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Effect of Temperature and Concentration of KBr or KNO_3 on the Volumetric and Transport Properties of Aqueous Solutions of Tripotassium Citrate

Mohammed Taghi Zafarani-Moattar* and Farhad Izadi

Physical Chemistry Department, University of Tabriz, Tabriz, Iran

S *Supporting Information*

ABSTRACT: Volumetric and transport properties of ternary aqueous solutions of tripotassium citrate in (KBr or KNO_3) + water have been determined in the different molality ranges of KBr and KNO_3 [(0.2, 0.3, 0.4, 0.5, 0.6, and 0.7) $mol \cdot kg^{-1}$] at T = (293.15 to 313.15) K. The apparent molar volume and apparent molar isentropic compressibility have been calculated from the measured density and speed of sound data for ternary aqueous solutions of tripotassium citrate. The apparent molar volume and apparent molar isentropic compressibility of ternary aqueous solutions of tripotassium citrate + (KBr or KNO_3) have been correlated with the Redlich−Mayer equation. Viscosity values of ternary aqueous solutions of tripotassium citrate + (KBr or KNO_3) have been fitted with the Jones−Dole equation. The results obtained have been interpreted in elucidating the effect of KBr or KNO_3 and temperature on the interaction of tripotassium citrate with water. Furthermore, density and viscosity values of ternary aqueous solutions of tripotassium citrate + (KBr or KNO_3) have successfully been predicted using the methods proposed by Laliberté et al. and Zafarani-Moattar et al. for mixtures of inorganic salts.

1. INTRODUCTION

The volumetric properties of electrolyte solutions have proven to be a very useful tool in elucidating the structural interactions (i.e., solute−solvent, solute−solute, and solvent−solvent) occurring in solution, because they may give us an indirect insight into the conformational features of the components in solution. The design and operation of industrial processes that involve electrolyte solutions require knowledge of rigorous models or experimental data to represent the nonideality of the mixtures. Accurate predictions of densities and viscosities of mixed electrolyte solutions are of great importance in industry.[1,2]

Citrate is biodegradable and nontoxic and could be discharged into biological wastewater treatment plants. Aqueous solutions of potassium citrate are of considerable significance in many other biochemical and chemical processes, and this salt is produced in large quantities and used in the food, cosmetic, pharmaceutical, and chemical industries.[3−6] Densities, speed of sound, and viscosity values of binary aqueous solutions of tripotassium citrate (K_3Cit) are available at very dilute concentrations.[7,8] These properties have been measured for this system over the entire concentration range at different temperatures in a previous work.[9] Volumetric and transport properties of binary aqueous solutions of potassium bromide (KBr) and potassium nitrate (KNO_3) are available in the literature. Density and viscosity values of binary aqueous solution of KBr and KNO_3 used in this work were taken from Supporting Information reported by Laliberté.[10,11]

In this work, volumetric and transport properties of ternary aqueous solutions of K_3Cit+ (KBr or KNO_3) have been determined in the different molality ranges of KBr and KNO_3 [(0.2, 0.3, 0.4, 0.5, 0.6, and 0.7) $mol \cdot kg^{-1}$] at T = (293.15, 298.15, 303.15, 308.15, and 313.15) K. Apparent molar volume (V_ϕ) and apparent molar isentropic compressibility (K_ϕ) values have been calculated from the density and speed of sound data for ternary aqueous solutions of K_3Cit + (KBr or KNO_3), and these quantities have been correlated with the Redlich−Mayer equation. Viscosity values of ternary aqueous solutions of K_3Cit + (KBr or KNO_3) have been fitted with the Jones−Dole equation.[12] Density and viscosity values of ternary aqueous solutions of tripotassium citrate + (KBr or KNO_3) have successfully been predicted on the basis of the Arrhenius type mixing rule using the methods proposed by Laliberté et al.[10,11] and Zafarani-Moattar and Majdan-Cegincara.[13]

2. EXPERIMENTAL PROCEDURE

2.1. Materials. Tripotassium citrate ($C_6H_5K_3O_7 \cdot H_2O$) with a minimum mass fraction purity of 0.99, potassium bromide with minimum mass fraction purity of 0.995, and potassium nitrate with minimum mass fraction purity of 0.99 were obtained from Merck. Tripotassium citrate was used without further purification. KBr and KNO_3 were dried in a free convection oven at about 110 °C for 24 h prior to use. The solutions were prepared using double-distilled−deionized water.

2.2. Apparatus and Procedure. The solutions were prepared by mass using an analytical balance (Shimatzu, 321 34553, Shimatzu Co., Japan) with an uncertainty of $\pm\ 1 \cdot 10^{-7}$ kg. Densities and speed of sounds were measured with a vibrating-tube densimeter (Anton Paar DSA-500, Austria). By this apparatus, the working temperature can be controlled within ± 0.001 K.

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Table 1. Experimental Density, d, Speed of Sound, u, Viscosity, η, Apparent Molar Volume, V_ϕ, and Apparent Molar Isentropic Compressibility, K_ϕ, Data for K_3Cit (1) + (KBr (2) or KNO_3 (3)) + H_2O (4) at Different Temperatures

m_1 $\text{mol}\cdot\text{kg}^{-1}$	$10^{-3}\cdot d$ $\text{kg}\cdot\text{m}^{-3}$	u $\text{m}\cdot\text{s}^{-1}$	η $\text{mPa}\cdot\text{s}$	$10^6\cdot V_\phi$ $\text{m}^3\cdot\text{mol}^{-1}$	$10^{13}\cdot K_\phi$ $\text{m}^3\cdot\text{mol}^{-1}\cdot\text{Pa}^{-1}$
T/K = 293.15					
m_2 = 0.2 $\text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.015189	1486.97	0.985		
0.2000	1.053197	1525.00	1.112	113.00 ± 0.09	−1.371 ± 0.003
0.4000	1.088467	1561.39	1.256	115.53 ± 0.04	−1.255 ± 0.001
0.6000	1.121330	1596.40	1.419	117.67 ± 0.03	−1.157 ± 0.001
0.7995	1.151926	1630.46	1.613	119.57 ± 0.02	−1.074 ± 0.001
1.0000	1.180499	1662.76	1.825	121.45 ± 0.01	−0.998 ± 0.001
1.2001	1.207569	1692.06	2.071	122.81 ± 0.01	−0.927 ± 0.001
m_2 = 0.3 $\text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.023339	1488.91	0.975		
0.2000	1.060984	1526.38	1.109	115.25 ± 0.09	−1.305 ± 0.003
0.4000	1.095967	1562.41	1.249	117.50 ± 0.04	−1.198 ± 0.001
0.6000	1.128530	1597.40	1.415	119.52 ± 0.03	−1.108 ± 0.001
0.8000	1.158726	1631.06	1.607	121.54 ± 0.02	−1.027 ± 0.001
1.0000	1.186999	1663.76	1.819	123.23 ± 0.01	−0.958 ± 0.001
1.2000	1.213639	1693.64	2.062	124.62 ± 0.01	−0.892 ± 0.001
m_2 = 0.4 $\text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.031606	1490.91	0.970		
0.2000	1.068612	1527.92	1.104	118.70 ± 0.09	−1.232 ± 0.003
0.4001	1.103065	1563.83	1.240	120.64 ± 0.04	−1.137 ± 0.001
0.6000	1.135097	1598.18	1.410	122.46 ± 0.02	−1.050 ± 0.001
0.8000	1.165092	1631.86	1.600	123.99 ± 0.02	−0.979 ± 0.001
1.0000	1.193199	1664.03	1.813	125.35 ± 0.01	−0.914 ± 0.001
1.2000	1.219595	1694.10	2.052	126.56 ± 0.01	−0.853 ± 0.001
m_2 = 0.5 $\text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.039705	1492.77	0.970		
0.2000	1.076290	1530.31	1.096	121.03 ± 0.08	−1.196 ± 0.003
0.4000	1.110395	1565.83	1.235	122.69 ± 0.04	−1.095 ± 0.001
0.6000	1.142197	1599.18	1.400	124.24 ± 0.02	−1.005 ± 0.001
0.8000	1.171892	1631.86	1.589	125.68 ± 0.02	−0.933 ± 0.001
1.0000	1.199599	1665.03	1.801	127.06 ± 0.01	−0.877 ± 0.001
1.2000	1.225573	1694.34	2.031	128.29 ± 0.01	−0.816 ± 0.001
m_2 = 0.6 $\text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.047694	1493.36	0.950		
0.2000	1.083904	1531.31	1.085	123.07 ± 0.08	−1.164 ± 0.003
0.4000	1.117642	1566.49	1.232	124.63 ± 0.04	−1.057 ± 0.001
0.6000	1.149060	1600.52	1.394	126.15 ± 0.02	−0.975 ± 0.001
0.8000	1.178429	1633.21	1.574	127.48 ± 0.02	−0.905 ± 0.001
1.0000	1.205829	1665.55	1.789	128.77 ± 0.01	−0.846 ± 0.001
1.2000	1.231557	1695.02	2.026	129.89 ± 0.01	−0.789 ± 0.001
m_2 = 0.7 $\text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.055698	1497.07	0.936		
0.2000	1.091370	1532.41	1.083	125.77 ± 0.08	−1.046 ± 0.002
0.4000	1.124606	1567.87	1.220	127.18 ± 0.04	−0.982 ± 0.001
0.6001	1.155637	1601.81	1.378	128.46 ± 0.02	−0.914 ± 0.001
0.8001	1.184571	1634.70	1.555	129.69 ± 0.02	−0.853 ± 0.001
1.0001	1.211891	1665.84	1.771	130.60 ± 0.01	−0.798 ± 0.001
1.2000	1.237331	1696.63	2.011	131.60 ± 0.01	−0.750 ± 0.001
T/K = 298.15					
m_2 = 0.2 $\text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.013796	1500.61	0.873		
0.2000	1.051639	1537.45	0.992	113.69 ± 0.09	−1.305 ± 0.003
0.4000	1.086705	1572.50	1.109	116.33 ± 0.04	−1.191 ± 0.001
0.6000	1.119378	1606.44	1.267	118.49 ± 0.03	−1.099 ± 0.001
0.7995	1.149831	1639.26	1.436	120.34 ± 0.02	−1.021 ± 0.001
1.0000	1.178269	1670.93	1.624	122.19 ± 0.01	−0.950 ± 0.001
1.2001	1.205333	1701.67	1.836	123.43 ± 0.01	−0.891 ± 0.001
m_2 = 0.3 $\text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.022076	1503.20	0.863		
0.2000	1.059468	1539.38	0.986	116.36 ± 0.09	−1.233 ± 0.003
0.4000	1.094167	1573.39	1.107	118.69 ± 0.04	−1.122 ± 0.001
0.6000	1.126430	1607.40	1.261	120.77 ± 0.03	−1.039 ± 0.001
0.8000	1.156726	1640.00	1.416	122.35 ± 0.02	−0.971 ± 0.001
1.0000	1.184859	1671.16	1.616	124.01 ± 0.01	−0.904 ± 0.001
1.2000	1.211429	1702.11	1.830	125.32 ± 0.01	−0.850 ± 0.001
m_2 = 0.4 $\text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.030172	1504.04	0.860		
0.2000	1.067069	1540.52	0.980	119.13 ± 0.09	−1.190 ± 0.003
0.4001	1.101344	1575.05	1.106	121.24 ± 0.04	−1.087 ± 0.001
0.6000	1.133259	1607.97	1.253	123.03 ± 0.02	−1.000 ± 0.001
0.8000	1.163131	1640.85	1.400	124.56 ± 0.02	−0.934 ± 0.001
1.0000	1.191092	1671.89	1.610	125.93 ± 0.01	−0.871 ± 0.001
1.2000	1.217329	1701.55	1.822	127.17 ± 0.01	−0.818 ± 0.001
m_2 = 0.5 $\text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.038336	1506.46	0.857		
0.2000	1.074768	1543.45	0.979	121.67 ± 0.08	−1.152 ± 0.002
0.4000	1.108695	1576.93	1.105	123.39 ± 0.04	−1.036 ± 0.001
0.6000	1.140300	1608.88	1.245	125.00 ± 0.02	−0.950 ± 0.001
0.8000	1.169802	1641.86	1.396	126.47 ± 0.02	−0.890 ± 0.001
1.0000	1.197499	1672.03	1.607	127.70 ± 0.01	−0.828 ± 0.001
1.2000	1.223548	1702.02	1.813	128.77 ± 0.01	−0.781 ± 0.001
m_2 = 0.6 $\text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.046105	1506.05	0.846		
0.2000	1.082110	1543.92	0.978	123.94 ± 0.08	−1.133 ± 0.002
0.4000	1.115634	1577.07	1.104	125.53 ± 0.04	−1.006 ± 0.001
0.6000	1.146832	1609.00	1.239	127.07 ± 0.02	−0.920 ± 0.001
0.8000	1.176075	1642.54	1.390	128.31 ± 0.02	−0.865 ± 0.001
1.0000	1.203469	1673.08	1.603	129.44 ± 0.01	−0.806 ± 0.001
1.2000	1.229108	1703.17	1.807	130.52 ± 0.01	−0.756 ± 0.001
m_2 = 0.7 $\text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.054223	1509.08	0.832		
0.2000	1.089751	1544.29	0.975	126.36 ± 0.08	−1.019 ± 0.002
0.4000	1.122809	1577.78	1.100	127.86 ± 0.04	−0.935 ± 0.001
0.6001	1.153782	1610.71	1.233	129.00 ± 0.02	−0.872 ± 0.001
0.8001	1.182596	1643.20	1.388	130.23 ± 0.02	−0.817 ± 0.001

