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Solubilities, Densities, and Refractive Indices for the Ternary Systems Glycerin + MCl + H_2O (M = Na, K, Rb, Cs) at (298.15 and 308.15) K

Rui Meng, Shu'ni Li,* Quanguo Zhai, Yucheng Jiang, Hong Lei, Huiying Zhang, and Mancheng Hu*

Key Laboratory of Macromolecular Science of Shaanxi Province, School of Chemistry & Chemical Engineering, Shaanxi Normal University, Xi'an, Shaanxi, 710062, P. R. China

ABSTRACT: Data are presented on the solubilities, densities, and refractive indices of the four ternary systems glycerin + MCl + H_2O (M = Na, K, Rb, Cs) at (298.15 and 308.15) K and atmospheric pressure. The mass fraction of glycerin in the salt-free mixed solvent extends from 0 to 1.0. In all cases, the experiment values of solubilities were found to decrease with an increase of the concentration of glycerin. The measured solubilities, densities, and refractive indices of saturated solutions as well as densities and refractive indices of unsaturated solutions were fitted with available empirical correlations, respectively.

INTRODUCTION

Addition of an organic solvent to an aqueous salt solution normally reduces the solubility of the salt. This process has a number of potential advantages over alternative crystallization techniques because it creates the possibility of carrying out the operation at room temperature, producing crystals of high purity. More particularly, knowledge of the solubility, density, refractive indices of the salt solutions is fundamentally important for the design of the crystallization process. Previously, many research groups have studied the effects on the solubilities of various salts by the addition of organic solvents to their aqueous solutions. For instance, Gomis et al. and his co-workers[1−4] studied ternary systems of alcohol (1-propanol, 2-propanol, 2-pentanol, 3-pentanol, 2-methyl-2-butanol, 2-methyl-1-butanol, and 1-pentanol) + MCl (M = Na, K) + water at 298.15 K. Takiyama et al.[5] studied the morphology of NaCl crystals produced by drowning-out with ethanol at 313.15 K. They found that operational conditions and degree of supersaturation greatly influenced the crystal form. Wagner et al.[6] measured solubilities of NaCl in different mixtures of solvents, including water + cyclohexane, water + cyclohexanol, and water + benzyl alcohol at 298.15 K. Galleguillos et al.[7] measured solubilities, densities, and refractive indices data for the ternary systems KCl + C_2H_5OH + H_2O and NaCl + C_2H_5OH + H_2O at (298.15 and 313.15 K). Also, data are also presented on the equilibrium of the quaternary system KCl + NaCl + C_2H_5OH + H_2O at (298.15 and 313.15) K. Lopes et al.[8] obtained solubilities of sodium chloride and potassium chloride in water + ethanol mixtures from (298 to 323) K. Pinho et al.[9] measured the solubility of NaCl, NaBr, KCl, and KBr in the pure solvents of water, methanol, and ethanol, and their binary mixed solvents over the whole solvent composition range. In all cases, the presence of organic solvent significantly reduces the solubility of the salts in aqueous solution.

In previous work, our research groups have focused on the solubility of rare alkali metal (Rb and Cs) salts in water + organic solvent.[10−18] The organic solvents are mainly some aliphatic alcohols that are completely miscible or partly miscible with water, such as methanol, ethanol, 1-propanol, 2-propanol, ethylene glycol, as well as PEG [poly-(ethylene glycol)]. To extend our work, we study the glycerin + MCl + H_2O (M = Na, K, Rb, Cs) systems at saturated and unsaturated conditions in this work. Our goal was to enrich the available data on water + alkali chloride + organic systems. The study could be used to apply the salting-out technique and to develop thermodynamic models for the effects of salts.

EXPERIMENTAL SECTION

Materials. Analytical grade rubidium chloride and cesium chloride were supplied by the Shanghai China Lithium Industrial Co. with purity of 99.5%, respectively. Glycerin, sodium chloride, and potassium chloride were supplied by the Sinopharm Chemical Reagent Co., Ltd. with purity of 99.8 %, 99.5 %, and 99.5 %, respectively. All of the chemicals used for this study were used without further purification. To avoid water salt contamination, salts were dried at 393.15 K in a drying stove for more than 24 h before use. All solutions were prepared using deionized water.

Apparatus and Procedure. The apparatus and approach used in the experiment have been described in previous paper,[10,11] so only a brief description of experiment procedure is present here. The procedure consists on the preparation of a saturated solution and unsaturated solution. The equilibrium cell is charged with known masses with an accuracy of $\pm 1 \cdot 10^{-4}$ g (Mettler Toledo AL204), and to reach solution equilibrium conditions, stirring is carried out for at least 24 h at the working temperature. Then the solution is allowed to settle at least 24 h before sampling with a heated syringe.

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Table 1. Mass Fraction, Density (ρ), and Refractive Index (n_D) for the Glycerin (1) + NaCl/KCl/RbCl/CsCl (2) + H_2O (3)) Systems at (298.15 and 308.15) K

