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Article

Thermodynamic Description of Aqueous Mixtures of New Antifreezes

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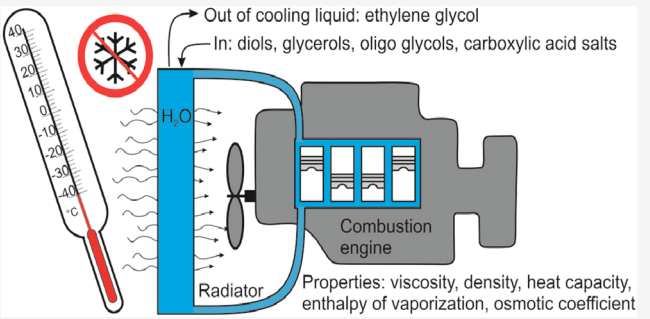
ABSTRACT: Combustion engines for motor vehicles will still play a substantial role for many years further. As a consequence of the power increase leading to high local surface temperatures of up to 260 °C in a modern engine, the requirements on coolants regarding stability and ability to maintain the dissipation of heat are also increased. In search for possible alternatives to currently applied freezing point depressants (coolants) and to create an overview about antifreezes in use, thermodynamic properties, namely, freezing points, densities, viscosities, specific isobaric heat capacity, boiling point curve data, and osmotic coefficient values were measured, generalized, and systemized with interpolated data from the literature for aqueous solutions of four molecule classes studied in wide temperature (from −45 to 95 °C) and composition ranges and are given as a database. The nonaqueous components are diols: 1,2-propanediol and 1,3-butanediol; glycols: ethylene glycol, diethylene glycol, triethylene glycol, and tetraethylene glycol; glycerols: glycerol and diglycerol; and carbonic acid salts: potassium acetate, sodium propionate, and potassium propionate. Conclusions have been drawn about the applicability of liquids with the best flow behavior and thermal properties as antifreezes, which fulfill the technical requirements but also those of sustainable chemistry including low toxicity, efficiency, and cheapness.



1. INTRODUCTION

Freezing point depressants are used in a multitude of applications. In wintertime, basic rock salt is used to deice roads, and antifreeze solutions keep the windscreens from freezing. An additional automotive application is the use of heat transfer fluids for cooling internal combustion engines to prevent excessive wear or cracking. The freezing point depressant prevents the coolant from solidifying and damaging the engine when low temperatures prevail. In most liquid-cooled engines, aqueous solutions of ethylene glycol (EG) are applied. In addition to a depressant, major components are corrosion inhibitors and pH stabilizers.[1,2]

The combustion inside an engine produces a vast amount of heat. The coolant carries surplus heat to a radiator, where the heat is transferred to the ambient air. In traditional engines, combustion leads to wall temperatures of the cooling jacket of up to 160 °C.[3] Due to the rising performance in modern engines, the thermal loads are increased, and hotspots of about 260 °C are reported.[4] At these temperatures, EG is not stable anymore and starts to degrade to carboxylic acids, which decreases the pH and promotes corrosion.[5] To counter the corrosiveness, additional inhibitors and pH stabilizers must be added to the mixture. However, the additives are partially consumed over time, and the coolant has to be replaced in regular intervals.[6,7]

The frequent exchange of EG containing coolants has several drawbacks. Its toxicity poses a health threat to the operator.[8,9] Especially since the exchange of the coolant mixture can be performed by laymen. Further, as a petroleum-based compound, it is not of a sustainable nature.[8,10−12]

Due to these shortcomings, a thermally more stable, less toxic, and sustainable antifreeze component is desirable. The alternatives should achieve a freezing point reduction similar to that of EG and meet the following requirements: the antifreeze should not decompose under working conditions. Neither the possible decomposition products nor itself should be corrosive to the building material of the engine. For proper flow behavior, a low-viscosity coolant solution is needed. High heat capacity and high thermal conductivity are favorable for efficient heat exchange. To achieve a fast heat-up of the engine, it would be beneficial if the cooling mixture had a lower specific heat capacity at ambient temperature and a higher one at elevated temperatures. Moreover, the antifreeze should be

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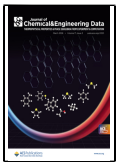

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Table 1. Chemicals Used in This Work[a]

chemical name	CAS reg. no.	M/g mol^{-1}	Source	mass fraction, provided by the suppliers
ethylene glycol	107−21−1	62.07	Sigma-Aldrich	≥0.990
diethylene glycol	111−46−6	106.12	Sigma-Aldrich	0.985
triethylene glycol	112−27−6	150.17	Sigma-Aldrich	≥0.990
tetraethylene glycol	112−60−7	194.23	Sigma-Aldrich	0.990
1,2-propanediol	57−55−6	76.09	Carl-Roth	0.995
1,3-butanediol	107−88−0	90.12	Carl-Roth	0.990
glycerol	56−81−5	92.09	Sigma-Aldrich	0.990
diglycerol	627−82−7	166.17	TCI Europe	0.800[b]
potassium acetate	127−08−2	98.14	Sigma-Aldrich	0.990
sodium propionate	137−40−6	96.06	Sigma-Aldrich	0.990
potassium propionate	327−62−8	112.17	TCI Europe	0.980
Millipore water	7732−18−50	18.02		

[a]All chemicals have been dried and stored over anhydrous silica gel in a nitrogen atmosphere. [b]This work: HPLC-ESI-MS confirmed that a diglycerol content of 80% and triglycerols were identified to be the main components of the remaining 20%; GC-MS revealed a 3:1 ratio for two diglycerol isomers, which could not be further identified; the third diglycerol isomer and monomeric glycerol occurred only in trace amounts.

Table 2. Values of Freezing Point, T_f, of Glycol Aqueous Solutions at Pressure P = 100 kPa[a]

ethylene glycol		diethylene glycol		triethylene glycol		tetraethylene glycol	
m/mol kg^{-1}	T_f/°C	m/mol kg^{-1}	T_f /°C[b]	m/mol kg^{-1}	T_f/°C	m/mol kg^{-1}	T_f/°C
1.9989	−4.03	1.9995	−6.62	2.0000	−5.20	1.0000	−2.52
3.9979	−8.29	3.9996	−14.28	3.9985	−12.76	1.9996	−5.99
5.9547	−12.79	5.9984	−22.53	5.9993	−21.39	3.9991	−14.63
7.9963	−17.71	8.0001	−29.21	7.9992	−30.35	5.9962	−26.26
9.9983	−22.30	10.0000	−37.32	9.9874	−40.12	7.9986	−40.43
12.0414	−26.92	11.9888	−45.00				
13.9897	−31.26						
15.9988	−36.48						
17.6256	−40.20						

[a]Standard uncertainties u are $u(m)$ = 0.00003 mol/kg and $u(T_f)$ = 0.01 °C. [b]$u(T_f)$ = 4 °C, $u(P)$ = 1 kPa.

nontoxic, non-inflammable, readily available, and originate from a regenerative source.[13]

An already known alternative is propylene glycol. It is less toxic but also based on petroleum.[14,15] Due to the so far successful application of EG, either alternatives are not investigated, or there is no knowledge about their physicochemical properties and corrosion behavior. Thus, potentially relevant alternatives and the physicochemical properties of their aqueous solutions should be studied.

In this study, we aim to give an overview of the applicability of several possible and known compounds. In the first step, the candidates are studied with respect to their capabilities to reduce the freezing point of an aqueous solution to −40 °C, and the concentration needed to reach this temperature is determined. Aqueous solutions of four molecule classes are studied in a wide temperature and composition ranges, namely, diols: 1,2-propanediol (propylene glycol, ProDiol) and 1,3-butanediol (1,3-dihydroxybutane, BuDiol); glycols: ethylene glycol (EG), diethylene glycol (1,5-dihydroxy-3-oxapentane, DiEG), triethylene glycol (1,8-dihydroxy-3,6-dioxaoctane, TriEG) and tetraethylene glycol (1,11-dihydroxy-3,6,9-trioxadecane, TeEG); glycerols: glycerol and diglycerol (4-oxaheptane-1,2,6,7-tetrol); and carbonic acid salts: potassium acetate (KAc), sodium propionate (NaPro) and potassium propionate (KPro). These compounds are widely industrially used, e.g., tri- and tetraethylene glycol in separation processes such as the removal of water from natural gas.[16,17] Diols can promote micellization[18,19] and could stabilize the protein structure in water.[20] Diols and glycols are used in the design of separation processes and connection of these systems to biological processes.[21]

As a following step, the prepared solutions are checked for important properties like density, viscosity, osmotic coefficients, and specific heat capacity over wide temperature and composition ranges. Densities and viscosities are measured at m = (2 to 14) mol kg^{-1} for the solutions of the diols and glycols and at m = (2 to 5) mol kg^{-1} for the solutions of the salts in the temperature range from −35 to 95 °C; osmotic coefficient values at m = (2 to 24) mol kg^{-1} for aqueous solutions of the diols and glycols and at m = (0.002 to 5) mol kg^{-1} for the solutions of the salts are measured at a temperature of 25 °C and supplemented with data from the literature. Osmotic coefficient values are given at 25 °C, except for TriEG (t = 24.45 °C) and TetEG (t = 24.95 °C). Specific isobaric heat capacity and isobaric heat capacity per unit volume values are determined for m = (2 to 14) mol kg^{-1} for the solutions of the diols and glycols and at m = (2 to 5) mol kg^{-1} for the solutions of the salts in the temperature range from 25 to 95 °C. From the data obtained, the thermal conductivity can be calculated. The pressure dependence of the boiling point is determined, from which the specific heat of vaporization is calculated. Finally, we draw conclusions on the applicability of liquids with the best flow behavior and thermal properties as antifreezes.

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Table 3. Values of Freezing Point, T_f, of Polyol Aqueous Solutions at Pressure P = 100 kPa[a]

1,2-propanediol		1,3-butanediol		glycerol		diglycerol	
m/mol kg^{-1}	T_f/°C	m/mol kg^{-1}	T_f/°C	m/mol kg^{-1}	T_f/°C	m/mol kg^{-1}	T_f/°C
1.9998	−3.98	2.9990	−6.54	0.9994	−1.93	1.9996	−4.60
3.9958	−8.99	5.9991	−13.71	1.9979	−3.97	4.0002	−10.50
5.9968	−14.85	8.9981	−21.27	2.9978	−6.30	5.9989	−16.60
7.9993	−20.12	11.9964	−27.68	3.9988	−8.54	7.9956	−23.60
10.0002	−26.42	14.9855	−33.39	4.9996	−11.03	9.9888	−31.00
11.9923	−30.76	17.5244	−40.20	5.9983	−13.44	11.9349	−40.40
13.9894	−36.70			7.9974	−18.10		
14.9298	−40.20			10.0014	−23.79		
				11.9989	−28.41		
				13.9879	−32.53		
				15.9936	−36.53		
				17.1421	−40.40		

[a]Standard uncertainties u are $u(m)$ = 0.00003 mol/kg, $u(T_f)$ = 0.01 °C, and $u(P)$ = 1 kPa.

2. EXPERIMENTAL METHODS AND SOURCES OF RELIABLE THERMODYNAMIC DATA

2.1. Materials

1,2-Propanediol 99.5% and 1,3-butanediol 99% were purchased from Carl-Roth. Ethylene glycol (1,2-ethanediol) 99%, diethylene glycol

Table 4. Values of Freezing Point, T_f, of Carbonic Acid Salt Aqueous Solutions at Pressure P = 100 kPa[a]

potassium acetate		sodium propionate		potassium propionate	
m/mol kg^{-1}	T_f/°C	m/mol·kg^{-1}	T_f/°C	m/mol·kg^{-1}	T_f/°C
0.9998	−3.87	0.9999	−4.11	1.0000	−4.42
2.0000	−8.72	2.0001	−9.11	1.9993	−9.89
2.991	−14.38	2.9996	−16.66	2.9991	−16.34
3.9938	−21.02	3.9984	−23.96	3.9977	−24.33
4.9983	−28.05	4.9871	−32.71	4.9978	−32.74
5.9979	−36.41	5.9989	−40.00	5.6901	−40.00
6.3722	−40.10			6.0003	−43.52

[a]Standard uncertainties u are $u(m)$ = 0.00003 mol/kg, $u(T_f)$ = 0.01 °C, and $u(P)$ = 1 kPa.

