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Viscosity of Selected Binary, Ternary, and Quaternary Liquid Mixtures

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Kinematic viscosities of eight binary, five ternary, and one quaternary liquid mixtures are reported for a wide range of temperatures and composition. The data were correlated with the McAllister equation and also with two modified versions of the McAllister equation.

Introduction

The solution to many engineering problems involving heat transfer, mass transfer, and fluid flow requires quantitative information on the viscosities of pure liquids or liquid mixtures.

In the present work, kinematic viscosities of eight binary, five ternary, and one quaternary liquid mixtures containing strongly polar components, including water, are presented. For comparison, the viscosity data were correlated by means of the methods of Soliman (*1*), McAllister (*2*), and Dizechi (*3*).

Experimental Section

Kinematic viscosities were measured with a Schott automated viscosity measuring system. This system consists of a basic control unit including a printer, a measuring stand, a constant-temperature-bath circulator, and calibrated capillary viscometers, depending on the measuring range. The capillary viscometers were calibrated by Schott Glaswerke, Mainz, FRG. The calibration was checked with distilled water and was found to produce measured values that agreed with published data within 0.4%. For instance, the "accepted" value of the viscosity of water at 25 °C is $\nu = 0.8929 \times 10^{-6}$ m^2/s, while the value obtained with the Schott automated viscosity measuring system is $\nu = 0.8950 \times 10^{-6}$ m^2/s. Thus the deviation of the measured value from the standard value is less than 0.3%. For measurements at and below ambient temperature, this system was used in conjunction with a Model RTE-4 Neslab cooling system. Bath temperatures can be set and held constant to about ±0.01 °C. Time measurements were made automatically with the help of a quartz timer with a resolution of 0.01 s and two light barriers across the viscometer. The light barriers detect the passing meniscus of the studied fluid and provide the start-stop signals for the time measurement. In any series of experiments at identical conditions, the deviation of measured values from the "average" value was found to be smaller than 0.5%.

Liquid mixtures were prepared with a Mettler precision digital balance Model PE 400 with an accuracy of $\pm 10^{-2}$ g. The buoyancy effect was neglected, since errors in measured mass fractions introduced by this effect were much smaller than 0.1%. Fluids used in this work were all purified, Spector AR. Evaporation rates were negligible in transferring the mixtures into capillary viscometers.

Results and Discussion

Viscosities of the following mixtures were measured at different temperatures, and the results of these measurements are listed in Table I: 2-propanol–water mixtures at 25 °C, 1-propanol–water mixtures at 25 °C; methanol–water mixtures at 25 °C, ethanol–water mixtures at 25 °C; ethanol–methanol mixtures at 25 °C; 2-propanol–1-propanol mixtures at 10, 20, 30, 40, and 50 °C; 2-propanol–methanol mixtures at 10, 20, 30, 40, and 50 °C; 2-propanol–ethanol mixtures at 10, 20, 30, 40, and 50 °C; 2-propanol–1-propanol–water mixtures at 10, 20, 30, 40, and 50 °C; ethanol–methanol–water mixtures at 25 °C; 2-propanol–ethanol–methanol mixtures at 10, 20, 30, 40, and 50 °C; 2-propanol–ethanol–water mixtures at 10, 20, 30, 40, and 50 °C; 2-propanol–methanol–water mixtures at 10, 20, 30, 40, and 50 °C; and 2-propanol–ethanol–methanol–water mixtures at 10, 20, 25, 30, 40, and 50 °C.

The binary and ternary data were correlated by means of the McAllister equation of viscosity (*2*), which for *n*-component mixtures reads

$$\ln \nu_{\mathrm{m}} = \sum_{i=1}^{n} x_i^3 \ln \nu_i M_i - \ln M_{\mathrm{av}} + 3\sum_{i=1}^{n} \sum_{\substack{j=1 \\ i \neq j}}^{n} x_i^2 x_j \ln \nu_{ij} M_{ij} + \sum_{i=1}^{n} \sum_{\substack{j=1 \\ i \neq j}}^{n} \sum_{\substack{k=1 \\ j \neq k}}^{n} x_i x_j x_k \ln \nu_{ijk} M_{ijk} \quad (1)$$

where

$$M_{\mathrm{av}} = \sum_{i=1}^{n} x_i M_i$$

$$M_{ij} = (2M_i + M_j)/3$$

$$M_{ijk} = (M_i + M_j + M_k)/3$$

For comparison, the data were also correlated with the modified McAllister equation proposed by Dizechi (*3*):