w'_1[a]	w_2[a]	n_D	ρ / $g \cdot cm^{-3}$	w'_1[a]	w_2[a]	n_D	ρ / $g \cdot cm^{-3}$
298.15 K glycerin (1) + NaCl (2) + H_2O (3)							
0.0000	0.2634	1.37958	1.19791	0.6005	0.1421	1.42968	1.24231
0.1015	0.2394	1.38651	1.20311	0.6997	0.1206	1.44084	1.25399
0.2002	0.2188	1.39383	1.20899	0.7996	0.1035	1.45288	1.26734
0.3001	0.2010	1.40177	1.21579	0.8969	0.0848	1.46521	1.28591
0.3785	0.1839	1.40859	1.22191	1.0000	0.0702		
0.5003	0.1584	1.41996	1.23265				
298.15 K glycerin (1) + KCl (2) + H_2O (3)							
0.0000	0.2605	1.36905	1.17747	0.6007	0.1302	1.42382	1.22703
0.1021	0.2388	1.37615	1.18284	0.7001	0.1089	1.43545	1.23788
0.2001	0.2190	1.38426	1.18898	0.8000	0.0909	1.44803	1.25441
0.2988	0.1942	1.39282	1.19551	0.8997	0.0765	1.46192	1.27092
0.4003	0.1657	1.40159	1.20223	1.0000	0.0696		
0.5000	0.1493	1.41263	1.21499				
298.15 K glycerin (1) + RbCl (2) + H_2O (3)							
0.0000	0.4854	1.38851	1.50084	0.6001	0.2942	1.43189	1.40661
0.1013	0.4483	1.39396	1.48460	0.7023	0.2502	1.44173	1.39381
0.2015	0.4258	1.40014	1.46868	0.7973	0.2129	1.45257	1.38467
0.3004	0.3921	1.40650	1.45311	0.8998	0.1783	1.46477	1.37540
0.3999	0.3570	1.41422	1.43674	1.0000	0.1420		
0.4999	0.3247	1.42266	1.42170				
298.15 K glycerin (1) + CsCl (2) + H_2O (3)							
0.0000	0.6547	1.41963	1.92428	0.5984	0.4644	1.44872	1.69844
0.1032	0.6289	1.42306	1.89174	0.6999	0.4180	1.45639	1.65321
0.2006	0.5956	1.42697	1.85674	0.7973	0.3767	1.46407	1.61234
0.3001	0.5684	1.43148	1.82094	0.8998	0.3294	1.47261	1.57132
0.4003	0.5394	1.43654	1.78082	1.0000	0.2824		
0.4994	0.5056	1.44242	1.74053				

w'_1[a]	w_2[a]	n_D	ρ / $g \cdot cm^{-3}$	w'_1[a]	w_2[a]	n_D	ρ / $g \cdot cm^{-3}$
308.15 K glycerin (1) + NaCl (2) + H_2O (3)							
0.0000	0.2653	1.37822	1.19382	0.6006	0.1443	1.42819	1.23773
0.1017	0.2476	1.38510	1.19897	0.7003	0.1197	1.43901	1.24890
0.2029	0.2228	1.39239	1.20475	0.7997	0.1069	1.45044	1.26143
0.3045	0.2054	1.40029	1.21160	0.8994	0.0915	1.46321	1.27591
0.4001	0.1869	1.40887	1.21919	1.0000	0.0752		
0.4983	0.1622	1.41824	1.22780				
308.15 K glycerin (1) + KCl (2) + H_2O (3)							
0.0000	0.2863	1.36932	1.18336	0.6008	0.1371	1.42278	1.22798
0.1000	0.2521	1.37627	1.18958	0.7001	0.1192	1.43457	1.24008
0.2005	0.2307	1.38417	1.19266	0.7998	0.1000	1.44680	1.25363
0.3001	0.2088	1.39263	1.20157	0.8992	0.0856	1.45948	1.26825
0.4001	0.1802	1.40255	1.20913	1.0000	0.0738		
0.4999	0.1603	1.41208	1.21726				
308.15 K glycerin (1) + RbCl (2) + H_2O (3)							
0.0000	0.5049	1.38949	1.51957	0.6005	0.3045	1.43104	1.41972
0.1002	0.4796	1.39476	1.50060	0.7007	0.2678	1.44031	1.40499
0.2008	0.4368	1.40025	1.48271	0.7998	0.2238	1.45134	1.39166
0.3001	0.4056	1.40708	1.47033	0.8988	0.1876	1.46315	1.38040
0.4007	0.3658	1.41395	1.45464	1.0000	0.1511		
0.5027	0.3359	1.42146	1.43682				
308.15 K glycerin (1) + CsCl (2) + H_2O (3)							
0.0000	0.6678	1.42093	1.94899	0.6004	0.4826	1.44901	1.71735
0.1001	0.6466	1.42372	1.91640	0.7003	0.4402	1.45599	1.67206
0.2007	0.6237	1.42727	1.88091	0.8001	0.3883	1.46435	1.62270
0.3000	0.5940	1.43177	1.84573	0.9001	0.3342	1.47366	1.57540
0.4002	0.5650	1.43670	1.80525	1.0000	0.2847		
0.5005	0.5297	1.44240	1.76148				

[a] w'_1 is the mass fraction of glycerin in the salt-free mixed solvent, and w_2 is the mass fraction of salts in the mixed solution.

The solubility of salts in the saturated solution was obtained by titration method with mercury nitrate,[20] and the uncertainty in the measurement was ± 0.5%. The amounts of glycerin was determined using oxidation with $K_2Cr_2O_7$,[21,22] and the uncertainty was ± 0.5%. The concentration of water was obtained by mss balance. Refractive indices and densities of both saturated and unsaturated solutions were determined using RXA 170 instrument and DMA 4500 apparatus (Anton Paar). Calibration was performed under atmospheric pressure using air and double-distilled water. The uncertainty of the refractive index and density are $\pm 1.0 \cdot 10^{-3}$ and $\pm 1.5 \cdot 10^{-3}$ $g \cdot cm^{-3}$, respectively. The measurement temperature was controlled to within ± 0.01 K.