98.5%, glycerol 99.5%, potassium acetate 99.0%, sodium propionate 99.0%, triethylene glycol 99%, and tetraethylene glycol 99% were obtained from Sigma-Aldrich. Diglycerol ≥80.0% and potassium propionate 98.0% were acquired from TCI Europe. All chemicals were used as received. KPro solutions are dried and stored under anhydrous silica gel in a nitrogen gas atmosphere. Water content in EG is determined coulometrically by Karl Fischer titration with the use of an 899 Coulometer of Metrohm. (water content equals 0.0554% (σ = 0.0005, δ = 0.9%)). The suppliers and purities are given in Table 1. The solutions were prepared on the basis of Millipore water with a specific conductivity of $5.5 \cdot 10^{-8}$ S cm^{-1} by weighing on a Mettler Toledo XS105 balance (max 40 g) with an accuracy of 0.00001 g or Scaltech (max 220 g) with an accuracy of 0.0001 g or Sartorius, M-power, Mettler Toledo PJ 3000, dependent on the solution amount. The resulting solutions were transparent and protected from CO_2 exposure.

2.2. Freezing Point

The freezing point was determined by a method closely resembling ASTM D-1177−22 (International Standard "Standard Test Method for Freezing Point of Aqueous Engine Coolants"), covering the test method for the determination of the freezing point of a coolant solution in the laboratory. The test method involves the determination of the time−temperature curve prior to freezing and the determination of the horizontal or flattened portion of the freezing curve. The freezing point is taken as the intersection of projections of the cooling curve and freezing curve. If the solution supercools, the freezing point is the

Table 5. Required Mole Fraction x_2 and Mass Fraction ω to Achieve a Freezing Point of around −40 °C in Aqueous Solution[a,b]

compound	x_2	ω	T_f/°C	T_b/°C	$\overline{\Delta_{vap}h}$/ kJ mol^{-1}
ethylene glycol	0.241	0.522	−40.2	109.3	43.6
diethylene glycol	0.178	0.560	−45.0	109.8	42.7
triethylene glycol	0.152	0.600	−40.7	106.2	43.4
tetraethylene glycol	0.126	0.610	−40.3	105.4	43.9
1,2-propanediol	0.212	0.532	−40.2	105.9	43.1
1,3-butanediol	0.240	0.613	−40.2	106.3	44.4
glycerol	0.236	0.613	−40.4	110.7	43.0
diglycerol	0.177	0.665	−40.4	108.4	43.9
potassium acetate	0.103	0.384	−40.1	109.8	43.9
sodium propionate	0.093	0.389	−40.0	107.1	44.0
potassium propionate	0.096	0.361	−40.6	107.6	44.2

[a]The actual freezing point T_f and boiling point T_b at pressure P = 100 kPa, and the appertaining molar heat of vaporization $\overline{\Delta_{vap}h}$ for the aqueous solutions. [b]Standard uncertainties u are $u(m)$ = 0.00003 mol/kg, $u(T_f)$ = 0.01 °C, $u(T_b)$ = 0.01 °C, $u\left(\overline{\Delta_{vap}h}\right)$ = 0.1 kJ·mol^{-1}, and $u(P)$ = 1 kPa.

maximum temperature reached after supercooling.[22] Solutions were cooled under intensive stirring using a dry ice−ethanol cooling mixture, while temperature was measured with an accuracy of 0.01 K by the use of a KT 1000 Precision Thermometer PT 100−4 from Technische Werkstätte Schrödle (Germany). Temperature calibration was performed with water and aqueous sodium chloride solutions of two concentrations. The measurements were repeated at least three times. The mean freezing points at the required concentrations are given in Tables 2, 3, and 4. An overview of the compounds studied due to compositions (mole fraction x and mass fraction ω) to achieve a freezing point around −40 °C in aqueous solution and of respective boiling points is given in Table 5.

2.3. Density

The density measurements were performed using an Anton Paar oscillating tube densitometer. A DMA 5000 M was used for the measurements from 5 to 85 °C. The temperature was controlled with an accuracy of 0.01 K. For density determination, five measurements were carried out in each case, and the mean value of the results was then calculated. Water and air were selected as the calibration substances. The density values are given in Tables 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, and 16.

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Table 6. Density, ρ, of the Aqueous Solutions Given Minimal Freezing Point (Compositions Shown in Table 5) at Temperatures from 5 to 95 °C and Pressure P = 100 kPa[a]

	ρ/g cm^{-3}					
temperature/°C	5.0	25.0	45.0	65.0	85.0	95.0
ethylene glycol	1.080	1.065	1.053	1.039	1.024	1.017
diethylene glycol	1.097	1.081	1.068	1.053	1.036	1.028
triethylene glycol	1.106	1.089	1.074	1.059	1.043	1.033
tetraethylene glycol	1.111	1.093	1.078	1.063	1.046	1.036
1,2-propanediol	1.054	1.037	1.023	1.007	0.989	0.981
1,3-butanediol	1.036	1.018	1.004	0.989	0.973	0.964
glycerol	1.169	1.155	1.143	1.130	1.115	1.108
diglycerol	1.197	1.181	1.169	1.156	1.142	1.133
potassium acetate	1.216	1.203	1.193	1.181	1.167	1.160
potassium propionate	1.175	1.157	1.144	1.130	1.115	1.106

[a]Standard uncertainties u are $u(\rho)$ = 0.002 g cm^{-3}, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 7. Ethylene Glycol: Density, ρ, of Aqueous Solutions with Different Molalities from 5 to 85 °C and Pressure P = 100 kPa[a]

Temp./°C	5.0	15.0	25.0	35.0	45.0	55.0	65.0	75.0	85.0
m/mol kg^{-1}	ρ/g cm^{-3}								
1.9989	1.016	1.015	1.012	1.008	1.004	0.998	0.994		
3.9979	1.028	1.026	1.022	1.018	1.013	1.008	1.003		
5.9547	1.039	1.036	1.032	1.027	1.022	1.017	1.010		
7.9963	1.048	1.044	1.040	1.035	1.030	1.024	1.018		
9.9983	1.056	1.052	1.047	1.042	1.036	1.030	1.024	1.017	
12.0414	1.062	1.058	1.053	1.047	1.041	1.035	1.029	1.022	
13.9897	1.067	1.062	1.057	1.051	1.046	1.039	1.033	1.026	
15.9988	1.072	1.067	1.062	1.056	1.050	1.043	1.036	1.029	1.020

[a]Standard uncertainties u are $u(m)$ = 0.00003 mol/kg, $u(\rho)$ = 0.002 g cm^{-3}, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 8. Diethylene Glycol: Density, ρ, of Aqueous Solutions with Different Molalities from 5 to 85 °C and Pressure P = 100 kPa[a]

Temp./°C	5.0	15.0	25.0	35.0	45.0	55.0	65.0	75.0	85.0
m/mol kg^{-1}	ρ/g cm^{-3}								
1.9995	1.025	1.023	1.020	1.016	1.012	1.006	1.001		
3.9996	1.044	1.040	1.036	1.031	1.026	1.021	1.015		
5.9984	1.058	1.053	1.049	1.043	1.038	1.032	1.025	1.018	
8.0001	1.068	1.063	1.058	1.052	1.046	1.039	1.033	1.025	
10.0000	1.076	1.071	1.065	1.059	1.053	1.046	1.040	1.033	1.025
11.9888	1.082	1.076	1.070	1.064	1.058	1.051	1.044	1.037	1.029

[a]Standard uncertainties u are $u(m)$ = 0.00003 mol/kg, $u(\rho)$ = 0.002 g cm^{-3}, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 9. Triethylene Glycol: Density, ρ, of Aqueous Solutions with Different Molalities from 5 to 85 °C and Pressure P = 100 kPa[a]

Temp./°C	5.0	15.0	25.0	35.0	45.0	55.0	65.0	75.0	85.0
m/mol kg^{-1}	ρ/g cm^{-3}								
2.0000	1.039	1.036	1.032	1.028	1.023	1.017	1.011	1.005	
3.9985	1.065	1.060	1.055	1.050	1.044	1.037	1.031	1.024	
5.9993	1.082	1.077	1.071	1.064	1.058	1.051	1.044	1.036	1.028
7.9992	1.094	1.088	1.081	1.075	1.068	1.060	1.053	1.045	1.037
9.9874	1.102	1.096	1.089	1.082	1.074	1.067	1.059	1.051	1.044

[a]Standard uncertainties u are $u(m)$ = 0.00003 mol/kg, $u(\rho)$ = 0.002 g cm^{-3}, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 10. Tetraethylene Glycol: Density, ρ, of Aqueous Solutions with Different Molalities from 5 to 85 °C and Pressure P = 100 kPa[a]

Temp./°C	5.0	15.0	25.0	35.0	45.0	55.0	65.0	75.0	85.0
m/mol kg^{-1}	ρ/g cm^{-3}								
1.9996	1.049	1.046	1.041	1.036	1.031	1.025	1.019	1.012	
3.9991	1.079	1.073	1.067	1.061	1.055	1.048	1.041	1.033	
5.9962	1.096	1.090	1.083	1.076	1.069	1.062	1.054	1.046	1.038
7.9986	1.107	1.100	1.093	1.085	1.078	1.070	1.062	1.054	1.046

[a]Standard uncertainties u are $u(m)$ = 0.00003 mol/kg, $u(\rho)$ = 0.002 g cm^{-3}, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

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Table 11. 1,2-Propanediol: Density, ρ, of Aqueous Solutions with Different Molalities from 5 to 85 °C and Pressure P = 100 kPa[a]

Temp./°C	5.0	15.0	25.0	35.0	45.0	55.0	65.0	75.0	85.0
m/mol kg^{-1}	ρ/g cm^{-3}								
1.9998	1.012	1.010	1.007	1.003	0.999	0.994	0.988		
3.9957	1.022	1.019	1.016	1.011	1.006	1.000	0.994		
5.9968	1.032	1.028	1.023	1.018	1.012	1.006	0.999	0.992	
7.9993	1.038	1.033	1.027	1.022	1.015	1.009	1.002	0.994	
10.0002	1.042	1.037	1.031	1.025	1.018	1.011	1.004	0.996	0.987
11.9923	1.046	1.040	1.034	1.027	1.020	1.013	1.006	0.998	0.990
13.9894	1.048	1.042	1.036	1.029	1.022	1.014	1.007	0.999	0.991

[a]Standard uncertainties u are $u(m)$ = 0.00003 mol/kg, $u(\rho)$ = 0.002 g cm^{-3}, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 12. 1,3-Butanediol: Density, ρ, of Aqueous Solutions with Different Molalities from 5 to 85 °C and Pressure P = 100 kPa[a]

Temp./°C	5.0	15.0	25.0	35.0	45.0	55.0	65.0	75.0	85.0
m/mol kg^{-1}	ρ/g cm^{-3}								
2.9990	1.011	1.009	1.005	1.001	0.996	0.991	0.985		
5.9991	1.021	1.017	1.012	1.007	1.001	0.995	0.988	0.981	
8.9981	1.026	1.021	1.015	1.009	1.003	0.996	0.988	0.981	
11.9964	1.029	1.023	1.017	1.011	1.004	0.996	0.989	0.982	0.971
14.9855	1.031	1.024	1.018	1.011	1.004	0.997	0.989	0.982	0.972
17.9820	1.031	1.024	1.018	1.011	1.004	0.997	0.989	0.982	0.974

[a]Standard uncertainties u are $u(m)$ = 0.00003 mol/kg, $u(\rho)$ = 0.002 g cm^{-3}, $u(T)$ = 0.01 °C, aand $u(P)$ = 1 kPa.

