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Article

Density, Viscosity, and Excess Molar Volume of the Binary Mixtures of 1-Propanol, 1,2-Propanediol, 1,3-Propanediol, and Glycerol

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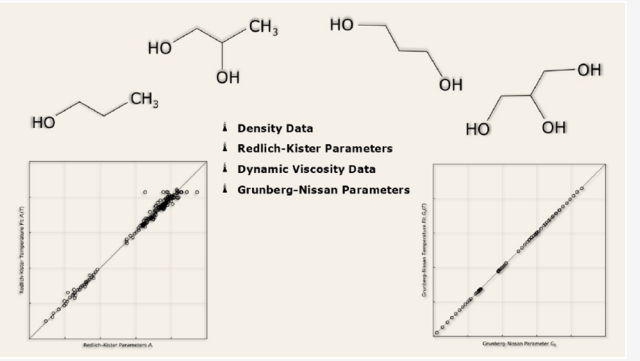
ABSTRACT: Experimental data on the density and dynamic viscosity of binary mixtures of 1-propanol, 1,2-propanediol, 1,3-propanediol, and glycerol at temperatures from 293.15 to 343.15 K are reported. For the majority of binary mixtures, this is the first time that the dynamic viscosity has been measured. Excess molar volumes are calculated and fitted to Redlich−Kister polynomials. The mixture viscosity has been fitted using the Grunberg−Nissan mixing rule to a maximum mean relative deviation of MRD = 1.10%.



INTRODUCTION

Glycerol (1,2,3-propanetriol) is a versatile substance that has experienced a rise in interest from different industrial sectors due to its solvent properties as well as low toxicity. While the synthesis of glycerol in the past was associated with high cost, the increasing production of biofuels has provided an increasingly cheap global feedstock of glycerol, which is produced as a coproduct in the production of biofuels from fats.[1,2] Due to the reported negative effects of the global use of fossil fuels, demand and interest in biofuels are projected to keep growing in the years to come.[3] Therefore, glycerol has experienced ever-growing interest as the so-called platform chemical recently.

The synthesis of 1-propanol from glycerol is not only interesting due to the ability to use 1-propanol as an advanced biofuel,[4] but also because of the possibility of providing biobased 1-propanol at a larger scale. The successful synthesis of 1-propanol from glycerol has been reported in the literature. In a biotechnological process outlined by Siebert and Wendisch,[5] the synthesis of 1-propanol and 1,2-propanediol is described. Both Bhanuchander et al.[6] and Gatti et al.[7] describe a process for the production of 1-propanol from glycerol, where both 1,2-propanediol and 1,3-propanediol are produced as intermediaries. Implementation of such processes will, in any case, require the handling of mixtures of these components, be it inside the reactor vessel or during the purification of the reaction products. In these matters, information on properties like density and viscosity of the mixtures of 1-propanol (1), 1,2-propanediol (2), 1,3-propanediol (3), and glycerol (4) will prove necessary for an accurate and efficient equipment design. While some attention has previously been paid to the density of the binary mixtures,[8−12] the viscosity of the binary mixtures has been more scarcely investigated.[11,13,14]

EXPERIMENTAL AND COMPUTATIONAL METHODS

Substances and Analysis. The substances used in the experiments for this work are listed in Table 1. Refractive indices were measured with an Atago PAL-RI refractometer. The substances were analyzed for their purity and, as satisfactory correspondence with the listed literature values was determined, used without additional purification steps. The measured mixture samples were prepared with a sample volume of 20 mL each and mixed using a vortex mixer to ensure complete mixing. After mixing, the samples were allowed to rest for 72 h to allow for the settling of the fluid and degassing of air trapped during the mixing process.

Density and Dynamic Viscosity Measurements. The density ρ and dynamic viscosity η of the samples were measured with an Anton Paar Stabinger SVM3000 Viscometer in 5 K intervals between 293.15 and 343.15 K. The apparatus uses a rotational viscometer with a free-floating measuring rotor, as well as a vibrating U-tube densimeter for simultaneous density and viscosity measurements from the same sample. The apparatus was calibrated using the reference fluids of a viscosity

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Table 1. Properties of the Chemicals Used for the Density and Viscosity Measurements[a]

substance	index	CAS Nr.	supplier	purity ($\%_{mb}$[I])	refractive index n_D[II] meas. @589 nm	refractive index n_D[II] lit.	water content w_{H_2O} ($\%_{mb}$[I]) (KF-titration)
1-propanol	(1)	71−23−8	Carl Roth	≥99.5%	1.3837	1.38369[15]	0.039%
1,2-propanediol	(2)	57−55−6	Carl Roth	≥99.5%	1.4322	1.4324[16]	0.035%
1,3-propanediol	(3)	504−63−2	Carl Roth	≥98%[III]	1.4397	1.4398[16]	0.062%
glycerol	(4)	112−27−6	Carl Roth	≥99%	1.4733	1.47301[15]	0.032%

[a]Standard uncertainties: $u(n_D)$ = 0.001, $u(w_{H_2O})$ = 0.005%; [I]mb ··· mass based; [II]@589 nm, 98 kPa, 25 °C [III]Manufacturer charge analysis certificate states 100%.

Table 2. Densities and Viscosities of the Pure Substances as Measured at an Ambient Pressure (98 kPa), Listed Together with Values from the Literature for the Relevant Temperature Range[a]