■ RESULTS AND DISCUSSION

The experimental values of solubility, density, and refractive index for saturated solutions of glycerin + MCl + H_2O (M = Na, K, Rb, Cs) ternary systems at (298.15 and 308.15) K are given in Table 1. The values for solubility, refractive index and density of the four systems in pure water at 298.15 K are compared with the literature data,[7,14,23] which are listed in Table 2. Figure 1a

Table 2. Solubility (S), Density (ρ), and Refractive Index (n_D) in Pure Water (w %) at 298.15 K

salt	this work: S / w %	this work: ρ / $g \cdot cm^{-3}$	this work: n_D	literature: S / w %	literature: ρ / $g \cdot cm^{-3}$	literature: n_D
NaCl	26.34	1.19791	1.37958	26.28,[7] 26.45[23]	1.19778[7]	1.37950[7]
KCl	26.05	1.17747	1.36905	26.67,[7] 26.22[23]	1.17720[7]	1.36890[7]
RbCl	48.54	1.50084	1.38851	48.52,[14] 48.42[23]	1.50185[14]	1.38738[14]
CsCl	65.47	1.92428	1.41963	65.56,[14] 65.64[23]	1.92536[14]	1.41937[14]

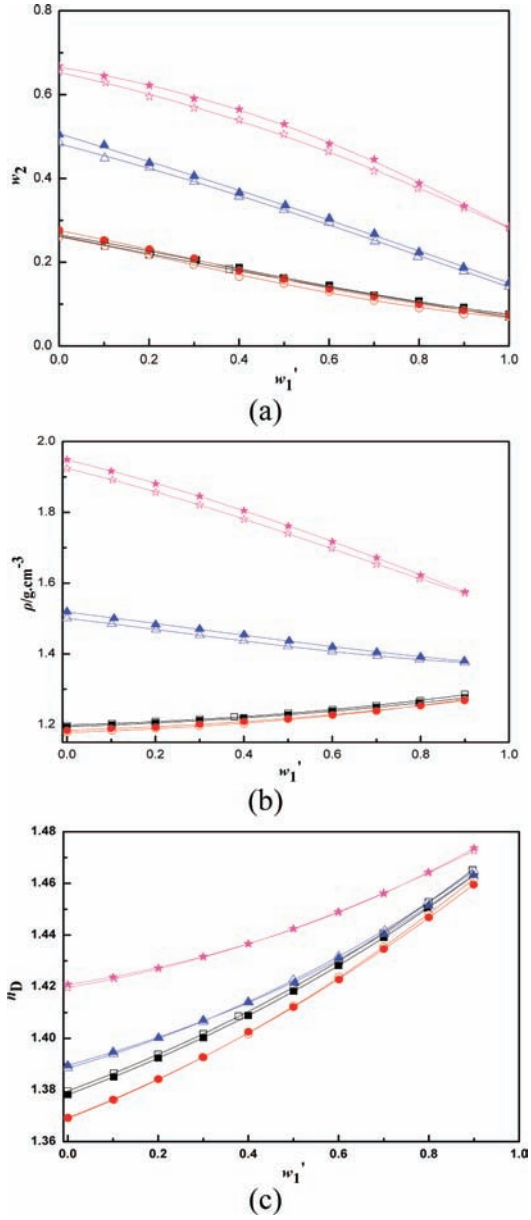

Figure 1. Solubilities (a), densities (b), and refractive indices (c) for the ternary systems of glycerin (1) + MCl (2) (M = Na, K, Rb, Cs) + H_2O (3) at (298.15 and 308.15) K (□, NaCl; ○, KCl; Δ, RbCl; ☆, CsCl at 298.15 K and ■, NaCl; ●, KCl; ▲, RbCl; ★, CsCl at 308.15 K. The solid lines in the figure are calculated from eq 1. (w_1' is the mass fraction of glycerin in the salt-free mixed solvent and w_2 is the mass fraction of salts in the mixed solution).

presents the solubility diagrams for the four ternary systems at (298.15 and 308.15) K. It can be observed that at a fixed temperature the solubility of all salts decreased with the addition of glycerin. Besides, the solubility of all salts increased with temperature increased, but in the glycerin + NaCl + H_2O system, no appreciable effect was observed of temperature on solubility. This phenomenon tends to be similar with the ethylene glycol + MCl (M = Na, K, Rb, Cs) + H_2O at (288.15 and 308.15 K).[18] Figure 1b presents the density diagrams for the four ternary systems at (298.15 and 308.15 K). It can be observed that the density for the glycerin + CsCl/RbCl + H_2O systems significantly

Table 3. Values of Parameters of eq 1[a]

system	A	B	C	D	δ
mass fraction					
298.15 K glycerin (1) + NaCl (2) + H_2O (3)	−1.3380	−0.8408	−0.1198	−0.3658	0.0081
298.15 K glycerin (1) + KCl (2) + H_2O (3)	−1.3507	−0.6088	−1.3512	0.6233	0.0182
298.15 K glycerin (1) + RbCl (2) + H_2O (3)	−0.7265	−0.6488	0.0024	−0.5790	0.0079
298.15 K glycerin (1) + CsCl (2) + H_2O (3)	−0.4241	−0.4106	−0.0458	0.3848	0.0039
308.15 K glycerin (1) + NaCl (2) + H_2O (3)	−1.3275	−0.6452	−0.7069	0.1001	0.0135
308.15 K glycerin (1) + KCl (2) + H_2O (3)	−1.2742	−0.7957	−0.8058	0.2627	0.0095
308.15 K glycerin (1) + RbCl (2) + H_2O (3)	−0.6747	−0.7856	0.2901	−0.7217	0.0096
308.15 K glycerin (1) + CsCl (2) + H_2O (3)	−0.4045	−0.2910	−0.1560	−0.4078	0.0034
density					
298.15 K glycerin (1) + NaCl (2) + H_2O (3)	0.1805	0.0416	0.0211	0.0186	0.0001
298.15 K glycerin (1) + KCl (2) + H_2O (3)	0.1636	0.0351	0.0470	0.0090	0.0007
298.15 K glycerin (1) + RbCl (2) + H_2O (3)	0.4058	−0.0981	−0.0463	0.0543	0.0003
298.15 K glycerin (1) + CsCl (2) + H_2O (3)	0.6543	−0.1491	−0.1303	0.0506	0.0005
308.15 K glycerin (1) + NaCl (2) + H_2O (3)	0.1771	0.0404	0.0248	0.0140	0.0001
308.15 K glycerin (1) + KCl (2) + H_2O (3)	0.1686	0.0368	0.0338	0.0119	0.0006
308.15 K glycerin (1) + RbCl (2) + H_2O (3)	0.4175	−0.1045	−0.0249	0.0253	0.0011
308.15 K glycerin (1) + CsCl (2) + H_2O (3)	0.6670	−0.1533	−0.0992	0.0075	0.0004
refractive index					
298.15 K glycerin (1) + NaCl (2) + H_2O (3)	0.3217	0.0486	0.0136	0.0083	0.0001
298.15 K glycerin (1) + KCl (2) + H_2O (3)	0.3141	0.0493	0.0262	−0.00001	0.0002
298.15 K glycerin (1) + RbCl (2) + H_2O (3)	0.3282	0.0363	0.0225	0.0036	0.0001
298.15 K glycerin (1) + CsCl (2) + H_2O (3)	0.3504	0.0208	0.0222	0.0003	0.0001
308.15 K glycerin (1) + NaCl (2) + H_2O (3)	0.3208	0.0473	0.0179	0.0039	0.0000
308.15 K glycerin (1) + KCl (2) + H_2O (3)	0.3143	0.0488	0.0273	−0.2009	0.0001
308.15 K glycerin (1) + RbCl (2) + H_2O (3)	0.3290	0.0353	0.0166	0.0089	0.0002
308.15 K glycerin (1) + CsCl (2) + H_2O (3)	0.3512	0.0188	0.0204	0.0041	0.0001