Table 13. Glycerol: Density, ρ, of Aqueous Solutions with Different Molalities from 5 to 85 °C and Pressure P = 100 kPa[a]

Temp./°C	5.0	15.0	25.0	35.0	45.0	55.0	65.0	75.0	85.0
m/mol kg^{-1}	ρ/g cm^{-3}								
0.9994	1.021	1.019	1.017	1.014	1.009	1.005	0.999		
1.9979	1.039	1.037	1.034	1.030	1.026	1.021	1.015		
2.9978	1.055	1.053	1.049	1.045	1.040	1.035	1.029		
3.9988	1.071	1.068	1.064	1.060	1.055	1.049	1.044		
4.9996	1.082	1.079	1.075	1.070	1.065	1.060	1.054		
5.9983	1.093	1.089	1.085	1.080	1.075	1.069	1.063		
7.9974	1.112	1.108	1.103	1.098	1.093	1.087	1.081	1.074	
10.0014	1.128	1.123	1.118	1.112	1.107	1.102	1.095	1.089	
11.9989	1.140	1.136	1.130	1.125	1.119	1.113	1.107	1.099	
13.9879	1.151	1.146	1.141	1.135	1.129	1.123	1.117	1.110	1.102
15.9936	1.163	1.158	1.152	1.146	1.140	1.134	1.128	1.121	1.115

[a]Standard uncertainties u are $u(m)$ = 0.00003 mol/kg, $u(\rho)$ = 0.002 g cm^{-3}, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 14. Potassium Acetate: Density, ρ, of Aqueous Solutions with Different Molalities from 5 to 85 °C and Pressure P = 100 kPa[a]

Temp./°C	5.0	15.0	25.0	35.0	45.0	55.0	65.0	75.0	85.0
m/mol kg^{-1}	ρ/g cm^{-3}								
2.0000	1.085	1.082	1.079	1.075	1.071	1.066	1.061		
2.9991	1.120	1.117	1.113	1.109	1.104	1.099	1.094	1.085	
3.9938	1.151	1.148	1.144	1.139	1.134	1.129	1.123	1.117	
4.9983	1.179	1.175	1.170	1.166	1.160	1.155	1.150	1.144	1.137
5.9979	1.203	1.199	1.194	1.189	1.184	1.178	1.172	1.166	1.160

[a]Standard uncertainties u are $u(m)$ = 0.00003 mol/kg, $u(\rho)$ = 0.002 g cm^{-3}, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

2.4. Viscosity

The viscosity was investigated using two types of viscometers. From 5 to 95 °C, an automated microviscometer (Anton Paar, AMVn) was used. The accuracy of time measurement is 0.001 s, and the temperature is internally controlled with an accuracy of 0.01 K. Per type of solution and temperature, 20 individual measurements have been performed. From −35 to 5 °C, an Ubbelohde capillary viscometer (Schott) was used. The capillaries were suspended in a thermostated bath, ensuring a temperature accuracy of 0.01 K. An automatic viscosity measurement system (Schott, AVS/G) and a custom stand were used for the flow time measurements with an accuracy of 0.01 s. With this method, five individual measurements were performed per type of solution and temperature. For both systems, the measurements have a standard uncertainty smaller than 0.5%. The dynamic viscosity as a function of temperature is depicted in Tables 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, and 27.

2.5. Specific Heat Capacity and Isobaric Heat Capacity Per Unit Volume

The specific isobaric heat capacity was determined in a temperature range from 0 to 100 °C by differential scanning calorimetry (PerkinElmer, DSC 8000) through at least three measurements with a standard uncertainty smaller than 0.04 kJ K^{-1} kg^{-1}. A standardized

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Table 15. Sodium Propionate: Density, ρ, of Aqueous Solutions with Different Molalities from 5 to 85 °C and Pressure P = 100 kPa[a]

Temp./°C	5.0	15.0	25.0	35.0	45.0	55.0	65.0	75.0	85.0
m/mol kg^{-1}	ρ/g cm^{-3}								
2.0001	1.075	1.072	1.068	1.064	1.059	1.053	1.047		
2.9996	1.105	1.101	1.097	1.092	1.086	1.080	1.074	1.067	
3.9984	1.131	1.126	1.121	1.116	1.110	1.103	1.097	1.090	1.082
4.9871	1.152	1.147	1.141	1.135	1.128	1.122	1.114	1.107	1.100
5.9989	1.171	1.165	1.159	1.152	1.146	1.139	1.131	1.124	1.117

[a]Standard uncertainties u are $u(m)$ = 0.00003 mol/kg, $u(\rho)$ = 0.002 g cm^{-3}, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 16. Potassium Propionate: Density, ρ, of Aqueous Solutions with Different Molalities from 5 to 85 °C and Pressure P = 100 kPa[a]

Temp./°C	5.0	15.0	25.0	35.0	45.0	55.0	65.0	75.0	85.0
m/mol kg^{-1}	ρ/g cm^{-3}								
1.9993	1.084	1.081	1.078	1.073	1.069	1.063	1.057		
2.9991	1.117	1.114	1.109	1.105	1.100	1.094	1.088	1.081	
3.9977	1.145	1.141	1.136	1.131	1.125	1.119	1.113	1.107	
4.9978	1.170	1.165	1.160	1.154	1.148	1.142	1.136	1.129	1.122
6.0003	1.190	1.185	1.179	1.174	1.167	1.161	1.154	1.148	1.141

[a]Standard uncertainties u are $u(m)$ = 0.00003 mol/kg, $u(\rho)$ = 0.002 g cm^{-3}, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 17. Dynamic Viscosity, η, of the Aqueous Solutions Given Minimal Freezing Point (Compositions Shown in Table 5) at Temperatures from −35 to 95 °C and Pressure P = 100 kPa[a]

	η/mPa s								
temperature/°C	−35.0	−20.0	−5.0	5.0	25.0	45.0	65.0	85.0	95.0
ethylene glycol	78.29	24.84	9.610	6.866	3.371	1.937	1.248	0.874	0.748
triethylene glycol	514.8	116.8	38.74	20.78	8.294	4.138	2.408	1.565	1.309
tetraethylene glycol	939.1	187.3	56.55	31.69	11.68	5.567	3.137	1.977	1.676
1,2-propanediol	513.0	103.2	29.40	15.83	5.965	2.898	1.673	1.098	0.917
1,3-butanediol	1671	261.0	70.21	33.27	11.00	4.803	2.544	1.545	1.249
glycerol	710.6	152.2	46.53	23.61	9.178	4.509	2.603	1.688	1.402
diglycerol	6622	936.2	214.6	96.33	28.08	11.54	5.880	3.540	2.748
potassium acetate	41.97	10.02	6.485	5.027	2.778	1.759	1.228	0.924	0.846
sodium propionate	967.8	138.5	35.39	17.95	6.831	3.438	2.063	1.387	1.179
potassium propionate	77.34	21.98	8.954	6.511	3.346	2.021	1.362	0.992	0.867

[a]Standard uncertainties u are $u_r(\eta)$ = 0.01, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 18. Ethylene Glycol: Dynamic Viscosity, η, of Aqueous Solutions with Different Molalities from −35 to 85 °C at Pressure P = 100 kPa[a]

Temp./°C	−30.0	−25.0	−15.0	−5.0	5.0	15.0	25.0	35.0	45.0	55.0	65.0	75.0	85.0
m/mol kg^{-1}	η/mPa s												
1.9989					2.099	1.532	1.169	0.922	0.746	0.616	0.518		
3.9979					2.678	1.918	1.439	1.116	0.888	0.723	0.598		
5.9547				4.992	3.256	2.307	1.712	1.315	1.036	0.835	0.684		
7.9963				6.015	3.891	2.725	2.002	1.524	1.194	0.956	0.777		
9.9983			11.79	7.060	4.369	3.210	2.293	1.731	1.347	1.071	0.868	0.709	
12.0414			13.87	8.005	4.749	3.448	2.578	1.941	1.501	1.187	0.954	0.778	
13.9897		28.27	15.18	9.00	5.517	3.990	2.856	2.123	1.635	1.286	1.029	0.835	
15.9988	46.17	32.09	17.20	10.10	6.127	4.396	3.159	2.316	1.775	1.390	1.108	0.895	0.732

[a]Standard uncertainties u are $u_r(\eta)$ = 0.01, $u(m)$ = 0.00003 mol/kg, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

measurement procedure was applied including three isothermal steps at 0, 50, and 100 °C for 5 min and two heating steps with a heating rate of 10 °C min^{-1}. Aluminum pans with a volume of 50 μL are used in combination with sealable aluminum covers with an uncertainty in mass of 1%. Two empty sample crucibles served for baseline measurements, and a crucible holding a sapphire crystal was used for calibration. 15 μL of each solution was placed into the pan, weighed, and measured instead of the sapphire crystal. Specific heat capacities (c_p) are summarized in Tables 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, and 38. The isobaric heat capacity per unit volume was calculated by multiplying the specific heat capacity (c_p) and density (ρ) data and is given in Table 39.

2.6. Boiling Point and Enthalpy of Vaporization

To calculate the enthalpy of vaporization, the vapor pressure curve was collected by measuring the boiling point at reduced pressures. The custom apparatus consisted of a reflux condenser and a two-necked 250 mL flask equipped with a magnetic stirrer bar and immersed in a heating bath. The reduced pressure was generated by a liquid jet pump and

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Table 19. Diethylene Glycol: Dynamic Viscosity, η, of Aqueous Solutions with Different Molalities from −35 to 85 °C at Pressure P = 100 kPa[a]

Temp./°C	−35.0	−25.0	−15.0	−5.0	5.0	15.0	25.0	35.0	45.0	55.0	65.0	75.0	85.0
m/mol kg^{-1}	η/mPa s												
1.9995					2.558	1.837	1.384	1.075	0.861	0.707	0.590		
3.9996				5.619	3.622	2.550	1.882	1.438	1.127	0.903	0.736		
5.9984			12.58	7.580	4.807	3.323	2.411	1.814	1.405	1.117	0.900	0.735	
8.0001			16.24	9.47	5.748	4.143	3.035	2.192	1.681	1.317	1.051	0.848	
10.0000		37.67	19.81	11.55	6.913	4.892	3.519	2.559	1.948	1.517	1.202	0.967	0.789
11.9888	99.82	44.71	23.27	13.45	7.917	5.588	3.980	2.892	2.187	1.692	1.334	1.067	0.865

[a]Standard uncertainties u are $u_r(\eta)$ = 0.01, $u(m)$ = 0.00003 mol/kg, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 20. Triethylene Glycol: Dynamic Viscosity, η, of Aqueous Solutions with Different Molalities from −35 to 85 °C at Pressure P = 100 kPa[a]

Temp./°C	−35.0	−25.0	−15.0	−5.0	5.0	15.0	25.0	35.0	45.0	55.0	65.0	75.0	85.0
m/mol kg^{-1}	η/mPa s												
2.0000					3.661	2.563	1.886	1.441	1.132	0.909	0.744	0.616	
3.9985				10.93	6.766	4.531	3.201	2.358	1.796	1.398	1.112	0.897	
5.9993			34.02	18.40	10.13	6.897	4.783	3.455	2.401	1.837	1.434	1.184	0.952
7.9992		120.8	53.46	27.56	14.56	9.581	6.453	4.545	3.307	2.479	1.906	1.491	1.187
9.9874	509.9	183.4	78.05	38.02	19.34	12.43	8.222	5.683	4.081	3.007	2.280	1.762	1.386

[a]Standard uncertainties u are $u_r(\eta)$ = 0.01, $u(m)$ = 0.00003 mol/kg, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 21. Tetraethylene Glycol: Dynamic Viscosity, η, of Aqueous Solutions with Different Molalities from −35 to 85 °C at Pressure P = 100 kPa[a]

Temp./°C	−35.0	−25.0	−15.0	−5.0	5.0	15.0	25.0	35.0	45.0	55.0	65.0	75.0	85.0
m/mol kg^{-1}	η/mPa s												
1.9996					4.539	3.298	2.382	1.799	1.402	1.111	0.898	0.736	
3.9991				17.73	9.840	6.721	4.676	3.389	2.331	1.791	1.406	1.176	
5.9962			58.26	32.95	16.75	10.87	7.256	5.045	3.647	2.742	2.090	1.624	1.288
7.9986	854.7	281.4	112.8	52.27	25.26	16.70	10.10	6.943	4.890	3.565	2.670	2.049	1.602

[a]Standard uncertainties u are $u_r(\eta)$ = 0.01, $u(m)$ = 0.00003 mol/kg, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 22. 1,2-Propandiol: Dynamic Viscosity, η, of Aqueous Solutions with Different Molalities from −35 to 85 °C at Pressure P = 100 kPa[a]

Temp./°C	−30.0	−25.0	−15.0	−5.0	5.0	15.0	25.0	35.0	45.0	55.0	65.0	75.0	85.0
m/mol kg^{-1}	η/mPa s												
1.9998					2.642	1.854	1.374	1.057	0.838	0.681	0.563		
3.9957					4.165	2.779	1.971	1.465	1.125	0.888	0.715		
5.9968				10.78	6.321	4.034	2.750	1.978	1.480	1.140	0.914	0.730	
7.9993			29.11	14.57	7.664	5.032	3.423	2.390	1.759	1.323	1.031	0.816	
10.0002			38.13	18.56	9.557	6.141	4.078	2.819	2.048	1.539	1,186	0.934	0.748
11.9923		119.9	48.20	23.03	11.43	7.232	4.746	3.290	2.332	1.730	1.319	1.025	0.815
13.9894	257.8	151,0	58.10	27.45	13.74	8.500	5.459	3.696	2.631	1.940	1.451	1.142	0.902

[a]Standard uncertainties u are $u_r(\eta)$ = 0.01, $u(m)$ = 0.00003 mol/kg, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 23. 1,3-Butanediol: Dynamic Viscosity, η, of Aqueous Solutions with Different Molalities from −35 to 85 °C at Pressure P = 100 kPa[a]

