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Measurement and Correlation of High Pressure Densities of Ionic Liquids, 1-Ethyl-3-methylimidazolium L-Lactate ([emim][Lactate]), 2-Hydroxyethyl-trimethylammonium L-Lactate ([$(C_2H_4OH)(CH_3)_3N$][Lactate]), and 1-Butyl-3-methylimidazolium Chloride ([bmim][Cl])

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S *Supporting Information*

ABSTRACT: Pressure−density−temperature ($p\rho T$) data for three types of ionic liquid, [emim][lactate], [$(C_2H_4OH)(CH_3)_3N$][lactate], and [bmim][Cl], were measured with a bellows type dilatometer at temperatures up to 452 K and at pressures up to 200 MPa. The Tait equation could correlate the density data as a function of temperature and pressure to within an average relative deviation (ARD) of 0.014 %. The thermal expansion coefficients and the isothermal compressibilities were estimated with the Tait equation, and their variations with temperature and pressure, $d\alpha_p/dT$ and dk_T/dp, were found to be negative. Lattice type equations of state were applied to correlate the data. The densities could be correlated to within an ARD of 0.21 % and 0.067 %, for the Sanchez−Lacombe equation of state and the ε-modified Sanchez−Lacombe equation of state, respectively. The equation-of-state parameters determined should be useful in estimating the properties of mixtures and in estimating the phase equilibria of binary and multicomponent systems.

INTRODUCTION

Ionic liquids have attracted much attention as green solvents, since they have unique physical properties such as low vapor pressures, low melting points, and relatively good chemical stability. Ionic liquids are being proposed for use as solvents in processes for gas separation,[1−5] metal extraction,[6,7] chemical synthesis,[8] battery electrolyte, and solar energy systems. In these applications, many types of physical properties such as density, viscosity, thermal conductivity, and phase equilibria are required, and the ILThermo database has been very instrumental in promoting scientific development.[9,10]

Pressure−density−temperature ($p\rho T$) data are fundamental for estimating fluid thermal expansion and compressibility required for almost any practical application. The availability of $p\rho T$ data provides a basis for other theories such as the determination of equation-of-state parameters. From the equation of state, phase equilibria can be calculated for many systems over wide ranges of pressure and temperature. Although many $p\rho T$ data have been reported for ionic liquids,[11−14] the number of different ionic liquids that have been studied is still very small.

In this work, pressure−density−temperature ($p\rho T$) data are reported for three ionic liquids, namely, 1-ethyl-3-methylimidazolium L-lactate ([emim][lactate]), 2-hydroxyethyl-trimethylammonium L-lactate ([$(C_2H_4OH)(CH_3)_3N$][lactate]), and 1-butyl-3-methylimidazolium chloride ([bmim][Cl]). The [emim][lactate] and [$(C_2H_4OH)(CH_3)_3N$][lactate] are functional ionic liquids that have been studied for possible chiral separations,[15] and [bmim][Cl] is a solvent being studied by many researchers for cellulose processing and modification. The $p\rho T$ data were measured with a bellows type dilatometer at temperatures up to 452 K and at pressures up to 200 MPa. The data are analyzed with the Tait equation, and equation-of-state parameters are reported for lattice-type equations of state.

MATERIALS AND METHODS

The [emim][lactate], CAS number 878132-19-5, was obtained from Sigma-Aldrich (USA) with a stated purity of 98 %; [$(C_2H_4OH)(CH_3)_3N$][lactate], CAS number 888724-51-4, was obtained from Sigma-Aldrich (USA) with a stated purity of 95 %, and [bmim][Cl], CAS number 79917-90-1, was obtained from Fluka (Tokyo) with a stated purity of 98 %. The ionic liquids were degassed under vacuum at 70 °C for 12 h before use and handled to minimize contamination. The water mass fraction was measured with Karl Fischer titration and was 0.051 %, 0.56 %, and 0.21 % for [emim][lactate], [$(C_2H_4OH)(CH_3)_3N$][lactate], and [bmim][Cl], respectively.