$$\ln \nu_{\mathrm{m}} = \frac{1}{t + C_{\mathrm{av}}} \sum_{i=1}^{n} (t + C_i) x_i^3 \ln \nu_i M_i - \ln M_{\mathrm{av}} + \frac{3}{t + C_{\mathrm{av}}} \sum_{i=1}^{n} \sum_{\substack{j=1 \\ i \neq j}}^{n} (t + C_{ij}) x_i^2 x_j \ln \nu_{ij} M_{ij} + \frac{1}{t + C_{\mathrm{av}}} \sum_{i=1}^{n} \sum_{\substack{j=1 \\ i \neq j \neq k}}^{n} \sum_{k=1}^{n} (t + C_{ijk}) x_i x_j x_k \ln \nu_{ijk} M_{ijk} \quad (2)$$

where

$$C_i = 239 + Zt_{b,i}$$

$$C_{\mathrm{av}} = \sum_{i=1}^{n} x_i C_i$$

$$C_{ij} = (2C_i + C_j)/3$$

$$C_{ijk} = (C_i + C_j + C_k)/3$$

The data were also correlated with a modified form of the McAllister equation recently proposed by Soliman (*1*). The general form of the equation is as follows:

$$\ln \nu_{\mathrm{m}} = \sum_{i=1}^{n} x_i^3 \ln \nu_i + 3 \sum_{i=1}^{n-1} \sum_{j=i+1}^{n} x_i x_j \ln \nu_{ij} + \sum_{i=1}^{n-1} \sum_{j=i+1}^{n} (x_i + x_j)^3 B_{ij} x_i x_j \Big/ \left[\left(\frac{M_i}{M_j} \right)^2 x_i + x_j \right] + 6\sum_{i=1}^{n-2} \sum_{j=i+1}^{n-1} \sum_{k=i+2}^{n} x_i x_j x_k \ln \nu_{ijk} \quad (3)$$

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Table I. Viscosities of Binary, Ternary, and Quaternary Mixtures

x_1^a	$10^6\upsilon$, m^2/s	x_1^a	$10^6\upsilon$, m^2/s	x_1^a	$10^6\upsilon$, m^2/s	x_1^a	$10^6\upsilon$, m^2/s
2-Propanol–Water, Temperature = 25 °C							
0.00	0.8950	0.30	3.5346	0.60	3.0249	0.90	2.6541
0.10	2.5942	0.40	3.4555	0.70	2.8429	1.00	2.6541
0.20	3.3129	0.50	3.2230	0.80	2.6951		
1-Propanol–Water, Temperature = 25 °C							
0.00	0.8950	0.30	3.0744	0.60	2.8308	0.90	2.52
0.10	2.263	0.40	3.0703	0.70	2.6842	1.00	2.4618
0.20	2.8884	0.50	2.938	0.80	2.5709		
Methanol–Water, Temperature = 25 °C							
0.00	0.8950	0.30	1.7215	0.60	1.3670	0.90	0.8539
0.10	1.3721	0.40	1.6592	0.70	1.1929	1.00	0.7042
0.20	1.6492	0.50	1.5260	0.80	1.0188		
Ethanol–Water, Temperature = 25 °C							
0.00	0.8950	0.30	2.6280	0.60	2.1170	0.90	1.4940
0.10	1.9974	0.40	2.5368	0.70	1.9352	1.00	1.4023
0.20	2.5269	0.50	2.3067	0.80	1.7519		
Ethanol–Methanol, Temperature = 25 °C							
0.00	0.7042	0.30	0.8790	0.60	1.0802	0.90	1.3193
0.10	0.7566	0.40	0.9386	0.70	1.1576	1.00	1.4023
0.20	0.8106	0.50	1.0057	0.80	1.2294		
2-Propanol–1-Propanol, Temperature = 10 °C							
0.00	3.5000	0.2199	3.6503	0.5201	3.8312	0.8573	4.0740
0.0871	3.5500	0.3692	3.7367	0.6019	3.8887	0.9438	4.1377
0.1464	3.6203	0.4513	3.7932	0.7416	4.0111	1.00	4.1940
2-Propanol–1-Propanol, Temperature = 20 °C							
0.00	2.7578	0.2199	2.8077	0.5201	2.9242	0.8573	3.0064
0.0871	2.7662	0.3692	2.8868	0.6019	2.9829	0.9438	3.0463
0.1464	2.7918	0.4513	2.9022	0.7416	3.0006	1.00	3.0808
2-Propanol–1-Propanol, Temperature = 30 °C							
0.00	2.1799	0.2199	2.2086	0.5201	2.2530	0.8573	2.2813
0.0871	2.1914	0.3692	2.2310	0.6019	2.2886	0.9438	2.2888
0.1464	2.2052	0.4513	2.2428	0.7416	2.2698	1.00	2.3054
2-Propanol–1-Propanol, Temperature = 40 °C							
0.00	1.7634	0.2199	1.7790	0.5201	1.7896	0.8573	1.7806
0.0871	1.7818	0.3692	1.7725	0.6019	1.7879	0.9438	1.7664
0.1464	1.7927	0.4513	1.7991	0.7416	1.7915	1.00	1.7629
2-Propanol–1-Propanol, Temperature = 50 °C							
0.00	1.4505	0.2199	1.4413	0.5201	1.4135	0.8573	1.3941
0.0871	1.4488	0.3692	1.4375	0.6019	1.4140	0.9438	1.3896
0.1464	1.4490	0.4513	1.4287	0.7416	1.4017	1.00	1.3777
2-Propanol–Methanol, Temperature = 10 °C							
0.00	0.8540	0.0	1.1278	0.60	1.7235	0.90	3.1614
0.10	0.9198	0.40	1.2824	0.70	2.0713	1.00	4.1940
0.20	1.0081	0.50	1.4735	0.80	2.5238		
2-Propanol–Methanol, Temperature = 20 °C							
0.00	0.7450	0.30	1.0132	0.60	1.4984	0.90	2.4602
0.10	0.8175	0.40	1.1463	0.70	1.7503	1.00	3.0808
0.20	0.9047	0.50	1.3049	0.80	2.0498		
2-Propanol–Methanol, Temperature = 30 °C							
0.00	0.6600	0.30	0.9054	0.60	1.2796	0.90	1.9312
0.10	0.7311	0.40	1.0159	0.70	1.4527	1.00	2.3054
0.20	0.8117	0.50	1.1395	0.80	1.6620		
2-Propanol–Methanol, Temperature = 40 °C							
0.00	0.5810	0.30	0.7918	0.60	1.0765	0.90	1.5254
0.10	0.6434	0.40	0.8793	0.70	1.1954	1.00	1.7629
0.20	0.7144	0.50	0.9731	0.80	1.3445		
2-Propanol–Methanol, Temperature = 50 °C							
0.00	0.5160	0.30	0.6598	0.60	0.8659	0.90	1.1985
0.10	0.5570	0.40	0.7215	0.70	0.9561	1.00	1.3771
0.20	0.6046	0.50	0.7898	0.80	1.0633		
2-Propanol–Ethanol, Temperature = 10 °C							
0.00	1.8200	0.30	1.8278	0.60	2.2712	0.90	3.4413
0.10	1.7816	0.40	1.9197	0.70	2.5502	1.00	4.1940
0.20	1.7802	0.50	2.0651	0.80	2.9226		