Temp./°C	−30.0	−25.0	−15.0	−5.0	5.0	15.0	25.0	35.0	45.0	55.0	65.0	75.0	85.0
m/mol kg^{-1}	η/mPa s												
2.9990					3.270	2.316	1.719	1.320	1.040	0.838	0.686		
5.9991				15.44	8.168	5.327	3.597	2.478	1.813	1.370	1.063	0.841	
8.9981			57.10	27.03	14.37	8.423	5.352	3.606	2.553	1.875	1.409	1.094	
11.9964			89.44	40.40	18.85	11.38	7.157	4.740	3.279	2.362	1.749	1.335	1.037
14.9855		339.5	126.8	55.30	24.84	14.63	9.015	5.871	4.054	2.852	2.084	1.566	1.203
17.9820	808.3	454.4	165.2	71.08	31.30	18.00	10.89	7.102	4.773	3.339	2.420	1.801	1.371

[a]Standard uncertainties u are $u_r(\eta)$ = 0.01, $u(m)$ = 0.00003 mol/kg, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

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Table 24. Glycerol: Dynamic Viscosity, η, of Aqueous Solutions with Different Molalities from −35 to 85 °C at Pressure P = 100 kPa[a]

Temp./°C	−30.0	−25.0	−15.0	−5.0	5.0	15.0	25.0	35.0	45.0	55.0	65.0	75.0	85.0
m/mol kg^{-1}	η/mPa s												
0.9994					1.979	1.453	1.116	0.886	0.722	0.601	0.507		
1.9979					2.440	1.769	1.341	1.051	0.845	0.694	0.580		
2.9978					2.998	2.140	1.600	1.239	0.985	0.800	0.664		
3.9988					3.756	2.637	1.944	1.486	1.168	0.937	0.767		
4.9996				7.016	4.458	3.091	2.253	1.706	1.329	1.059	0.858		
5.9983				8.489	5.328	3.640	2.620	1.963	1.516	1.197	0.963		
7.9974				12.27	7.432	4.945	3.482	2.558	1.941	1.511	1.200	0.968	
10.0014			31.21	16.95	9.474	6.462	4.496	3.231	2.414	1.852	1.451	1.158	
11.9989			44.13	23.04	12.35	8.256	5.634	4.009	2.967	2.289	1.837	1.347	
13.9879		137.9	60.07	30.34	15.67	10.23	6.859	4.802	3.490	2.599	1.987	1.557	1.237
15.9936	326.8	198.6	81.23	39.80	19.89	12.56	8.215	5.688	4.081	3.031	2.310	1.798	1.424

[a]Standard uncertainties u are $u_r(\eta)$ = 0.01, $u(m)$ = 0.00003 mol/kg, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 25. Potassium Acetate: Dynamic Viscosity, η, of Aqueous Solutions with Different Molalities from −35 to 85 °C at Pressure P = 100 kPa[a]

Temp./°C	−30.0	−25.0	−15.0	−5.0	5.0	15.0	25.0	35.0	45.0	55.0	65.0	75.0	85.0
m/mol kg^{-1}	η/mPa s												
2.0000					2.341	1.745	1.352	1.078	0.881	0.732	0.617		
2.9991				3.912	2.756	2.052	1.587	1.279	1.038	0.860	0.772	0.612	
3.9938			7.517	4.974	3.310	2.449	1.882	1.490	1.208	0.994	0.833	0.703	
4.9983			9.149	5,857	4.019	2.939	2.241	1.758	1.412	1.155	0.953	0.803	0.681
5.9979	26.81	19.25	11.03	6.974	4.558	3.486	2.593	2.028	1.628	1.329	1.105	0.920	0.776

[a]Standard uncertainties u are $u_r(\eta)$ = 0.01, $u(m)$ = 0.00003 mol/kg, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 26. Sodium Propionate: Dynamic Viscosity, η, of Aqueous Solutions with Different Molalities from −35 to 85 °C at Pressure P = 100 kPa[a]

Temp./°C	−35.0	−25.0	−15.0	−5.0	5.0	15.0	25.0	35.0	45.0	55.0	65.0	75.0	85.0
m/mol kg^{-1}	η/mPa s												
2.0001					3.955	2.774	2.040	1.551	1.159	0.935	0.768		
2.9996				9.297	5.726	3.840	2.736	2.040	1.573	1.244	1.004	0.825	
3.9984			29.48	15.17	8.232	5.564	3.881	2.855	2.151	1.629	1.291	1.034	0.843
4.9871			49.52	23.45	11.92	7.684	5.153	3.616	2.678	2.057	1.598	1.274	1.032
5.9989	967.5	246.1	83.81	36.55	17.45	10.73	6.939	4.811	3.462	2.591	2.000	1.562	1.250

[a]Standard uncertainties u are $u_r(\eta)$ = 0.01, $u(m)$ = 0.00003 mol/kg, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 27. Potassium Propionate: Dynamic Viscosity, η, of Aqueous Solutions with Different Molalities from −35 to 85 °C at Pressure P = 100 kPa[a]

Temp./°C	−35.0	−25.0	−15.0	−5.0	5.0	15.0	25.0	35.0	45.0	55.0	65.0	75.0	85.0
m/mol kg^{-1}	η/mPa s												
1.9993					2.751	2.030	1.545	1.215	0.979	0.801	0.669		
2.9991				5.696	3.653	2.621	1.967	1.529	1.221	0.993	0.822	0.688	
3.9977			11.77	7.111	4.420	3.318	2.434	1.868	1.470	1.185	0.972	0.805	
4.9978		28.08	14.86	8.855	5.479	4.029	3.017	2.235	1.758	1.411	1.154	0.955	0.798
6.0003	82.15	35.96	18.63	10.89	6.606	4.805	3.547	2.672	2.084	1.662	1.348	1.108	0.920

[a]Standard uncertainties u are $u_r(\eta)$ = 0.01, $u(m)$ = 0.00003 mol/kg, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

measured with an accuracy of 250 Pa by a differential vacuum gauge. The apparatus and pump were connected via a 5 L pressure compensation flask. The boiling temperature was measured 3 min after a constant reflux in the vapor phase was reached directly above the liquid by a digital thermometer with 0.01 K accuracy. The boiling points T_b at 100 kPa and the molar heat of vaporization $\overline{\Delta_{vap}h}$ for the aqueous solutions are presented in Table 5; boiling point curve data are presented in Tables 40, 41, and 42.

2.7. Osmotic Coefficients

2.7.1. Vapor Pressure Measurements of the Aqueous Solutions of Ethylene Glycol and Potassium Propionate. By vapor pressure measurements, concentration-dependent activities of water, activity coefficients of water, and the corresponding osmotic coefficients of aqueous ethylene glycol and potassium propionate have been determined, all of which are directly related to the chemical potentials of the different species and therefore to their Gibbs energies. Vapor pressure measurements were applied to the systems ethylene glycol + water and potassium propionate + water, varying the concentrations m (EG) = 2.0 to 24.0 mol kg^{-1} and m (KPro) =

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Table 28. Specific Heat Capacity, c_p, of Aqueous Solutions Given Minimal Freezing Point (Compositions Shown in Table 5) from 25 to 95 °C at Pressure P = 100 kPa[a]

	c_p/J g^{-1} K^{-1}				
temperature/°C	25.0	45.0	65.0	85.0	95.0
ethylene glycol	3.15	3.27	3.38	3.51	3.57
diethylene glycol	3.10	3.23	3.35	3.47	3.52
triethylene glycol	3.08	3.20	3.32	3.44	3.49
tetraethylene glycol	3.12	3.23	3.34	3.46	3.50
1,2-propanediol	3.35	3.48	3.60	3.72	3.78
1,3-butanediol	3.33	3.50	3.66	3.81	3.87
glycerol	2.99	3.11	3.23	3.34	3.39
diglycerol	2.88	2.99	3.11	3.23	3.29
potassium acetate	2.93	2.97	3.02	3.09	3.11
sodium propionate	3.63	3.65	3.67	3.70	3.72
potassium propionate	3.34	3.39	3.43	3.50	3.53

[a]Standard uncertainties u are $u_r(c_p)$ = 0.01, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 29. Ethylene Glycol: Specific Heat Capacity, c_p, of Aqueous Solutions with Different Molalities from 25 to 95 °C at Pressure P = 100 kPa[a]

Temp./°C	25.0	45.0	65.0	85.0	95.0
m/mol kg^{-1}	c_p/J g^{-1} K^{-1}				
3.9979	3.768	3.828	3.894	3.908	3.968
7.9963	3.517	3.613	3.667	3.747	3.829
12.0414	3.298	3.410	3.477	3,576	3.657

[a]Standard uncertainties u are u(m) = 0.00003 mol/kg, $u_r(c_p)$ = 0.01, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 30. Diethylene Glycol: Specific Heat Capacity, c_p, of Aqueous Solutions with Different Molalities from 25 to 95 °C at Pressure P = 100 kPa[a]

Temp./°C	25.0	45.0	65.0	85.0	95.0
m/mol kg^{-1}	c_p/J g^{-1} K^{-1}				
3.9996	3.662	3.748	3.813	3.839	3.911
5.9984	3.356	3.473	3.547	3.602	3.668
10.0000	3.161	3.277	3.379	3.468	3.524

[a]Standard uncertainties u are u(m) = 0.00003 mol/kg, $u_r(c_p)$ = 0.01, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 31. Triethylene Glycol: Specific Heat Capacity, c_p, of Aqueous Solutions with Different Molalities from 25 to 95 °C at Pressure P = 100 kPa[a]

Temp./°C	25.0	45.0	65.0	85.0	95.0
m/mol kg^{-1}	c_p/J g^{-1} K^{-1}				
2.0000	3.716	3.785	3.842	3.916	3.954
5.9993	3.290	3.416	3.517	3.600	3.669
8.0001	3.127	3.244	3.359	3.444	3.512

[a]Standard uncertainties u are $u(m)$ = 0.00003 mol/kg, $u_r(c_p)$ = 0.01, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

0.002 to 2.0 mol kg^{-1} at t = 25 °C. Vapor pressure osmometry (VPO) measurements were undertaken with the help of a K-7000 Osmometer from Knauer GmbH (Germany). With this VPO apparatus, the vapor pressure is determined by using two thermistors to measure voltage changes caused by the changes in temperature due to condensation or evaporation of the solvent. A comparison between results obtained with this equipment and those from direct vapor pressure lowering using a classical precise equipment[23] has been done elsewhere[24] and showed good agreement between both techniques. For each solution, at least

Table 32. Tetraethylene Glycol: Specific Heat Capacity, c_p, of Aqueous Solutions with Different Molalities from 25 to 95 °C at Pressure P = 100 kPa[a]

Temp./°C	25.0	45.0	65.0	85.0	95.0
m/mol kg^{-1}	c_p/J g^{-1} K^{-1}				
1.9996	3.566	3.644	3.704	3.752	3.782
3.9991	3.299	3.412	3.506	3,547	3.604
5.9962	3.203	3.302	3,396	3.449	3,520

[a]Standard uncertainties u are u(m) = 0.00003 mol/kg, $u_r(c_p)$ = 0.01, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 33. 1,2-Propandiol: Specific Heat Capacity, c_p, of Aqueous Solutions with Different Molalities from 25 to 95 °C at Pressure P = 100 kPa[a]

Temp./°C	25.0	45.0	65.0	85.0	95.0
m/mol kg^{-1}	c_p/J g^{-1} K^{-1}				
3.9957	3.878	3.934	3.976	4.007	4.066
7.9993	3.608	3.717	3.782	3.849	3.907
11.9923	3.466	3.587	3.682	3.741	3.780

[a]Standard uncertainties u are $u(m)$ = 0.00003 mol/kg, $u_r(c_p)$ = 0.01, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 34. 1,3-Butanediol: Specific Heat Capacity, c_p, of Aqueous Solutions with Different Molalities from 25 to 95 °C at Pressure P = 100 kPa[a]

Temp./°C	25.0	45.0	65.0	85.0	95.0
m/mol kg^{-1}	c_p/J J g^{-1} K^{-1}				
2.9990	4.029	4.087	4.128	4.166	4.223
8.9981	3.546	3.677	3.771	3.820	3.902
14.9855	3.168	3.336	3.461	3.531	3.614

[a]Standard uncertainties u are $u(m)$ = 0.00003 mol/kg, $u_r(c_p)$ = 0.01, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 35. Glycerol: Specific Heat Capacity, c_p, of Aqueous Solutions with Different Molalities from 25 to 95 °C at Pressure P = 100 kPa[a]

Temp./°C	25.0	45.0	65.0	85.0	95.0
m/mol kg^{-1}	c_p/J J g^{-1} K^{-1}				
3.9988	3.440	3.499	3.559	3.590	3.665
7.9974	3.282	3.363	3.478	3.511	3.584
11.9989	3.061	3.145	3.240	3.332	3.389

[a]Standard uncertainties u are $u(m)$ = 0.00003 mol/kg, $u_r(c_p)$ = 0.01, $u(T)$ = 0.01 °C, and u(P) = 1 kPa.