[a] $\delta = [\sum(\ln Y_{cal} - \ln Y_{exp})^2/N]^{0.5}$, where N is the number of experimental points.

Table 4. Density (ρ) and Refractive Index (n_D) for the Glycerin (1) + NaCl (2) + H_2O (3) Unsaturated System at (298.15 and 308.15) K

w_1	w_2	n_D	ρ / $g \cdot cm^{-3}$	w_1	w_2	n_D	ρ / $g \cdot m^{-3}$
298.15 K $w_1/w_3 = 1/9$							
0.0996	0.0239	1.34869	1.03794	0.0871	0.1273	1.36587	1.11181
0.0952	0.0465	1.35232	1.05361	0.0858	0.1443	1.36860	1.12385
0.0951	0.0692	1.35633	1.07008	0.0841	0.1613	1.37187	1.13791
0.0927	0.0872	1.35922	1.08276	0.0825	0.1711	1.37357	1.14557
0.0898	0.1072	1.36267	1.09777	0.0807	0.1927	1.37745	1.16251
298.15 K $w_1/w_3 = 3/7$							
0.2941	0.0199	1.37331	1.08488	0.2679	0.1074	1.38720	1.14664
0.2882	0.0388	1.37625	1.09783	0.2630	0.1233	1.38960	1.15810
0.2833	0.0566	1.37919	1.11052	0.2586	0.1379	1.39173	1.16796
0.2774	0.0740	1.38178	1.12248	0.2544	0.1526	1.39386	1.17816
0.2729	0.0914	1.38459	1.13488	0.2492	0.1691	1.39677	1.19141
298.15 K $w_1/w_3 = 5/5$							
0.4922	0.0156	1.39981	1.13389	0.4558	0.0879	1.40984	1.18088
0.4825	0.0313	1.40165	1.14360	0.4485	0.1032	1.41175	1.19196
0.4765	0.0467	1.40369	1.15342	0.4411	0.1177	1.41377	1.20209
0.4680	0.0623	1.40616	1.16451	0.4369	0.1262	1.41485	1.20754
0.4626	0.0744	1.40784	1.17261	0.4323	0.1364	1.41629	1.21464
298.15 K $w_1/w_3 = 7/3$							
0.6905	0.0134	1.42817	1.18555	0.6508	0.0699	1.43490	1.22100
0.6824	0.0235	1.42893	1.19086	0.6416	0.0817	1.43606	1.22823
0.6756	0.0348	1.43075	1.19884	0.6376	0.0883	1.43726	1.23317
0.6675	0.0461	1.43193	1.20589	0.6320	0.0976	1.43823	1.23718
0.6590	0.0569	1.43279	1.21088	0.6245	0.1075	1.43961	1.24583
298.15 K $w_1/w_3 = 9/1$							
0.8907	0.0087	1.45809	1.23603	0.8554	0.0466	1.46164	1.25816
0.8828	0.0173	1.45910	1.24129	0.8506	0.0542	1.46253	1.26289
0.8717	0.0313	1.45999	1.24878	0.8415	0.0630	1.46317	1.26785
0.8787	0.0238	1.46001	1.24558	0.8373	0.0680	1.46361	1.27079
0.8642	0.0387	1.46115	1.25377	0.8276	0.0798	1.46460	1.27785