Table I (Continued)

x_1^a	$10^6\nu$, m^2/s	x_1^a	$10^6\nu$, m^2/s	x_1^a	$10^6\nu$, m^2/s	x_1^a	$10^6\nu$, m^2/s
2-Propanol–Ethanol, Temperature = 20 °C							
0.00	1.5270	0.30	1.7117	0.60	2.0884	0.90	2.7367
0.10	1.5719	0.40	1.8125	0.70	2.2640	1.00	3.0808
0.20	1.6304	0.50	1.9379	0.80	2.4752		
2-Propanol–Ethanol, Temperature = 30 °C							
0.00	1.2800	0.30	1.5906	0.60	1.9148	0.90	2.2047
0.10	1.3897	0.40	1.6970	0.70	2.0214	1.00	2.3054
0.20	1.4926	0.50	1.8027	0.80	2.1135		
2-Propanol–Ethanol, Temperature = 40 °C							
0.00	1.0790	0.30	1.4878	0.60	1.7634	0.90	1.8011
0.10	1.2253	0.40	1.5974	0.70	1.8074	1.00	1.7629
0.20	1.3608	0.50	1.6913	0.80	1.8170		
2-Propanol–Ethanol, Temperature = 50 °C							
0.00	0.9200	0.30	1.3165	0.60	1.5433	0.90	1.4561
0.10	1.0650	0.40	1.4192	0.70	1.5554	1.00	1.3771
0.20	1.1999	0.50	1.4942	0.80	1.5272		