substance	T/K	ρ/kg m^{-3} meas.	ρ/kg m^{-3} lit.	η/mPas meas.	η/mPas lit.
1-propanol	293.15	803.62	803.767[24]	2.2093	2.2085[24]
	298.15	799.71	799.6;[8] 799.755[24]	1.9595	1.9503[24]
	303.15	795.71	795.5;[8] 795.717[24]	1.7430	1.7270[24]
	308.15	791.50	791.5;[8] 791.638[24]	1.5543	1.5419[24]
	313.15	787.40	787.3;[8] 787.521[24]	1.3924	1.3783[24]
	318.15	783.19	783.3;[8] 783.357[24]	1.2474	1.2365[24]
	323.15	778.99	779.0;[8] 779.137[24]	1.1223	1.1241[24]
	328.15	774.78	774.9[8]	1.0117	
	333.15	770.38	770.8[8]	0.91327	
	338.15	765.97		0.82516	
	343.15	761.56		0.74764	
1,2-propanediol	293.15	1036.8	1036.247[24]	58.974	58.753[24]
	298.15	1033.2	1032.5;[8] 1032.558;[24] 1032.7[11]	43.498	43.437;[24] 43.45[11]
	303.15	1029.5	1029.2;[8] 1028.881;[24] 1028.8[11]	32.787	32.754;[24] 33.89[11]
	308.15	1025.8	1025.3;[8] 1025.137;[24] 1025.2[11]	25.184	25.156;[24] 25.22[11]
	313.15	1022.0	1021.5;[8] 1021.351[24]	19.682	19.646[24]
	318.15	1018.2	1018.0;[8] 1017.524[24]	15.638	15.581[24]
	323.15	1014.4	1014.0;[8] 1013.656[24]	12.611	12.535[24]
	328.15	1010.5	1010.2[8]	10.310	
	333.15	1006.5	1006.2[8]	8.5372	
	338.15	1002.5		7.1527	
	343.15	998.65		6.0670	
1,3-propanediol	293.15	1053.4	1052.864[24]	51.788	51.913[24]
	298.15	1050.3	1049.9;[8] 1049.753;[24] 1049.6[11]	40.875	41.041;[24] 41.96[11]
	303.15	1047.2	1047.0;[8] 1046.635;[24] 1046.2[11]	32.720	32.843;[24] 33.57[11]
	308.15	1044.1	1044.1;[8] 1043.497;[24] 1043.1[11]	26.501	26.580;[24] 27.21[11]
	313.15	1041.0	1040.8;[8] 1040.347[24]	21.684	21.737[24]
	318.15	1037.9	1037.8;[8] 1037.175[24]	17.950	17.950[24]
	323.15	1034.7	1034.8;[8] 1033.981[24]	14.992	14.958[24]
	328.15	1031.5	1031.4[8]	12.634	
	333.15	1028.2	1028.2[8]	10.735	
	338.15	1024.9		9.1943	
	343.15	1021.7		7.9767	
glycerol	293.15	1261.2		1424.9	1412[25]
	298.15	1258.1	1258.9;[8] 1257.8[26]	912.35	943.8[26]
	303.15	1254.9	1255.6;[8] 1254.7[26]	601.43	612;[25] 628.9[26]
	308.15	1251.6	1252.7;[8] 1252.2[26]	407.13	431.9[26]
	313.15	1248.4	1249.5;[8] 1249.4[26]	282.83	284;[25] 295.9[26]
	318.15	1245.1	1246.2;[8] 1245.8[26]	201.04	185.8[26]
	323.15	1242.0	1242.9;[8] 1242.3[26]	146.12	142;[25] 151.2[26]
	328.15	1238.8	1239.6;[8] 1239.3[26]	108.41	108.4[26]
	333.15	1235.6	1236.0;[8] 1236.1[26]	81.931	81.3;[25] 84.8[26]
	338.15	1232.4	1232.8[26]	62.997	62.2[26]
	343.15	1229.1		49.225	50.6[25]

[a]Standard uncertainties: $u(T)$ = 0.01 K, $u_c(\rho)$ = 0.31 kg m^{-3}, and $u_r(\eta)$ = 0.01022.

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Table 3. Parameters and Mean Relative Deviation MRD of the PPDS9 Equation (6) Used for Calculations of the Pure Substances' Dynamic Viscosity η

substance	T_c/K	ρ_c/kg m^{-3}	A	B	C	D	MRD(η_i)
1-propanol	536.750	274.000	816.271	−549.210	696.984	−232.082	-4.62×10^{-10}
1,2-propanediol	as published in Stephan et al.[17]						-5.78×10^{-4}
1,3-propanediol	as published in Stephan et al.[17]						2.78×10^{-4}
glycerol	as published in Stephan et al.[17]						9.71×10^{-5}

Table 4. Parameters and Mean Relative Deviation MRD of the PPDS9 Equation (6) Used for Calculation of the Pure Substances' Dynamic Viscosity η

substance	A	B	C/K	D/K	E/Pas	MRD(η_i)
1-propanol	4.15014	2.10781	4.45634×10^{2}	1.59288×10^{1}	2.85079×10^{-5}	-7.00×10^{-7}
1,2-propanediol	9.79638×10^{-2}	1.58796×10^{-2}	1.48885×10^{4}	1.41581×10^{2}	3.40266×10^{-5}	-2.97×10^{-4}
1,3-propanediol	-2.36036×10^{-1}	9.17477×10^{-1}	1.17135×10^{3}	8.46455×10^{1}	1.47718×10^{-4}	-5.65×10^{-5}
glycerol	−5.46504	8.70127	5.22371×10^{2}	7.25889×10^{1}	3.80078×10^{-2}	6.13×10^{-5}

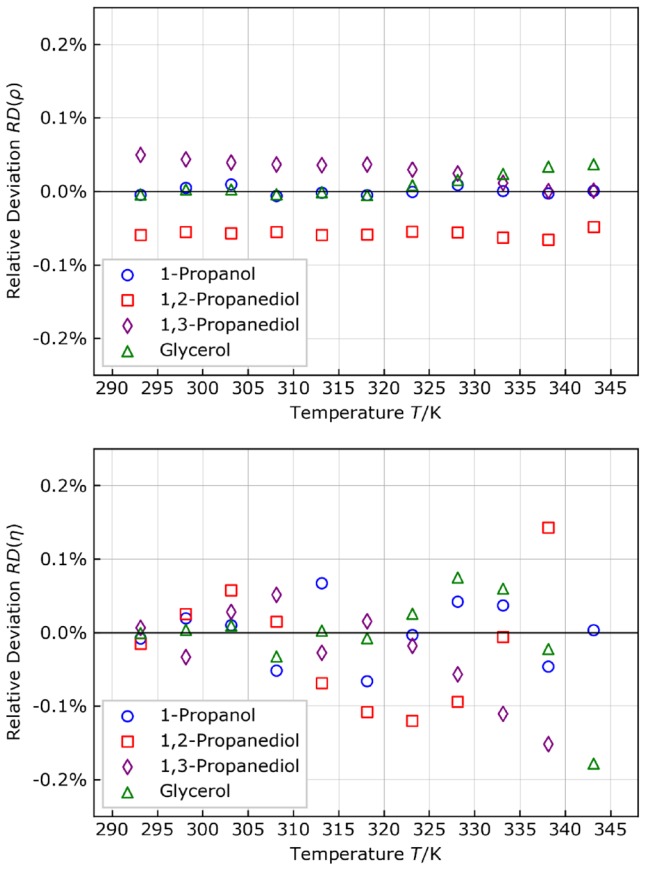

Figure 1. Relative deviation between the pure components' density ρ_i and dynamic viscosity η_i measured in the laboratory and the values calculated by the PPDS equations PPDS10 (eq 3) and PPDS9 (eq 6).

standard set supplied by the manufacturer, Anton Paar. To prevent bubble formation in the apparatus, the samples were carefully injected at 343.15 K.

Density and Excess Molar Volume Calculations. The excess molar volume V_m^E of the samples is defined as the deviation of the mixture's molar volume from the molar volume expected according to ideal mixing. The molar volume V_m of the sample is calculated from the sample's measured density ρ using the molar masses M_i and the molar fractions x_i of its constituents (eq 1).

$$V_m = \frac{\sum_i x_i M_i}{\rho} \tag{1}$$

The excess molar volume is calculated by subtracting the molar volume determined by the assumption of ideal mixing from the measured molar volume of the mixture (eq 2).[8]

$$V_m^E = V_m - \sum \frac{M_i}{\rho_i} x_i \tag{2}$$

The density of the pure substances ρ_i is calculated using the PPDS10 equation (3). Even though parameters for this equation have been published,[17] for substances (1) and (2), the parameters A_i, B_i, C_i, and D_i were fitted to achieve a more satisfactory representation in the relevant temperature range, with $\rho_{c,i}$ and $T_{c,i}$ being the critical density and the critical temperature of the respective substances.

