* Obtained for Andrade's Equation

pure substances	A	B	r/%	$10^6\sigma$/m^2·s^{-1}	T/K
palmitic	−19.558	2718.3	0.3	0.02	338−373
stearic	−19.647	2835.0	0.2	0.02	348−373
oleic	−20.241	2945.4	1.5	0.42	303−373

$$r° = -\ln\left[x_1 + (1 - x_1)\frac{M_2}{M_1}\right] + 3{x_1}^2(1 - x_1)\ln\left(\frac{2}{3} + \frac{M_2}{3M_1}\right) + 3{x_1}^2(1 - x_1)^2\ln\left(\frac{1}{3} + \frac{2M_2}{3M_1}\right) + (1 - x_1)^3\ln\left(\frac{M_2}{M_1}\right) \quad (4)$$

where ν_m, ν_1, and ν_2 are the kinematic viscosities of the mixture and the pure components, respectively, ν_{12} and ν_{21} are the pseudokinematic viscosities of McAllister's method, M is the molecular mass, and x_i is the mole fraction of component i. The term $r°$ is approximately zero when the molecular mass ratio is approximately unity.

The temperature dependence of the two pseudokinematic viscosities ν_{12} and ν_{21} is also given by Andrade's equation:

$$\ln \nu_{ij} = A_{ij} + \frac{B_{ij}}{T} \quad (5)$$

For each binary system, the Andrade parameters of eq 5, A_{ij} and B_{ij} (i and j = 1, 2; $i \neq j$), are obtained using the method of least-squares, from measurements of kinematic viscosity at several temperatures and compositions and eq 3.

McAllister's equation can be applied for the ternary mixture (Serrano et al., 1990). The equation has the following form:

$$\ln \nu_m = {x_1}^3 \ln \nu_1 + {x_2}^3 \ln \nu_2 + {x_3}^3 \ln \nu_3 + 3{x_1}^2x_2 \ln \nu_{12} + 3{x_1}^2x_3 \ln \nu_{13} + 3{x_2}^2x_1 \ln \nu_{21} + 3{x_2}^2x_3 \ln \nu_{23} + 3{x_3}^2x_1 \ln \nu_{31} + 3{x_3}^2x_2 \ln \nu_{32} + 6x_1x_2x_3 \ln \nu_{123} + r° \quad (6)$$

To determine the pseudokinematic viscosity of the ternary interaction ν_{123}, the procedure explained according to eq 5 was also applied, and $r°$ was set to zero for the same reason as that for binary mixtures.

Teja and Rice's method for estimating the kinematic viscosities of liquid mixtures, which is based on the model of General Corresponding States (GCS), is reported by Reid et al. (1987) and Teja and Rice (1981). The equation has the following form:

$$\ln(\nu\xi) = \ln(\nu\xi)^{r_1} + \frac{\omega - \omega^{r_1}}{\omega^{r_1} - \omega^{r_1}}[\ln(\nu\xi)^{r_2} - \ln(\nu\xi)^{r_1}] \quad (7)$$

where ν is the kinematic viscosity of the mixture or of the reference substances 1 and 2, ω is the acentric factor of the mixture or of the reference substances 1 and 2, and ξ is a parameter, defined as $\xi = V_c^{2/3}/(T_cM)^{1/2}$ for the mixture or for the reference substances 1 and 2. V_c and T_c are the critical mole volume and temperature for the mixture or for the reference substances 1 and 2. The numbers 1 and 2 refer to the two fluids that need to be defined and that have known temperature-dependent properties.