Table 1. Continued

m_1 $\mathrm{mol\cdot kg^{-1}}$	$10^{-3}\cdot d$ $\mathrm{kg\cdot m^{-3}}$	u $\mathrm{m\cdot s^{-1}}$	η $\mathrm{mPa\cdot s}$	$10^{6}\cdot V_{\phi}$ $\mathrm{m^3\cdot mol^{-1}}$	$10^{13}\cdot K_{\phi}$ $\mathrm{m^3\cdot mol^{-1}\cdot Pa^{-1}}$
1.0001	1.209758	1674.30	1.587	131.17 ± 0.01	−0.767 ± 0.001
1.2000	1.235303	1703.55	1.791	132.01 ± 0.01	−0.719 ± 0.001
T/K = 303.15					
$m_2 = 0.2\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.012300	1512.48	0.782		
0.2000	1.049999	1548.28	0.899	114.29 ± 0.09	−1.251 ± 0.003
0.4000	1.085029	1582.79	1.003	116.67 ± 0.04	−1.149 ± 0.001
0.6000	1.117591	1615.18	1.133	118.87 ± 0.03	−1.054 ± 0.001
0.7995	1.147901	1647.27	1.289	120.79 ± 0.02	−0.980 ± 0.001
1.0000	1.176300	1678.28	1.458	122.59 ± 0.01	−0.914 ± 0.001
1.2001	1.203315	1708.10	1.648	123.80 ± 0.01	−0.857 ± 0.001
$m_2 = 0.3\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.020507	1514.18	0.772		
0.2000	1.057747	1549.42	0.897	116.99 ± 0.09	−1.184 ± 0.003
0.4000	1.092267	1583.39	1.000	119.39 ± 0.04	−1.088 ± 0.001
0.6000	1.124530	1616.40	1.127	121.22 ± 0.03	−1.008 ± 0.001
0.8000	1.154626	1647.46	1.282	122.92 ± 0.02	−0.936 ± 0.001
1.0000	1.182799	1678.76	1.447	124.43 ± 0.01	−0.875 ± 0.001
1.2000	1.209412	1708.23	1.646	125.64 ± 0.01	−0.820 ± 0.001
$m_2 = 0.4\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.028555	1516.04	0.769		
0.2000	1.065314	1551.53	0.895	119.70 ± 0.09	−1.140 ± 0.002
0.4001	1.099480	1584.63	0.997	121.75 ± 0.04	−1.036 ± 0.001
0.6000	1.131285	1617.20	1.120	123.53 ± 0.02	−0.960 ± 0.001
0.8000	1.161058	1648.90	1.277	125.04 ± 0.02	−0.894 ± 0.001
1.0000	1.188880	1678.90	1.435	126.45 ± 0.01	−0.834 ± 0.001
1.2000	1.215236	1708.56	1.644	127.51 ± 0.01	−0.783 ± 0.001
$m_2 = 0.5\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.036786	1517.15	0.765		
0.2000	1.073072	1553.01	0.893	122.28 ± 0.08	−1.101 ± 0.002
0.4000	1.106895	1586.23	0.995	123.91 ± 0.04	−1.001 ± 0.001
0.6000	1.138397	1618.58	1.118	125.49 ± 0.02	−0.925 ± 0.001
0.8000	1.167822	1648.86	1.276	126.92 ± 0.02	−0.855 ± 0.001
1.0000	1.195409	1678.03	1.426	128.17 ± 0.01	−0.795 ± 0.001
1.2000	1.221249	1708.94	1.643	129.33 ± 0.01	−0.751 ± 0.001
$m_2 = 0.6\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.044604	1517.90	0.760		
0.2000	1.079290	1555.73	0.891	124.71 ± 0.08	−1.066 ± 0.002
0.4000	1.113833	1587.56	0.993	126.16 ± 0.04	−0.968 ± 0.001
0.6000	1.144873	1619.92	1.116	127.72 ± 0.02	−0.893 ± 0.001
0.8000	1.173936	1650.57	1.275	129.01 ± 0.02	−0.826 ± 0.001
1.0000	1.201303	1680.55	1.423	130.02 ± 0.01	−0.772 ± 0.001
1.2000	1.227006	1709.16	1.641	130.96 ± 0.01	−0.723 ± 0.001
$m_2 = 0.7\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.052518	1520.44	0.740		
0.2000	1.087952	1553.85	0.881	126.73 ± 0.08	−0.956 ± 0.002
0.4000	1.120899	1586.60	0.990	128.28 ± 0.04	−0.889 ± 0.001
0.6001	1.151711	1619.29	1.113	129.51 ± 0.02	−0.835 ± 0.001
0.8001	1.180386	1650.03	1.271	130.78 ± 0.02	−0.778 ± 0.001
1.0001	1.207480	1680.21	1.422	131.67 ± 0.01	−0.731 ± 0.001
1.2000	1.232980	1709.51	1.617	132.46 ± 0.01	−0.689 ± 0.001
T/K = 308.15					
$m_2 = 0.2\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.010632	1523.42	0.698		
0.2000	1.048148	1558.28	0.802	115.06 ± 0.09	−1.202 ± 0.003
0.4000	1.082959	1590.71	0.901	117.54 ± 0.04	−1.090 ± 0.001
0.6000	1.115440	1623.05	1.028	119.56 ± 0.03	−1.011 ± 0.001
0.7995	1.145752	1653.95	1.162	121.30 ± 0.02	−0.940 ± 0.001
1.0000	1.174110	1684.53	1.311	123.03 ± 0.01	−0.879 ± 0.001
1.2001	1.201175	1713.44	1.487	124.13 ± 0.01	−0.825 ± 0.001
$m_2 = 0.3\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.018897	1524.57	0.688		
0.2000	1.055956	1559.39	0.802	117.76 ± 0.09	−1.151 ± 0.002
0.4000	1.090367	1591.39	0.900	120.00 ± 0.04	−1.040 ± 0.001
0.6000	1.122530	1623.40	1.022	121.77 ± 0.03	−0.966 ± 0.001
0.8000	1.152606	1654.46	1.159	123.35 ± 0.02	−0.900 ± 0.001
1.0000	1.180749	1684.76	1.307	124.80 ± 0.01	−0.843 ± 0.001
1.2000	1.206976	1713.79	1.479	126.25 ± 0.01	−0.790 ± 0.001
$m_2 = 0.4\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.026732	1526.42	0.682		
0.2000	1.063355	1560.82	0.801	120.24 ± 0.09	−1.093 ± 0.002
0.4001	1.097413	1592.35	0.897	122.24 ± 0.04	−0.987 ± 0.001
0.6000	1.129113	1624.54	1.019	124.00 ± 0.02	−0.921 ± 0.001
0.8000	1.158816	1655.20	1.155	125.47 ± 0.02	−0.859 ± 0.001
1.0000	1.186571	1685.41	1.300	126.86 ± 0.01	−0.805 ± 0.001
1.2000	1.212659	1713.90	1.470	128.07 ± 0.01	−0.754 ± 0.001
$m_2 = 0.5\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.035001	1527.70	0.678		
0.2000	1.071163	1561.53	0.800	122.77 ± 0.08	−1.033 ± 0.002
0.4000	1.104895	1593.93	0.894	124.34 ± 0.04	−0.951 ± 0.001
0.6000	1.136257	1625.18	1.011	125.99 ± 0.02	−0.880 ± 0.001
0.8000	1.165642	1655.86	1.151	127.34 ± 0.02	−0.822 ± 0.001
1.0000	1.193109	1685.83	1.295	128.61 ± 0.01	−0.771 ± 0.001
1.2000	1.218962	1714.26	1.460	129.69 ± 0.01	−0.723 ± 0.001
$m_2 = 0.6\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.042987	1527.77	0.674		
0.2000	1.078643	1562.42	0.799	125.41 ± 0.08	−1.009 ± 0.002
0.4000	1.111892	1594.00	0.891	126.85 ± 0.04	−0.912 ± 0.001
0.6000	1.142836	1625.74	1.008	128.32 ± 0.02	−0.848 ± 0.001
0.8000	1.171855	1656.97	1.148	129.50 ± 0.02	−0.795 ± 0.001
1.0000	1.198884	1686.08	1.291	130.73 ± 0.01	−0.741 ± 0.001
1.2000	1.224614	1714.94	1.453	131.54 ± 0.01	−0.698 ± 0.001
$m_2 = 0.7\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.050610	1529.56	0.664		
0.2000	1.085927	1562.91	0.797	127.28 ± 0.08	−0.940 ± 0.002
0.4000	1.118802	1595.29	0.882	128.65 ± 0.04	−0.871 ± 0.001
0.6001	1.149507	1626.31	1.000	129.92 ± 0.02	−0.808 ± 0.001
0.8001	1.178099	1657.55	1.145	131.18 ± 0.02	−0.759 ± 0.001
1.0001	1.205051	1686.95	1.275	132.13 ± 0.01	−0.711 ± 0.001
1.2000	1.230391	1715.66	1.442	132.97 ± 0.01	−0.669 ± 0.001

