Viscosities of Fatty Acids, Triglycerides, and Their Binary Mixtures

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ABSTRACT: Viscosity data have been obtained as a function of temperature for seven fatty acids (pelargonic, capric, lauric, myristic, palmitic, stearic, and oleic) and four triglycerides (tricaprilin, tripalmitin, tristearin, and triolein) and their binary mixtures at temperatures from above their melting points to 90°C. The viscosity measurements were performed by using Cannon Fenske glass capillary kinematic viscometers. Modified versions of the Andrade equation were used to correlate the kinematic viscosities of pure fatty acids and pure triglycerides. The MacAllister method was used for their binary mixtures. The correlation constants are valuable for designing or evaluating chemical process equipment, such as heat exchangers, reactors, distillation columns, and process piping.

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KEY WORDS: Fatty acid mixtures, fatty acids, kinematic viscosity, triglyceride mixtures, triglycerides, viscosity.

Deacidification of edible oils can be accomplished by steam refining, caustic refining, or by liquid–liquid extraction (1,2). To get a more quantitative insight into the various factors that determine the efficiency of refining processes, the heat and mass transfer mechanism should be analyzed. For this purpose, knowledge of triglyceride viscosities is fairly important. Viscosities of fatty acids and their mixtures are important in the design of process equipment for the oil and fatty acid industry. For example, viscosity is an essential parameter in estimating the efficiency of distillation columns for separation of fatty acids (3). Viscosity data are also required for the design of heat transfer equipment, process piping, reactors, stripping columns, deodorizers, liquid extractors, and other units for the oil industry.

EXPERIMENTAL PROCEDURES

Viscosity data were obtained for a number of fatty acids (pelargonic, $C_{9:0}$; capric, $C_{10:0}$; lauric, $C_{12:0}$; myristic, $C_{14:0}$; palmitic, $C_{16:0}$; stearic, $C_{18:0}$; and oleic, $C_{18:1}$) and their binary mixtures ($C_{9:0}/C_{10:0}$, $C_{10:0}/C_{12:0}$, $C_{12:0}/C_{14:0}$, $C_{14:0}/C_{16:0}$, $C_{16:0}/C_{18:0}$, and $C_{18:1}/C_{18:0}$), and for triglycerides (tricaprilin, tripalmitin, tristearin, and triolein) and their binary mixtures (tricaprilin/tripalmitin, tricaprilin/tristearin, tripalmitin/tristearin, and triolein/tristearin). All chemicals used were obtained from Sigma (St. Louis, MO). The acids had high purity (99–100%), except for $C_{9:0}$ and $C_{18:1}$, which were 90 and 95% pure, respectively. The triglycerides had the following purities: 97–98 (tricaprilin), 90 (tripalmitin and tristearin), and 99% (triolein). The solutions were prepared by mass, accurate to ±0.1 mg, by using an analytical balance (Sartorius, Goettingen, Germany). They were weighed in three different mass ratios, approximately 1:1, 1:3, and 3:1 of the first and second compounds, respectively. Viscosities were obtained at temperatures above the melting point of each substance up to 90°C. They were measured with glass capillary kinematic viscometers from Cannon Fenske (Cannon Instrument Co., Pittsburgh, PA, and Cole Parmer Instrument Co., Niles, IL) in a constant-temperature bath (Cole Parmer Instrument Co.). This assembly maintains a temperature uniformity of ±0.05°C. Thermometers (Cole Parmer Instrument Co.) with subdivisions of 0.1°C were used for monitoring bath temperature. Variations in viscosity as a function of temperature and composition were measured. Data were correlated by using the Statistical Analysis System (SAS, Cary, NC) package.

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RESULTS AND DISCUSSION

Kinematic viscosities ν, expressed in centistokes, were calculated from the measured flow time θ and instrument constant c by using the following equation:

$$\nu = c\theta \qquad [1]$$

The values for c are provided by the viscometer manufacturer. The viscometer constants were corrected for effects of temperature. Final results for the kinematic viscosities of fatty acids and triglycerides are tabulated in Tables 1 and 2, respectively. For the fatty acid mixtures, the results are presented in Tables 3 and 4, and in Table 5 are tabulated the kinematic viscosities of the triglyceride mixtures .