Table 36. Potassium Acetate: Specific Heat Capacity, c_p, of Aqueous Solutions with Different Molalities from 25 to 95 °C at Pressure P = 100 kPa[a]

Temp./°C	25.0	45.0	65.0	85.0	95.0
m/mol kg^{-1}	c_p/J g^{-1} K^{-1}				
2.0000	3.448	3.469	3.528	3.534	3.596
2.9991	3.311	3.354	3.390	3.404	3.464
4.9983	3.044	3.077	3.089	3.098	3.130

[a]Standard uncertainties u are u(m) = 0.00003 mol/kg, $u_r(c_p)$ = 0.01, $u(T)$ = 0.01 °C, and u(P) = 1 kPa.

nine determinations (zero point adjustment was performed after three and six measurements) were performed. Panel readings for KPro solutions were in the range from 0.3 to 330 (standard deviation σ = 0.04 to 1.5; relative error δ = 0.1 to 2%), and readings for EG solutions were

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Table 37. Sodium Propionate: Specific Heat Capacity, c_p, of Aqueous Solutions with Different Molalities from 25 to 95 °C at Pressure P = 100 kPa[a]

Temp./°C	25.0	45.0	65.0	85.0	95.0
m/mol kg^{-1}	c_p/J J g^{-1} K^{-1}				
2.0010	3.729	3.754	3.788	3.808	3.843
2.9996	3.615	3.638	3.673	3.681	3.706
4.9871	3.526	3.530	3.536	3.543	3.554

[a]Standard uncertainties u are $u(m)$ = 0.00003 mol/kg, $u_r(c_p)$ = 0.01, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

Table 38. Potassium Propionate: Specific Heat Capacity, c_p, of Aqueous Solutions with Different Molalities from 25 to 95 °C at Pressure P = 100 kPa[a]

Temp./°C	25.0	45.0	65.0	85.0	95.0
m/mol kg^{-1}	c_p/J g^{-1} K^{-1}				
1.9939	3.579	3.605	3.639	3.642	3.693
2.9919	3.322	3.349	3.420	3.425	3.490
4.9978	3.061	3.074	3.118	3.122	3.144

[a]Standard uncertainties u are u(m) = 0.00003 mol/kg, $u_r(c_p)$ = 0.01, $u(T)$ = 0.01 °C, and u(P) = 1 kPa.

Table 39. Isobaric Heat Capacity Per Unit Volume, c_p/v, of Aqueous Solutions Given Minimal Freezing Point (Compositions Shown in Table 5) from 25 to 95 °C at Pressure P = 100 kPa[a]

	c_v/J cm^{-3} K^{-1}				
temperature/°C	25.0	45.0	65.0	85.0	95.0
ethylene glycol	3.35	3.44	3.51	3.60	3.63
diethylene glycol	3.35	3.45	3.53	3.60	3.62
triethylene glycol	3.35	3.44	3.51	3.59	3.61
tetraethylene glycol	3.41	3.48	3.55	3.62	3.63
1,2-propanediol	3.47	3.56	3.63	3.68	3.71
1,3-butanediol	3.40	3.51	3.62	3.70	3.73
glycerol	3.45	3.55	3.64	3.73	3.76
diglycerol	3.40	3.50	3.59	3.69	3.73
potassium acetate	3.52	3.55	3.57	3.60	3.61
sodium propionate	4.20	4.17	4.14	4.13	4.11
potassium propionate	3.93	3.93	3.95	3.97	3.98

[a]Standard uncertainties u are $u_r(c_p/v)$ = 0.01, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

in the range from 60 to 1120 (standard deviation σ = 0.1 to 7; relative error δ = 0.1 to 2%).

2.7.2. Activity of Water and Osmotic Coefficient. In the molality scale, the osmotic coefficient ϕ is defined as

$$\phi = -\frac{1000}{M_1 \sum_i \nu_i m_i} \ln a_1 \quad (1)$$

where m is the solute molality, ν is the stoichiometric coefficient of the solute, M_1, in g/mol, is the molar mass of the solvent, and i means each component except solvent 1. The Knauer osmometer was calibrated using aqueous NaCl solutions, yielding a function that correlates the panel readings to m (NaCl). Osmotic coefficient values ϕ of EG and KPro solutions of concentrations m (EG) and m (KPro) are obtained via water activities of the solutions under investigation. From eq 1, for two solutions (the EG or KPro solution and NaCl calibration solution) with equal solvent activities, we obtain the following relationship for the osmotic coefficients of the EG and KPro solutions

$$\phi_{solute} = \frac{\nu_{NaCl} m_{NaCl}}{\nu_{solute} m_{solute}} \phi_{NaCl} \quad (2)$$

where m_{NaCl} is the molality of a NaCl solution showing the same panel reading as the EG or KPro solution (isopiestic concentrations are presented in Table 43). m_{solute} represents the molality of ethylene glycol and potassium propionate. The stoichiometric coefficients (van't Hoff factor) are ν_{NaCl} = 2, ν_{KPro} = 2, and ν_{EG} = 1. The osmotic coefficient ϕ_{NaCl} is calculated with a set of equations developed by Gibbard and Scatchard,[25] and the procedure is described in detail in ref 26. The activity of water, a_1, was calculated from experimentally determined osmotic coefficients. The corresponding data for the activities of water can be found in Table 44.

The uncertainties of density, heat capacity, osmotic coefficient, and water activity are given in Tables 6−16, 28−38, and 43−47. In vapor pressure calculations, the mean value of MW from 9 measurements is used. According to the law of error distribution

$$S_\phi = \sqrt{\left(\frac{\partial f}{\partial m}\right)^2 S_m^{\,2} + \left(\frac{\partial f}{\partial \phi_{ref}}\right)^2 S_{\phi_{ref}}^{\,2}} \quad (3)$$

where S_m is the standard deviation of the solution molality. $S_{\phi ref}$ is the standard deviation of the osmotic coefficient of NaCl solutions used as an external standard. For low concentrations in the case of potassium propionate solutions: $m \sim 0.168$ mol kg^{-1}; $S_m \sim 0.00003$; $\phi \sim 1.0087$; and $S_{\phi ref} \sim 0.001$, i.e., S_ϕ reaches 0.001005. For high concentrations: $m \sim 2$ mol kg^{-1}; $S_m \sim 0.00003$; $\phi \sim 1.26$; and $S_{\phi ref} \sim 0.001$, i.e., S_ϕ reaches 0.000787. For low concentrations in the case of ethylene glycol solutions: $m \sim 3.86$ mol kg^{-1}; $S_m \sim 0.00003$; $\phi \sim 0.9704$; and $S_{\phi ref} \sim 0.001$, i.e., S_ϕ reaches 0.00103. For high concentrations: $m \sim 20.03$ mol kg^{-1}; $S_m \sim 0.00003$; $\phi \sim 0.8187$; and $S_{\phi ref} \sim 0.001$, i.e., S_ϕ reaches 0.00121. That is, the discrepancy in ϕ is stipulated by the error in molality at low concentrations and by the error in the standard osmotic coefficient at high concentrations.

2.8. Osmotic Coefficients from Literature Data

2.8.1. Sodium Propionate. The osmotic and activity coefficients of sodium propionate up to molalities of 3.0 mol kg^{-1} are reported by Robinson and Stokes.[27] In the work of Bonner,[28] the data of osmotic and activity coefficients are extended for sodium formate to a molality of 3.5 mol kg^{-1} and for sodium propionate to a concentration of 10.0 mol kg^{-1} (i.e., to the limit of solubility of these salts so that they might be used in the preparation of a table of free energies of formation in the aqueous solution of organic compounds related to the citric acid cycle). The data from these two publications are employed in this work.

2.8.2. Potassium Acetate. The osmotic coefficients of KAc in the molality range from m = (0.001 to 3.5) mol kg^{-1} are taken from a review by Hamer and Wu.[29]

The values of the osmotic coefficient ϕ of carbonic acid salt solutions: KAc, KPro, and NaPro at t = 25 °C are presented in Table 45.

2.8.3. Triethylene Glycol and Tetraethylene Glycol. The vapor−liquid equilibrium data in aqueous solutions of tetraethylene glycol[16] and triethylene glycol[17] are presented in tabular form in the papers of Herskowitz and Gottlieb. The activity of water was measured over a wide range of mole fractions by an isopiestic method with LiCl as the reference electrolyte. The measurements were performed at 24.45 and 59.45 °C for TriEG and at 24.95 °C for TeEG. We have treated the osmotic coefficient values on the basis of the 24.45 and 24.95 °C measurements.

The values of the osmotic coefficients ϕ of glycol solutions, including ethylene glycol (measured in this work at t = 25 °C), are given in Table 46.

2.8.4. 1,2-Propanediol and 1,3-Butanediol. Water activity, osmotic coefficients, solute activity coefficients, and pairwise Gibbs energy coefficients for solute−solute interactions of aqueous solutions of 1,3-butanediol (13BD) and five other butanediols, i.e., 1,2-butanediol, 1,4-butanediol, 2,3-butanediol, 1,2,4-butanetriol, and 1,2,3,4-butanetetrol, have been determined in a study of Romero and Paez with isopiestic measurements at 25 °C.[21] Vapor pressure osmometry at 25 °C with diluted sucrose solutions as a primary standard and urea as a secondary standard was applied by Borghesani et al. to study a further set of aliphatic diols, i.e., 1,3-propanediol, 1,2-propanediol, 1,2-butanediol, 1,3-butanediol, 1,2-butanediol, 2,3-

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Table 40. Boiling Point Curve Data of Glycol Aqueous Solutions[a,b]

ethylene glycol		diethylene glycol		triethylene glycol		tetraethylene glycol	
T_b/°C	p/kPa	T_b/°C	p/kPa	T_b/°C	p/kPa	T_b/°C	p/kPa
57.2	11.60	56.2	11.73	54.6	11.47	54.4	11.60
61.4	14.27	60.5	14.40	59.40	14.4	58.8	14.27
65.0	16.93	64.2	17.06	63.0	17.06	62.9	17.20
68.6	19.86	67.2	19.73	65.8	19.73	65.6	19.60
71.4	22.40	70.2	22.40	68.4	22.13	68.4	22.00
76.1	27.46	75.2	27.73	73.2	27.46	73.2	27.33
80.4	32.93	79.8	33.60	77.6	33.06	77.2	32.93
83.8	37.86	83.2	38.40	81.2	38.40	80.6	38.26
87.0	43.33	86.9	43.73	84.4	43.73	83.6	43.60
92.6	53.99	92.4	54.39	90.0	54.39	89.0	53.99
97.3	64.66	97.0	65.06	94.6	65.06	93.8	64.93
101.4	75.33	101.2	75.73	98.4	75.73	97.6	75.59
105.0	85.99	104.2	86.39	102.0	86.39	101.2	86.52
107.4	96.66	106.8	97.06	105.2	97.06	104.2	96.92

[a]Measured boiling point T_b and corresponding pressure p of the aqueous solutions with minimal freezing point (compositions shown in Table 5). [b]Standard uncertainties u are $u(T_b)$ = 0.1 °C and $u(p)$ = 0.25 kPa.