Table 1. Continued

m_1	$10^{-3}\cdot d$	u	η	$10^6\cdot V_\phi$	$10^{13}\cdot K_\phi$
$\mathrm{mol\cdot kg^{-1}}$	$\mathrm{kg\cdot m^{-3}}$	$\mathrm{m\cdot s^{-1}}$	$\mathrm{mPa\cdot s}$	$\mathrm{m^3\cdot mol^{-1}}$	$\mathrm{m^3\cdot mol^{-1}\cdot Pa^{-1}}$
$T/\mathrm{K} = 313.15$					
$m_2 = 0.2\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.008742	1532.35	0.622		
0.2000	1.046084	1566.18	0.727	115.78 ± 0.09	−1.157 ± 0.003
0.4000	1.080813	1597.77	0.827	118.05 ± 0.04	−1.053 ± 0.001
0.6000	1.113293	1629.57	0.929	119.88 ± 0.03	−0.981 ± 0.001
0.7995	1.143241	1659.00	1.042	121.96 ± 0.02	−0.906 ± 0.001
1.0000	1.171599	1688.92	1.190	123.56 ± 0.01	−0.849 ± 0.001
1.2001	1.198543	1717.19	1.340	124.67 ± 0.01	−0.797 ± 0.001
$m_2 = 0.3\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.016994	1532.80	0.612		
0.2000	1.053923	1566.67	0.727	118.27 ± 0.09	−1.112 ± 0.002
0.4000	1.088207	1598.39	0.825	120.52 ± 0.04	−1.012 ± 0.001
0.6000	1.120330	1629.40	0.926	122.16 ± 0.03	−0.941 ± 0.001
0.8000	1.150356	1659.46	1.039	123.69 ± 0.02	−0.873 ± 0.001
1.0000	1.178409	1689.06	1.187	125.16 ± 0.01	−0.818 ± 0.001
1.2000	1.204703	1717.52	1.333	126.50 ± 0.01	−0.767 ± 0.001
$m_2 = 0.4\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.024777	1534.70	0.608		
0.2000	1.061293	1568.05	0.726	120.65 ± 0.09	−1.054 ± 0.002
0.4001	1.095226	1599.46	0.820	122.70 ± 0.04	−0.962 ± 0.001
0.6000	1.126887	1630.92	0.920	124.36 ± 0.03	−0.897 ± 0.001
0.8000	1.156455	1659.76	1.037	125.89 ± 0.02	−0.829 ± 0.001
1.0000	1.184160	1689.87	1.183	127.24 ± 0.01	−0.780 ± 0.001
1.2000	1.210150	1717.99	1.323	128.47 ± 0.01	−0.732 ± 0.001
$m_2 = 0.5\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.032880	1535.45	0.605		
0.2000	1.068894	1568.72	0.725	123.37 ± 0.08	−1.006 ± 0.002
0.4000	1.102545	1599.75	0.818	124.79 ± 0.04	−0.919 ± 0.001
0.6000	1.133897	1631.18	0.918	126.29 ± 0.02	−0.859 ± 0.001
0.8000	1.163192	1660.06	1.030	127.67 ± 0.02	−0.796 ± 0.001
1.0000	1.190609	1690.13	1.175	128.92 ± 0.01	−0.750 ± 0.001
1.2000	1.216476	1718.16	1.321	129.95 ± 0.01	−0.704 ± 0.001
$m_2 = 0.6\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.041011	1535.92	0.602		
0.2000	1.076533	1569.74	0.725	125.95 ± 0.08	−0.976 ± 0.002
0.4000	1.109646	1600.10	0.812	127.41 ± 0.04	−0.877 ± 0.001
0.6000	1.140560	1631.49	0.912	128.73 ± 0.02	−0.821 ± 0.001
0.8000	1.169537	1661.94	1.028	129.86 ± 0.02	−0.770 ± 0.001
1.0000	1.196429	1691.84	1.170	131.15 ± 0.01	−0.723 ± 0.001
1.2000	1.222219	1718.48	1.317	131.85 ± 0.01	−0.676 ± 0.001
$m_2 = 0.7\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.048619	1538.14	0.595		
0.2000	1.083798	1570.11	0.725	127.76 ± 0.08	−0.895 ± 0.002
0.4000	1.116570	1600.76	0.808	129.15 ± 0.04	−0.825 ± 0.001
0.6001	1.147249	1631.75	0.903	130.29 ± 0.02	−0.776 ± 0.001
0.8001	1.175736	1662.54	1.022	131.57 ± 0.02	−0.731 ± 0.001
1.0001	1.202572	1692.27	1.167	132.55 ± 0.01	−0.689 ± 0.001
1.2000	1.227943	1719.99	1.310	133.31 ± 0.01	−0.648 ± 0.001
$T/\mathrm{K} = 293.15$					

Table 1. Continued

m_1	$10^{-3}\cdot d$	u	η	$10^6\cdot V_\phi$	$10^{13}\cdot K_\phi$
$\mathrm{mol\cdot kg^{-1}}$	$\mathrm{kg\cdot m^{-3}}$	$\mathrm{m\cdot s^{-1}}$	$\mathrm{mPa\cdot s}$	$\mathrm{m^3\cdot mol^{-1}}$	$\mathrm{m^3\cdot mol^{-1}\cdot Pa^{-1}}$
$m_3 = 0.2\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.010621	1490.65	0.980		
0.1500	1.039472	1519.25	1.088	111.49 ± 0.12	−1.415 ± 0.004
0.3002	1.066788	1547.06	1.196	113.49 ± 0.06	−1.323 ± 0.002
0.4498	1.092547	1573.15	1.316	115.31 ± 0.04	−1.233 ± 0.001
0.6000	1.117053	1599.93	1.448	116.99 ± 0.03	−1.167 ± 0.001
0.7499	1.140178	1624.91	1.592	118.63 ± 0.02	−1.098 ± 0.001
0.9000	1.162337	1650.59	1.753	119.93 ± 0.02	−1.045 ± 0.001
$m_3 = 0.3\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.016689	1493.50	0.977		
0.1500	1.045154	1521.83	1.083	114.39 ± 0.12	−1.352 ± 0.004
0.3000	1.072098	1549.76	1.193	116.17 ± 0.06	−1.273 ± 0.002
0.4500	1.097607	1576.45	1.313	117.84 ± 0.04	−1.193 ± 0.001
0.6000	1.121853	1603.23	1.447	119.28 ± 0.03	−1.129 ± 0.001
0.7500	1.144798	1628.21	1.591	120.72 ± 0.02	−1.063 ± 0.001
0.9001	1.166446	1652.18	1.750	122.22 ± 0.02	−1.002 ± 0.001
$m_3 = 0.4\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.022681	1497.36	0.973		
0.1500	1.050841	1525.02	1.080	116.70 ± 0.12	−1.278 ± 0.004
0.3000	1.077477	1552.00	1.190	118.43 ± 0.06	−1.199 ± 0.002
0.4502	1.102814	1578.38	1.310	119.84 ± 0.04	−1.130 ± 0.001
0.6002	1.126844	1604.38	1.444	121.14 ± 0.03	−1.070 ± 0.001
0.7500	1.149536	1629.51	1.588	122.50 ± 0.02	−1.013 ± 0.001
0.9018	1.171390	1654.99	1.745	123.75 ± 0.02	−0.963 ± 0.001
$m_3 = 0.5\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.028599	1501.03	0.970		
0.1500	1.056408	1528.09	1.077	119.24 ± 0.12	−1.208 ± 0.004
0.3000	1.082677	1554.50	1.186	120.96 ± 0.05	−1.134 ± 0.002
0.4500	1.107664	1580.98	1.304	122.23 ± 0.03	−1.077 ± 0.001
0.6000	1.131344	1606.38	1.438	123.50 ± 0.02	−1.018 ± 0.001
0.7500	1.153936	1631.91	1.583	124.56 ± 0.02	−0.969 ± 0.001
0.9000	1.175546	1656.57	1.735	125.45 ± 0.02	−0.923 ± 0.001
$m_3 = 0.6\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.034609	1504.40	0.967		
0.1500	1.062135	1531.64	1.065	121.30 ± 0.11	−1.171 ± 0.003
0.2999	1.088109	1558.41	1.166	122.95 ± 0.05	−1.102 ± 0.002
0.4500	1.112847	1584.09	1.286	124.15 ± 0.03	−1.036 ± 0.001
0.5974	1.135907	1609.52	1.414	125.29 ± 0.02	−0.985 ± 0.001
0.7502	1.158528	1635.32	1.563	126.50 ± 0.02	−0.935 ± 0.001
0.9048	1.180576	1659.66	1.730	127.29 ± 0.01	−0.885 ± 0.001
$m_3 = 0.7\ \mathrm{mol\cdot kg^{-1}}$					
0.0000	1.040367	1507.61	0.963		
0.1500	1.067557	1534.21	1.047	123.62 ± 0.11	−1.105 ± 0.003
0.2999	1.093268	1560.71	1.145	125.00 ± 0.05	−1.049 ± 0.002
0.4501	1.117630	1586.76	1.261	126.35 ± 0.03	−0.992 ± 0.001
0.6001	1.140718	1611.95	1.398	127.53 ± 0.02	−0.939 ± 0.001
0.8333	1.174687	1650.88	1.620	128.77 ± 0.02	−0.872 ± 0.001
0.9052	1.184628	1662.11	1.700	129.17 ± 0.01	−0.851 ± 0.001
$T/\mathrm{K} = 298.15$					
$m_3 = 0.2\ \mathrm{mol\cdot kg^{-1}}$					