$$\rho_i = \rho_{c,i} + A_i\left(1 - \frac{T}{T_{c,i}}\right)^{0.35} + B_i\left(1 - \frac{T}{T_{c,i}}\right)^{2/3} + C_i\left(1 - \frac{T}{T_{c,i}}\right) + D_i\left(1 - \frac{T}{T_{c,i}}\right)^{4/3} \tag{3}$$

To fit the so-determined excess molar volumes on the isotherms, the Redlich−Kister polynomial was used, in which the polynomial parameters A_i are fitted to describe the binary mixtures' behavior (eq 4).[18]

$$V_m^E = x(1-x)\sum_{i=0}^{n} A_i(2x-1)^i \tag{4}$$

To reduce the number of necessary parameters as well as to enable calculations at varying temperatures, the Redlich−Kister parameters were then fitted to a temperature-dependent polynomial (eq 5).

$$A_i = \sum_{k=0}^{m} a_{i,k} T^k \tag{5}$$

Viscosity Calculations. To describe the temperature dependency of the dynamic viscosity of the pure components, the PPDS9 equation, which is reported to show comparatively high accuracy, was used (eq 6).[19,20]

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Table 5. Measured Values of the Density ρ of the Binary Mixtures at an Ambient Pressure (98 kPa)[a]

		density ρ/kg m^{-3} of the binary mixtures										
x_1	T /K	293.15	298.15	303.15	308.15	313.15	318.15	323.15	328.15	333.15	338.15	343.15
1-Propanol (1) + 1,2-Propanediol (2)												
0.1005		1014.0	1010.6	1006.5	1003.0	998.76	995.21	991.72	986.99	984.01	979.49	975.32
0.1998		990.77	987.15	983.09	980.29	976.24	971.75	968.62	964.44	959.62	955.65	951.83
0.3004		967.96	963.81	959.71	956.66	952.08	948.65	944.67	940.65	935.88	932.06	927.59
0.4000		944.60	940.81	937.27	933.19	929.36	925.09	920.67	917.10	912.39	908.82	904.71
0.5000		921.51	917.08	913.80	908.88	905.31	901.70	897.74	893.13	889.57	885.06	880.41
0.6006		897.44	893.47	889.66	885.20	881.90	877.25	873.75	869.90	865.50	861.45	856.15
0.7003		873.97	870.07	866.33	862.14	858.69	853.71	849.67	845.58	842.04	837.36	832.72
0.8003		850.30	846.27	842.69	838.97	834.89	830.07	826.60	821.78	817.51	813.19	808.61
0.9001		827.46	823.70	818.70	815.64	810.54	806.59	802.59	798.53	793.63	790.08	785.88
1-Propanol (1) + 1,3-Propanediol (3)												
0.1008		1030.1	1026.9	1023.7	1020.5	1017.3	1014.0	1010.8	1007.5	1004.1	1001.0	997.55
0.2001		1006.1	1002.9	999.85	996.55	993.24	989.94	986.54	983.13	979.73	976.22	972.72
0.3002		982.43	979.03	975.72	972.32	968.91	965.51	962.01	958.50	955.00	951.39	947.79
0.3998		957.90	954.49	950.99	947.48	943.88	940.38	936.77	933.17	929.46	925.76	922.05
0.5003		932.67	929.17	925.66	922.06	918.55	914.85	911.25	907.54	903.74	900.03	896.13
0.6004		907.44	903.74	900.03	896.33	892.52	888.82	885.01	881.21	877.30	873.40	869.29
0.6999		881.71	878.01	874.30	870.50	866.69	862.89	858.98	855.08	851.17	847.07	842.86
0.8000		856.18	852.37	848.57	844.66	840.76	836.86	832.85	828.75	824.74	820.54	816.23
0.9000		830.15	826.34	822.44	818.43	814.43	810.32	806.42	802.01	797.51	793.20	788.80
1-Propanol (1) + Glycerol (4)												
0.1004		1219.7	1216.3	1213.0	1209.8	1206.6	1203.2	1200.1	1196.7	1193.3	1189.7	1186.3
0.2009		1174.7	1171.5	1168.2	1165.2	1161.8	1158.1	1155.0	1151.6	1147.9	1145.0	1141.1
0.3004		1129.8	1126.4	1123.0	1119.4	1116.1	1112.7	1108.9	1105.7	1102.2	1098.0	1094.7
0.4006		1083.7	1080.3	1076.5	1073.3	1069.5	1066.0	1062.5	1058.7	1055.0	1051.0	1047.3
0.5005		1038.1	1034.3	1030.5	1027.1	1023.7	1019.4	1015.6	1012.2	1008.1	1004.0	1000.4
0.5991		992.82	989.12	985.67	981.86	978.19	974.27	970.29	966.15	962.45	958.39	954.37
0.7007		946.41	942.66	938.74	935.06	930.92	927.31	923.14	919.20	915.30	910.64	906.51
0.8009		899.86	896.23	892.24	888.58	884.56	880.68	876.42	871.91	868.23	863.68	859.77
0.8992		853.38	848.95	845.26	841.11	837.20	833.43	829.39	825.00	820.44	816.53	812.15
1,2-Propanediol (2) + 1,3-Propanediol (3)												
0.0999		1052.6	1049.1	1046.0	1043.0	1039.9	1036.5	1033.2	1029.9	1026.5	1023.2	1019.8
0.2000		1051.6	1048.4	1045.2	1042.0	1038.7	1035.4	1032.1	1028.8	1025.4	1021.9	1018.5
0.2999		1050.5	1047.3	1044.0	1040.7	1037.4	1034.0	1030.7	1027.3	1023.8	1020.3	1016.8
0.3999		1048.8	1045.6	1042.4	1039.0	1035.7	1032.4	1028.9	1025.5	1022.1	1018.5	1014.9
0.5000		1047.5	1044.2	1040.8	1037.4	1034.0	1030.5	1027.0	1023.4	1019.8	1016.2	1012.6
0.5998		1046.1	1043.0	1039.7	1036.3	1032.8	1029.3	1025.7	1022.2	1018.6	1014.8	1010.9
0.6999		1044.5	1041.1	1037.7	1034.2	1030.6	1027.1	1023.5	1019.8	1016.1	1012.2	1008.2
0.7997		1042.8	1039.3	1035.7	1032.2	1028.5	1024.9	1021.2	1017.4	1013.6	1009.7	1005.6
0.8993		1039.9	1036.3	1032.7	1029.0	1025.3	1021.6	1017.8	1014.0	1010.1	1006.2	1002.2
1,2-Propanediol (2) + Glycerol (4)												
0.1005		1239.7	1237.0	1233.8	1230.7	1227.1	1224.2	1221.0	1217.9	1214.0	1210.7	1207.6
0.2018		1218.2	1215.2	1211.6	1208.7	1205.5	1201.9	1198.7	1195.6	1192.1	1188.6	1185.5
0.3008		1196.3	1193.3	1190.3	1187.2	1183.6	1180.1	1176.8	1173.3	1170.2	1166.9	1163.1
0.4019		1174.0	1170.8	1167.8	1164.4	1161.1	1157.9	1154.6	1150.5	1147.4	1143.6	1140.0
0.5018		1151.6	1148.4	1145.3	1141.7	1138.3	1135.0	1131.5	1127.8	1124.8	1121.1	1117.2
0.6008		1129.2	1126.1	1122.6	1119.2	1115.8	1112.4	1109.3	1105.6	1102.2	1098.4	1094.6
0.7007		1106.5	1103.1	1100.0	1096.0	1092.6	1089.2	1086.2	1082.0	1078.9	1074.8	1071.2
0.7993		1083.3	1080.3	1076.6	1073.6	1069.9	1066.4	1063.1	1059.2	1055.9	1052.3	1048.1
0.8996		1060.6	1057.1	1053.3	1050.2	1046.5	1042.9	1038.9	1035.5	1032.0	1027.8	1024.2
1,3-Propanediol (3) + Glycerol (4)												
0.1006		1241.0	1237.8	1234.2	1231.2	1228.3	1225.1	1221.6	1218.6	1215.1	1212.0	1208.8
0.2007		1220.0	1216.8	1213.6	1210.9	1207.3	1204.7	1201.2	1198.3	1194.8	1191.9	1188.8
0.3010		1200.1	1196.5	1193.5	1190.9	1187.3	1184.6	1181.5	1178.1	1174.6	1171.9	1168.3
0.4002		1179.3	1175.9	1173.3	1170.1	1166.8	1164.0	1160.5	1157.8	1154.2	1151.1	1147.9
0.4999		1158.7	1155.3	1152.1	1149.2	1146.4	1143.1	1140.4	1136.7	1133.6	1130.5	1127.1
0.6002		1138.1	1134.9	1131.9	1128.7	1126.0	1123.0	1119.9	1116.4	1113.4	1110.6	1107.0
0.6987		1117.1	1114.3	1111.0	1108.2	1105.3	1102.3	1099.0	1096.2	1092.9	1089.6	1086.5
0.7999		1095.8	1092.9	1089.7	1087.1	1083.7	1081.0	1077.6	1074.6	1071.4	1068.6	1065.2