Table 41. Boiling Point Curve Data of Polyol Solutions[a,b]

1,2-propanediol		1,3-butanediol		glycerol		diglycerol	
T_b/°C	p/kPa	T_b/°C	p/kPa	T_b/kPa	p/kPa	T_b/°C	p/kPa
54.3	11.47	56.0	11.60	56.4	11.47	56.4	11.73
58.4	14.13	60.2	14.27	61.6	13.87	61.0	14.13
62.0	16.80	63.8	17.20	65.3	16.53	64.8	16.80
65.2	19.46	66.6	19.73	68.6	19.2	68.0	19.46
68.0	22.13	69.4	22.26	71.4	21.86	70.8	22.13
72.9	27.46	74.2	27.60	76.8	27.46	75.6	27.46
76.8	32.80	78.0	32.93	81.4	33.33	79.6	32.80
80.4	38.13	81.4	38.26	85.0	38.13	83.2	38.13
83.8	43.46	84.4	43.33	88.0	42.93	86.4	43.46
89.4	54.13	90.2	54.26	93.4	53.59	91.8	54.13
94.2	64.79	94.7	64.93	98.4	64.53	96.3	64.79
98.2	75.46	98.6	75.59	102.4	75.19	100.3	75.46
101.7	86.12	102.4	86.26	105.9	85.86	103.9	86.12
105.0	96.79	105.2	96.92	108.5	96.52	107.3	96.79

[a]Measured boiling point T_b and corresponding pressure p of the aqueous solutions with minimal freezing point (compositions shown in Table 5). [b]Standard uncertainties u are $u(T_b)$ = 0.1 °C and $u(p)$ = 0.25 kPa

Table 42. Boiling Point Curve Data of Carbonic Acid Salt Solutions[a,b]

potassium acetate		sodium propionate		potassium propionate	
T_b/°C	p/kPa	T_b/°C	p/kPa	T_b/°C	p/kPa
57.4	11.33	56.4	11.47	54.2	10.93
61.8	14.00	60.4	14.13	59.6	13.60
65.4	16.67	64.0	16.80	63.0	16.27
68.8	19.33	67.2	19.46	66.2	18.93
71.5	22	70	22.13	69.2	21.86
76.4	27.33	74.9	27.46	74.4	27.20
80.4	32.66	78.8	32.80	78.4	32.26
84.0	38.00	82.5	38.13	82.0	37.73
87.2	43.33	85.5	43.46	85.0	42.80
93.1	53.73	91.2	54.13	90.6	53.59
97.6	64.66	95.8	64.79	95.4	64.26
101.6	75.33	99.7	75.46	99.2	74.93
105.4	85.99	103.3	86.12	102.4	85.59
108.7	96.66	106.4	96.79	105.0	96.26

[a]Measured boiling point T_b and corresponding pressure p of the aqueous solutions with minimal freezing point (compositions shown in Table 5). [b]Standard uncertainties u are $u(T_b)$ = 0.1 °C and $u(p)$ = 0.25 kPa.

butanediol, 1,5-pentanediol, and 1,6-hexanediol.[18] In Table 47, the osmotic coefficients of 1,2-propanediol and 1,3-butanediol are presented, which have been processed and interpolated from Borghesani's work.[18]

3. RESULTS AND DISCUSSION

3.1. Freezing Point

All 11 compounds investigated were able to induce a sufficient freezing point depression of −40 °C. In a preliminary study, other promising candidates had been found to be not sufficient because of rapid decomposition under the influence of heat and pressure, i.e., 1-ethyl-3-methyl-imidazolium-ethylsulfate, potassium formate, dimethyl sulfoxide, and trimethylamine N-oxide, or due to a lack of solubility in water, i.e., sodium citrate, sodium dihydrogen phosphate, sodium nitrate, and sodium succinate, or due to eutectic points being above −40 °C, i.e., 1,4-butanediol and dipotassium hydrogen phosphate.[30]

For the 11 compounds investigated, the mean freezing point depression was determined as a function of concentration by at least three measurements per concentration until a freezing point of −40 °C was reached. Compilations of the results are given in Tables 2−4 and Figures 1, 2, and 3. The standard deviation of the freezing point is between 1 and 1.2 °C for the solutions of 1,2-propanediol, triethylene glycol, and tetraethylene glycol and below 0.6 °C for the rest. The measured freezing points for the aqueous solutions of ethylene glycol, diethylene glycol, triethylene glycol, and 1,2-propanediol are in good agreement with literature data, whereas the freezing point for the tetraethylene glycol solution shows a significant deviation.[31−34] In Figures 1−3, experimental results are compared with calculations for ideal behavior considering heterogeneous solvent solid−liquid equilibrium using

$$T_f = \frac{1}{\frac{-\ln\left(\frac{1000}{1000 + m \cdot M_1}\right) \cdot R}{\Delta_f H_1^0} + \frac{1}{T_f^0}} \quad (4)$$

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Table 43. Values of the Osmotic Coefficient ϕ of NaCl Aqueous Solutions[25] Isopiestic with Ethylene Glycol and Potassium Propionate Aqueous Solutions at T = 25.0 °C and Pressure P = 100 kPa[a,b]

ethylene glycol			potassium propionate		
m(EG)/ mol kg^{-1}	ϕ (NaCl)	m(NaCl)/ mol kg^{-1}	m(KPro)/ mol kg^{-1}	ϕ (NaCl)	m(NaCl)/ mol kg^{-1}
1.07660	0.9233	0.55942	0.00288	0.9816	0.00279
2.17527	0.9410	1.12191	0.00777	0.9720	0.00752
3.86008	0.9790	1.91313	0.01639	0.9644	0.01367
4.80145	1.0022	2.32217	0.04156	0.9517	0.03240
7.11740	1.0594	3.22131	0.16829	0.9258	0.18336
9.00940	1.1175	4.04676	0.45972	0.9230	0.54124
11.39059	1.1722	4.88897	0.91330	0.9387	1.06447
15.94997	1.2472	5.73568	1.40410	0.9699	1.74103
17.98655	1.2775	6.10783	1.93068	1.0083	2.42365
20.03013	1.3025	6.32819			
20.65378	1.2956	6.41204			

[a]The reference data on NaCl solutions are from ref 25. [b]Standard uncertainties u are $u(m)$ = 0.00003 mol kg^{-1}, $u(\phi)$ = 0.001, u(T) = 0.01 °C, and u(P) = 1 kPa.

Table 44. Experimental Values of Activity of Water, a_1, of Ethylene Glycol and Potassium Propionate Aqueous Solutions at T = 25.0 °C and Pressure P = 100 kPa[a]

ethylene glycol		potassium propionate	
m(EG)/mol kg^{-1}	a_1	m(KPro)/mol kg^{-1}	a_1
1.0766	0.9816	0.0029	1.0000
2.1753	0.9627	0.0078	0.9997
3.8601	0.9347	0.0164	0.9994
4.8015	0.9196	0.0416	0.9989
7.1174	0.8843	0.1683	0.9939
9.0094	0.8496	0.4597	0.9822
11.3910	0.8173	0.9133	0.9646
15.9500	0.7728	1.4040	0.9410
17.9866	0.7549	1.9307	0.9157
20.0301	0.7442		
20.6538	0.7401		

[a]Standard uncertainties u are $u(m)$ = 0.00003 mol kg^{-1}, u (a_1) = 0.0004, u(T) = 0.01 °C, and u(P) = 1 kPa.

where m is the molality of the solute (mol kg^{-1}), M_1 is the molar mass of water (g mol^{-1}), T_f^0 is the freezing temperature of water, R is the universal gas constant, and $\Delta_f H_1^0$ is the molar enthalpy of fusion of water. The value 6010 J mol^{-1}[35] for the molar enthalpy of fusion of water is used in calculations.

This equation could be derived from the equality of the chemical potential of the solvent in two phases (solid phase (s) as pure solvent and liquid phase (l) as solution)

$$\mu_1^{s}(p, T) = \mu_1^{L}(p, T) \quad (5)$$

$$\mu_1^{0,s}(p, T) = \mu_1^{0,L}(p, T) + RT \ln a_1 \quad (6)$$

The freezing point curve of ethylene glycol shows a clear difference from the theoretical curve. The deviation of the experimental freezing point from the theoretical one increases in the series KAc + water < NaPro + water < KPro + water; EG + water < DiEG + water ≈ TriEG + water < TeEG + water; and BuDiol + water < Gly + water < ProDiol + water < DiGly + water, which testifies the strengthening of nonelectrostatic intermolecular bonds due to the interaction of oppositely charged or polarized regions—hydrogen bond—and short-range interactions that arise from temporary fluctuations in electron distribution and hydrophobic interactions in the series from KAc to KPro; EG to TeEG; and BuDiol to DiGly mixtures.

3.2. Density

Density data were collected from 5 to 85 °C. Data points for 95 °C were obtained by linear extrapolation. The fit equation yielded an accuracy of R^2 > 0.995 in the correlation coefficient for all solutions. For the measurements at 5 °C, the standard deviation is smaller than $1{\cdot}10^{-4}$ g cm^{-3}, while for the measurements above 5 °C, it is $1{\cdot}10^{-6}$ g cm^{-3}. The density as a function of temperature for solutions giving the maximum freezing point depression is depicted in Figure 4 and summarized in Table 6. Ethylene glycol, diethylene glycol, triethylene glycol, and glycerol values match with data found in the literature.[36−38] The data on the concentration dependence of our measurements are in good agreement with the literature data. The comparison, for example, of ethylene glycol and 1,2-propanediol is presented in the Supporting Information. The densities of 1,2-propanediol and 1,3-butanediol are smaller (−2% to − 6%), while the values for the oligomeric glycols (1 to 4%), glycerols (8−12%), and salts (8−14%) are larger than the densities of the EG solution. Tables 7−16 summarize density data for solutions of different concentrations as a function of temperature.

3.3. Viscosity

Dynamic viscosity data were collected from −35 to 95 °C. To convert kinematic to dynamic viscosity, density data were used. The dynamic viscosity as a function of the temperature of antifreeze mixtures resulting in the maximum freezing point depression is presented in Figure 5 and summarized in Table 17. For the measurements below 0 °C, the standard deviation is 0.5% or lower, with the exception of the value of diglycerol at −35 °C. The mixture of water and diglycerol showed increased deviations in flow time during the measurements and partially solidified during the last three measurements. The freezing at this temperature implies that the measured freezing point of −40.4 °C is of a metastable nature, and the true point is at −35 °C or higher. For the measurements above 0 °C, the standard deviation is 2% or lower.

At −20 °C, the data point for potassium acetate seems to be off; thus, the absolute values for −20 and −35 °C should be considered with caution. Further, the two data sets for 1,2-propanediol do not seem to match each other, indicated by the discrepancy between −5 and 5 °C. The viscosities of EG, diethylene glycol, triethylene glycol, and glycerol are in accordance with data found in the literature.[33,36,37] All studied mixtures except the solutions of potassium acetate and potassium propionate have a substantially higher dynamic viscosity than that of EG for temperatures of 5 °C and below. The propionate salt solutions show a large difference in viscosity, although their mole fraction is almost the same. The higher viscosity of sodium propionate may be a consequence of the more pronounced hydration of the sodium ion. The higher amount of interacting water increases the effective concentration of salt, which causes a rise in viscosity. The bigger hydration shell of sodium ions compared with the potassium ions increases their resistance to flow. Also, the influence of the hydration shell on the viscosity of the propionate solutions should be studied by including the lithium salt. Disregarding the salts, the viscous behavior of all other solutions can be explained by their chain length. Within the homologous series of glycols and glycerols,

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Table 45. Values of Osmotic Coefficient ϕ of Carbonic Acid Salt Aqueous Solutions: Potassium Acetate,[29] Potassium Propionate[this work], and Sodium Propionate[27,28] at T = 25.0°C and Pressure P = 100 kPa[a]

potassium acetate		potassium propionate		sodium propionate	
m(salt)/mol kg^{-1}	ϕ	m(salt)/mol kg^{-1}	ϕ	m(salt)/mol kg^{-1}	ϕ
0.001	0.989	0.0029	0.9517	0.001	0.989
0.002	0.984	0.0078	0.9399	0.002	0.984
0.005	0.977	0.0164	0.8048	0.005	0.977
0.010	0.969	0.0416	0.7420	0.010	0.970
0.020	0.961	0.1683	1.0087	0.020	0.961
0.050	0.949	0.4597	1.0867	0.050	0.950
0.100	0.943	0.9133	1.0941	0.100	0.945
0.200	0.944	1.4040	1.2026	0.200	0.947
0.300	0.950	1.9307	1.2657	0.300	0.954
0.400	0.958			0.400	0.964
0.500	0.967			0.500	0.975
0.600	0.977			0.600	0.986
0.700	0.987			0.700	0.998
0.800	0.997			0.800	1.010
0.900	1.008			0.900	1.022
1.000	1.018			1.000	1.034
1.200	1.039			1.200	1.059
1.400	1.060			1.400	1.083
1.600	1.081			1.600	1.107
1.800	1.103			1.800	1.130
2.000	1.124			2.000	1.152
2.500	1.176			2.500	1.206
3.000	1.230			3.000	1.255
3.500	1.286			3.500	1.302
				4.000	1.342
				5.000	1.404
				6.000	1.452
				7.000	1.470
				8.000	1.472
				9.000	1.476
				10.000	1.486
				10.200	1.488

[a]Standard uncertainties u are $u(m)$ = 0.00003 mol kg^{-1}, $u(\phi)$ = 0.002, $u(T)$ = 0.01 °C, and u(P) = 1 kPa.

viscosity increases with increasing possibilities for spatial interference. 1,2-Propanediol and 1,3-butanediol show the same behavior. The high viscosities at low temperatures for 1,3-butanediol, diglycerol, sodium propionate, and tetraethylene glycol are disadvantageous for an application as a coolant solution. Tables 18−27 summarize dynamic viscosity data for solutions of different concentrations as a function of temperature.