Viscosity-temperature correlations. For the pure substances, the Andrade correlation (Equation 2) and some of its modified versions (Equation 3, in general form) were used to correlate the data for viscosities as a function of temperature (4,5):

$$\ln \nu = A + B/T \qquad [2]$$

TABLE 1
Kinematic Viscosity of Fatty Acids

Temperature (°C)	Viscosity (cSt)						
	$C_{9:0}$	$C_{10:0}$	$C_{12:0}$	$C_{14:0}$	$C_{16:0}$	$C_{18:0}$	$C_{18:1}$
20	9.0158						
30	6.7089						26.8958
40	5.9730	6.9018					19.6931
50	4.1463	5.2488	8.0213				14.8302
60	3.3802	4.2557	6.3157	8.5344			10.1226
70	2.7299	3.5086	5.1075	6.7729	9.0718		8.0418
75					8.3829	9.7008	
80	2.3223	2.7332	4.1202	5.4988	7.4113	8.6771	6.5485
85					6.7140	7.8771	
90	1.9957	2.3317	3.5288	4.5312	6.0323	7.0287	5.4307
Literature value[a]	3.47	4.28	6.28	8.71	8.99	8.72	6.58
Reference temperature[a]	60	60	60	60	71.1	82.2	82.2

[a]Data from References 5 and 6.

$$\ln \nu = A + B/(T + C) + DT + E/T^2 \quad [3]$$

where A, B, C, D, and E are parameters which should be fitted to the experimental data. In both equations, temperature T is in K.

The best results of the regression analysis for kinematic viscosities of fatty acids and triglycerides are tabulated in Tables 6 and 7, respectively. Figure 1 shows the experimental and predicted viscosities from the correlations for pure fatty acids, and Figure 2 shows these results for triglycerides. In these figures, lines represent the best fitted correlations compared to the experimental points. For all fatty acid data, the mean deviation r, calculated according to Equation 4, is less than 2.51%, except for $C_{9:0}$, which has a mean deviation of 3.16%. The higher standard deviation σ is 0.5835 cSt, calculated according to Equation 5. For triglycerides, the mean deviation r is less than 0.8570%. The higher standard deviation σ is 0.2334 cSt.

$$r = \left[\left(\Sigma \left|\nu_{exp} - \nu_{cal}\right| / \nu_{exp}\right)/n\right]100 \quad [4]$$

$$\sigma = \left[\Sigma\left(\nu_{exp} - \nu_{cal}\right)^2/(n - p)\right]^{1/2} \quad [5]$$

where ν_{exp} and ν_{calc} are the experimental and calculated values for the kinematic viscosities, respectively, n is the number of experimental points, and p is the number of adjusted parameters.

Viscosity indices. Because triglycerides have excellent lubricity, vegetable oil derivatives can be used as components of lubricant oils. Basu *et al.* (7) studied the potential of several glycol esters of fatty acids as lubricants. The viscosity index is a measure that is widely used to indicate the temperature dependence of the kinematic viscosity for lubricating oils. This index is defined by ASTM Standard D2270-93 (8) for oils that exhibit a kinematic viscosity greater than or equal to 2.0 cSt at 100°C. Viscosity indices for oleic acid, tri-

TABLE 2
Kinematic Viscosity of Triglycerides

Temperature (°C)	Viscosity (cSt)			
	Tricaprilin	Tripalmitin	Tristearin	Triolein
20				86.6958
30	17.3424			56.6127
40	12.3109			38.6284
50	9.1460			27.6632
60	7.1029			20.8588
70	5.6760	17.3069		15.3199
75		15.4161	18.7471	
80	4.7348	13.1755	16.3086	12.2278
85		11.6050	14.4477	
90	3.7795	10.3734	12.9165	9.9367

TABLE 3
Kinematic Viscosities of Fatty Acid Mixtures[a]