x_1^a	x_2^b	$10^6\nu$, m^2/s	x_1^a	x_2^b	$10^6\nu$, m^2/s	x_1^a	x_2^b	$10^6\nu$, m^2/s
2-Propanol–1-Propanol–Water, Temperature = 10 °C								
0.0953	0.7510	3.9858	0.4492	0.1641	4.9214	0.6813	0.1890	4.0713
0.1084	0.4885	4.3675	0.4644	0.4510	3.9084	0.7543	0.1285	4.0687
0.2029	0.6994	3.7259	0.5312	0.3260	4.0512	0.8475	0.0560	4.0704
0.3630	0.2480	4.9143						
2-Propanol–1-Propanol–Water, Temperature = 20 °C								
0.0953	0.7510	2.9216	0.4487	0.1615	3.4513	0.6970	0.1860	3.0139
0.1858	0.4994	3.1947	0.4644	0.4510	2.9650	0.7543	0.1285	3.0153
0.2029	0.6994	2.9060	0.5312	0.3260	3.0109	0.8475	0.0560	3.0055
0.3630	0.2480	3.4089						
2-Propanol–1-Propanol–Water, Temperature = 30 °C								
0.0953	0.7510	2.2985	0.4487	0.1615	2.4944	0.6970	0.1860	2.2605
0.1858	0.4994	2.3860	0.4644	0.4510	2.2478	0.7543	0.1285	2.2745
0.2029	0.6994	2.2507	0.5312	0.3260	2.2640	0.8475	0.0560	2.2615
0.3630	0.2480	2.4801						
2-Propanol–1-Propanol–Water, Temperature = 40 °C								
0.0833	0.6276	1.8528	0.4136	0.1050	1.9886	0.7124	0.1709	1.7574
0.1079	0.2598	2.0116	0.5417	0.1005	1.8854	0.8531	0.1187	1.751
0.3476	0.3427	1.8536	0.6010	0.0885	1.8852	0.2217	0.6931	1.7928
2-Propanol–1-Propanol–Water, Temperature = 50 °C								
0.0833	0.6276	1.4662	0.4136	0.1050	1.5381	0.7124	0.1709	1.3862
0.1079	0.2498	1.5712	0.5417	0.1005	1.4682	0.8531	0.1187	1.3785
0.3476	0.3427	1.4633	0.6010	0.0885	1.4417	0.2217	0.6931	1.4369
Ethanol–Methanol–Water, Temperature = 25 °C								
0.10	0.10	2.1774	0.10	0.50	1.5380	0.50	0.10	1.8691
0.20	0.30	1.8605	0.20	0.70	0.9712	0.40	0.20	1.7585
0.40	0.30	1.4809	0.30	0.40	1.4460	0.50	0.40	1.2094
0.80	0.050	1.5130						
2-Propanol–Ethanol–Methanol, Temperature = 10 °C								
0.10	0.10	1.0316	0.10	0.50	1.5666	0.50	0.10	1.7860
0.20	0.30	1.5491	0.20	0.70	1.8557	0.40	0.20	1.8205
0.40	0.30	1.9896	0.30	0.40	1.9053	0.50	0.40	2.1952
0.80	0.05	2.6830						
2-Propanol–Ethanol–Methanol, Temperature = 20 °C								
0.10	0.10	0.8940	0.10	0.50	1.2976	0.50	0.10	1.4911
0.20	0.30	1.2616	0.20	0.70	1.6108	0.40	0.20	1.4836
0.40	0.30	1.6107	0.30	0.40	1.5438	0.50	0.40	1.8949
0.80	0.05	2.1635						
2-Propanol–Ethanol–Methanol, Temperature = 30 °C								
0.10	0.10	0.7639	0.10	0.50	1.0159	0.50	0.10	1.1841
0.20	0.30	0.9601	0.20	0.70	1.3475	0.40	0.20	1.1180
0.40	0.30	1.2016	0.30	0.40	1.1548	0.50	0.40	1.5633
0.80	0.05	1.7272						
2-Propanol–Ethanol–Methanol, Temperature = 40 °C								
0.10	0.10	0.6436	0.10	0.50	0.7908	0.50	0.10	0.9081
0.20	0.30	0.7089	0.20	0.70	1.1161	0.40	0.20	0.8105
0.40	0.30	0.861	0.30	0.40	0.8312	0.50	0.40	1.2635
0.80	0.05	1.3666						

Table I (Continued)