3.4. Specific Heat Capacity and Isobaric Heat Capacity Per Unit Volume

Specific isobaric heat capacities as a function of temperature for solutions resulting in maximal freezing point depression are presented in Figure 6 and summarized in Table 28. Tables 29−38 summarize specific heat capacity data for solutions of different concentrations as a function of temperature. Isobaric heat capacity per unit volume was calculated from the density (Table 6) and specific heat capacity data (Table 28). Figure 7 and Table 39 present the data as a function of temperature for solutions giving the maximum freezing point depression. The values were recorded from 25 to 95 °C with a standard deviation of 0.01 J g^{-1} K^{-1} or lower. The values of the specific heat capacity of EG and diethylene glycol are within the range of the published data.[39] The propionate solutions have an isobaric heat capacity per unit volume higher by at least 10%, while all other mixtures are within a 5% difference from the values of EG.

3.5. Boiling Point and Enthalpy of Vaporization

Boiling point data were determined by one measurement per solution resulting in the maximum freezing point depression from 10 to 100 kPa and are presented in Table 5. All solutions have boiling points in the range from 105 to 111 °C. Errors for the boiling point are estimated to be smaller than 1%. The boiling point of glycerol is in good accordance with the literature.[38] In contrast to the freezing point depression, the decreasing mole fractions within the homologous series of glycols and glycerols are reflected by a reduced boiling point elevation. The enthalpy of vaporization of all solutions is approximately 10% higher than that of pure water.[40]

3.6. Osmotic Coefficients

The values of activities of water and osmotic coefficients of the salts, diols, and glycerols in aqueous solutions are presented in Figures 8, 9, and 10.

In the osmotic coefficients versus molality curves of the diols (Figure 9), the minimum is observed at m = 4.0 mol kg^{-1} for 1,2-propanediol and at m = 2.5 mol kg^{-1} for 1,3-butanediol, i.e., the minimum is shifted to lower molalities with the increase of the hydrocarbon chain length of the diol (as it should be also the

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Table 46. Values of Osmotic Coefficient ϕ of Glycol Aqueous Solutions: Ethylene Glycol[this work] at T = 25.00 °C, Triethylene Glycol[17] at T = 24.45°C, and Tetraethylene Glycol[16] at T = 24.95°C and Pressure P = 100 kPa[a]

ethylene glycol		triethylene glycol		tetraethylene glycol	
x_2	ϕ	x_2	ϕ	x_2	ϕ
0.0190	0.9596	0.0541	1.209	0.0707	1.127
0.0377	0.9707	0.0549	1.257	0.1093	1.195
0.0650	0.9704	0.0641	1.245	0.1292	1.204
0.0796	0.9694	0.0657	1.251	0.1423	1.215
0.1136	0.9590	0.1026	1.337	0.1500	1.268
0.1396	1.0038	0.1051	1.344	0.1545	1.238
0.1703	0.9836	0.1077	1.346	0.1638	1.210
0.2232	0.8971	0.1157	1.326	0.1837	1.202
0.2447	0.8676	0.1575	1.348	0.2415	1.129
0.2652	0.8187	0.1657	1.344	0.2489	1.112
0.2712	0.8087	0.1894	1.352	0.2657	1.108
		0.2187	1.345	0.3016	1.070
		0.2505	1.311	0.3197	1.053
		0.3057	1.262	0.3647	0.9989
		0.3898	1.147	0.3689	0.9948
		0.3996	1.135	0.4131	0.9406
		0.4967	1.005	0.4588	0.8778
		0.5001	0.9932	0.4817	0.8618
		0.5449	0.9318	0.4886	0.8471
		0.5809	0.8785	0.6898	0.5756
		0.6731	0.7290	0.7434	0.4982
		0.7838	0.5354		
		0.8062	0.4941		

[a]Standard uncertainties u are $u(m)$ = 0.00003 mol kg^{-1}, $u(\phi)$ = 0.001, $u(T)$ = 0.05 °C, and $u(P)$ = 1 kPa.

Table 47. Values of Osmotic Coefficient ϕ of Diol Aqueous Solutions[18] at Temperature T = 25.0 °C and Pressure P = 100 kPa[a]

m(diol)/mol kg^{-1}	1,2-propanediol	1,3-butanediol
0.010	0.9995	0.9987
0.050	0.9975	0.9943
0.100	0.9950	0.9889
0.200	0.9901	0.9781
0.500	0.9763	0.9498
1.000	0.9560	0.9130
1.500	0.9391	0.8898
2.000	0.9256	0.8800
2.500	0.9155	0.8838
3.000	0.9088	0.9010
3.500	0.9055	0.9318
4.000	0.9056	0.9760
4.500	0.9091	1.0338
5.000	0.9160	1.1050
5.500	0.9263	1.1898

[a]Standard uncertainties u are $u(m)$ = 0.00003 mol kg^{-1}, $u(\phi)$ = 0.001, $u(T)$ = 0.01 °C, and $u(P)$ = 1 kPa.

case for the concentration dependencies of other thermodynamic properties of such substances (dH^{mix}, dH^{dil}, dV^{excess})).

Osmotic coefficients of glycols pass through maxima that indicate a nonelectrostatic interaction mechanism in these solutions, as opposed to electrolyte solutions. They also have the maxima shifted to the region of lower molalities with an increase in the hydrocarbon chain length of the glycol (Figure 10).

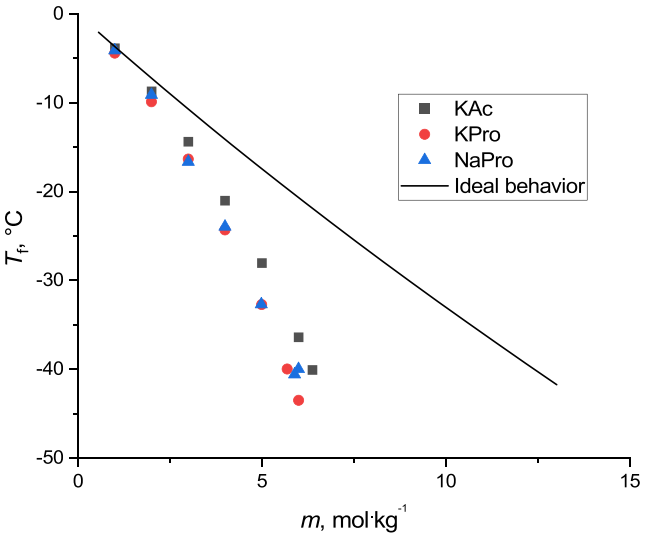

Figure 1. Experimental (points) and theoretical (line) freezing points calculated for the case of an ideal solution for carbonic acid salt aqueous solutions: KAc (■), KPro (●), and NaPro (▲).

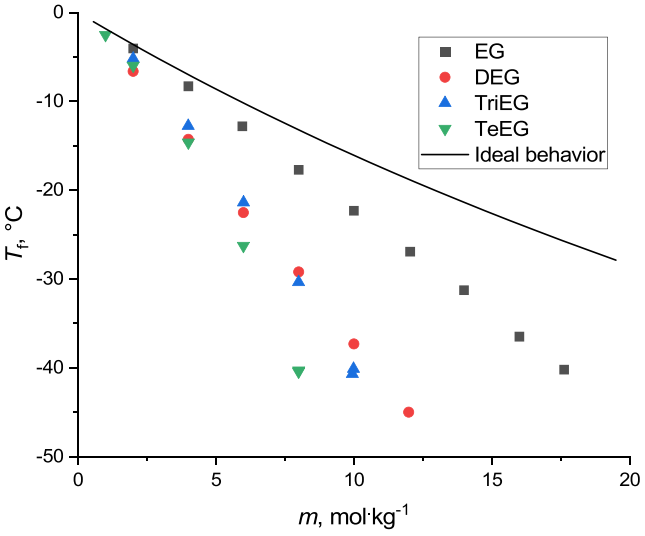

Figure 2. Experimental (points) and theoretical (line) freezing points calculated for the case of ideal solution for aqueous glycol solutions: ethylene glycol (■), diethylene glycol (●), triethylene glycol (▲), and tetraethylene glycol (▼) solutions.

Osmotic coefficients decrease in the following series: TriEG + water > TeEG + water > EG + water, which is in line with the freezing point change, where tetra- and triethylene glycols show a clear difference to the theoretical curve, which could be explained by strong intermolecular interactions in the case of tetra- and triethylene glycol. The deviation of the experimental freezing point from the theoretical one increases in the order similar to osmotic coefficient changes: EG + water < DiEG + water < TriEG + water < TeEG + water (Figure 2).

In ethylene glycol solutions, hydrogen bonds are formed between an electron pair of the hydroxyl group and a polar-bonded H atom of another hydroxyl group of ethylene glycol or a water molecule.[41] However, since single bonds are freely rotatable, no permanent hydrate shell could be formed in ethylene glycol solutions. The formation of hydrogen bonds again seems to play a role in explaining the difference in the discrepancies from the theoretical freezing point. Also, the more

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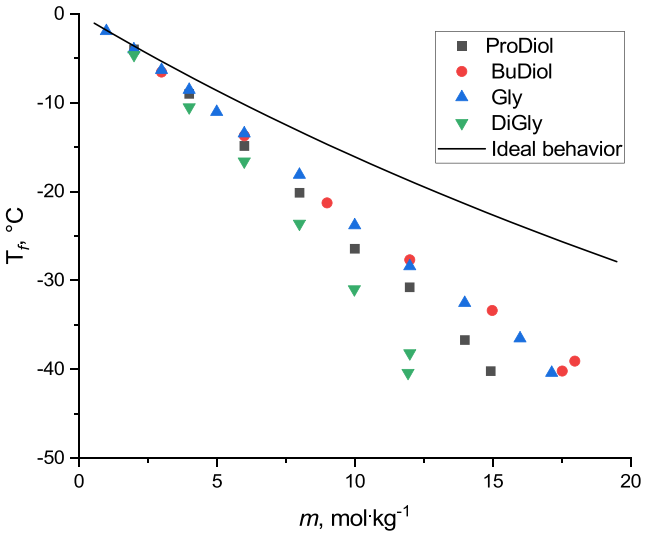

Figure 3. Experimental (points) and theoretical (line) freezing points calculated for the case of ideal solution for diol and glycerol aqueous solutions: 1,2-propanediol (■), 1,3-butanediol (●), glycerol (▲), and diglycerol (▼).

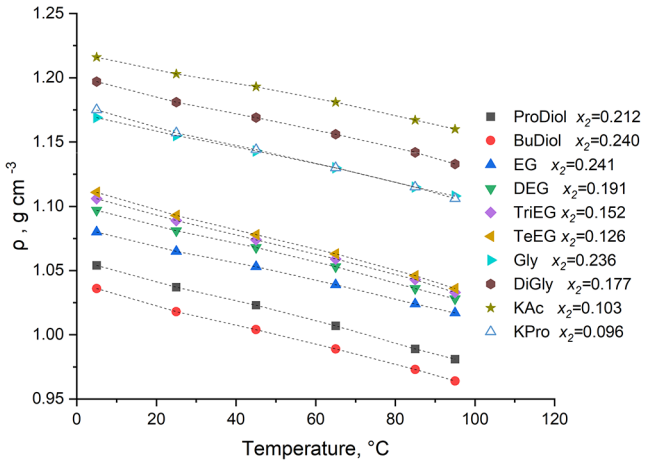

Figure 4. Temperature dependence of the density of the aqueous solutions of 1,2-propanediol (■), 1,3-butanediol (●), ethylene glycol (▲), diethylene glycol (▼), triethylene glycol (♦), tetraethylene glycol (◀), glycerol (▶), diglycerol (⬣), potassium acetate (★), and potassium propionate (Δ) (x_2— mole fraction of the nonaqueous component, points—experimental data, and lines—spline).

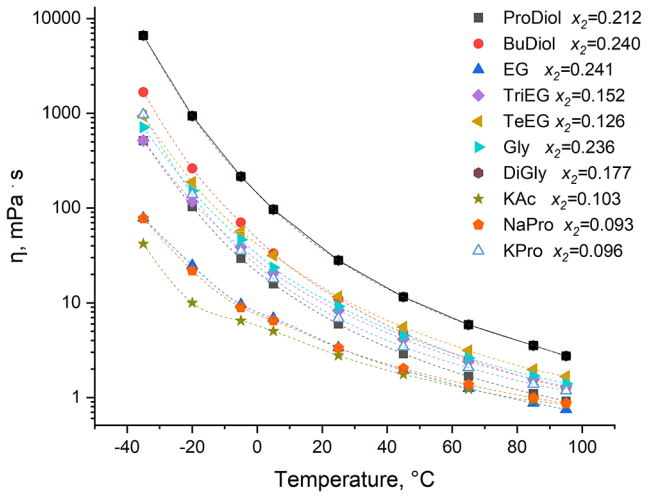

Figure 5. Temperature dependence of the dynamic viscosity of the aqueous solutions of 1,2-propanediol (■), 1,3-butanediol (●), ethylene glycol (▲), triethylene glycol (♦), tetraethylene glycol (◀), glycerol (▶), diglycerol (⬣), potassium acetate (★), sodium propionate (⬟), and potassium propionate (Δ) (x_2—mole fraction of the nonaqueous component, points—experimental data, and lines—spline).