Temperature (°C)	$C_{9:0}/C_{10:0}$		$C_{10:0}/C_{12:0}$		$C_{12:0}/C_{14:0}$	
	x_1	ν (cSt)	x_1	ν (cSt)	x_1	ν (cSt)
40	0.2653	6.6844				
50	0.2653	5.2900	0.2823	7.2843		
60	0.2653	4.2677	0.2823	5.7620	0.2730	8.2267
70	0.2653	3.5175	0.2823	4.6610	0.2730	6.5152
80	0.2653	2.8791	0.2823	3.8540	0.2730	5.3903
90	0.2653	2.4554	0.2823	3.1057	0.2730	4.4050
40	0.5158	6.3415				
50	0.5158	5.0429	0.5520	6.6136		
60	0.5158	4.1504	0.5520	5.2787	0.5308	7.5788
70	0.5158	3.3931	0.5520	4.3178	0.5308	6.0331
80	0.5158	2.7163	0.5520	3.6102	0.5308	4.9317
90	0.5158	2.3257	0.5520	2.9577	0.5308	4.1255
40	0.7602	5.9834				
50	0.7602	4.7469	0.7789	5.9881		
60	0.7602	3.8827	0.7789	4.7914	0.7742	6.9701
70	0.7602	3.2205	0.7789	3.9253	0.7742	5.5782
80	0.7602	2.6187	0.7789	3.2371	0.7742	4.5891
90	0.7602	2.2375	0.7789	2.7454	0.7742	3.8235

[a]The variable x_1 represents the mole fraction of the component with the lower molecular mass.

TABLE 4
Kinematic Viscosities of Fatty Acid Mixtures[a]

Temperature	$C_{14:0}/C_{16:0}$		$C_{16:0}/C_{18:0}$		$C_{18:1}/C_{18:0}$	
(°C)	x_1	ν (cSt)	x_1	ν (cSt)	x_1	ν (cSt)
70	0.2732	8.4036				
75			0.2715	9.7194	0.2455	9.8888
80	0.2732	6.6319	0.2715	8.6018	0.2455	8.7067
85			0.2715	7.7339	0.2455	7.8148
90	0.2732	5.4247	0.2715	6.9813	0.2455	7.0352
70	0.5277	7.9055				
75			0.5270	9.3893	0.5044	9.0949
80	0.5277	6.4139	0.5270	8.3745	0.5044	8.1687
85			0.5270	7.5352	0.5044	7.3439
90	0.5277	5.2754	0.5270	6.8412	0.5044	6.6754
70	0.7705	7.5926				
75			0.7685	8.8609	0.7519	8.6365
80	0.7705	6.3927	0.7685	7.9138	0.7519	7.8252
85			0.7685	7.1343	0.7519	7.0993
90	0.7705	5.3005	0.7685	6.4445	0.7519	6.3596

[a]The variable x_1 represents the mole fraction of the component with the lower molecular mass.

caprilin, and triolein were calculated from their kinematic viscosities at 40 and 100°C by following the procedure specified in the ASTM standard. The viscosities at 100°C were obtained by extrapolation by using Equation 3 and parameters given in Tables 6 and 7. The viscosity indices are tabulated in Table 8. Their values indicate that the viscosities of oleic acid, tricaprilin, and triolein exhibit a higher temperature dependence than the viscosities reported by Basu *et al.* (7) for glycol diesters of fatty acids.

Binary mixtures. The binary kinematic viscosity data were correlated by using the cubic equation of MacAllister (9):

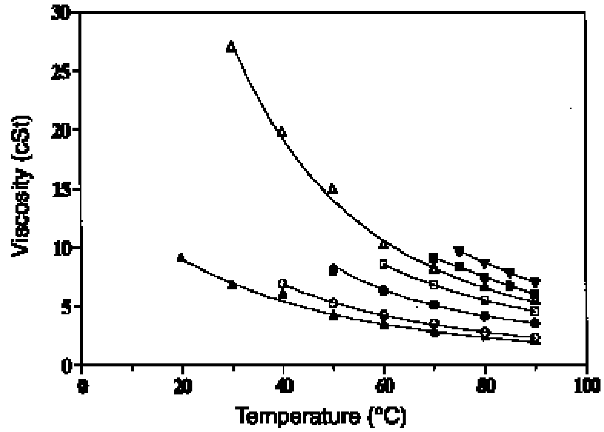

FIG. 1. Viscosity vs. temperature for fatty acids. The lines are correlation derivatives, and the points are experimental data. (▲), Pelargonic acid; (○), capric acid; (●), lauric acid; (□), myristic acid; (△), oleic acid; (■), palmitic acid; (▼), stearic acid.