w_1	w_2	n_D	ρ / $g \cdot cm^{-3}$	w_1	w_2	n_D	ρ / $g \cdot m^{-3}$
308.15 K $w_1/w_3 = 1/9$							
0.0975	0.0253	1.34715	1.03423	0.0875	0.1262	1.36501	1.10642
0.0967	0.0467	1.35155	1.04985	0.0857	0.1442	1.36688	1.11824
0.0945	0.0679	1.35459	1.06484	0.0843	0.1627	1.37142	1.13379
0.0953	0.0886	1.35910	1.08025	0.0829	0.1784	1.37423	1.14581
0.0892	0.1079	1.36112	1.09279	0.0801	0.2013	1.37712	1.16309
308.15 K $w_1/w_3 = 3/7$							
0.2941	0.0207	1.37358	1.08246	0.2685	0.1081	1.38628	1.14063
0.2878	0.0391	1.37553	1.09302	0.2633	0.1234	1.38953	1.15236
0.2836	0.0565	1.37857	1.10731	0.2586	0.1384	1.39181	1.16323
0.2783	0.0751	1.3813	1.11796	0.2540	0.1525	1.39295	1.17226
0.2729	0.0910	1.38376	1.12923	0.2492	0.1693	1.39566	1.18395
308.15 K $w_1/w_3 = 5/5$							
0.4918	0.0163	1.39811	1.12895	0.4557	0.0896	1.40942	1.17662
0.4866	0.0318	1.40208	1.13977	0.4499	0.1011	1.40992	1.18580
0.4758	0.0484	1.40250	1.14985	0.4426	0.1132	1.41282	1.19292
0.4692	0.0607	1.40549	1.15627	0.4300	0.1394	1.41499	1.21113
0.4615	0.0766	1.40636	1.16850				
308.15 K $w_1/w_3 = 7/3$							
0.6905	0.0116	1.42623	1.17873	0.6632	0.0522	1.43230	1.20322
0.6780	0.0305	1.42867	1.19090	0.6456	0.0754	1.43528	1.21845
0.6659	0.0488	1.43090	1.20249	0.6352	0.0913	1.43742	1.22914
308.15 K $w_1/w_3 = 9/1$							
0.8918	0.0086	1.45645	1.23057	0.8566	0.0478	1.46170	1.25322
0.8840	0.0159	1.45841	1.23399	0.8456	0.0603	1.46288	1.25910
0.8763	0.0256	1.45980	1.24054	0.8388	0.0679	1.46354	1.26501
0.8616	0.0420	1.46131	1.24976	0.8294	0.0780	1.46437	1.27082

decreased with increasing mass fraction of glycerin in the solvent and increased with the temperature increased, but for the glycerin + NaCl/KCl + H_2O systems, the density increases with the addition of glycerin and densities are not sensitive to an increase of the temperature. In our opinion, this phenomenon is mainly ascribed to two factors. The salt content should be the key factor that influenced the density for the glycerin + CsCl/RbCl + H_2O ternary systems, but for the glycerin + NaCl/KCl + H_2O systems, mixed solvent composition should be the key factor. The refractive index of the four ternary systems at (298.15 and 308.15 K) is illustrated in figure 1 (c). It can be seen that the refractive increases with increasing mass fraction of glycerin. However, the refractive is not sensitive to an increase of temperature. This phenomenon is agreement with refractive index data for glycerin-water saturated with sodium chloride at 20 °C reported by Chen et al.[19] But for the ethanol + NaCl/KCl + H_2O systems reported by Galleguillos et al.,[7] the density and refractive index decreased with increasing mass fraction ethanol of in the solvent. It can be explained by the smaller density of ethanol than that of glycerin.

The reliability of the measured mass fraction of salts, the refractive index, and density in saturated solutions were correlated, respectively, according to the equation[24]

$$\ln Y = A + Bw_1' + Cw_1'^2 + Dw_1'^3 \quad (1)$$

where w_1' is the mass fraction of glycerin in the salt-free mixed solvent and Y represents the mass fraction of salts (w_2) in the mixed solution, the density (ρ), the refractive index (n_D) of the ternary systems. A, B, C, and D are empirical constants whose values are listed in Table 3, together with the corresponding standard deviations of the fit. From the obtained standard deviations, we conclude that eq 1 can be successfully used to correlate the solubility, density, and refractive index data.

Table 5. Density (ρ) and Refractive Index (n_D) for the Glycerin (1) + KCl (2) + H_2O (3) Unsaturated System at (298.15 and 308.15) K