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Table 5. continued

density ρ/kg m^{-3} of the binary mixtures												
x_1	T /K	293.15	298.15	303.15	308.15	313.15	318.15	323.15	328.15	333.15	338.15	343.15
0.9004		1074.5	1071.0	1068.0	1065.0	1062.2	1059.5	1056.0	1053.3	1049.8	1046.5	1043.4

[a]Standard uncertainties: $u(T)$ = 0.01 K, $u(x)$ = 6.74 × 10^{-5}, and $u_c(\rho)$ = 0.31 kg m^{-3}.

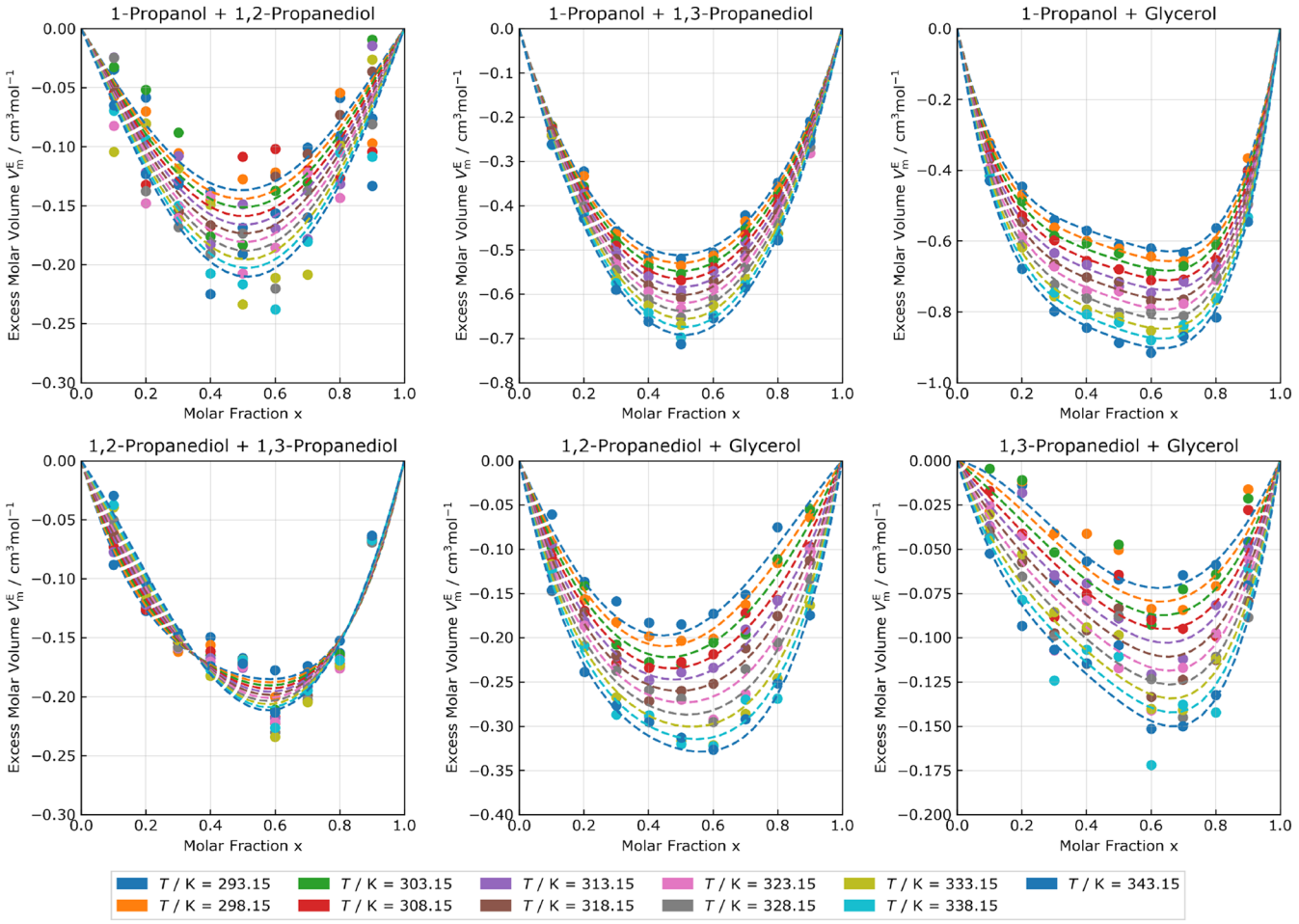

Figure 2. Excess molar volume of the mixtures over the molar fractions at different temperatures. Solid symbols denote the values calculated from measured densities. Dashed lines show the course of the Redlich−Kister temperature-dependent polynomial fitted to the values.