Table 1. Continued

m_1 $\text{mol}\cdot\text{kg}^{-1}$	$10^{-3}\cdot d$ $\text{kg}\cdot\text{m}^{-3}$	u $\text{m}\cdot\text{s}^{-1}$	η $\text{mPa}\cdot\text{s}$	$10^6\cdot V_\phi$ $\text{m}^3\cdot\text{mol}^{-1}$	$10^{13}\cdot K_\phi$ $\text{m}^3\cdot\text{mol}^{-1}\cdot\text{Pa}^{-1}$
0.0000	1.009415	1504.00	0.871		
0.1500	1.038147	1531.68	0.967	112.17 ± 0.12	−1.347 ± 0.004
0.3002	1.065297	1558.53	1.062	114.33 ± 0.06	−1.257 ± 0.002
0.4498	1.090949	1584.38	1.168	116.07 ± 0.04	−1.179 ± 0.001
0.6000	1.115407	1609.91	1.285	117.63 ± 0.03	−1.112 ± 0.001
0.7499	1.138542	1633.91	1.412	119.12 ± 0.02	−1.047 ± 0.001
0.9000	1.160502	1659.12	1.552	120.55 ± 0.02	−0.997 ± 0.001
$m_3 = 0.3\ \text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.015467	1506.99	0.869		
0.1500	1.043810	1534.42	0.965	115.09 ± 0.12	−1.285 ± 0.004
0.3000	1.070618	1560.76	1.060	116.91 ± 0.06	−1.198 ± 0.002
0.4500	1.096047	1586.95	1.164	118.49 ± 0.04	−1.131 ± 0.001
0.6000	1.120203	1612.23	1.282	119.89 ± 0.03	−1.068 ± 0.001
0.7500	1.143078	1636.91	1.409	121.29 ± 0.02	−1.010 ± 0.001
0.9001	1.164806	1660.72	1.545	122.61 ± 0.02	−0.956 ± 0.001
$m_3 = 0.4\ \text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.02125	1510.14	0.867		
0.1500	1.049288	1537.24	0.959	117.39 ± 0.12	−1.228 ± 0.003
0.3000	1.075803	1563.48	1.056	119.12 ± 0.06	−1.149 ± 0.002
0.4502	1.100969	1589.65	1.157	120.64 ± 0.04	−1.087 ± 0.001
0.6002	1.124855	1615.14	1.277	121.95 ± 0.03	−1.029 ± 0.001
0.7500	1.147435	1639.60	1.408	123.28 ± 0.02	−0.973 ± 0.001
0.9018	1.169293	1662.91	1.537	124.40 ± 0.02	−0.919 ± 0.001
$m_3 = 0.5\ \text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.027126	1513.37	0.862		
0.1500	1.054811	1540.10	0.952	119.94 ± 0.12	−1.169 ± 0.003
0.3000	1.080977	1566.00	1.050	121.60 ± 0.05	−1.094 ± 0.002
0.4500	1.105864	1591.38	1.155	122.85 ± 0.03	−1.033 ± 0.001
0.6000	1.129474	1616.38	1.276	124.07 ± 0.02	−0.978 ± 0.001
0.7500	1.151936	1641.51	1.400	125.17 ± 0.02	−0.932 ± 0.001
0.9000	1.173430	1664.63	1.532	126.07 ± 0.02	−0.883 ± 0.001
$m_3 = 0.6\ \text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.032835	1517.00	0.862		
0.1500	1.060218	1543.38	0.950	122.10 ± 0.11	−1.114 ± 0.003
0.2999	1.086080	1568.78	1.046	123.66 ± 0.05	−1.041 ± 0.002
0.4500	1.110731	1594.12	1.141	124.79 ± 0.03	−0.987 ± 0.001
0.5974	1.133676	1618.92	1.275	125.94 ± 0.02	−0.940 ± 0.001
0.7502	1.156277	1643.72	1.397	127.04 ± 0.02	−0.891 ± 0.001
0.9048	1.178268	1667.92	1.528	127.80 ± 0.01	−0.847 ± 0.001
$m_3 = 0.7\ \text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.038556	1520.05	0.859		
0.1500	1.065619	1545.61	0.930	124.33 ± 0.11	−1.045 ± 0.003
0.2999	1.091217	1571.22	1.023	125.67 ± 0.05	−0.994 ± 0.002
0.4501	1.115515	1596.51	1.116	126.91 ± 0.03	−0.944 ± 0.001
0.6001	1.138579	1620.82	1.250	127.98 ± 0.02	−0.894 ± 0.001
0.8333	1.172456	1658.02	1.450	129.20 ± 0.02	−0.829 ± 0.001
0.9052	1.182361	1669.58	1.503	129.60 ± 0.01	−0.812 ± 0.001
$T/\text{K} = 303.15$					
$m_3 = 0.2\ \text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.007803	1515.58	0.778		
0.1500	1.036418	1542.67	0.866	112.81 ± 0.12	−1.298 ± 0.004
0.3002	1.063416	1569.27	0.946	115.09 ± 0.06	−1.217 ± 0.002
0.4498	1.088974	1593.75	1.037	116.75 ± 0.04	−1.131 ± 0.001
0.6000	1.113207	1619.00	1.140	118.48 ± 0.03	−1.070 ± 0.001
0.7499	1.136282	1642.46	1.252	119.87 ± 0.02	−1.008 ± 0.001
0.9000	1.158360	1666.29	1.373	121.05 ± 0.02	−0.958 ± 0.001
$m_3 = 0.3\ \text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.013822	1518.72	0.776		
0.1500	1.042068	1545.57	0.863	115.60 ± 0.12	−1.239 ± 0.003
0.3000	1.068768	1571.06	0.943	117.47 ± 0.06	−1.151 ± 0.002
0.4500	1.094097	1596.15	1.033	119.05 ± 0.04	−1.082 ± 0.001
0.6000	1.118203	1619.93	1.137	120.38 ± 0.03	−1.017 ± 0.001
0.7500	1.140978	1644.91	1.247	121.80 ± 0.02	−0.969 ± 0.001
0.9001	1.162656	1669.07	1.370	123.09 ± 0.02	−0.922 ± 0.001
$m_3 = 0.4\ \text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.019623	1522.03	0.775		
0.1500	1.047515	1548.39	0.861	118.22 ± 0.12	−1.175 ± 0.003
0.3000	1.073949	1573.98	0.941	119.75 ± 0.06	−1.102 ± 0.002
0.4502	1.098986	1598.93	1.031	121.31 ± 0.04	−1.037 ± 0.001
0.6002	1.122767	1623.76	1.134	122.61 ± 0.03	−0.983 ± 0.001
0.7500	1.145364	1647.20	1.245	123.78 ± 0.02	−0.929 ± 0.001
0.9018	1.167054	1669.53	1.367	124.99 ± 0.02	−0.876 ± 0.001
$m_3 = 0.5\ \text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.025396	1524.67	0.774		
0.1500	1.052995	1550.91	0.858	120.39 ± 0.12	−1.130 ± 0.003
0.3000	1.079077	1575.90	0.937	122.05 ± 0.05	−1.051 ± 0.002
0.4500	1.103864	1599.98	1.028	123.34 ± 0.03	−0.986 ± 0.001
0.6000	1.127444	1624.98	1.130	124.47 ± 0.02	−0.941 ± 0.001
0.7500	1.149836	1649.51	1.240	125.58 ± 0.02	−0.897 ± 0.001
0.9000	1.171162	1672.32	1.361	126.59 ± 0.02	−0.851 ± 0.001
$m_3 = 0.6\ \text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.031201	1528.36	0.773		
0.1500	1.058498	1553.92	0.856	122.56 ± 0.11	−1.065 ± 0.003
0.2999	1.084279	1578.22	0.933	124.12 ± 0.05	−0.991 ± 0.002
0.4500	1.108823	1602.83	1.022	125.30 ± 0.03	−0.941 ± 0.001
0.5974	1.131625	1626.41	1.125	126.54 ± 0.02	−0.893 ± 0.001
0.7502	1.154181	1650.56	1.238	127.57 ± 0.02	−0.849 ± 0.001
0.9048	1.176093	1674.52	1.354	128.32 ± 0.01	−0.810 ± 0.001
$m_3 = 0.7\ \text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.036791	1530.91	0.772		
0.1500	1.063782	1555.97	0.835	124.70 ± 0.11	−1.011 ± 0.003
0.2999	1.089258	1580.99	0.908	126.20 ± 0.05	−0.959 ± 0.002
0.4501	1.113501	1605.12	1.004	127.37 ± 0.03	−0.905 ± 0.001
0.6001	1.136481	1628.78	1.101	128.45 ± 0.02	−0.857 ± 0.001
0.8333	1.170293	1665.03	1.289	129.61 ± 0.02	−0.796 ± 0.001
0.9052	1.180155	1676.01	1.339	130.02 ± 0.01	−0.778 ± 0.001
$T/\text{K} = 308.15$					
$m_3 = 0.2\ \text{mol}\cdot\text{kg}^{-1}$					
0.0000	1.006091	1525.97	0.672		
0.1500	1.034543	1552.29	0.782	113.75 ± 0.12	−1.246 ± 0.003