$$\ln \nu_m = x_1^3 \ln \nu_1 + 3x_1^2(1 - x_1)\ln \nu_{12} + 3x_1(1 - x_1)^2 \ln \nu_{21} + (1 - x_1)^3 \ln \nu_2 + r^0 \quad [6]$$

where r^0 is defined as:

$$r^0 = -\ln [x_1 + (1 - x_1)(M_2/M_1) + 3x_1^2(1 - x_1)\ln (2/3 + M_1/3M_2) + 3x_1(1 - x_1)^2 \ln (1/3 + 2/3\,(M_2/M_1))] + (1 - x_1)^3 \ln (M_2/M_1) \quad [7]$$

The mole fraction of the component with the lower molecular mass is x_1, and M_1 and M_2 are the molecular masses of both components. The kinematic viscosities of the mixtures and pure substances are represented, respectively, by ν_m, ν_1, and

TABLE 5
KinematicViscosities of Triglyceride Mixtures[a]

Temperature	Tricaprilin/tripalmitin		Tricaprilin/tristearin		Tripalmitin/tristearin		Triolein/tristearin	
(°C)	x_1	ν (cSt)	x_1	ν (cSt)	x_1	ν (cSt)	x_1	ν (cSt)
70	0.3746	13.4583						
75	0.3746	11.7847	0.3908	13.9865	0.2688	18.4255	0.2509	17.9429
80	0.3746	10.3905	0.3908	12.3157	0.2688	16.1902	0.2509	15.8549
85	0.3746	9.2749	0.3908	10.9739	0.2688	14.2827	0.2509	14.0841
90	0.3746	8.2871	0.3908	9.8135	0.2688	12.7447	0.2509	12.3673
70	0.6247	10.2601						
75	0.6247	9.0904	0.6527	10.0849	0.5246	17.6775	0.5045	16.7529
80	0.6247	8.1441	0.6527	9.1773	0.5246	15.5041	0.5045	14.8047
85	0.6247	7.3108	0.6527	8.0759	0.5246	13.7426	0.5045	13.1119
90	0.6247	6.4489	0.6527	7.3371	0.5246	12.2992	0.5045	11.7559
70	0.8368	7.5221						
75	0.8368	6.8934	0.8209	7.7684	0.7638	16.5723	0.7498	15.0578
80	0.8368	6.1371	0.8209	6.9627	0.7638	14.5748	0.7498	13.3848
85	0.8368	5.5109	0.8209	6.2712	0.7638	12.9343	0.7498	12.0014
90	0.8368	5.0264	0.8209	5.6972	0.7638	11.5051	0.7498	10.8063

[a]The variable x_1 represents the mole fraction of the component with the lower molecular mass.

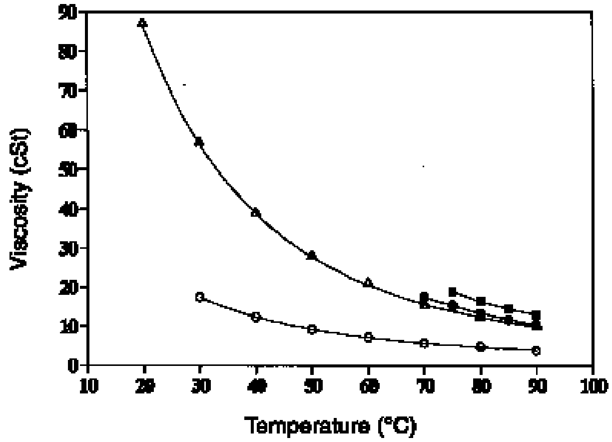

FIG. 2. Viscosity vs. temperature for triglycerides. The lines are correlation derivatives, and the points are experimental data. (○), Tricaprilin; (△), triolein; (▲), tripalmitin; (■), tristearin.

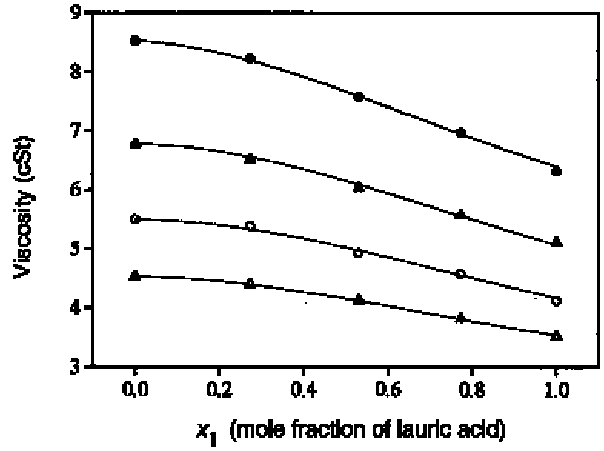

FIG. 3. Viscosities for the mixture lauric acid/myristic acid at various temperatures. The lines are correlation derivatives, and the points are experimental data. (●), 60°C; (▲), 70°C; (○), 80°C; (△), 90°C.