Table 6. Redlich−Kister Parameter Set Fitted to the Molar Excess Volumes Determined from the Mixture Density Data

Redlich−Kister	A_0		A_1		A_2		
binary mixture	a_{01}	a_{00}	a_{11}	a_{10}	a_{21}	a_{20}	MRD
(1) + (2)	−5.84609 × 10^{-3}	1.16556	−1.44190 × 10^{-3}	4.39022 × 10^{-1}	1.96890 × 10^{-4}	7.47562 × 10^{-2}	−5.65 × 10^{-5}
(1) + (3)	−1.44219 × 10^{-2}	2.17949	−3.12226 × 10^{-3}	9.42722 × 10^{-1}	2.42996 × 10^{-3}	−1.05952	−1.50 × 10^{-4}
(1) + (4)	−2.22403 × 10^{-2}	4.12525	−1.11188 × 10^{-3}	−2.50749 × 10^{-1}	−1.61669 × 10^{-2}	2.52908	−8.77 × 10^{-5}
(2) + (3)	−1.85010 × 10^{-3}	−1.86891 × 10^{-1}	−3.59451 × 10^{-3}	9.20255 × 10^{-1}	9.95531 × 10^{-3}	−3.30523	−7.34 × 10^{-6}
(2) + (4)	−1.05196 × 10^{-2}	2.30538	−7.81660 × 10^{-3}	2.50582	−1.63740 × 10^{-2}	4.95029	−9.75 × 10^{-6}
(3) + (4)	−5.57093 × 10^{-3}	1.36875	−1.86813 × 10^{-3}	3.64342 × 10^{-1}	−8.33986 × 10^{-3}	2.49315	−1.60 × 10^{-6}

$$\eta_i = E_i \exp\left(A_i\left(\frac{C_i - T}{T - D_i}\right)^{1/3} + B_i\left(\frac{C_i - T}{T - D_i}\right)^{4/3}\right) \quad (6)$$

The effect of the molar composition on the liquid mixtures' viscosities was fitted using the Grunberg−Nissan mixing rule (eq 7),[21] along all isotherms.

$$\ln \eta_m = \sum_i x_i \ln \eta_i + \frac{1}{2}\sum_i \sum_{j=i} x_i x_j G_{ij} \quad (7)$$

The Grunberg−Nissan mixing rule does not include the temperature dependency of its parameter G_{ij}. To make the method more usable in calculations at varying temperatures, the temperature dependency of the parameter was fitted with eq 8, using polynomial parameters Γ_k.

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$$G_{ij} = \sum_{k=0}^{n} \Gamma_k T^k \tag{8}$$

Parameter Regression. For all regression procedures, an objective function OF corresponding to the average absolute relative deviation was used, as seen in eq 9. For evaluating the accuracy of the description of the mixtures' behavior over the temperature and composition ranges, the mean relative deviation MRD was used (eq 10).

$$\mathrm{OF} = \frac{1}{n} \sum_{i}^{n} \left| \frac{\varepsilon_{\mathrm{exp},i} - \varepsilon_{\mathrm{cal},i}}{\varepsilon_{\mathrm{exp},i}} \right| \tag{9}$$

$$\mathrm{MRD} = \frac{1}{n} \sum_{i}^{n} \frac{\varepsilon_{\mathrm{exp},i} - \varepsilon_{\mathrm{cal},i}}{\varepsilon_{\mathrm{exp},i}} \tag{10}$$

Measurement Uncertainties. For the uncertainty of the molar composition, Gaussian error propagation was applied to the mass-based sample preparation procedure, arriving at an uncertainty of $u(x) = 6.74 \times 10^{-5}$ for the molar compositions of the mixtures. The uncertainty of the density and dynamic viscosity measurements was determined according to NIST TN 1297.[22] For the density values, a combined uncertainty of $u_c(\rho) = 0.31$ kg m^{-3} was determined; for the dynamic viscosity values, a combined relative uncertainty of $u_r(\eta) = 0.01022$ was obtained, taking into account a minimum uncertainty for the viscosity of water as a reference fluid of $u_r(\eta) = 0.01$ reported by Huber et al.[23]

RESULTS AND DISCUSSION

Pure Substances. The measurement results for the densities ρ_i and viscosities η_i of the pure substances are listed in Table 2, together with reference values from the literature. The measured values show satisfactory agreement with the data from the literature.

For the calculation of the molar excess volume of the mixtures, the PPDS10 equation for the density of liquids was used. The parameters published for this equation in Stephan et al.[17] showed satisfactory congruency with our measured pure component data in every case but for 1-propanol, where the equation was refitted to enhance the representation of the substance density in the relevant temperature range. The parameters found by this refitting, as well as the *MRD* values of all parameter sets, are given in Table 3.

For subsequent calculations, the PPDS9 equation for viscosities of saturated liquids, as described in eq 6, was fitted to the pure substance data. The parameters for the pure substances' viscosities are listed in Table 4. As indicated by the values of the MRD, the temperature dependence of dynamic viscosities η_i can be well described by this approach. The accuracy of the pure component parameter fit can also be observed in Figure 1, where the deviations of the substances' calculated dynamic viscosities from the values measured in this work are plotted.

Mixture Density and Excess Molar Volume. The measured results of the density of the mixtures are listed in Table 5. The molar excess volumes V_m^E of the mixtures were calculated from the measured density values according to eq 2 and are plotted in Figure 2.

Data on the mixture density have been previously reported. Li et al.[8] reported the data for all the mixtures with glycerol. Our data show good agreement with the reported trends. For the mixture of 1-propanol + 1,2-propanediol, the most extensive data set has been reported by Zorebski and Przybyla[10] and agrees well with our data. The mixture of 1-propanol + 1,3-propanediol has been investigated by Alavianmehr et al.,[12] and their excess molar volume shows a trend congruent to that of our data. The excess molar volume of a mixture of 1,2-propanediol + 1,3-propanediol was reported by Moosavi et al.[11] Their reported density data agree well with our measurements. However, there is no clear temperature dependence of the excess molar volume, as can be seen from Figure 2. This can be observed in the plot of the mixture's excess molar volume in Figure 2. The lines in the diagrams of Figure 2 present the course of the Redlich−Kister polynomial. All used substances are known to show self-associating behavior due to hydrogen bonding.[27−30] As reported by Usacheva et al.[28] and Jensen et al.,[30] alkanediols and alkanetriols show the formation of network clusters through H bonds, while alkanols with only one hydroxy group form chain-like structures. As can be observed in Figure 2, all mixtures show overassociation. The mixtures containing 1-propanol show excess molar volumes at larger scales than the mixtures of diols and triols. The mixture of 1-propanol and glycerol shows the largest excess molar volume, conceivably because the difference in behavior of the two pure components is the largest, and therefore, the deviation from ideal mixture behavior is also the highest.

For the temperature dependency of its parameters A_i, a polynomial as described in eq 5 with $m = 1$ was used. In Table 6, the values for the temperature-dependent Redlich−Kister polynomial with $n = 1$, $m = 1$ are listed together with the achieved *MRD* for the mixtures. The goodness of fit can be seen by the small values of MRD as well as by the distribution along the line of unity in the deviation diagram in Figure 3. Some deviations remain visible, which could be avoided by extending the temperature polynomial (eq 5) to $m = 2$ or even $m = 3$; however, the resulting increase in accuracy would scarcely justify the increase in the number of mixture parameters.