w_1	w_2	n_D	ρ / $g \cdot cm^{-3}$	w_1	w_2	n_D	ρ / $g \cdot cm^{-3}$
			298.15 K $w_1/w_3 = 1/9$				
0.1048	0.0233	1.34846	1.03708	0.0875	0.1257	1.36178	1.10431
0.0957	0.0454	1.35056	1.04961	0.0863	0.1442	1.36434	1.11753
0.0937	0.0676	1.35364	1.06425	0.0846	0.1612	1.36668	1.12949
0.0915	0.0877	1.35666	1.07887	0.0825	0.1778	1.36885	1.14107
0.0895	0.1072	1.35941	1.09234	0.0812	0.1924	1.37070	1.15099
			298.15 K $w_1/w_3 = 3/7$				
0.2933	0.0194	1.37240	1.08225	0.2683	0.1079	1.38286	1.13810
0.2904	0.0378	1.37491	1.09424	0.2634	0.1189	1.38407	1.14493
0.2852	0.0553	1.37699	1.10515	0.2597	0.1349	1.38638	1.15658
0.2782	0.0724	1.37876	1.11542	0.2553	0.1494	1.38838	1.16583
0.2733	0.0894	1.38079	1.12632	0.2519	0.1625	1.39038	1.17710
			298.15 K $w_1/w_3 = 5/5$				
0.4924	0.0148	1.39899	1.13169	0.4603	0.0785	1.40567	1.17043
0.4859	0.0278	1.39982	1.13852	0.4561	0.0905	1.40711	1.17829
0.4799	0.0408	1.40169	1.14739	0.4483	0.1009	1.40781	1.18413
0.4732	0.0540	1.40280	1.15470	0.4430	0.1139	1.40959	1.19331
0.4672	0.0662	1.40431	1.16257	0.4352	0.1296	1.41098	1.20266
			298.15 K $w_1/w_3 = 7/3$				
0.6925	0.0106	1.42804	1.18367	0.6587	0.0571	1.43160	1.20982
0.6851	0.0212	1.42887	1.18958	0.6537	0.0659	1.43270	1.21564
0.6789	0.0295	1.42937	1.19396	0.6480	0.0741	1.43275	1.21917
0.6705	0.0388	1.43010	1.19924	0.6417	0.0833	1.43371	1.22510
0.6654	0.0486	1.43124	1.20543	0.6326	0.0976	1.43486	1.23372
			298.15 K $w_1/w_3 = 9/1$				
0.8928	0.0079	1.45847	1.23581	0.8588	0.0455	1.46036	1.25546
0.8864	0.0152	1.45892	1.23944	0.8524	0.0512	1.46079	1.25697
0.8798	0.0224	1.45896	1.24321	0.8455	0.0590	1.46116	1.26119
0.8730	0.0299	1.45968	1.24716	0.8380	0.0686	1.46162	1.26827
0.8668	0.0367	1.45989	1.25014	0.8359	0.0708	1.46186	1.26868
			308.15 K $w_1/w_3 = 1/9$				
0.0963	0.0400	1.34831	1.04253	0.0862	0.1388	1.36125	1.10763
0.0927	0.0771	1.35289	1.06563	0.0833	0.1675	1.36461	1.12544
0.0890	0.1090	1.35714	1.08689	0.0807	0.2000	1.36909	1.14846
			308.15 K $w_1/w_3 = 3/7$				
0.2930	0.0293	1.37099	1.08271	0.2667	0.1090	1.38068	1.13342
0.2854	0.0498	1.37421	1.09721	0.2565	0.1463	1.38537	1.15867
0.2750	0.0867	1.37837	1.11986	0.2497	0.1692	1.38751	1.17191
			308.15 K $w_1/w_3 = 5/5$				
0.4865	0.0295	1.39832	1.12373	0.4383	0.1061	1.40484	1.17925
0.4714	0.0582	1.40132	1.15254	0.4369	0.1266	1.40800	1.19247
0.4575	0.0849	1.40400	1.16876	0.4281	0.1394	1.40922	1.20214
			308.15 K $w_1/w_3 = 7/3$				
0.6922	0.0108	1.42604	1.17849	0.6592	0.0587	1.42967	1.20524
0.6859	0.0201	1.42648	1.18160	0.6483	0.0671	1.42982	1.20811
0.6789	0.0292	1.42733	1.18804	0.6478	0.0743	1.43130	1.21478
0.6652	0.0483	1.42866	1.19808				
			308.15 K $w_1/w_3 = 9/1$				
0.8899	0.0094	1.45585	1.22859	0.8433	0.0614	1.45919	1.25771
0.8832	0.0189	1.45649	1.23328	0.8384	0.0685	1.45983	1.26172
0.8762	0.0264	1.45828	1.24888	0.8308	0.0763	1.46002	1.26585
0.8521	0.0528	1.45875	1.25332	0.8238	0.0832	1.46019	1.26682

Densities and refractive indexes in the four ternary systems were measured in undersaturated solutions in completion of this study. The experiment was carried out varying the concentration of the salts in the solutions using the same mass ratios of glycerin and water. From Tables 4 to 7, we can observe the values of density and refractive index for the glycerin + CsCl + H_2O ternary system increased with increasing the content of salt but decreased with increasing of temperatures. And the density and refractive index of the solutions also increased with increasing mass fraction ratios of glycerin + water, similar phenomenon was observed at the glycerin + NaCl + H_2O, glycerin + KCl + H_2O, and glycerin + RbCl + H_2O ternary systems in unsaturated solutions. However, the density of the ethanol + NaCl/KCl + H_2O systems reported by Galleguillos et al.[7] decreased with increasing mass fraction ratios of ethanol + water.

The reliability of the measured refractive index and density of the four ternary systems in the unsaturated solutions are ascertained by the following equations[7]

$$Y = (A_0 + A_1 w_2 + A_2 w_1 + A_3 w_1 w_2 + A_4 w_2 w_1^2) \times \exp[A_5(w_1/w_3) + A_6(w_1/w_3)^2] \quad (2)$$

where Y represents density (ρ) or the refractive index (n_D) of the ternary system, where w_1, w_2, and w_3 is the mass fractions of glycerin, salt, and water in the mixed solution, respectively. The coefficients of eq 2 (A_i, i = 1 to 6) along with the corresponding standard deviations for the investigated systems are given in table 8. On the basis of the obtained standard deviations, it can be seen that eq 2 are satisfactory for the systems in this work.

■ CONCLUSION

The solubility, refractive index, and density of the four ternary systems MCl + glycerin + MCl + H_2O (M = Na, K, Rb, Cs) have been determined at different temperatures, respectively. For the

Table 6. Density (ρ) and Refractive Index (n_D) for the Glycerin (1) + RbCl (2) + H_2O (3) Unsaturated System at (298.15 and 308.15) K

w_1	w_2	n_D	ρ / $g\cdot cm^{-3}$	w_1	w_2	n_D	ρ / $g\cdot cm^{-3}$
			298.15 K $w_1/w_3 = 1/9$				
0.0976	0.0317	1.34730	1.04455	0.0811	0.1972	1.36361	1.18723
0.0932	0.0741	1.35133	1.07812	0.0724	0.2773	1.37203	1.26738
0.0865	0.1418	1.35764	1.13444	0.0692	0.3097	1.37546	1.30044
			298.15 K $w_1/w_3 = 3/7$				
0.2829	0.0582	1.37472	1.11524	0.2310	0.2318	1.39008	1.26911
0.2674	0.1074	1.37927	1.15706	0.2271	0.2432	1.39173	1.28306
0.2424	0.1937	1.38647	1.23267	0.2151	0.2821	1.39477	1.32104
			298.15 K $w_1/w_3 = 5/5$				
0.4746	0.0498	1.40120	1.16184	0.4165	0.1668	1.40966	1.26107
0.4536	0.0927	1.40437	1.19666	0.3930	0.2125	1.41311	1.30272
0.4324	0.1347	1.40722	1.23162	0.3766	0.2467	1.41586	1.33610
			298.15 K $w_1/w_3 = 7/3$				
0.6727	0.0391	1.42969	1.20766	0.6025	0.1397	1.43427	1.28837
0.6473	0.0757	1.43140	1.23649	0.5918	0.1551	1.43616	1.30400
0.6241	0.1074	1.43289	1.26155	0.5832	0.1663	1.43677	1.3138
			298.15 K $w_1/w_3 = 9/1$				
0.8695	0.0297	1.45819	1.25323	0.8011	0.1097	1.46232	1.31787
0.8430	0.0618	1.45853	1.27548	0.7792	0.1336	1.46335	1.33323
0.8215	0.0865	1.46138	1.2988	0.7615	0.1508	1.46381	1.35266