ν_2, while ν_{12} and ν_{21} are the pseudokinematic viscosities of the mixture in the MacAllister method. Pure-liquid kinematic viscosities of fatty acids and triglycerides were calculated by using the correlations presented in Tables 6 and 7. For the two pseudokinematic viscosities of the mixture, ν_{12} and ν_{21}, a temperature dependence of the same form as the Andrade equation was adopted (9):

$$\ln \nu_{ij} = A_{ij} + B_{ij}/T \quad [8]$$

In Equation 8, A_{ij} and B_{ij} are fitted parameters. The results of regression analysis by the MacAllister method are presented in Tables 9 and 11 for fatty acid mixtures and triglyceride mixtures, respectively. The dependence of viscosity on composition and temperature is plotted in Figure 3 for the mixture $C_{12:0}/C_{14:0}$, and in Figure 4 for the mixture tripalmitin/tristearin. The MacAllister equation describes the kinematic viscosities of fatty acid mixtures with a mean deviation below 1.05%. The higher standard deviation is 0.1126 cSt for the mixture $C_{18:1}/C_{18:0}$. For triglyceride mixtures, the mean deviation is less than 0.66%. The higher standard deviation is 0.0895 cSt for the mixture triolein/tristearin.

Dynamic viscosities. The dynamic viscosities can be calculated from kinematic viscosities by means of the following equation:

$$\mu = \rho\nu \quad [9]$$

where μ is the dynamic viscosity expressed in centipoise, and ρ is the density expressed in g/cm^3.

TABLE 6
Correlations for Viscosities of Pure Fatty Acids

Fatty acids	Constants						
	A	*B*	*C*	*D*	*E*	*r* (%)	σ (cSt)
$C_{9:0}$	−5.749326	2329.100067	0	0	0	3.16	0.2586
$C_{10:0}$	−11.404554	3377.368186	0	0.00812	0	1.33	0.0878
$C_{12:0}$	−0.68689935	253.0359629	−233.486574	0	0	1.72	0.0596
$C_{14:0}$	−2.880546	1046.499036	−124.717354	0	0	2.8E-4	0.0074
$C_{16:0}$	−27.841	18476.727	0	0	−2802192.736	0.63	0.0929
$C_{18:0}$	10.3767703	−125.8355809	0	−0.0222529	0	0.32	0.0589
$C_{18:1}$	−22.540759	5635.462033	0	0.023952	0	2.51	0.5835

TABLE 7
Correlations for Viscosities of Pure Triglycerides

Triglyceride	Constants[a]					
	A	*B*	*C*	*E*	*r* (%)	σ (cSt)
Tricaprilin	−1.7800282	581.801325	−177.4168083	0	0.78	0.0718
Tripalmitin	−6.638964	3256.938437	0	0	0.86	0.1724
Tristearin	20.472	−15737.119	0	3352188.033	0.08	0.0274
Triolein	1.5554	−2182.0021	0	888879.2445	0.66	0.2334

[a]Parameter D (Equation 3) was set equal to zero for all triglycerides.

TABLE 8
Viscosity Index for Fatty Compounds

Compound	Viscosity index
Oleic acid	146
Tricaprilin	145
Triolein	191

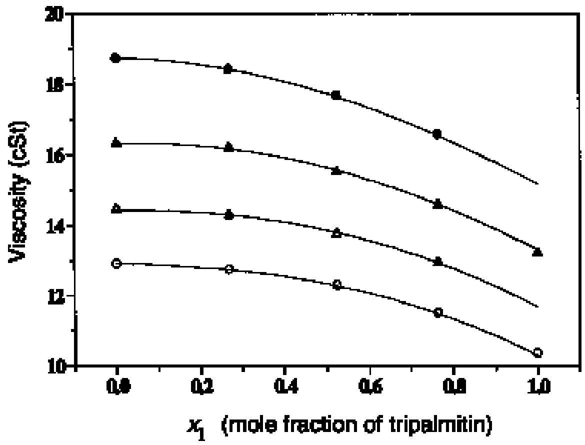

FIG. 4. Viscosities for the mixture tripalmitin/tristearin at various temperatures. The lines are correlation derivatives, and the points are experimental data. (●), 75°C; (▲), 80°C; (△), 85°C; (○), 90°C.