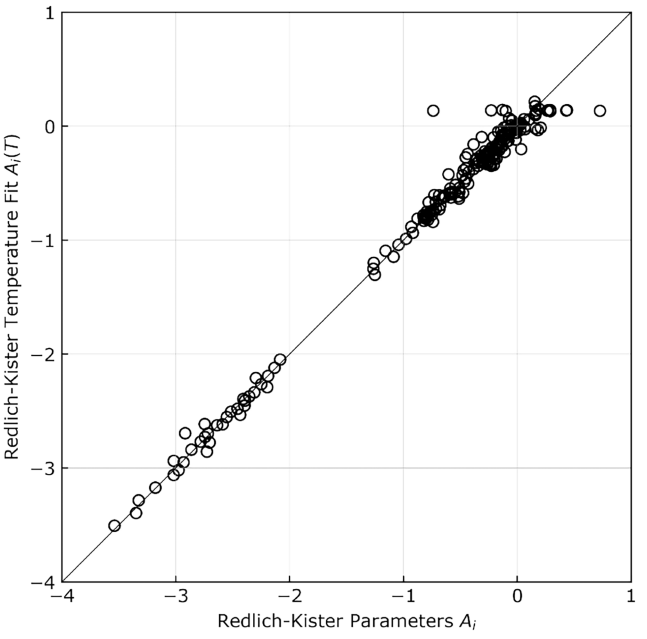

Figure 3. Deviation plot showing the deviation between the Redlich−Kister parameters individually fitted to the isotherms and the Redlich−Kister parameters produced by the temperature polynomial in eq 5.

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Table 7. Measured Values of the Dynamic Viscosity η of the Binary Mixtures at Ambient Pressure (98 kPa)[a]

		viscosity η/mPas of the binary mixtures										
x_1	T /K	293.15	298.15	303.15	308.15	313.15	318.15	323.15	328.15	333.15	338.15	343.15
1-Propanol (1) + 1,2-Propanediol (2)												
0.1005		42.124	31.630	24.261	18.919	15.028	12.027	9.7826	8.0503	6.6983	5.6280	4.7750
0.1998		28.906	22.078	17.268	13.707	11.108	8.9883	7.4096	6.1741	5.1948	4.4098	3.7735
0.3004		20.066	15.681	12.495	10.087	8.2216	6.8121	5.6884	4.7962	4.0792	3.4969	3.0210
0.4000		14.109	11.333	9.1901	7.5378	6.1858	5.2338	4.4231	3.7705	3.2399	2.7996	2.4426
0.5000		9.8489	8.1298	6.7107	5.5968	4.6378	4.0021	3.4301	2.9642	2.5808	2.2620	1.9948
0.6006		6.9910	5.9112	4.9556	4.1919	3.5132	3.0708	2.6559	2.3118	2.0242	1.7811	1.5774
0.7003		5.1217	4.4043	3.7499	3.2152	2.7328	2.4105	2.1058	1.8495	1.6321	1.4476	1.2928
0.8003		3.7805	3.2962	2.8491	2.4767	2.1387	1.9024	1.6801	1.4906	1.3278	1.1867	1.0655
0.9001		2.8518	2.5226	2.2108	1.9468	1.7028	1.5291	1.3626	1.2187	1.0933	0.98214	0.88544
1-Propanol (1) + 1,3-Propanediol (3)												
0.1008		42.876	33.999	27.352	22.242	18.275	15.133	12.648	10.657	9.0475	7.7322	6.6554
0.2001		33.333	26.595	21.565	17.662	14.652	12.173	10.232	8.6664	7.3903	6.3413	5.4671
0.3002		25.696	20.598	16.841	13.898	11.663	9.7087	8.2099	6.9922	5.9929	5.1666	4.4776
0.3998		19.106	15.605	12.893	10.738	9.0004	7.6197	6.4838	5.5521	4.7811	4.1358	3.5869
0.5003		13.734	11.410	9.5291	8.0060	6.7313	5.7871	4.9652	4.2864	3.7166	3.2450	2.8627
0.6004		9.6355	8.1679	6.8946	5.8604	4.9388	4.3146	3.7349	3.2505	2.8424	2.4961	2.2088
0.6999		6.7842	5.8558	5.0009	4.2960	3.6466	3.2222	2.8116	2.4644	2.1684	1.9146	1.6990
0.8000		4.7331	4.1311	3.5615	3.0857	2.6468	2.3500	2.0638	1.8205	1.6105	1.4299	1.2745
0.9000		3.2558	2.8811	2.5229	2.2178	1.9352	1.7349	1.5428	1.3766	1.2314	1.1031	0.99199
1-Propanol (1) + Glycerol (4)												
0.1004		747.08	494.54	335.95	233.80	166.43	121.01	89.738	67.792	52.106	40.701	32.275
0.2009		392.10	267.13	186.42	133.06	96.987	72.101	54.596	42.058	32.925	26.165	21.087
0.3004		207.11	145.18	104.05	76.149	56.824	43.183	33.380	26.217	20.900	16.895	13.835
0.4006		108.91	78.562	57.842	43.410	33.168	25.770	20.338	16.288	13.225	10.875	9.0505
0.5005		57.382	42.592	32.210	24.788	19.391	15.402	12.410	10.134	8.3793	7.0096	5.9281
0.5991		30.486	23.276	18.074	14.258	11.416	9.2677	7.6216	6.3442	5.3409	4.5440	3.9043
0.7007		15.888	12.488	9.9649	8.0647	6.6135	5.4909	4.6117	3.9154	3.3579	2.9071	2.5390
0.8009		8.3546	6.7578	5.5394	4.5974	3.8603	3.2768	2.8099	2.4326	2.1247	1.8713	1.6609
0.8992		4.4472	3.6998	3.1137	2.6489	2.2763	1.9747	1.7282	1.5250	1.3561	1.2147	1.0953
1,2-Propanediol (2) + 1,3-Propanediol (3)												
0.0999		56.415	44.153	35.053	28.154	22.843	18.757	15.536	12.980	10.932	9.2782	7.9311
0.2000		58.386	45.389	35.803	28.591	23.071	18.854	15.549	12.941	10.862	9.1888	7.8396
0.2999		60.006	46.353	36.339	28.863	23.177	18.855	15.488	12.845	10.747	9.0650	7.7088
0.3999		61.117	46.914	36.588	28.912	23.115	18.728	15.323	12.666	10.564	8.8855	7.5347
0.5000		61.984	47.309	36.696	28.845	22.932	18.458	15.085	12.464	10.403	8.7616	7.3841
0.5998		62.516	47.316	36.596	28.691	22.748	18.276	14.825	12.238	10.135	8.4536	7.1056
0.6999		62.865	47.256	36.370	28.260	22.292	17.885	14.456	11.820	9.7650	8.1456	6.8564
0.7997		63.043	46.946	35.789	27.704	21.775	17.331	13.971	11.399	9.4043	7.8386	6.5886
0.8993		62.467	46.307	35.007	26.933	21.045	16.704	13.435	10.943	9.0123	7.5012	6.3041
1,2-Propanediol (2) + Glycerol (4)												
0.1005		1110.8	718.49	478.04	326.62	228.78	163.90	120.03	89.634	68.142	52.680	41.326
0.2018		852.95	557.49	374.51	258.23	182.40	131.75	97.198	73.117	55.958	43.529	34.374
0.3008		657.68	434.30	294.70	205.12	146.25	106.50	79.234	60.038	46.277	36.224	28.894
0.4019		487.55	326.05	223.94	157.66	113.55	83.586	62.767	47.983	37.267	29.399	23.612
0.5018		355.98	242.21	168.87	120.50	87.747	65.265	49.385	37.972	29.614	22.579	18.228
0.6008		255.84	176.33	124.49	89.928	66.266	49.924	38.261	29.810	23.571	18.899	15.341
0.7007		179.35	125.61	90.062	65.992	49.260	37.597	29.145	22.945	18.320	14.820	12.126
0.7993		128.27	91.267	66.409	49.332	37.289	28.804	22.572	17.949	14.467	11.808	9.7453
0.8996		90.986	65.798	48.619	36.635	28.068	21.936	17.381	13.962	11.357	9.3463	7.7782
1,3-Propanediol (3) + Glycerol (4)												
0.1006		872.23	575.45	389.76	270.92	192.81	140.20	104.04	78.699	60.571	47.362	37.610
0.2007		620.81	418.75	289.59	205.09	148.46	109.69	82.565	63.243	49.240	38.897	31.273
0.3010		462.94	317.96	224.07	161.28	118.44	88.676	67.548	52.323	41.139	32.791	26.469
0.4002		326.95	230.12	165.47	121.36	90.623	68.947	53.279	41.791	33.233	26.763	21.802
0.4999		233.94	168.29	123.42	92.191	69.970	54.089	42.395	33.694	27.120	22.081	18.172
0.6002		162.26	119.48	89.532	68.215	52.697	41.442	32.980	26.573	21.655	17.832	14.817
0.6987		121.54	91.085	69.359	53.829	42.162	33.587	27.039	22.018	18.116	15.051	12.619
0.7999		93.809	71.473	55.282	43.360	34.394	27.721	22.555	18.539	15.386	12.884	10.879
0.9004		70.783	54.863	43.130	34.333	27.617	22.530	18.537	15.396	12.898	10.889	9.2738