Table 1. Continued

m_1	$10^{-3} \cdot d$	u	η	$10^6 \cdot V_\phi$	$10^{13} \cdot K_\phi$
$\text{mol} \cdot \text{kg}^{-1}$	$\text{kg} \cdot \text{m}^{-3}$	$\text{m} \cdot \text{s}^{-1}$	$\text{mPa} \cdot \text{s}$	$\text{m}^3 \cdot \text{mol}^{-1}$	$\text{m}^3 \cdot \text{mol}^{-1} \cdot \text{Pa}^{-1}$
0.3002	1.061469	1577.40	0.853	115.74 ± 0.06	−1.158 ± 0.002
0.4498	1.086882	1601.82	0.935	117.47 ± 0.04	−1.087 ± 0.001
0.6000	1.111036	1625.95	1.024	119.13 ± 0.03	−1.025 ± 0.001
0.7499	1.134107	1649.31	1.132	120.38 ± 0.02	−0.970 ± 0.001
0.9000	1.156087	1672.50	1.234	121.58 ± 0.02	−0.922 ± 0.001
$m_3 = 0.3\ \text{mol} \cdot \text{kg}^{-1}$					
0.0000	1.011973	1528.82	0.670		
0.1500	1.040103	1555.02	0.778	116.23 ± 0.12	−1.196 ± 0.003
0.3000	1.066698	1579.96	0.852	118.07 ± 0.06	−1.112 ± 0.002
0.4500	1.091947	1604.15	0.930	119.59 ± 0.04	−1.043 ± 0.001
0.6000	1.116003	1627.03	1.018	120.85 ± 0.03	−0.979 ± 0.001
0.7500	1.138778	1650.01	1.119	122.18 ± 0.02	−0.926 ± 0.001
0.9001	1.160397	1673.88	1.225	123.46 ± 0.02	−0.885 ± 0.001
$m_3 = 0.4\ \text{mol} \cdot \text{kg}^{-1}$					
0.0000	1.017788	1531.37	0.669		
0.1500	1.045566	1556.96	0.775	118.84 ± 0.12	−1.130 ± 0.003
0.3000	1.071871	1582.08	0.850	120.42 ± 0.06	−1.065 ± 0.002
0.4502	1.096831	1605.93	0.924	121.90 ± 0.04	−0.997 ± 0.001
0.6002	1.120566	1630.50	1.016	123.12 ± 0.03	−0.950 ± 0.001
0.7500	1.143057	1652.85	1.115	124.32 ± 0.02	−0.895 ± 0.001
0.9018	1.164806	1674.59	1.210	125.37 ± 0.02	−0.845 ± 0.001
$m_3 = 0.5\ \text{mol} \cdot \text{kg}^{-1}$					
0.0000	1.023575	1534.27	0.678		
0.1500	1.051066	1559.74	0.770	120.98 ± 0.12	−1.086 ± 0.003
0.3000	1.077077	1584.00	0.844	122.53 ± 0.05	−1.011 ± 0.002
0.4500	1.101814	1607.88	0.923	123.75 ± 0.03	−0.954 ± 0.001
0.6000	1.125344	1631.38	1.010	124.85 ± 0.02	−0.904 ± 0.001
0.7500	1.147646	1654.51	1.108	125.99 ± 0.02	−0.858 ± 0.001
0.9000	1.168859	1677.91	1.206	127.04 ± 0.02	−0.820 ± 0.001
$m_3 = 0.6\ \text{mol} \cdot \text{kg}^{-1}$					
0.0000	1.029215	1536.77	0.666		
0.1500	1.056393	1562.00	0.766	123.21 ± 0.11	−1.039 ± 0.003
0.2999	1.082110	1586.22	0.837	124.61 ± 0.05	−0.971 ± 0.002
0.4500	1.106555	1610.25	0.920	125.81 ± 0.03	−0.919 ± 0.001
0.5974	1.129365	1633.10	1.004	126.90 ± 0.02	−0.870 ± 0.001
0.7502	1.151900	1657.53	1.102	127.89 ± 0.02	−0.831 ± 0.001
0.9048	1.173701	1679.64	1.198	128.70 ± 0.01	−0.786 ± 0.001
$m_3 = 0.7\ \text{mol} \cdot \text{kg}^{-1}$					
0.0000	1.034869	1539.38	0.665	125.55 ± 0.11	−0.978 ± 0.003
0.1500	1.061709	1563.96	0.754	126.79 ± 0.05	−0.927 ± 0.002
0.2999	1.087123	1588.27	0.805	127.98 ± 0.03	−0.878 ± 0.001
0.4501	1.111255	1612.19	0.900	128.93 ± 0.02	−0.832 ± 0.001
0.6001	1.134218	1635.29	0.990	130.05 ± 0.02	−0.772 ± 0.001
0.8333	1.167944	1670.64	1.130	130.36 ± 0.01	−0.755 ± 0.001
0.9052	1.177874	1681.39	1.183	125.55 ± 0.11	−0.978 ± 0.003
$T/\text{K} = 313.15$					
$m_3 = 0.2\ \text{mol} \cdot \text{kg}^{-1}$					
0.0000	1.004283	1534.64	0.595		
0.1500	1.032598	1560.35	0.710	114.51 ± 0.12	−1.205 ± 0.003
0.3002	1.059388	1585.02	0.774	116.51 ± 0.06	−1.123 ± 0.002
0.4498	1.084610	1608.79	0.848	118.37 ± 0.04	−1.051 ± 0.001
0.6000	1.108770	1632.35	0.930	119.78 ± 0.03	−0.993 ± 0.001
0.7499	1.131726	1654.98	1.029	121.04 ± 0.02	−0.939 ± 0.001
0.9000	1.153556	1677.95	1.119	122.28 ± 0.02	−0.894 ± 0.001
$m_3 = 0.3\ \text{mol} \cdot \text{kg}^{-1}$					
0.0000	1.009980	1537.54	0.593		
0.1500	1.037991	1562.68	0.719	116.87 ± 0.12	−1.143 ± 0.003
0.3000	1.064498	1587.06	0.785	118.62 ± 0.06	−1.070 ± 0.002
0.4500	1.089647	1611.05	0.846	120.15 ± 0.04	−1.010 ± 0.001
0.6000	1.113603	1633.53	0.925	121.41 ± 0.03	−0.949 ± 0.001
0.7500	1.136278	1656.41	1.015	122.75 ± 0.02	−0.900 ± 0.001
0.9001	1.157937	1678.88	1.113	123.89 ± 0.02	−0.857 ± 0.001
$m_3 = 0.4\ \text{mol} \cdot \text{kg}^{-1}$					
0.0000	1.015895	1539.73	0.585		
0.1500	1.043535	1564.97	0.710	119.61 ± 0.12	−1.102 ± 0.003
0.3000	1.069751	1589.19	0.777	121.05 ± 0.06	−1.030 ± 0.002
0.4502	1.094688	1612.41	0.840	122.34 ± 0.04	−0.965 ± 0.001
0.6002	1.118424	1635.66	0.922	123.44 ± 0.03	−0.915 ± 0.001
0.7500	1.140817	1658.25	1.010	124.69 ± 0.02	−0.868 ± 0.001
0.9018	1.162351	1679.28	1.109	125.90 ± 0.02	−0.817 ± 0.001
$m_3 = 0.5\ \text{mol} \cdot \text{kg}^{-1}$					
0.0000	1.021558	1542.48	0.577		
0.1500	1.048893	1567.29	0.698	121.86 ± 0.11	−1.047 ± 0.003
0.3000	1.074817	1591.00	0.765	123.20 ± 0.05	−0.978 ± 0.002
0.4500	1.099494	1614.38	0.830	124.30 ± 0.03	−0.924 ± 0.001
0.6000	1.122894	1637.38	0.917	125.46 ± 0.02	−0.876 ± 0.001
0.7500	1.145236	1660.51	1.006	126.42 ± 0.02	−0.836 ± 0.001
0.9000	1.166368	1682.16	1.107	127.49 ± 0.02	−0.793 ± 0.001
$m_3 = 0.6\ \text{mol} \cdot \text{kg}^{-1}$					
0.0000	1.027232	1544.85	0.569		
0.1500	1.054265	1569.17	0.689	124.03 ± 0.11	−0.994 ± 0.003
0.2999	1.079893	1593.05	0.759	125.26 ± 0.05	−0.939 ± 0.002
0.4500	1.104278	1616.50	0.826	126.36 ± 0.03	−0.889 ± 0.001
0.5974	1.127052	1640.00	0.915	127.36 ± 0.02	−0.851 ± 0.001
0.7502	1.149572	1663.82	0.991	128.27 ± 0.02	−0.812 ± 0.001
0.9048	1.171247	1686.10	1.106	129.14 ± 0.01	−0.769 ± 0.001
$m_3 = 0.7\ \text{mol} \cdot \text{kg}^{-1}$					
0.0000	1.032757	1547.68	0.563		
0.1500	1.059488	1571.46	0.690	126.15 ± 0.11	−0.940 ± 0.003
0.2999	1.084815	1594.95	0.753	127.33 ± 0.05	−0.890 ± 0.002
0.4501	1.108933	1618.30	0.815	128.35 ± 0.03	−0.846 ± 0.001
0.6001	1.131858	1641.06	0.900	129.26 ± 0.02	−0.804 ± 0.001
0.8333	1.165439	1675.64	1.050	130.45 ± 0.02	−0.747 ± 0.001
0.9052	1.175331	1685.71	1.105	130.77 ± 0.01	−0.729 ± 0.001

The apparatus was calibrated at each temperature with double-distilled−deionized water and dry air. For pure water, the values (998.203, 997.043, 995.647, 994.028, and 992.216) $\text{kg} \cdot \text{m}^{-3}$ for density and (1482.76, 1496.96, 1508.85, 1520.15, and 1529.56) $\text{m} \cdot \text{s}^{-1}$ for ultrasonic velocity were obtained, respectively, at T = (293.15, 298.15, 303.15, 308.15, and 313.15) K.

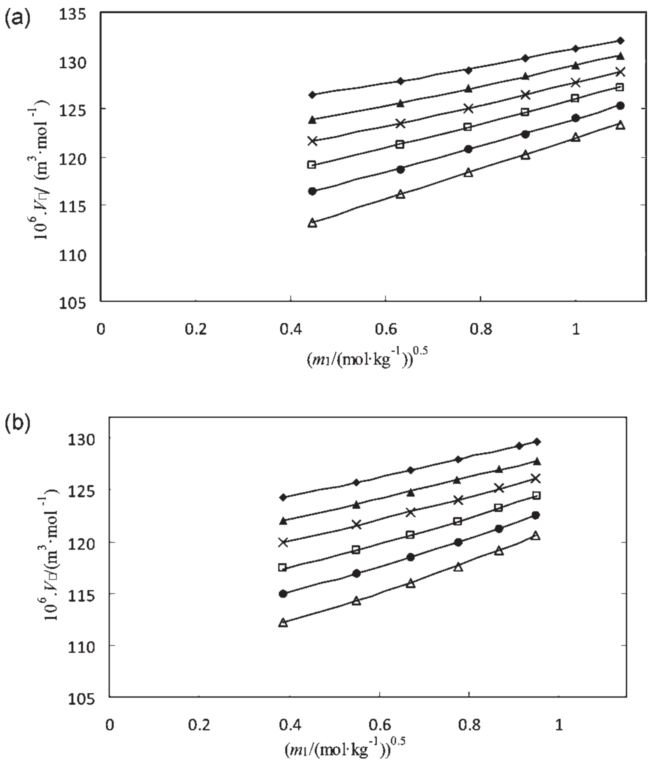

Figure 1. Plot of the apparent molar volume, V_ϕ, a: $K_3Cit + KBr + H_2O$; b: $K_3Cit + KNO_3 + H_2O$, against the square root of K_3Cit molality, $m_1^{0.5}$, at $T = 298.15$ K in aqueous solutions of KNO_3 or KBr at different molalities: △, 0.2; ●, 0.3; □, 0.4; ×, 0.5; ▲, 0.6; ♦, 0.7; solid line, eq 5.

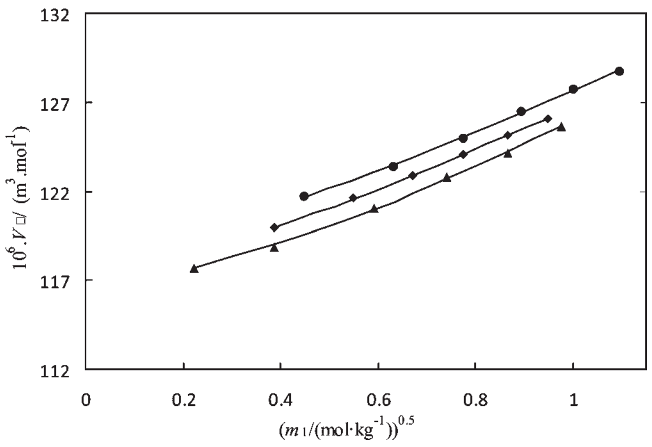

Figure 2. Plot of apparent molar molar volume of K_3Cit, V_ϕ, against the square root of K_3Cit molality, $m_1^{0.5}$, at $T = 298.15$ K in aqueous solutions of, ●, KBr ($m_2 = 0.5$ mol·kg^{-1}); ♦, KNO_3 ($m_3 = 0.5$ mol·kg^{-1}); ▲, KCl (0.55 mol·kg^{-1}, taken from ref 9); solid line, eq 5.

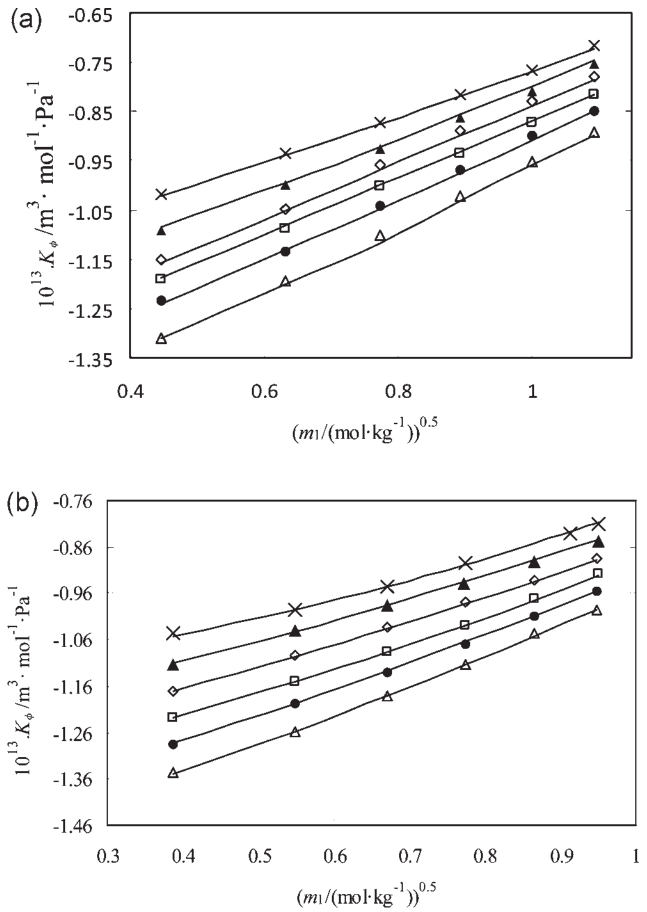

Figure 3. Plot of apparent molar isentropic compressibility of K_3Cit, K_ϕ, a: $K_3Cit + KBr + H_2O$; b: $K_3Cit + KNO_3 + H_2O$, against square root of K_3Cit molality, $m_1^{0.5}$, at $T = 298.15$ K: in aqueous solutions of KNO_3 at different molalities: △, 0.2; ●,0.3; □, 0.4; ◇, 0.5; ▲, 0.6; ×, 0.7; solid line, eq 10.