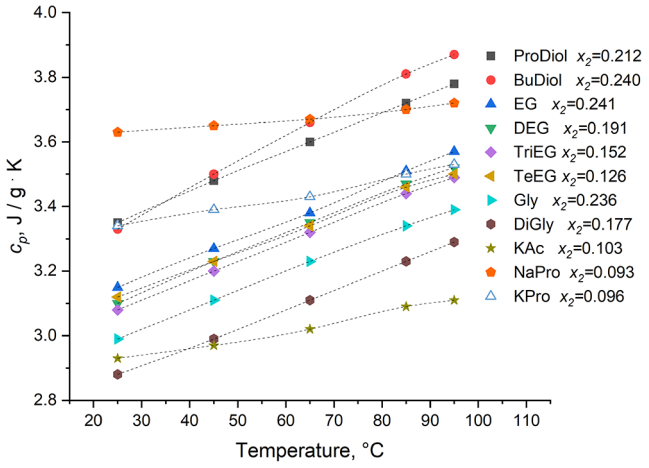

Figure 6. Temperature dependence of the specific heat capacity of the aqueous solutions of 1,2-propanediol (■), 1,3-butanediol (●), ethylene glycol (▲), diethylene glycol (▼), triethylene glycol (♦), tetraethylene glycol (◀), glycerol (▶), diglycerol (⬣), potassium acetate (★), sodium propionate (⬟), and potassium propionate (Δ) (x_2—mole fraction of nonaqueous component, points—experimental data, and lines—spline).

favorable formation of H-bond bridges in glycol solutions with longer hydrocarbon radical intramolecular carbon chains could already promote weak van der Waals forces, in addition to dipole−dipole interactions. Thus, the cluster formation is more strongly expressed in ethylene glycol solutions in comparison with glycols with a longer hydrocarbon chain. Also, in the case of very high concentrations of nonaqueous component glycol (with 10 molal and higher concentrations, the systems approach mole fractions of glycols approaching 1), water is not really the solvent anymore (region of small water additions).

Osmotic coefficients of the considered salts pass through minima and then strongly increase and exceed the value of unity. So, NaPro and KAc show positive deviations from ideality as well. As we can see from the activity coefficients of the salts (Figure11, activity coefficients of LiAc are from ref 42 calculated with the use of database ref 43), the activity coefficients of the lithium salt—lithium acetate—also pass through a minimum just as the activity coefficients of potassium acetate and sodium propionate, but then increase considerably less in comparison with potassium and sodium salts, perhaps, because of association of the lithium salt. For the lithium salt, the electrostatic contribution is predominant, which means an attractive interaction. For sodium and potassium salts, the repulsive interaction due to nonelectrostatic effects prevails, especially when electrostatic screening effects are important. This evidence is revealed in the high concentration range: at molalities from 1.5 to 2 mol/kg, the activity coefficient is larger than 1. Also, in the same concentration range (m > 1.5−2 mol/kg), the strong freezing point T_f deviation for sodium and potassium acetate and propionate solutions is striking.

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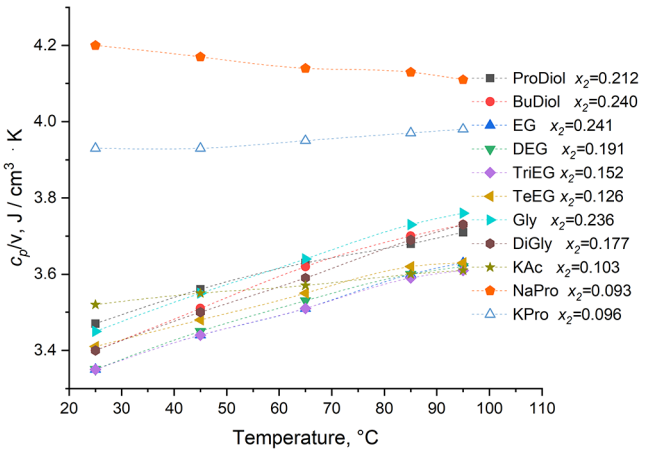

Figure 7. Temperature dependence of the isobaric heat capacity per unit volume of the aqueous solutions of 1,2-propanediol (■), 1,3-butanediol (●), ethylene glycol (▲), diethylene glycol (▼), triethylene glycol (♦), tetraethylene glycol (◀), glycerol (▶), diglycerol (⬣), potassium acetate (★), sodium propionate (⬟), and potassium propionate (Δ) (x_2—mole fraction of nonaqueous component, points—experimental data, and lines—spline).

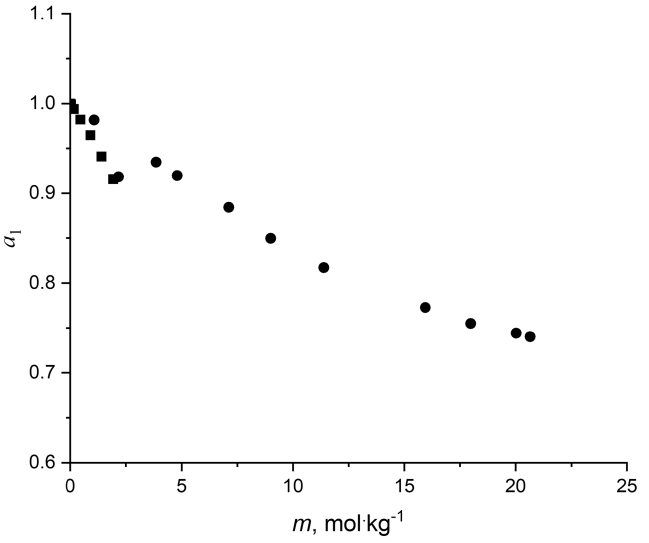

Figure 8. Water activity, a_1, of aqueous ethylene glycol (●) and potassium propionate (■) solutions at a temperature T = 25.00 °C.

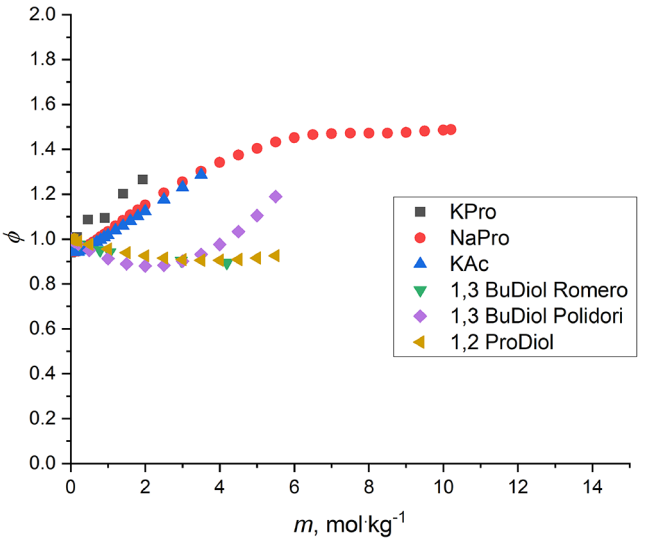

Figure 9. Osmotic coefficients ϕ of aqueous potassium propionate (■), sodium propionate (●), potassium acetate (▲), 1,2-propanediol (▲), and 1,3-butanediol solutions (▼, ♦) at temperature T = 25.00 °C.

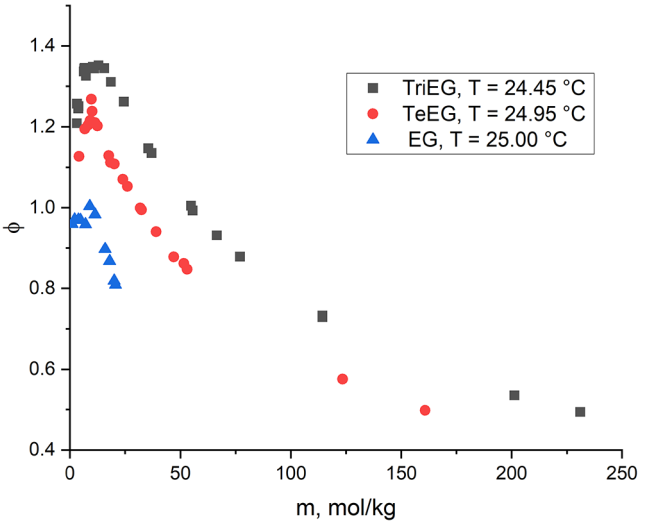

Figure 10. Osmotic coefficients, ϕ, of EG (▲) at temperature T = 25.00 °C, of TriEG (■), and TeEG (●) aqueous solutions at temperature T = 24.45 and 24.95 °C.

4. CONCLUSIONS

In this study, we aimed to check possible alternatives for EG as freezing point depressants in heat transfer liquids and to create an overview of the crucial physicochemical properties of these alternatives and already known depressants.

Ten of the preselected compounds form an aqueous solution with a freezing point of −40 °C and are qualified for further investigations. Their boiling point and heat of vaporization are in the same order as those for EG. The relatively high dynamic viscosity at low temperatures of the mixtures of tetraethylene glycol, sodium propionate, 1,3-butanediol, and especially diglycerol is disadvantageous for applications as heat transfer fluids. All other studied substances show a higher viscosity than EG at low temperatures, with the exception of the potassium salts. The potassium propionate solution excels with regard to its isobaric heat capacity per unit volume. Potassium acetate shows the best thermal conductivity. Additionally, their viscosity is favorable over that of EG at low temperatures. To conclude, the potassium salts are found to be promising in terms of flow behavior and thermal properties. Nevertheless, all other candidates also show reasonable thermal behavior and viscosities.

First hints regarding the stability of the new depressants were obtained from a pressure-cooker experiment, which showed lower stability of the diols and glycerols and higher stability of tri- and tetraethylene glycol, potassium acetate, and sodium propionate compared to ethylene glycol. However, these properties should be evaluated in more detail in future studies.[30] To obtain an in-depth understanding of the involved mechanisms, a systematic investigation of the glycol series is necessary. The influence of the hydration shell on the viscosity of the propionate solutions should be studied by including the lithium salt.

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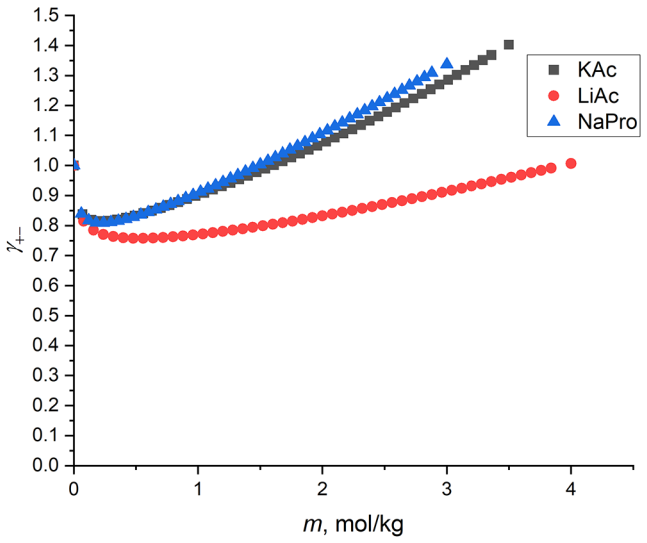

Figure 11. Activity coefficients, $\gamma_{\pm}$, of aqueous KAc (■), LiAc (●), and NaPro (▲) solutions at temperature T = 25.00 °C.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.jced.5c00403.

Comparison of reported experimental data with literature ones (PDF)

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E.N.T.: Methodology, investigation, measurements, formal analysis, data curation, and writing—original draft. T.L.: Investigation, measurements, formal analysis, and data curation. K.B.: Investigation, measurements, and data curation. J.S.: Investigation, measurements, and data curation. S.S.: Investigation, measurements, and data curation. W.K.: Writing—review and editing, validation, supervision, and project administration. R.M.: Writing—review and editing, methodology, validation, supervision, and conceptualization

Notes

The authors declare no competing financial interest.

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