x_1^a	x_2^b	$10^6\upsilon$, m^2/s	x_1^a	x_2^b	$10^6\upsilon$, m^2/s	x_1^a	x_2^b	$10^6\upsilon$, m^2/s
2-Propanol–Ethanol–Methanol, Temperature = 50 °C								
0.10	0.10	0.5498	0.10	0.50	0.6680	0.50	0.10	0.7241
0.20	0.30	0.5783	0.20	0.70	0.9521	0.40	0.20	0.6470
0.40	0.30	0.6905	0.30	0.40	0.6734	0.50	0.40	1.0593
0.80	0.05	1.0820						
2-Propanol–Ethanol–Water, Temperature = 10 °C								
0.10	0.10	6.8621	0.10	0.50	3.6596	0.50	0.10	4.5445
0.20	0.30	4.6240	0.20	0.70	2.1371	0.40	0.20	4.1756
0.40	0.30	3.4137	0.30	0.40	3.2981	0.50	0.40	2.6126
0.80	0.05	3.7921						
2-Propanol–Ethanol–Water, Temperature = 20 °C								
0.10	0.10	4.2295	0.10	0.50	2.8197	0.50	0.10	3.3397
0.20	0.30	3.3954	0.20	0.70	1.8814	0.40	0.20	3.1754
0.40	0.30	2.7653	0.30	0.40	2.6943	0.50	0.40	2.3180
0.80	0.05	2.8849						
2-Propanol–Ethanol–Water, Temperature = 25 °C								
0.10	0.10	3.5517	0.10	0.50	2.3608	0.50	0.10	2.8160
0.20	0.30	2.8331	0.20	0.70	1.7130	0.40	0.20	2.6664
0.40	0.30	2.3843	0.30	0.40	2.3032	0.50	0.40	2.1126
0.80	0.05	2.5300						
2-Propanol–Ethanol–Water, Temprature = 30 °C								
0.10	0.10	2.8309	0.10	0.50	2.1577	0.50	0.10	2.4992
0.20	0.30	2.4978	0.20	0.70	1.6442	0.40	0.20	2.4176
0.40	0 30	2.2074	0.30	0.40	2.1536	0.50	0.40	2.0187
0.80	0.05	2.2317						
2-Propanol–Ethanol–Water, Temperature = 40 °C								
0.10	0.10	2.0054	0.10	0.50	1.6550	0.50	0.10	1.8996
0.20	0.30	1.8637	0.20	0.70	1.4336	0.40	0.20	1.8529
0.40	0.30	1.7551	0.30	0.40	1.7088	0.50	0.40	1.7437
0.80	0.05	1.7585						
2-Propanol–Ethanol–Water, Temperature = 50 °C								
0.10	0.10	1.4817	0.10	0.50	1.2917	0.50	0.10	1.4505
0.20	0.30	1.4074	0.20	0.70	1.2071	0.40	0.20	1.4117
0.40	0.30	1.3633	0.30	0.40	1.3329	0.50	0.40	1.4502
0.80	0.05	1.3888						
2-Propanol–Methanol–Water, Temperature = 10 °C								
0.10	0.10	5.1866	0.10	0.50	2.2702	0.50	0.10	4.2170
0.20	0.30	3.3886	0.20	0.70	1.2712	0.40	0.20	3.5244
0.40	0.30	2.6859	0.30	0.40	2.3181	0.50	0.40	1.9220
0.80	0.05	3.6349						
2-Propanol–Methanol–Water, Temperature = 20 °C								
0.10	0.10	3.3455	0.10	0.50	1.8269	0.50	0.10	3.1435
0.20	0.30	2.6007	0.20	0.70	1.1187	0.40	0.20	2.7405
0.40	0.30	2.2147	0.30	0.40	1.9420	0.50	0.40	1.6667
0.80	0.05	2.7607						
2-Propanol–Methanol–Water, Temperature = 25 °C								
0.10	0.10	2.7897	0.10	0.50	1.6272	0.50	0.10	2.7330
0.20	0.30	2.2811	0.20	0.70	1.0400	0.40	0.20	2.4345
0.40	0.30	1.9870	0.30	0.40	1.7635	0.50	0.40	1.5325
0.80	0.05	2.4200						
2-Propanol–Methanol–Water, Temperature = 30 °C								
0.10	0.10	2.3073	0.10	0.50	1.4685	0.50	0.10	2.3588
0.20	0.30	1.9891	0.20	0.70	0.9892	0.40	0.20	2.1183
0.40	0.30	1.7919	0.30	0.40	1.6015	0.50	0.40	1.4149
0.80	0.05	2.1275						
2-Propanol–Methanol–Water, Temperature = 40 °C								
0.10	0.10	1.6717	0.10	0.50	1.2043	0.50	0.10	1.8191
0.20	0.30	1.5545	0.20	0.70	0.8653	0.40	0.20	1.6706
0.40	0.30	1.4656	0.30	0.40	1.3268	0.50	0.40	1.1903
0.80	0.05	1.6659						
2-Propanol–Methanol–Water, Temperature = 50 °C								
0.10	0.10	1.2879	0.10	0.50	0.9906	0.50	0.10	1.4235
0.20	0.30	1.2448	0.20	0.70	0.7243	0.40	0.20	1.3233
0.40	0.30	1.1841	0.30	0.40	1.0808	0.50	0.40	0.9641
0.80	0.05	1.3182						

Table I (Continued)