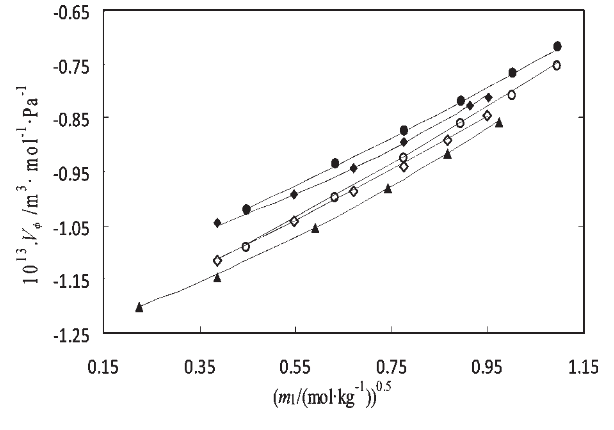

Figure 4. Plot of apparent molar isentropic compressibility of K_3Cit, V_ϕ, against square root of K_3Cit molality, $m_1^{0.5}$, at $T = 298.15$ K in aqueous solutions of, ●, KBr ($m_2 = 0.7$ mol·kg^{-1}); ♦, KNO_3 ($m_3 = 0.7$ mol·kg^{-1}); ○, KBr ($m_2 = 0.5$ mol·kg^{-1}); ◇, KNO_3 ($m_3 = 0.5$ mol·kg^{-1}); ▲, KCl (0.55 mol·kg^{-1}, taken from ref 9); solid line, eq 10.

The apparatus was also tested with the density of a known molality of aqueous NaCl using the data given by Pitzer et al.[14] The uncertainty in the measurement of density and speed of sound was estimated to be 0.003 kg·m^{-3} for density and 0.1 m·s^{-1} for ultrasonic velocity. The viscosities of solutions were determined with a suspended Ubbelohde-type viscometer at $T = (293.15$ to $313.15)$ K in which the temperature was controlled with a precision of 0.01 K with a thermostat (Julabo, MD-18V, Germany). The flow times were measured using a stopwatch. The precision of the stopwatch used was ± 0.01 s. The dynamic viscosity, η, was calculated by the following relation:

$$\eta = dK(t - \theta) \quad (1)$$

where t is the flow time; K is the viscometer constant; θ is the Hagenbach correction factor; and d is the density. The viscometer constant, K, is determined by calibrating at the working temperatures with distilled water using viscosity values from the literature.[15] The uncertainty for the dynamic viscosity determination was estimated to be ± 0.5 %. Each measurement was repeated five times.

3. RESULTS AND DISCUSSION

Density, speed of sound, and viscosity measurements for K_3Cit in aqueous solutions of (KBr or KNO_3) in the different molality ranges of KBr and KNO_3 [(0.2, 0.3, 0.4, 0.5, 0.6, and 0.7) $mol \cdot kg^{-1}$] at different K_3Cit concentrations were made at T = (293.15, 298.15, 303.15, 308.15, and 313.15) K. The values of density, d, speed of sound, u, and viscosity, η, for the ternary aqueous solution of K_3Cit + (KBr or KNO_3) are reported in Table 1. The apparent molar volumes and apparent molar isentropic compressibility of the K_3Cit in aqueous (KBr or KNO_3) solutions were computed from the measured density and speed of sound values using eqs 2 and 3, respectively:[16]

$$V_\phi = \frac{M_1}{d} - \frac{(d - d_0)}{m_1 d d_0} \quad (2)$$

$$K_\phi = \frac{(k_s d_0 - k_{s0} d)}{m_1 d d_0} + \frac{M_1 k_s}{d} \quad (3)$$

where M_1 is the molar mass of K_3Cit.; m_1 is the molality of the K_3Cit; d and d_0 are the densities of solution and solvent, respectively; k_s and k_{s0} are the isentropic compressibility of the solution and solvent, respectively. The (KBr or KNO_3) + water is considered as the solvent. Using the sound velocity and density values, the isentropic compressibility, k_s (kPa^{-1}), values were calculated for the investigated mixtures from the Laplace–Newton equation:

$$k_s = d^{-1}u^{-2} \quad (4)$$

The results obtained have also been reported in Table 1. In Figure 1, the concentration dependence of V_ϕ for K_3Cit in different molalities of aqueous solutions of KBr and KNO_3, respectively, has been shown at 298.15 K as an example. Similar behavior has been observed for other temperatures. From Table 1 and Figure 1, it can be seen that the apparent molar volumes of K_3Cit in aqueous KBr and KNO_3 solutions increase with an increase in the K_3Cit molality. In the electrolyte solutions, the solute–solute interactions are characterized by positive slopes of V_ϕ versus concentration plots.[17] This is attributed to the phenomenon, described in terms of destructive overlap of cospheres,[18,19] resulting in a net decrease of solvation, thereby increasing the solute volume. In Figure 2 the apparent molar volumes of K_3Cit + KBr + H_2O and K_3Cit + KNO_3 + H_2O have been plotted at the KBr and KNO_3 molality of 0.5 at T = 298.15 K. From Figure 2, it can be seen that there is a positive transfer of volume of K_3Cit from aqueous KNO_3 solution to an aqueous KBr solution. This indicates that the potassium citrate ions in aqueous KBr solutions are larger than in aqueous KNO_3 solutions. The strong interactions between KBr and the water molecule induce the dehydration of ions of K_3Cit more than

Table 2. Values of $V_\phi^0/(cm^3 \cdot mol^{-1})$ of K_3Cit at Different Temperatures and Different Concentrations of KBr or KNO_3

	T/K				
system	293.15	298.15	303.15	308.15	313.15
	V_ϕ^0				
K_3Cit + KBr (0.2 m) + H_2O	107.45	107.44	108.58	109.05	110.62
K_3Cit + KBr (0.3 m) + H_2O	111.53	110.99	111.45	113.23	113.79
K_3Cit + KBr (0.4 m) + H_2O	114.27	114.34	114.80	115.81	116.26
K_3Cit + KBr (0.5 m) + H_2O	118.11	117.81	118.98	119.36	120.6
K_3Cit + KBr (0.6 m) + H_2O	120.10	120.43	121.05	122.11	122.78
K_3Cit + KBr (0.7 m) + H_2O	122.77	123.10	122.92	124.16	124.74
K_3Cit + KNO_3 (0.2 m) + H_2O	107.63	107.98	107.71	109.14	109.67
K_3Cit + KNO_3 (0.3 m) + H_2O	111.55	111.85	112.17	112.82	113.43
K_3Cit + KNO_3 (0.4 m) + H_2O	113.84	113.95	115.19	115.53	117.57
K_3Cit + KNO_3 (0.5 m) + H_2O	115.37	116.39	117.01	118.22	119.70
K_3Cit + KNO_3 (0.6 m) + H_2O	117.75	118.71	118.91	120.04	121.57
K_3Cit + KNO_3 (0.7 m) + H_2O	119.90	120.92	120.85	122.18	123.50

Table 3. Fitting Parameters of eq 6 and Term $(\partial^2 V_\phi^0/\partial T^2)_P$ along with the Absolute Average Relative Deviation (AARD)

system	A_0	A_1	$10^3 \cdot A_2$	$100 \cdot AARD^a$	$10^3(\partial^2 V_\phi^0/\partial T^2)_P$ $cm^6 \cdot mol^{-2} \cdot K^{-2}$
K_3Cit + KBr (0.2 m) + H_2O	107.39 ± 0.09	0.02 ± 0.02	7.11 ± 1.00	0.46	9.26
K_3Cit + KBr (0.3 m) + H_2O	110.46 ± 0.12	0.09 ± 0.04	4.34 ± 1.75	0.27	8.68
K_3Cit + KBr (0.4 m) + H_2O	114.19 ± 0.13	0.03 ± 0.03	3.74 ± 1.58	0.16	4.62
K_3Cit + KBr (0.5 m) + H_2O	117.99 ± 0.19	0.0003 ± 0.04	6.54 ± 1.51	0.23	3.32
K_3Cit + KBr (0.6 m) + H_2O	120.05 ± 0.09	0.08 ± 0.02	3.20 ± 1.13	0.12	6.40
K_3Cit + KBr (0.7 m) + H_2O	122.81 ± 0.07	−0.01 ± 0.02	5.49 ± 1.25	0.22	10.98
K_3Cit + KNO_3 (0.2 m) + H_2O	107.67 ± 0.07	−0.0.01 ± 0.03	5.89 ± 1.47	0.27	1.94
K_3Cit + KNO_3 (0.3 m) + H_2O	111.55 ± 0.01	0.04 ± 0.003	2.71 ± 0.23	0.04	3.02
K_3Cit + KNO_3 (0.4 m) + H_2O	113.83 ± 0.04	0.01 ± 0.03	8.46 ± 1.28	0.26	17.20
K_3Cit + KNO_3 (0.5 m) + H_2O	115.46 ± 0.14	0.12 ± 0.03	4.31 ± 0.57	0.12	8.62
K_3Cit + KNO_3 (0.6 m) + H_2O	117.90 ± 0.24	0.06 ± 0.05	5.91 ± 1.44	0.20	11.82
K_3Cit + KNO_3 (0.7 m) + H_2O	120.06 ± 0.27	0.05 ± 0.05	5.71 ± 1.47	0.27	1.70

[a] AARD = $1/N\Sigma_i^N|(V_{\phi,\exp_i}{}^0 - V_{\phi,\mathrm{cal}_i}{}^0)/V_{\phi,\exp_i}{}^0|$.