[a]Standard uncertainties: $u(T) = 0.01$ K, $u(x) = 6.74 \times 10^{-5}$, and $u_r(\eta) = 0.01022$.

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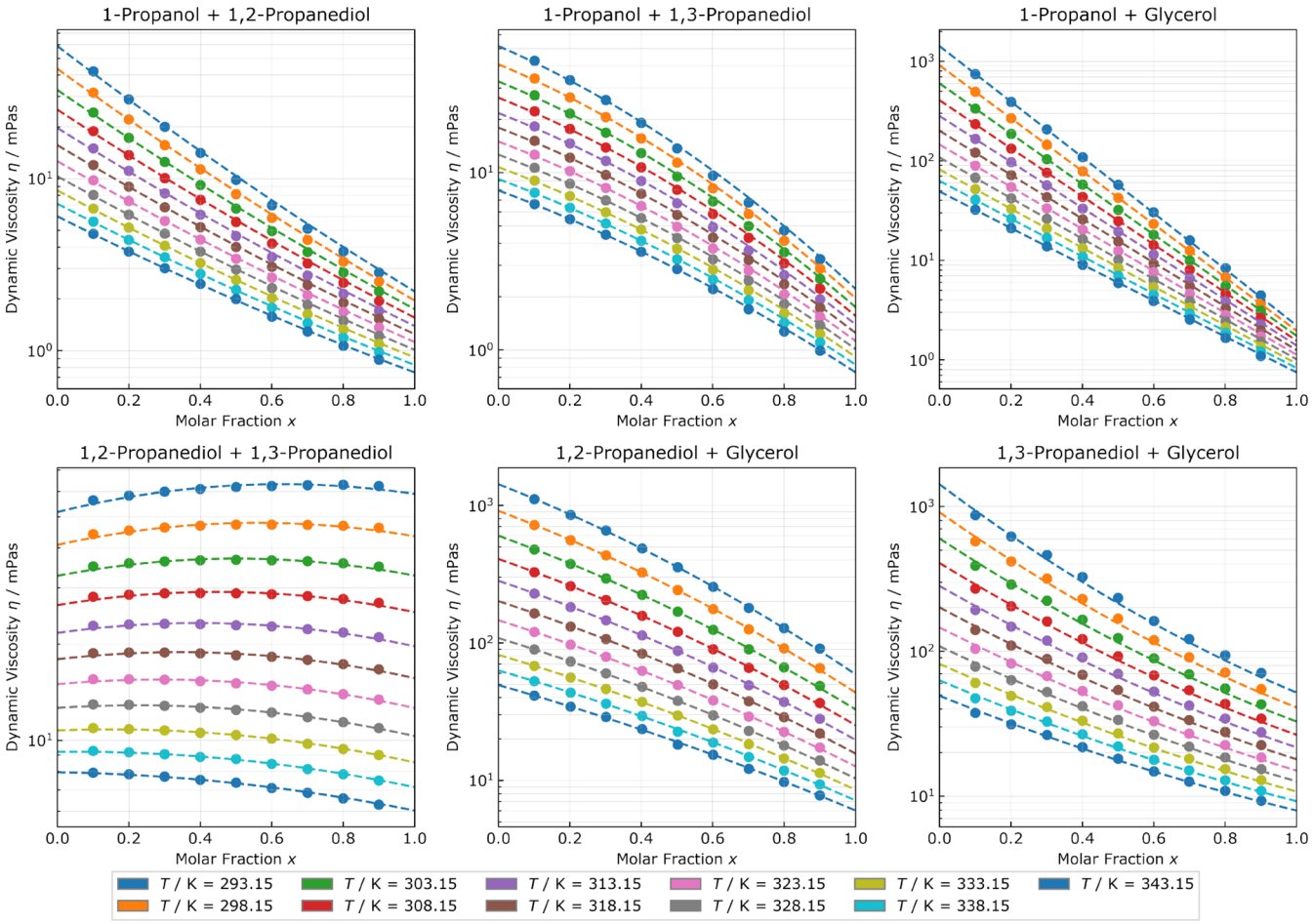

Figure 4. Dynamic viscosity of the binary mixtures over the composition in molar fractions at temperatures from 293.15 to 343.15 K. Solid symbols denote the measured values. Dashed lines indicate the course of the Grunberg−Nissan mixing rule fitted to the experimental data.

Using the determined Redlich−Kister parameter set, the deviation from the reported literature data was calculated. For the system of 1-propanol + 1,2-propanediol, a deviation of MRD = 0.07% from the data of Zorebski and Przybyla[10] was found, also MRD = 0.07% from the data by Zarei et al.[9] For the system of 1-propanol + 1,3-propanediol, the deviation is MRD = 0.10% from data by Alavianmehr et al.[12] For the system of 1-propanol + glycerol, a deviation of MRD = 0.09% was found for the data of Vicente et al.[31] In the case of 1,2-propanediol + 1,3-propanediol, a deviation of MRD = 0.12% from the data of Moosavi et al.[11] was determined; for the data of Rane et al.,[32] MRD = 0.10%. For the data of the system 1,3-propanediol + glycerol by Amireche and Hernández,[33] an MRD = 0.08% was achieved. Using the data of Li et al.[8] for 1-propanol + glycerol, an MRD = 0.12% was found; for 1,2-propanediol + glycerol, MRD = 0.04%, and for 1,3-propanediol + glycerol, MRD = 0.07%. In the case of the 1-propanol + 1,2-propanediol data by Pal et al.,[34] values of V_m^E were reported, for which MRD = 9.35% was found. Deviation plots for all literature data can be found in the Supporting Information.