x_1^a	x_2^b	x_3^c	$10^6 v$, m^2/s	x_1^a	x_2^b	x_3^c	$10^6 v$, m^2/s
		2-Propanol–Ethanol–Methanol–Water, Temperature = 10 °C					
0.1378	0.1264	0.1337	4.4339	0.0731	0.3822	0.2433	2.6387
0.2282	0.2846	0.2638	2.6540	0.0740	0.2142	0.2830	2.9446
0.1137	0.1846	0.1859	3.6265	0.1741	0.1911	0.3166	2.7677
0.1113	0.1983	0.1725	3.7598	0.0795	0.0860	0.4473	2.5276
0.1408	0.2440	0.2234	3.1210	0.1233	0.1054	0.4800	2.2689
0.1764	0.4999	0.1249	2.7125				
		2-Propanol–Ethanol–Methanol–Water, Temperature = 20 °C					
0.1378	0.1264	0.1337	3.2690	0.0731	0.3822	0.2433	2.0475
0.2282	0.2846	0.2638	2.0890	0.0740	0.2142	0.2830	2.2258
0.1137	0.1846	0.1859	2.7335	0.1741	0.1911	0.3166	2.1364
0.1408	0.2440	0.2234	2.3259	0.0795	0.0860	0.4473	2.0088
0.1764	0.4999	0.1249	2.1129	0.1233	0.1054	0.4800	1.8275
		2-Propanol–Ethanol–Methanol–Water, Temperature = 25 °C					
0.1378	0.1264	0.1337	2.6683	0.0731	0.3822	0.2433	1.8164
0.2282	0.2846	0.2638	1.7891	0.0740	0.2142	0.2830	1.9562
0.1137	0.1846	0.1859	2.2906	0.1741	0.1911	0.3166	1.8984
0.1113	0.1983	0.1725	2.3341	0.0795	0.0860	0.4473	1.7631
0.1408	0.2440	0.2234	2.0775	0.1233	0.1054	0.4800	1.5960
0.1764	0.4999	0.1249	1.8856				
		2-Propanol–Ethanol–Methanol–Water, Temperature = 30 °C					
0.1378	0.1264	0.1337	2.3034	0.0731	0.3822	0.2433	1.6172
0.2282	0.2846	0.2638	1.5822	0.0740	0.2142	0.2830	1.7178
0.1137	0.1846	0.1859	2.0042	0.1741	0.1911	0.3166	1.6710
0.1113	0.1983	0.1725	2.0499	0.0795	0.0860	0.4473	1.5871
0.1408	0.2440	0.2234	1.8194	0.1233	0.1054	0.4800	1.4498
1.1764	0.4999	0.1249	1.6714				
		2-Propanol–Ethanol–Methanol–Water, Temperature = 40 °C					
0.1378	0.1264	0.1337	1.6616	0.0731	0.3822	0.2433	1.3021
0.2282	0.2846	0.2638	1.2190	0.0740	0.2142	0.2830	1.3663
0.1137	0.1846	0.1859	1.5559	0.1741	0.1911	0.3166	1.3478
0.1113	0.1983	0.1725	1.5798	0.0795	0.0860	0.4473	1.2732
0.1408	0.2440	0.2234	1.4457	0.1233	0.1054	0.4800	1.1946
0.1764	0.4999	0.1249	1.3507				
		2-Propanol–Ethanol–Methanol–Water, Temperature = 50 °C					
0.1378	0.1264	0.1337	1.2656	0.0731	0.3822	0.2433	1.0705
0.2282	0.2846	0.2638	0.9850	0.0740	0.2142	0.2830	1.1102
0.1137	0.1846	0.1859	1.2452	0.1741	0.1911	0.3166	1.0399
0.1113	0.1983	0.1725	1.2676	0.0795	0.0860	0.4473	1.0414
0.1408	0.2440	0.2234	1.1762	0.1233	0.1054	0.4800	0.9664
0.1764	0.4999	0.1249	1.1097				

Data for the quaternary mixtures were correlated with eqs 2 and 3 only.

The constants v_{ij}, v_{ji}, v_{ijk}, and B_{ij} are found from experimental data of viscosities of mixtures with the help of the least-squares technique. Their values are assumed to be independent of concentration but not of temperature. The values of Z are found by using the optimization technique proposed by Dizechi (*4*). Even though Z is actually dependent on temperature, the dependency is so weak that selection of one value for Z is sufficient.

While eqs 1 and 3 have the same number of adjustable parameters, eq 2 introduces an additional parameter, Z.

Results of the comparison are listed in Table II, where D, the mean percentage deviation, is defined as

$$D = \frac{1}{m}\sum_{i=1}^{m}\frac{100|\text{Viscosity}_{i,\text{exp}} - \text{Viscosity}_{i,\text{calc}}|}{\text{Viscosity}_{i,\text{exp}}} \qquad (4)$$

and S, the standard percentage error is defined as follows:

$$S = \left\{\frac{1}{m}\sum_{i=1}^{m}\left[\frac{100(\text{Viscosity}_{i,\text{exp}} - \text{Viscosity}_{i,\text{calc}})}{\text{Viscosity}_{i,\text{exp}}}\right]^2\right\}^{1/2}$$

where m is the number of data points.

In order to correlate the quaternary mixture data, information of viscosities presented in refs 3 and 5 was used in addition to the information presented in this paper. Inspection of Table II shows that Soliman's equation is as good or better than the equation proposed by Dizechi even though it has fewer adjustable constants. Details of this comparison including obtained values for the coefficients v_{ij}, v_{ji}, v_{ijk}, B_{ij}, and Z can be found in ref 1.

Discussion and Conclusion

In general, more accurate correlating equations for the viscosity of liquid mixtures employ a large number of adjustable parameters, a fact that frequently reduces the usefulness of the equation. In addition, these parameters depend usually on either concentration or temperature or both. Improving the accuracy of a correlating equation, while at the same time reducing the number of adjustable parameters, is a worthwhile attempt. On the other hand, improving a correlating equation by accurately describing the dependence of the adjustable parameters is also a task worthwhile to undertake.