Table 4. Values of $K_\phi^0/(\mathrm{m^3 \cdot mol^{-1} \cdot Pa^{-1}})$ of K_3Cit at Different Temperatures and Different Concentrations of KBr and KNO_3

	T/K				
system	293.15	298.15	303.15	308.15	313.15
		$10^{13}\ K_\phi^0$			
K_3Cit + KBr (0.3 m) + H_2O	−1.532 ± 0.026	−1.486 ± 0.023	−1.437 ± 0.022	−1.426 ± 0.024	−1.405 ± 0.017
K_3Cit + KBr (0.4 m) + H_2O	−1.450 ± 0.012	−1.443 ± 0.006	−1.386 ± 0.003	−1.336 ± 0.014	−1.279 ± 0.002
K_3Cit + KBr (0.5 m) + H_2O	−1.450 ± 0.012	−1.428 ± 0.015	−1.344 ± 0.007	−1.292 ± 0.007	−1.234 ± 0.005
K_3Cit + KBr (0.6 m) + H_2O	−1.404 ± 0.016	−1.295 ± 0.002	−1.240 ± 0.001	−1.204 ± 0.008	−1.144 ± 0.010
K_3Cit + KBr (0.7 m) + H_2O	−1.171 ± 0.018	−1.200 ± 0.007	−1.080 ± 0.003	−1.081 ± 0.002	−1.045 ± 0.002
K_3Cit + KBr (0.2 m) + H_2O	−1.651 ± 0.014	−1.578 ± 0.007	−1.505 ± 0.017	−1.470 ± 0.006	−1.388 ± 0.009
K_3Cit + KNO_3 (0.2 m) + H_2O	−1.634 ± 0.018	−1.549 ± 0.010	−1.496 ± 0.022	−1.450 ± 0.004	−1.402 ± 0.009
K_3Cit + KNO_3 (0.3 m) + H_2O	−1.499 ± 0.009	−1.459 ± 0.009	−1.454 ± 0.007	−1.403 ± 0.012	−1.338 ± 0.022
K_3Cit + KNO_3 (0.4 m) + H_2O	−1.424 ± 0.003	−1.372 ± 0.008	−1.305 ± 0.006	−1.237 ± 0.007	−1.238 ± 0.008
K_3Cit + KNO_3 (0.5 m) + H_2O	−1.356 ± 0.003	−1.328 ± 0.003	−1.321 ± 0.006	−1.261 ± 0.002	−1.208 ± 0.004
K_3Cit + KNO_3 (0.6 m) + H_2O	−1.318 ± 0.006	−1.262 ± 0.008	−1.228 ± 0.007	−1.184 ± 0.004	−1.110 ± 0.006
K_3Cit + KNO_3 (0.7 m) + H_2O	−1.216 ± 0.013	−1.137 ± 0.009	−1.122 ± 0.014	−1.090 ± 0.009	−1.040 ± 0.002

Table 5. Viscosity *B*-Coefficients of K_3Cit in Aqueous (KBr or KNO_3) at Different Temperatures

	T/K				
system	293.15	298.15	303.15	308.15	313.15
K_3Cit + KBr (0.2 m) + H_2O	1.4683	1.4623	1.4331	1.4526	1.4084
K_3Cit + KBr (0.3 m) + H_2O	1.4700	1.4676	1.4346	1.4371	1.3907
K_3Cit + KBr (0.4 m) + H_2O	1.4799	1.4745	1.4402	1.4214	1.3844
K_3Cit + KBr (0.5 m) + H_2O	1.4712	1.4692	1.4425	1.4121	1.3813
K_3Cit + KBr (0.6 m) + H_2O	1.4817	1.4636	1.4485	1.4056	1.3903
K_3Cit + KBr (0.7 m) + H_2O	1.4775	1.4325	1.4196	1.3923	1.3890
K_3Cit + KNO_3 (0.2 m) + H_2O	1.2221	1.2101	1.1724	1.1067	1.0754
K_3Cit + KNO_3 (0.3 m) + H_2O	1.2373	1.2061	1.1818	1.1610	0.9578
K_3Cit + KNO_3 (0.4 m) + H_2O	1.2369	1.2197	1.1859	1.0556	0.992
K_3Cit + KNO_3 (0.5 m) + H_2O	1.2362	1.2344	1.1931	1.0960	1.0752
K_3Cit + KNO_3 (0.6 m) + H_2O	1.2924	1.2503	1.1909	1.0731	1.0654
K_3Cit + KNO_3 (0.7 m) + H_2O	1.3271	1.3262	1.3118	1.1719	1.1496

KNO_3, and therefore at high KBr concentrations, the water molecules around the K_3Cit become less, thereby increasing the solute volume. The apparent molar volumes of K_3Cit + KCl + H_2O taken from previous works[9] have also been plotted at a KCl molality of 0.55 at T = 298.15 K. It is obvious that V_ϕ of K_3Cit in aqueous KCl solutions are smaller than V_ϕ of K_3Cit in aqueous KBr and KNO_3 solutions considering that the V_ϕ values of K_3Cit in KCl with a molality of 0.55 $\mathrm{mol \cdot kg^{-1}}$ are smaller than those corresponding to a KCl molality of 0.55 $\mathrm{mol \cdot kg^{-1}}$.

The values of K_ϕ are plotted as a function of K_3Cit concentration at various KBr and KNO_3 concentrations at 298.15 K in Figure 3. Similar trends have been observed for other temperatures. From Table 1 and Figure 3, it is obvious that the value of apparent molar isentropic compressibility of K_3Cit in aqueous (KBr or KNO_3) solutions is negative, and it increases with increasing concentration of K_3Cit and temperature. Figure 3 also shows that at each temperature and at a constant salt concentration the value of K_ϕ of K_3Cit increases as the concentration of (KBr or KNO_3) increases. The strong interactions between (KBr or KNO_3) and water molecules induce the dehydration of ions, and therefore at high (KBr or KNO_3) concentrations, the water molecules around the K_3Cit become more compressible than those at lower KBr or KNO_3 concentrations. In Figure 4 the apparent molar volumes of K_3Cit + KBr + H_2O and K_3Cit + KNO_3 + H_2O have been plotted at the KBr and KNO_3 molalities of (0.5 and 0.7) $\mathrm{mol \cdot kg^{-1}}$, respectively, at T = 298.15 K. From Figure 4 it can be seen that the water molecules around the K_3Cit in aqueous KBr solution are more compressible than those in aqueous KNO_3 solution. This indicates that interactions between KBr and water molecules are stronger than the interaction of KNO_3 with water. When the concentration of KBr became larger, it is clear that the difference between compressibility of water molecules around K_3Cit in aqueous KBr and KNO_3 solutions became more obvious. The apparent molar isentropic compressibility of K_3Cit + KCl + H_2O taken from previous work[9] has also been plotted at a KCl molality of 0.55 at T = 298.15 K. It is obvious that water molecules around the K_3Cit in aqueous KCl solution are of lower compressibility than those for K_3Cit in aqueous KBr and KNO_3 solutions even at a KCl molality of 0.55.

The Redlich−Mayer-type equation in the form[20,21]

$$V_\phi = V_\phi^0 + S_V {m_1}^{0.5} + B_V m_1 \tag{5}$$

was used to obtain V_ϕ^0, the limiting apparent molar volume of K_3Cit at different temperatures and different (KBr or KNO_3) concentrations. In this equation, S_V and B_V are the empirical parameters which depend on solute, solvent, and temperature. The limiting apparent molar volume (V_ϕ^0), S_V, and B_V values have been obtained from the correlation of V_ϕ values with the Redlich−Mayer equation. The V_ϕ^0 values have been given in Table 2. The S_V and S_V values for KBr and KNO_3 along with the absolute average relative deviation (AARD) were reported in Tables S1 and S2 as Supporting Information. The value of the apparent molar volume is an important property. At infinite dilution, each ion is surrounded only by the solvent molecules and is infinitely distant from other ions. It follows that V_ϕ^0 is unaffected by ion−ion interaction, and it is a measure only of the ion−solvent interaction.[22,23]

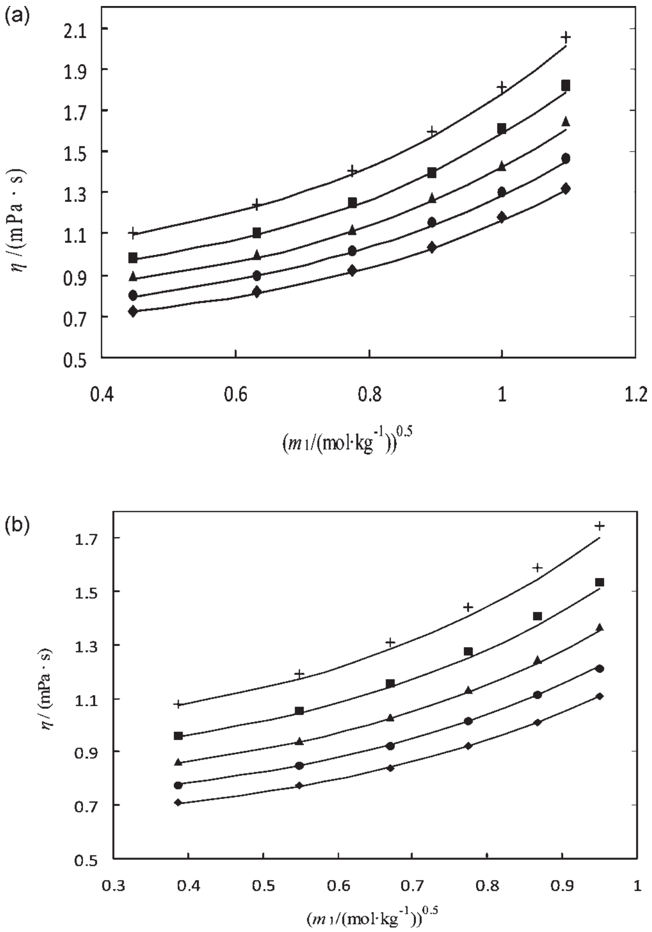

Figure 5. Plot of viscosity of K_3Cit, η, a: K_3Cit + KBr + H_2O; b: K_3Cit + KNO_3 + H_2O, against the square root of K_3Cit molality, $m_1{}^{0.5}$, in aqueous solution of KBr or KNO_3 (m_2, m_3 = 0.4 mol·kg^{-1}) at different temperatures: +, 293.15 K; ■, 298.15 K; ▲, 303.15 K; ●, 308.15; ◆, 313.15 K; solid line, eq 21.

The temperature dependency of V_ϕ^0 calculated from eq 5 was expressed in a polynomial form as follows

$$V_\phi^0 = A_0 + A_1(T - 293.15) + A_2(T - 293.15)^2 \qquad (6)$$

The parameters A_0, A_1, and A_2 of eq 6 were calculated from the correlation of V_ϕ^0 values at different temperatures and different (KBr or KNO_3) concentrations. The obtained parameters along with the standard deviation (σ) of V_ϕ^0 values have been given in Table 3. In fact, the apparent molar volume is equal to the infinite dilution partial molar volume; therefore, due to the additivity principle at infinite dilution, we have

$$V_\phi^0(K_3\text{Cit}) = 3V_\phi^0(K^+) + V_\phi^0(\text{Cit}^{3-}) \qquad (7)$$

The apparent molar volume of ions can be expressed as the sum of two contributions,[23]

$$V_\phi^0(\text{ion}) = V_\phi^0(\text{int}) + V_\phi^0(\text{elect}) \qquad (8)$$

where V_ϕ^0 (int) is the intrinsic (related to the size of the ions and to packing effects) apparent molar volume and V_ϕ^0 (elect) is the electrostriction apparent molar volume related to ion–solvent interactions (i.e., the decrease in volume due to hydration). The term $\partial V_\phi^0(\text{int})/\partial T$ contributes negligibly to the overall temperature dependence of V_ϕ^0 (ion), and therefore an increase in the V_ϕ^0 by increasing temperature occurs with a loss of hydration.