The variables related to the composition of the mixture are defined by the terms V_{cm}, T_{cm}, M_m, and ω_m. The rules suggested by several authors for these parameters of the mixture are

Table 5. Parameters Obtained for McAllister's Equation

binary mixtures	A_{12}	B_{12}	A_{21}	B_{21}	r/%	$10^6\sigma$/m^2·s^{-1}	T/K
palmitic + stearic	−19.6040	2766.3045	−19.3798	2709.7740	0.8	0.07	343−373
oleic + stearic	−18.6589	2413.3122	−20.4441	3102.6215	0.9	0.01	348−373
palmitic + oleic	−19.8169	2801.6949	−19.3978	2677.2858	0.6	0.05	338−373

Table 6. Parameters Obtained for McAllister's Equation

ternary mixture	A_{123}	B_{123}	r/%	$10^6\sigma$/m^2·s^{-1}	T/K
palmitic + stearic + oleic	−18.1505	2227.2146	1.1	0.09	348−373

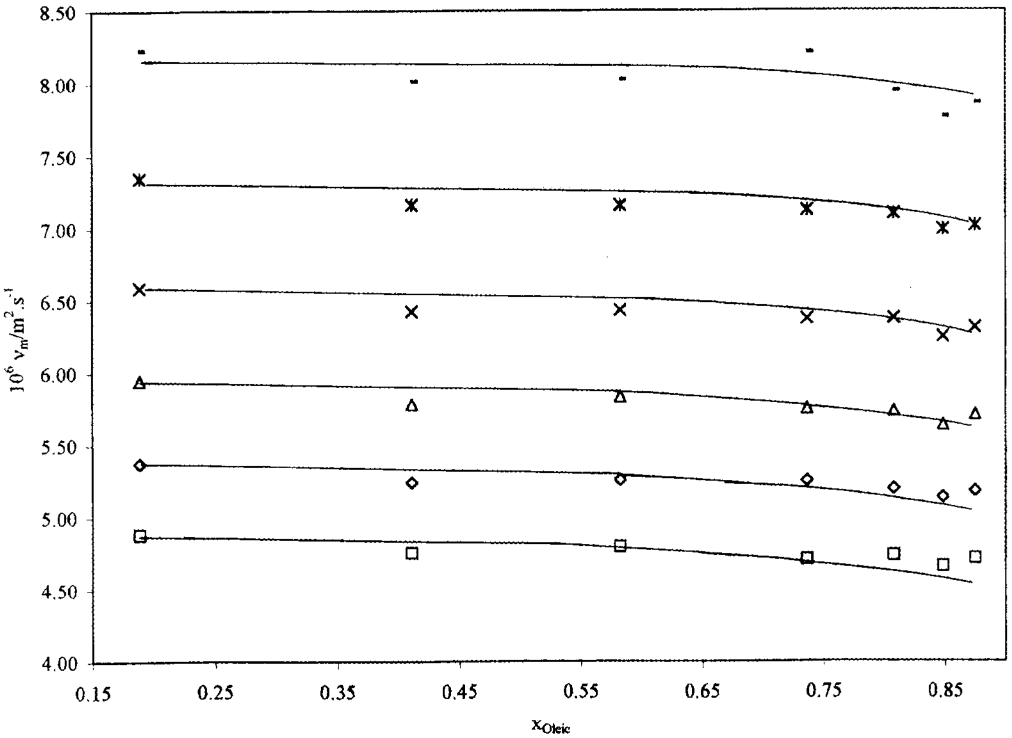

Figure 2. Estimated kinematic viscosity and the measurements for the ternary mixture from McAllister's equation when the oleic acid was increased with mass ratios from 3:1:1 to 3:1:30. Solid lines are correlations, and the points are experimental data. (□) 373 K; (◇) 368 K; (△) 363 K; (×) 358 K; (∗) 353 K; (-) 348 K.

Table 7. Constants *A* and *B* Obtained for Andrade's Equation

fatty acid	*A*	*B*	*r*/%	$10^6\sigma/m^2\cdot s^{-1}$	*T*/K
erucic	−19.750	2956.6	0.2	0.62	310−383

$$V_{cm} = \sum_i \sum_j x_i x_j V_{cij} \tag{8}$$

$$V_{c_{ij}} = \frac{(V_{ci}{}^{1/3} + V_{cj}{}^{1/3})^3}{8} \tag{9}$$

$$T_{cm} = \frac{\sum_i \sum_j x_i x_j T_{cij} V_{cij}}{V_{cm}} \tag{10}$$

$$T_{cij} V_{cij} = \varphi_{ij} (T_{ci} T_{cj} V_{ci} V_{cj})^{1/2} \tag{11}$$

$$M_m = \sum_i x_i M_i \tag{12}$$

$$\omega_m = \sum_i x_i \omega_i \tag{13}$$

In eq 11, φ_{ij} is an interaction parameter close to unity, which is determined by the kinematic viscosity experimental data.