w_1	w_2	n_D	ρ / $g\cdot cm^{-3}$	w_1	w_2	n_D	ρ / $g\cdot cm^{-3}$
			308.15 K $w_1/w_3 = 1/9$				
0.0926	0.0751	1.34977	1.07447	0.0758	0.2425	1.36658	1.22636
0.0872	0.1381	1.35587	1.12741	0.0722	0.2762	1.37000	1.25917
0.0811	0.1949	1.36140	1.17830	0.0692	0.3074	1.37342	1.29180
			308.15 K $w_1/w_3 = 3/7$				
0.2824	0.0609	1.37351	1.11376	0.2389	0.2055	1.38591	1.23886
0.2671	0.1143	1.37794	1.15834	0.2275	0.2427	1.38933	1.27480
0.2518	0.1613	1.38178	1.19728	0.2158	0.2821	1.39327	1.31644
			308.15 K $w_1/w_3 = 5/5$				
0.4755	0.0492	1.39925	1.15598	0.4158	0.1688	1.40795	1.25635
0.4548	0.0917	1.40243	1.19085	0.3965	0.2075	1.41090	1.29179
0.4331	0.1301	1.40485	1.22174	0.3761	0.2444	1.41363	1.32760
			308.15 K $w_1/w_3 = 7/3$				
0.6721	0.0391	1.42728	1.20170	0.6032	0.1381	1.43312	1.27166
0.6473	0.0740	1.42910	1.22871	0.5820	0.1684	1.43498	1.30950
0.6244	0.1083	1.43145	1.25763	0.5601	0.2007	1.43717	1.34030
			308.15 K $w_1/w_3 = 9/1$				
0.8687	0.0345	1.45675	1.23668	0.8025	0.1083	1.46054	1.31171
0.8478	0.0576	1.45819	1.27019	0.7726	0.1408	1.46188	1.33969
0.8236	0.0834	1.45908	1.29008	0.7636	0.1513	1.46226	1.34787

Table 7. Density (ρ) and Refractive Index (n_D) for the Glycerin (1) + CsCl (2) + H_2O (3) Unsaturated System at (298.15 and 308.15) K

w_1	w_2	n_D	ρ / $g\cdot cm^{-3}$	w_1	w_2	n_D	ρ / $g\cdot cm^{-3}$
			298.15 K $w_1/w_3 = 1/9$				
0.0939	0.0621	1.34926	1.07045	0.0733	0.2662	1.36829	1.27193
0.0911	0.1073	1.35330	1.11017	0.0701	0.3000	1.37260	1.31720
0.0890	0.1547	1.35783	1.15479	0.0672	0.3276	1.37506	1.34610
0.0804	0.1956	1.36111	1.19476	0.0649	0.3509	1.37786	1.37671
0.0757	0.2436	1.36588	1.24576	0.0618	0.3851	1.38236	1.42580
			298.15 K $w_1/w_3 = 3/7$				
0.2840	0.0532	1.37382	1.11393	0.2241	0.2527	1.39039	1.30851
0.2699	0.1050	1.37778	1.15920	0.2148	0.2835	1.39333	1.34462
0.2584	0.1456	1.38436	1.19742	0.2048	0.3133	1.39634	1.38157
0.2466	0.1828	1.38119	1.23445	0.1983	0.3386	1.39920	1.41500
0.2335	0.2187	1.38750	1.27240	0.1915	0.3647	1.40226	1.45000
			298.15 K $w_1/w_3 = 5/5$				
0.4745	0.0519	1.40101	1.16651	0.3836	0.2325	1.41417	1.34230
0.4545	0.0922	1.40364	1.20155	0.3683	0.2624	1.41657	1.37598
0.4340	0.1310	1.40647	1.23861	0.3556	0.2865	1.41833	1.38295
0.4140	0.1712	1.40933	1.27705	0.3436	0.3117	1.42160	1.43688
0.3982	0.2022	1.41155	1.30915	0.3330	0.3366	1.42293	1.46502
			298.15 K $w_1/w_3 = 7/3$				
0.6684	0.0414	1.42893	1.21184	0.5622	0.1945	1.43879	1.35675
0.6469	0.0749	1.43277	1.24380	0.5465	0.2187	1.43988	1.38295
0.6225	0.1088	1.43392	1.27230	0.5281	0.2456	1.44292	1.41656
0.6014	0.1406	1.43681	1.30718	0.5130	0.2670	1.44362	1.43873
0.5807	0.1702	1.43838	1.33510	0.5007	0.2847	1.44503	1.46009
			298.15 K $w_1/w_3 = 9/1$				
0.8680	0.0344	1.45933	1.26042	0.7520	0.1636	1.46571	1.38083
0.8449	0.0604	1.46016	1.28280	0.7299	0.1867	1.46641	1.39918
0.8189	0.0886	1.46157	1.30870	0.7149	0.2041	1.46782	1.42378
0.7947	0.1160	1.46305	1.33384	0.6951	0.2237	1.46845	1.44454
0.7696	0.1439	1.46371	1.35984	0.6721	0.2467	1.46932	1.47234