For determining the structure-making and structure-breaking capacities of the solute in different mixed solvents, the following equation of Hepler [24] was used

$$(\partial C_P/\partial T)_P = -(\partial^2 V_\phi^0/\partial T^2)_P \qquad (9)$$

The values in eq 9 are also listed in Table 3. It can be seen that $(\partial^2 V_\phi^0(\text{int})/\partial T^2)$ all are positive over the temperature range under investigation, indicating an obvious structure-making tendency of K_3Cit in aqueous (KBr or KNO_3) solutions.

The limiting apparent molar isentropic compressibility (K_ϕ^0) was obtained from the fitting of K_ϕ with an equation as[25]

$$K_\phi = K_\phi^0 + S_K m_1{}^{0.5} + B_K m_1 \qquad (10)$$

where S_K and B_K are empirical parameters. Values of K_ϕ^0 at different temperatures and (KBr or KNO_3) concentrations calculated from eq 10 are given in Table 4. The values of S_K and B_K along with the absolute average relative deviation (AARD) were reported in Tables S3 and S4 as Supporting Information. As can be seen from Table 4, K_ϕ^0 values are negative and increase with increasing temperature and (KBr or KNO_3) concentration. The negative values of K_ϕ and K_ϕ^0 of K_3Cit in aqueous (KBr or KNO_3) solutions indicate that the water molecules around the K_3Cit ions are less compressible than the water molecules in the bulk solution. By differentiating eq 8 with respect to pressure, we obtain

$$K_\phi^0(\text{ion}) = K_\phi^0(\text{int}) + K_\phi^0(\text{elect}) \qquad (11)$$

where $K_\phi^0(\text{int}) = -(\partial V_\phi^0(\text{int})/\partial P)_T$ is the intrinsic apparent molar compressibility and $K_\phi^0(\text{elect}) = -(\partial V_\phi^0(\text{elect})/\partial P)_T$ is the electrostriction apparent molar compressibility. Because the effect of pressure on the volume of crystals is small, one would expect $K_\phi^0(\text{int})$ to be positive and close to zero.[26] Thus $K_\phi^0(\text{ion})$ is due mainly to $K_\phi^0(\text{elect})$. In fact, the negative values of K_ϕ and K_ϕ^0 of K_3Cit in aqueous (KBr or KNO_3) solutions are attributed to the strong attractive interactions due to the hydration of ions at low temperatures. By increasing temperature, ion–solvent interactions are weakened, and therefore at high temperatures, the water molecules around the K_3Cit become more compressible than those at lower temperatures.

Viscosity values (η) of ternary aqueous solutions of K_3Cit + (KBr or KNO_3) have also been measured in this work. The results obtained are given in Table 1. The relative viscosities, η_r ($\eta_r = \eta/\eta_0$, where η and η_0 are the viscosities of solution and solvent, respectively), have been used to calculate the viscosity B-coefficients of K_3Cit using the Jones–Dole equation[12] where c is molarity:

$$\eta_r = 1 + Ac^{0.5} + Bc \qquad (12)$$

The values of the B coefficients have been reported in Table 5. In general, the viscosity B-coefficients reflect solute–solvent interactions.[27] It is well-established that the viscosity B-coefficient is a measure of solute–solvent interactions and is directly dependent on the size, shape, and charge of the solute molecules. The values of the viscosity B-coefficients for K_3Cit in water and in aqueous (KBr or KNO_3) solutions at the five different temperatures are positive indicating that the ion–solvent interactions are strong.[28] The dB/dT values, which give important information regarding the structure-making agent and structure-breaking role of the solute in solvent media, are a better criterion[29] than the B-coefficient. The negative dB/dT values of K_3Cit in water and aqueous (KBr or KNO_3) solutions reported in Table 5 show that K_3Cit acts as a structure-making agent.

3.1. Prediction of Density. To predict the density of ternary aqueous K_3Cit + (KBr or KNO_3) solutions the method proposed by Laliberté and Cooper [10] has been used. First the apparent specific volumes (V_{app}) of binary aqueous solutions have been calculated with the following equation,

$$V_{app} = 1 - \frac{\left(\frac{d(1-w_2)}{d_0}\right)}{dw_2} \tag{13}$$

where w_2 refers to the mass fraction of K_3Cit and d is the density of binary aqueous solution of K_3Cit, KBr, or KNO_3. Density values for binary aqueous solution of K_3Cit have been taken from ref 7. The calculated V_{app} values were correlated with the following equation:[11]

$$V_{app} = \frac{(1 - w_{H_2O}) + C_2 + C_3 t}{(C_0(1 - w_{H_2O}) + C_1)\exp(0.000001(t + C_4)^2)} \tag{14}$$

where C_0 to C_4 are adjustable parameters and t is the temperature in Celsius.

The parameters of this equation for the $K_3Cit + H_2O$ and $KNO_3 + H_2O$ systems were taken from our previous work[9] and the Supporting Information of ref 11, respectively. The obtained parameters of eq 14 (C_0 to C_4) for the binary aqueous solution of KBr are 0.012, $1.501 \cdot 10^4$, 4.379, $1.139 \cdot 10^{-3}$, and $4 \cdot 10^{-3}$, respectively, with an absolute average relative deviation (AARD) value of 0.085. Density values of K_3Cit + (KBr or KNO_3) solutions can be predicted with the equation:[11]

$$\ln \eta = w_{H_2O} \ln \eta_{H_2O} + w_{K_3Cit} \ln \eta_{K_3Cit/H_2O} + w_{KCl} \ln \eta_{KCl/H_2O} \tag{15}$$

where

$$d_{H_2O} = \frac{(((((-2.8054253 \times 10^{-10} t + 1.0556302 \cdot 10^{-7})t - 4.6170461 \cdot 10^{-5})t - 0.0079870401)t + 16.945176)t + 999.83952)}{1 + 0.01687985 t} \tag{16}$$

$V_{app,K_3Cit/H_2O}$ and $V_{app,(KBr\ or\ KNO_3)/H_2O}$ have been calculated at the corresponding mass fraction of the ternary solutions from the parameters of eq 14 obtained in this work for KBr and those reported previously for aqueous K_3Cit[9] and KNO_3.[10] The absolute average relative deviation obtained between predicted and measured density values (100 AARD = 0.04) for ternary aqueous solutions of K_3Cit + KBr and (100 AARD = 0.03) for ternary aqueous solutions of $K_3Cit + KNO_3$ at working temperatures indicating that the prediction method proposed by Laliberté and Cooper[11] is satisfactory for prediction of density of the investigated ternary systems, at different temperatures.

3.2. Prediction of Viscosity. To predict the viscosity of ternary aqueous K_3Cit + (KBr or KNO_3) solutions, first the solute viscosities, η_e, have been calculated from the data reported in refs 9 and 13 and Supporting Information of ref 10 with the following equation:

$$\eta_e = \left(\frac{\eta}{\eta_{H_2O}^{w_{H_2O}}}\right)^{1/w_2} \tag{17}$$

where

$$\eta_{H_2O} = \frac{t + 246}{(0.05594t + 5.2842)t + 137.37} \tag{18}$$

The η_e values obtained for the binary aqueous solutions of K_3Cit, KBr, and KNO_3 were correlated using empirical eq 19 proposed by Laliberté[10] and semiempirical eq 20 used by Zafarani-Moattar and Majdan-Cegincara:[13]

$$\eta_e = \frac{\exp((C_0(1 - w_{H_2O})^{C_1} + C_2)/(C_3 t + 1))}{C_4(1 - w_{H_2O})^{C_5} + 1} \tag{19}$$

$$\eta_e = C_0 \exp(C_1/t - C_2)\exp(C_3 m + C_4 m^2) \tag{20}$$

The parameters of eqs 19 and 20 for binary aqueous solutions of K_3Cit have been taken from ref 9. The parameters of eq 19 and 20 for binary aqueous solutions of KBr and KNO_3 were taken from refs 10 and 13, respectively. The viscosity values of ternary aqueous K_3Cit + (KBr or KNO_3) solutions were predicted by the Arrhenius type mixing rule as follows:

$$d_{K_3Cit + (KBr\ or\ KNO_3)/H_2O} = \frac{1}{\frac{w_{H_2O}}{d_{H_2O}} + w_{K_3Cit} V_{app,K_3Cit/H_2O} + w_{KCl} V_{app,(KBr\ or\ KNO_3)/H_2O}} \tag{21}$$

The estimated 100 AARD values between the measured viscosity values and predicted values by eq 21 for ternary aqueous K_3Cit + KBr solutions are 2.09 and 0.70 using the eqs of 19 and 20, respectively. Also, for ternary aqueous $K_3Cit + KNO_3$ solutions 100 AARD values of 2.24 and 1.14 are obtained by the equations proposed by Laliberté (eq 19) and Zafarani-Moattar and Majdan-Cegincara (eq 20), respectively. From the results obtained, it is obvious that both of these methods have good performance in the prediction of viscosity values of K_3Cit + (KBr or KNO_3) + H_2O, especially using the procedure given in our previous work.[13] To see the performance of eq 21 in the prediction of viscosity values of aqueous ternary system of K_3Cit + KCl in a better manner, experimental and predicted viscosity values have been plotted against the square root of K_3Cit molality at the KBr or KNO_3 concentration of 0.4 $mol \cdot kg^{-1}$ at different temperatures as in Figure 5. As can be seen from the figure it is also obvious that performance of eq 21 is good in the prediction of viscosity values of the investigated systems.

4. CONCLUSIONS

Density, speed of sound, and viscosity for ternary aqueous solutions of K_3Cit + (KBr or KNO_3) have been measured over the entire concentration range at T = (293.15, 298.15, 303.15, 308.15, and 313.15) K. The values of apparent molar volumes and apparent molar isentropic compressibilities of solutions were calculated from the measured data. The results show a positive transfer of volume of K_3Cit from an aqueous solution to an aqueous (KBr or KNO_3) solution. Also, the results show that the apparent molar volumes of tripotassiumm citrate increase as the concentration of potassium citrate increases. The apparent molar isentropic compressibility of K_3Cit in ternary solutions has negative values at each temperature. The negative values of apparent molar isentropic compressibility of K_3Cit imply that the water molecules around the K_3Cit ions are less compressible than the water molecules in the bulk solutions. The viscosity *B*-coefficient values of K_3Cit in the ternary aqueous solutions of K_3Cit + (KBr or KNO_3) show that K_3Cit acts as a structure-making agent. The viscosity and density values of ternary aqueous K_3Cit + (KBr or KNO_3) systems have been predicted using parameters obtained from the binary aqueous solutions. The results obtained show that the performance of prediction methods proposed by Laliberté and Cooper for density and Laliberté and Zafarani-Moattar and Majdan-Cegincara for viscosity is excellent for the ternary aqueous system containing an organic and an inorganic salt.

ASSOCIATED CONTENT

Ⓢ **Supporting Information.** Parameters of eqs 5 and 10 (four tables). This material is available free of charge via the Internet at http://pubs.acs.org.

AUTHOR INFORMATION

Corresponding Author

*Fax: +98 411 3340191. E-mail: zafarani47@yahoo.com.

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