To estimate the kinematic viscosity of the mixture at a certain temperature using eq 7, knowledge of the kinematic viscosity values of the reference substances is required. These values are determined according to Andrade's equation, using the reference temperature $T(T_c^{r_1}/T_{cm})$ for reference substance 1 and $T(T_c^{r_2}/T_{cm})$ for reference substance 2. The choice of the reference substances is preferably carried out from among the components of the mixture, although they may also be arbitrary (Teja and Rice, 1981).

The values of kinematic viscosity estimated for each equation are compared using the average deviation *r* and the standard deviation σ, by means of the expressions

$$r = \left[\frac{\sum(\nu - \nu_{est}|/\nu)}{n}\right]100 \tag{14}$$

$$\sigma = \left[\frac{\sum(\nu - \nu_{est})^2}{(n - p)}\right]^{1/2} \tag{15}$$

where ν and ν_{est} are the kinematic viscosity measurement and the estimated one, *n* is the number of experimental points, and *p* is the number of adjustment parameters.

Starting from the measurement, the values of the kinematic viscosity for pure components are presented in Table 1; these were obtained from the dynamic viscosity μ and density ρ measurements reported by Cedeño et al. (1999). The results of the measurements for binary and ternary mixtures are presented in Tables 2 and 3. In previous works, kinematic viscosities for the palmitic, stearic, and oleic acids and some binary mixtures were measured with a similar experimental technique. Although in the present work the measurements obtained by other authors are not compared, the results obtained were similar to the values published by Noureddini et al. (1992b) and Valeri and Meirelles (1997) (palmitic, stearic, and oleic acids and (palmitic + stearic) and (oleic + stearic) binary mixtures), the minimum average deviation being $r \cong 0.6\%$ for the case of stearic acid and the maximum average deviation being $r \cong 4.8\%$ for the case of the binary mixture (oleic + stearic).

The constants *A* and *B* of eq 2 for the palmitic, stearic, and oleic acids are presented in Table 4. The average deviation values *r* are <0.3%, except for oleic acid. The highest standard deviation value σ corresponds to oleic acid, being $0.42 \times 10^{-6}\ m^2\cdot s^{-1}$.