Density data for fatty acids as a function of temperature are available in the literature (6,10,11). Most authors suggest that the densities for fatty acids and triglycerides are linear with temperature (t, °C) by Equation 10:

$$\rho = b + mt \quad [10]$$

Table 10 gives the linear coefficients b and m for saturated fatty acids and for oleic acid as reported by Fisher (10) and by Noureddini *et al.* (6), respectively. For triglycerides, few density data have been published. The coefficients for tricaprilin and triolein, given in Table 11, were obtained by fitting a linear correlation to the experimental data reported by Rodriguez *et al.* (12) and Thomas (13), respectively. For tripalmitin and tristearin, Formo *et al.* (14) reported density values of 0.8663 g/cm^3 and 0.8632 g/cm^3, respectively, both at 80°C. For triglycerides, the change in density per degree Celsius is in the range 0.00067–0.00073 g/cm^3 (14).

For the fatty acid and triglyceride binary mixtures used in this work, there are no published density data available. Some authors have obtained good estimations by assuming ideal behavior for triglyceride mixtures (15) and for mixtures that contain canola oil and weakly polar solvents, such as acetone (16). The densities for binary mixtures can be estimated by the following equation:

$$M_m/\rho_m = x_1(M_1/\rho_1) + x_2(M_2/\rho_2) \quad [11]$$

TABLE 9
Correlations for Viscosities of Fatty Acid Mixtures

Mixture	A_{12}	B_{12}	A_{21}	B_{21}	r (%)	σ (cSt)
$C_{9:0}/C_{10:0}$	−5.174533	2214.839303	−5.164269	2210.377626	1.02	0.0496
$C_{10:0}/C_{12:0}$	−4.362779	2046.120792	−6.035633	2578.184018	0.71	0.0464
$C_{12:0}/C_{14:0}$	−5.273697	2442.203926	−5.365630	2495.440123	0.50	0.0414
$C_{14:0}/C_{16:0}$	−2.575806	1643.447623	−7.583463	3298.14179	1.02	0.0979
$C_{16:0}/C_{18:0}$	−2.430736	1631.684855	−7.632808	3461.462273	0.92	0.1053
$C_{18:1}/C_{18:0}$	−4.142343	2224.013461	−6.758281	3144.997316	1.05	0.1126

TABLE 10
Linear Correlations for the Fatty Acid and Triglyceride Densities (g/cm^3)

Compound	Intercept b	Slope m	Reference
Fatty acids			
$C_{9:0}$	0.92106	−7.9229E-04	10
$C_{10:0}$	0.91660	−7.8237E-04	10
$C_{12:0}$	0.90813	−7.4862E-04	10
$C_{14:0}$	0.90274	−7.2693E-04	10
$C_{16:0}$	0.89809	−7.0831E-04	10
$C_{18:0}$	0.89535	−6.9741E-04	10
$C_{18:1}$	0.90941	−6.9820E-04	6
Triglycerides			
Tricaprilin	0.97002	−7.8786E-04	12
Triolein	0.92880	−8.4000E-04	13

TABLE 11
Correlations for Viscosities of Triglyceride Mixtures

Mixture	A_{12}	B_{12}	A_{21}	B_{21}	r (%)	σ (cSt)
Tricaprilin/tripalmitin	−5.658522	2893.916251	−6.22857	3056.88376	0.51	0.0561
Tricaprilin/tristearin	−5.293304	2877.308473	−5.981152	3023.309139	0.66	0.0862
Tripalmitin/tristearin	−5.202612	2827.837226	−6.251716	3194.682166	0.17	0.0359
Triolein/tristearin	−4.772933	2609.13623	−6.639965	3328.00992	0.41	0.0895

where ρ_m, ρ_1, and ρ_2 are the densities for the mixture and for the pure substances, respectively, while M_m is the average molecular mass for the mixture. By using Equation 11 for the densities, the dynamic viscosities for the binary mixtures can be estimated from Equation 9.

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