The Soliman equation (*3*) was developed as an attempt to improve the accuracy of existing correlating equations for the viscosity of liquid mixtures by taking the first route: reducing the number of adjustable parameters. Inspection of Table II shows that Soliman's equation is as good or better than the equation proposed by Dizechi, even though it has fewer adjustable parameters.

One very important shortcoming of all three correlating equations studied here is the assumption of independence of

Table II. Comparison of McAllister's, Dizechi's, and Soliman's Equations

system	temp, °C	*D*, % *S*, %		
		McAllister	Dizechi	Soliman
2-propanol–water	25	22.26	0.90	0.44
		85.4	4.20	1.56
1-propanol–water	25	19.00	0.75	0.91
		65.68	2.78	3.09
methanol–water	25	5.12	0.33	0.60
		8.5	0.70	1.18
ethanol–water	25	15.05	3.11	1.30
		40.04	8.89	3.35
ethanol–methanol	25	0.17	0.17	0.37
		0.27	0.27	0.49
2-propanol–1-propanol	10	0.30	0.30	0.33
		1.40	1.41	1.88
	20	0.67	0.67	0.77
		2.93	2.93	3.13
	30	0.21	0.21	0.35
		0.77	0.77	0.95
	40	0.88	0.89	0.36
		2.07	2.08	0.92
	50	0.13	0.13	0.22
		0.25	0.25	0.38
2-propanol–methanol	10	0.60	0.78	0.53
		2.17	2.50	1.06
	20	0.54	0.70	0.50
		1.49	1.76	0.84
	30	0.39	0.55	0.35
		0.83	1.08	0.46
	40	0.42	0.57	0.32
		0.66	0.83	0.35
	50	0.41	0.52	0.23
		0.53	0.62	0.22
2-propanol–ethanol	10	0.25	0.28	0.21
		0.90	0.96	0.54
	20	0.21	0.22	0.16
		0.64	0.68	0.36
	30	0.34	0.27	0.33
		0.87	0.71	0.75
	40	0.20	0.14	0.21
		0.44	0.34	0.45
	50	0.28	0.24	0.29
		0.53	0.44	0.51
2-propanol–1-propanol–water	10	32.12	1.76	0.96
		168.51	11.28	5.95
	20	27.55	1.84	0.25
		113.94	6.89	1.09
	30	21.83	0.84	0.48
		63.57	2.94	1.63
	40	24.74	0.96	1.21
		62.61	2.32	4.43
	50	18.01	0.45	0.84
		36.81	0.87	2.11
ethanol–methanol–water	25	6.67	2.08	1.77
		15.05	4.66	3.84
2-propanol–ethanol–methanol	10	0.68	0.91	0.41
		2.16	2.11	0.97
	20	0.66	0.53	0.47
		1.56	1.33	0.82
	30	0.47	0.49	0.31
		0.80	1.04	0.42
	40	0.39	1.37	0.20
		0.54	1.75	0.21
	50	0.49	1.65	0.16
		0.62	1.70	0.15
2-propanol–ethanol–water	10	26.28	2.11	1.57
		171.20	27.41	8.04
	20	21.15	2.77	1.60
		84.62	16.49	5.96
	25	11.20	2.70	1.66
		39.71	12.84	5.31
	30	17.07	1.84	1.17
		58.67	8.67	3.23
	40	14.90	1.82	0.67
		39.18	5.34	1.46
	50	12.75	1.59	0.62
		26.02	3.30	1.07
2-propanol–methanol–water	10	23.98	1.73	2.04
		118.88	14.51	10.18
	20	17.02	1.99	2.01
		65.87	10.23	6.93
	25	8.10	1.77	1.72
		28.27	6.49	5.29
	30	15.30	1.14	1.33
		46.54	4.37	3.11
	40	13.52	1.03	0.77
		32.32	2.20	1.39
	50	11.17	0.95	0.78
		21.70	1.75	1.29
2-propanol–ethanol–methanol–water	10		3.13	3.04
			10.83	10.95
	20		2.63	1.84
			7.76	5.50
	25		3.70	3.32
			8.44	7.58
	30		3.29	2.14
			6.87	4.59
	40		6.50	4.26
			10.14	6.80
	50		3.86	5.44
			6.32	6.68

concentration for the adjustable parameters, even though they actually depend on concentration. It is recommended that this point as well as their dependence on temperature be investigated further in future studies.