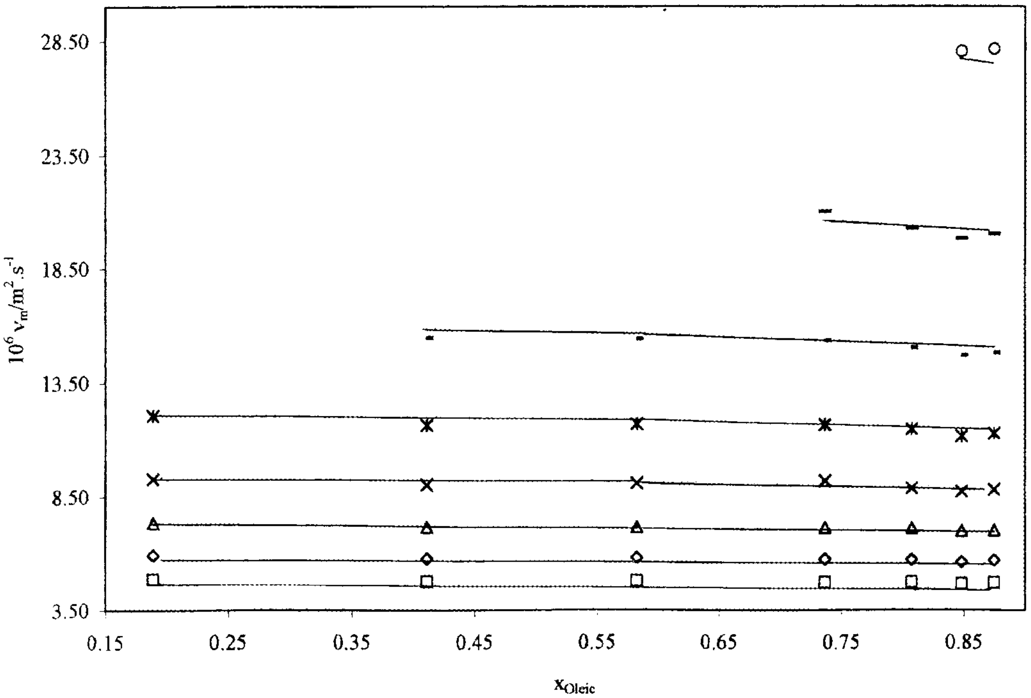

Figure 3. Estimated kinematic viscosity and the measurements for the ternary mixture from Teja and Rice's equation when the oleic fatty acid was increased with mass ratios from 3:1:1 to 3:1:30. Solid lines are correlations, and the points are experimental data. (□) 373 K; (◇) 363 K; (△) 353 K; (×) 343 K; (∗) 333 K; (-) 323 K; (−) 313 K; (○) 303 K.

Table 8. Interaction Parameter φ_{ij} Obtained for Teja and Rice's Equation

	$\varphi_{\text{palmitic-stearic}}$	$\varphi_{\text{palmitic-oleic}}$	$\varphi_{\text{stearic-oleic}}$	r/%	$10^6\sigma/m^2{\cdot}s^{-1}$	T/K
palmitic + stearic + oleic	1.0916	1.0387	0.9771	2.0	0.23	303−308

The constants A_{ij} and B_{ij} of eq 3 for the binary mixtures are presented in Table 5. Once the constants are substituted in the equation, r does not exceed 1% in any case. The estimated kinematic viscosity and the measurements for the binary mixtures as a function of the mole fraction are presented in Figure 1. A nearly lineal behavior of the kinematic viscosity is observed for the three binary mixtures, although small deviations are observed for the mixtures (palmitic + stearic) and (oleic + stearic). The values of the kinematic viscosity remain nearly constant with the mole fraction for mixtures (palmitic + oleic). The values of $r°$ were found to be around 1.0×10^{-3} in all the cases. The use of McAllister's equation is limited when the temperature is higher than the melting point of any component of the mixture. However, the mixture may be liquid at a lower temperature.

The interaction parameters A_{123} and B_{123} for the ternary mixtures (eq 6) are presented in Table 6. In the determination of these coefficients, the term $r°$ was given the value zero. The estimated kinematic viscosity and the measurements for the ternary mixtures as a function of the oleic mole fraction are presented in Figure 2 for the mass ratios (3:1:1), (3:1:3), (3:1:6), (3:1:12), (3:1:18), (3:1:24), and (3:1:30). A nearly lineal behavior is also observed when the oleic mole fraction increases. For the same reason as for the binary mixtures, the estimation of the viscosity by McAllister's equation is limited and cannot be applied when the temperature is lower than those of the solidification of the pure components.

One alternative to McAllister's equation is Teja and Rice's equation (eq 7), but reference substances must be selected. Oleic acid and erucic acid were chosen as reference substances. Measurements of density and dynamic viscosity for erucic acid are reported by Noureddini et al. (1992a,b). The values of the constants A and B for erucic acid, which were obtained according to eq 2, are presented in Table 7. The interaction parameters φ_{ij} for the ternary mixtures are presented in Table 8.

The estimated kinematic viscosity (eq 7) and the measurements for the ternary mixtures as a function of the oleic mole fraction are presented in Figure 3 for the mass ratios (3:1:1), (3:1:3), (3:1:6), (3:1:12), (3:1:18), (3:1:24), and (3:1:30). As in the application of McAllister's equation, a nearly lineal behavior can be observed, and furthermore, Teja and Rice's equation allows a wider temperature range than McAllister's equation.

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