w_1	w_2	n_D	ρ / $g\cdot cm^{-3}$	w_1	w_2	n_D	ρ / $g\cdot cm^{-3}$
			308.15 K $w_1/w_3 = 1/9$				
0.0940	0.0605	1.34765	1.06498	0.0716	0.2841	1.36738	1.27713
0.0881	0.1190	1.35206	1.11267	0.0667	0.3395	1.37295	1.33907
0.0838	0.1650	1.35609	1.15545	0.0637	0.3708	1.37569	1.36901
0.0799	0.2015	1.35972	1.19313	0.0612	0.3933	1.37718	1.39051
0.0755	0.2466	1.36497	1.24661				
			308.15 K $w_1/w_3 = 3/7$				
0.2843	0.0557	1.37195	1.11037	0.2325	0.2265	1.38531	1.27013
0.2684	0.1050	1.37372	1.14650	0.2216	0.2621	1.38746	1.30262
0.2544	0.1516	1.37991	1.19848	0.2145	0.2888	1.39172	1.34205
0.2429	0.1906	1.38173	1.23038	0.2014	0.3337	1.39513	1.39050
			308.15 K $w_1/w_3 = 5/5$				
0.4750	0.0521	1.39930	1.16167	0.3665	0.2670	1.41564	1.37785
0.4309	0.1363	1.40105	1.22455	0.3527	0.2929	1.41631	1.41162
0.4135	0.1719	1.40831	1.27522	0.3415	0.3168	1.41843	1.42252
0.3963	0.2066	1.40919	1.30427	0.3402	0.3175	1.41985	1.44163
0.3787	0.2424	1.41180	1.34142	0.3271	0.3458	1.42046	1.46438
			308.15 K $w_1/w_3 = 7/3$				
0.6474	0.0755	1.42921	1.23668	0.5269	0.2470	1.43961	1.40467
0.5972	0.1469	1.43253	1.30015	0.5054	0.2777	1.44198	1.44431
0.5627	0.1956	1.43622	1.35170	0.4837	0.3084	1.44409	1.48124
			308.15 K $w_1/w_3 = 9/1$				
0.8547	0.0497	1.45612	1.26462	0.7435	0.1739	1.46369	1.38509
0.8150	0.0938	1.45884	1.30539	0.7056	0.2151	1.46550	1.42804
0.7761	0.1361	1.46141	1.34672	0.6707	0.2539	1.46734	1.47108

Table 8. Values of Parameters of eq 2 for the Density and Refractive Index of the Unsaturated Ternary Systems at (298.15 and 308.15) K

system	A_0	A_1	A_2	A_3	A_4	A_5	A_6	δ^a
			density					
298.15 K glycerin (1) + NaCl (2) + H_2O (3)	0.9908	0.7806	0.2677	−0.0446	0.1697	−0.0008	0.0006	0.0009
298.15 K glycerin (1) + KCl (2) + H_2O (3)	0.9931	0.7239	0.2501	0.4086	−0.2899	0.0051	−0.0005	0.0020
298.15 K glycerin (1) + RbCl (2) + H_2O (3)	0.9825	0.9378	0.2493	0.2560	−0.2203	0.0069	−0.0005	0.0029
298.15 K glycerin (1) + CsCl (2) + H_2O (3)	0.9635	1.0989	0.2346	0.3271	−0.5105	0.0194	−0.0016	0.0067
308.15 K glycerin (1) + NaCl (2) + H_2O (3)	0.9900	0.7400	0.2476	0.1483	−0.0989	0.0038	−0.0003	0.0008
308.15 K glycerin (1) + KCl (2) + H_2O (3)	0.9930	0.6470	0.2078	0.4209	−0.4768	0.0146	−0.0011	0.0022
308.15 K glycerin (1) + RbCl (2) + H_2O (3)	0.9707	0.9686	0.2837	0.0616	0.2609	−0.0042	0.0003	0.0032
308.15 K glycerin (1) + CsCl (2) + H_2O (3)	0.9709	0.9759	0.1887	0.8120	−1.0540	0.0252	−0.0019	0.0064
			refractive index					
298.15 K glycerin (1) + NaCl (2) + H_2O (3)	1.3306	0.1923	0.1306	−0.0397	0.1210	0.0014	−0.0009	0.0005
298.15 K glycerin (1) + KCl (2) + H_2O (3)	1.3305	0.1597	0.1295	−0.0516	0.1141	0.0021	−0.0002	0.0004
298.15 K glycerin (1) + RbCl (2) + H_2O (3)	1.3319	0.0969	0.1125	0.1778	−0.1734	0.0058	−0.0004	0.0007
298.15 K glycerin (1) + CsCl (2) + H_2O (3)	1.3281	0.1098	0.1222	0.1077	−0.0619	0.0049	−0.0004	0.0009
308.15 K glycerin (1) + NaCl (2) + H_2O (3)	1.3308	0.1719	0.1214	0.1075	−0.0704	0.0035	−0.0002	0.0006
308.15 K glycerin (1) + KCl (2) + H_2O (3)	1.3340	0.0947	0.0905	0.3725	−0.4352	0.0101	−0.0008	0.0009
308.15 K glycerin (1) + RbCl (2) + H_2O (3)	1.3285	0.1102	0.1232	0.0827	−0.0278	0.0036	−0.0003	0.0002
308.15 K glycerin (1) + CsCl (2) + H_2O (3)	1.3290	0.0929	0.1129	0.1577	−0.0946	0.0054	−0.0004	0.0019

[a] $\delta = [\Sigma(Y_{cal} - Y_{exp})^2/N]^{0.5}$, where N is the number of experimental points.

saturated systems, the solubilities increased with the increase of temperature and decreased with increasing glycerin composition. The refractive increases with increasing mass fraction of glycerin, but the refractive are not sensitive to an increase of temperature. And For the unsaturated systems, the density and refractive index both increased with increasing the content of salt, and increased with the increasing of glycerin to water ratio.

AUTHOR INFORMATION

Corresponding Author

*E-mail: lishuni@snnu.edu.cn (S.L.); hmch@snnu.edu.cn (M.H.).

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