Glossary

B	binary interaction coefficient in eq 3
C	constant in eq 2
D	mean percentage deviation
m	number of experimental data points at a fixed temperature
M	molar mass
S	standard percentage error
t	temperature, °C
t_b	boiling temperature, °C
x_i	mole fraction of component *i*
Z	constant in eq 2
ν	kinematic viscosity, m^2/s
ν_m	kinematic viscosity of the mixture

Subscripts

1,2,3	components 1, 2, and 3, respectively
ij	refer to interaction of type *i*–*i*–*j*, etc.
123	refer to interaction between three molecules; one of component 1, one of component 2, and one of component 3
av	average
calc	calculated
exp	experimental

Superscripts

a	mole fraction of first-named component
b	mole fraction of second-named component
c	mole fraction of third-named component

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Measurements of the Critical Parameters and the Vapor–Liquid Coexistence Curve in the Critical Region of HCFC-123

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Measurements of the vapor–liquid coexistence curve in the critical region were made for HCFC-123 ($CHCl_2CF_3$) by visual observation of the vapor–liquid interface in an optical cell. Thirty densities of both saturated liquid and vapor were obtained in the range of temperatures from 401 K to the critical temperature corresponding to the density range between 203 and 1141 kg/m^3. The experimental uncertainty of the temperature measurements was estimated to be less than ±12 mK, while that of the density measurements was estimated to be between ±0.09% and ±0.53%. Not only the level where the meniscus disappeared but also the intensity of the critical opalescence were considered for the determination of the critical temperature T_c and the critical density ρ_c of HCFC-123 as 456.86 ± 0.02 K and 556 ± 3 kg/m^3, respectively. The critical exponent β was also determined, and the correlation of the saturated liquid density of HCFC-123 was established.

Introduction

HCFC-123 (2,2-dichloro-1,1,1-trifluoroethane, $CHCl_2CF_3$) attracts a great deal of attention as a stratospherically safe alternative to CFC-11 (trichlorofluoromethane, CCl_3F), which is mainly used as a blowing agent and as a refrigerant for turborefrigeration machines. A comparison of some general physicochemical properties of HCFC-123 with CFC-11 is shown in Table I. The critical parameters are essential and important in predicting various thermodynamic properties and in formulating equations of state. By means of visual observation of the meniscus in an optical cell, the critical temperature and the critical density of HCFC-123 were determined. The level where the meniscus disappeared and the intensity of the critical opalescence observed were the key factors for determining the critical parameters of HCFC-123. Because of its high critical temperature, we modified our original apparatus (*1*) so as to perform accurate measurements at even higher temperatures beyond 425 K.

The saturated liquid densities and saturated vapor densities near the critical point were also determined by the observing disappearance of the meniscus in the cell. The critical exponent β and the saturated liquid density correlation were subsequently determined on the basis of the present measurements with available other data.

Table I. General Properties of CFC-11 and HCFC-123[a]

substance	CFC-11	HCFC-123
chemical formula	CCl_3F	$CHCl_2CF_3$
molar mass, g/mol	137.368	152.931
critical temperature, K	471.15	456.86*
critical pressure, MPa	4.409	3.6655
critical density, kg/m^3	554	556*
critical volume, cm^3/mol	248	275
normal boiling point, K	297	301.5
freezing point, K	162	172
dipole moment, D	0.5	
ozone depletion potential	1.0	<0.05
global warming potential	0.4	<0.1

[a] Values marked with an asterisk were determined in the present work.

Experimental Section

The experimental apparatus and basic experimental procedures have been already described in the previous papers (*1*–*3*). By using the apparatus, we have already measured critical properties of HFC-23 (*1*), CFC-12 (*2*), HCFC-22 (*2*), Halon-1301 (*3*), CFC-114 (*4*), HFC-152a (*5*), CFC-115 (*6*), HFC-134a (*7*), and azeotropic mixture refrigerant 502 (*8*), respectively. The apparatus is composed of three vessels, i.e., an optical cell with two synthetic sapphire windows (15 mm in thickness) for observing the meniscus behavior of the sample fluid, an expansion vessel, and a supplying vessel.

In this study, two modifications were made for performing the measurements of HCFC-123. Firstly, we constructed new vessels so as to perform measurements at higher temperatures beyond 425 K. A new optical cell is shown in Figure 1. A gold packing (0.2 mm in thickness) and an aluminum packing (1 mm in thickness) between a sapphire window and the cell made of 304 stainless steel were used for the high-temperature sealing. The other two vessels made of 304 stainless steel were designed so as to withstand higher temperatures and pressures. These vessels were connected to high-temperature and -pressure valves (Sno-Trik Model SS-410-FP). The inner volumes of the respective three vessels were calibrated by using pure water, because its density value at room temperature (298.15 K) is well-known. The volumes are 10.956 ± 0.004, 6.249 ± 0.004, and 77.305 ± 0.004 cm^3, respectively.

Secondly, a new expansion procedure called "supplementary expansion" was introduced so as to improve the accuracy and ease of determining the various density values. This expansion procedure makes it possible to make subtle density changes for the measurements in the high-density region.

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