Mixture Viscosity. In Table 7, the measured values of dynamic viscosity η are listed. The dynamic viscosities of the present binary mixtures have not been as extensively investigated as the mixtures' densities and excess molar volumes. Prior reports include the dynamic viscosity of 1-propanol + glycerol at temperatures from 133.15 to 243.15 K with four molar fractions by Kono et al.,[13] of 1,2-propanediol + 1,3-propanediol at 298.15, 303.15, and 308.15 K by Moosavi et al.,[11] and of 1,2-propanediol + glycerol from 298.15 to 363.15 K by Spann et al.[14] The results presented by Kono et al.[13] for 1-propanol + glycerol are too far out of the temperature range investigated in this work to allow in depth comparison, but seem to reflect a similar trend as found in our measurements. The data reported for 1,2-propanediol + 1,3-propanediol do not fully agree with our measured results. The trends appear to be similar, while the measured values show some offset. However, this may be the result of the higher water content of the pure substances used by Moosavi et al.[11] Spann et al.[14] reported dynamic viscosity data for 1,2-propanediol + glycerol at only one 1,2-propanediol mass fraction of $w_2 = 0.6$, which corresponds to a molar fraction of $x_2 = 0.645$. The reported values fit well within the behavior described by our data.

In Figure 4, the dynamic viscosity data are plotted over the composition range, grouped by temperature. From these graphs, some interesting observations can be made; for example, the close to log−linearity of the dynamic viscosity over the composition in the 1-propanol + glycerol mixture, as well as a slight but observable nonideality of the 1,2-propanediol + 1,3-propanediol mixture's behavior. Another interesting phenomenon is observed regarding the deviation of the mixtures' dynamic viscosity from logarithmic ideal mixing. Such behavior would appear as linear connections of the pure substance's dynamic viscosity in the logarithmically scaled graphs of Figure 4. Interestingly, the mixtures of 1-propanol + 1,2-propanediol

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Table 8. Grunberg−Nissan Parameter Set Fitted to the Molar Excess Volumes Determined from the Mixture Density Data

Grunberg−Nissan	G_{12}			
binary mixture	Γ_0	Γ_1	Γ_2	MRD
(1) + (2)	−3.72593	1.99456×10^{-2}	-2.91200×10^{-5}	-1.50×10^{-2}
(1) + (3)	5.22929	-1.69414×10^{-2}	1.02817×10^{-5}	-1.53×10^{-2}
(1) + (4)	5.09910	-2.97938×10^{-2}	4.30122×10^{-5}	-1.20×10^{-2}
(2) + (3)	−7.52815	5.54816×10^{-2}	-9.57472×10^{-5}	2.49×10^{-3}
(2) + (4)	4.60279	-1.63957×10^{-2}	1.20454×10^{-5}	1.01×10^{-3}
(3) + (4)	-1.70582×10^{1}	9.35429×10^{-2}	-1.31600×10^{-4}	2.19×10^{-2}

and 1,3-propanediol + glycerol show the same sign in their deviation. This is also the case for 1-propanol + 1,3-propanediol and 1,2-propanediol + glycerol. This is remarkable because in both cases, the mixtures' constituents differ in the existence of one OH^- group. In the first case, the difference between the two mixing constituents is an OH^- group on the C_2 atom. In the second case, the differing OH^- group is on the C_3 atom. This may indicate that the position of the OH^- group has some influence on the branching of the mixture's H-bond network structures.

The parameters for the Grunberg−Nissan mixing rule are listed in Table 8. As can be observed by the values of the *MRD*, as well as by the deviation plot in Figure 5, the mixing rule is able to

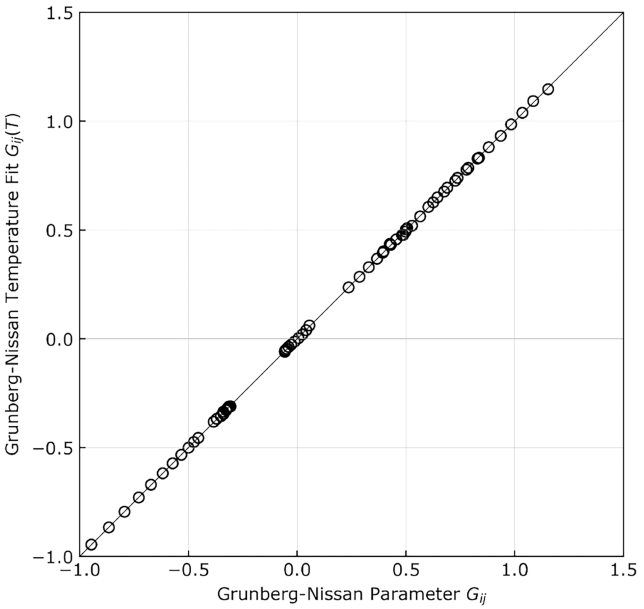

Figure 5. Deviation diagram illustrating the goodness of fit of the Grunberg−Nissan parameters' temperature polynomial.

describe the binary mixtures' viscosity with reasonable accuracy. As visible in the diagram, the temperature polynomial (eq 8) with $n = 2$ provides a very accurate fit for G_{12}.

The newly determined Grunberg−Nissan parameters were then used to determine mean relative deviations from the data published by Moosavi et al.[11] for which MRD = 5.65% and Spann et al.[14] for which MRD = 9.30%. This indicates that for the 1,2-propanediol + 1,3-propanediol system investigated by Moosavi et al.,[11] the reduced ambient pressure may have had some effect on the viscosity of the mixture. Regarding the mixture of 1,2-propanediol and glycerol, the deviation is possibly explained by the usage of a cone−plate rheometer by Spann et al.[14]; however, it is hard to reach a conclusion, as the viscosity of only one molar fraction was reported. Plots of the deviation from the literature data are included in the Supporting Information.

CONCLUSIONS

The density and dynamic viscosity of the binary systems of 1-propanol (1), 1,2-propanediol, 1,3-propanediol, and glycerol were measured and reported. In the case of the density, the binary mixtures have been investigated before; nevertheless, our data sets serve to clarify the present understanding of the mixture's behavior, as well as extend the covered temperature ranges for some mixtures. In the case of the dynamic viscosity, novel data sets were presented. Except for the systems of 1,2-propanediol + 1,3-propanediol and 1,2-propanediol + glycerol, where small portions of our region of focus had been covered, the data sets presented here provide first information on the binary mixtures' dynamic viscosity between 293.15 and 343.15 K. Using the measured density data, a set of parameters for the Redlich−Kister polynomial was generated to enable the description of the excess molar volume of the mixtures. Using the dynamic viscosity data, the Grunberg−Nissan mixing rule was fitted. The resulting parameter sets were reported, and their accuracy was deemed satisfactory. The Grunberg−Nissan mixing rule provided a representation of the mixtures' dynamic viscosity using only three fitted parameters. At a maximum MRD of 1.10% in the case of 1-propanol + glycerol, lower for others, the Grunberg−Nissan mixing rule is well below the expected error of 5% for polar mixtures.[21]

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.jced.5c00274.

Deviation of the newly provided parameters from all cited literature data (PDF)

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Author Contributions

M.V.: investigation, data acquisition, and drafting of the original manuscript. S.L.: supervision, data curation, writing, reviewing, and editing. T.W.: supervision, data curation, writing, reviewing, and editing.

Notes

The authors declare no competing financial interest.

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