Densities. Densities of ionic liquids (ILs) at 0.101 MPa were measured with a vibrating tube instrument (Anton Paar SVM 3000, Graz). The measurement temperature was (293 to 352) K for [emim][lactate] and [$(C_2H_4OH)(CH_3)_3N$][lactate] and (348 to 375) K for [bmim][Cl]. High pressure density data of the ionic liquids at elevated pressures were measured with a

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bellows apparatus. The $p\rho T$ apparatus consisted of a bellows dilatometer, a pressure vessel, multiple temperature control sections, a hand pump, pressure gauges, and a section for displacement measurement. The change of volume for the ionic liquid was determined from the change in length of the bellows, as measured by a linear variable differential transformer (LVDT). Measurements were replicated by measuring volume displacements upon increasing and decreasing pressure. More details on the precise procedure are described in a previous paper.[16] The uncertainty of pressure measurement was ± 0.1 MPa

Table 1. Densities of Ionic Liquids[a] at 0.101 MPa Measured with a Vibrating Tube Instrument

T/K	ρ/kg·m^3		
	[emim][lactate]	[$(C_2H_4OH)(CH_3)_3N$][lactate]	[bmim][Cl]
293.15	1149.7	1143.2	
298.15	1146.1	1139.9	
303.15	1142.5	1136.8	
308.15	1139.0	1133.6	
313.15	1135.6	1130.5	
318.15	1132.2	1127.3	
323.15	1128.8	1124.2	
328.15	1125.4	1121.1	
333.15	1122.1	1118.1	
338.15	1118.7	1115.1	
343.15	1115.4	1112.2	
348.15	1112.1	1109.3	1055.7
353.15	1108.8	1106.4	1053.0
358.15			1050.4
363.15			1047.7
368.15			1045.1
373.15			1042.6

[a] 1-ethyl-3-methylimidazolium L-lactate ([emim][lactate]); 2-hydroxyethyl-trimethylammonium L-lactate ([$(C_2H_4OH)(CH_3)_3N$][lactate]); 1-butyl-3-methylimidazolium chloride ([bmim][Cl]).

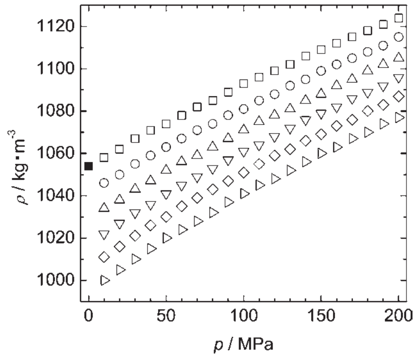

Figure 1. Density of 1-butyl-3-methylimidazolium chloride, [bmim][Cl], versus pressure at various temperatures; □, 352.1 K; ○, 372.4 K; △, 392.3 K; ▽, 412.3 K; ◇, 432.2 K; right-pointing triangle, 452.0 K; ■, measured with a vibrating tube instrument.

Table 2. Experimental Densities (ρ) for 1-Ethyl-3-methylimidazolium L-Lactate, [emim][Lactate]

	T/K						
	312.6	332.1	352.1	372.2	392.4	412.4	432.2
p/MPa	ρ/kg·m^{-3}						
0.1	1136[a]						
10	1140	1126	1113	1101	1088	1076	1064
20	1144	1131	1118	1105	1093	1081	1069
30	1148	1135	1122	1110	1098	1086	1074
40	1151	1139	1126	1114	1102	1090	1079
50	1155	1142	1130	1118	1106	1095	1084
60	1158	1146	1134	1122	1110	1099	1088
70	1162	1149	1137	1126	1114	1103	1093
80	1165	1153	1141	1129	1118	1107	1097
90	1168	1156	1144	1133	1122	1111	1101
100	1172	1159	1148	1137	1126	1115	1105
110	1175	1163	1151	1140	1129	1119	1109
120	1178	1166	1154	1143	1133	1122	1112
130	1181	1169	1157	1147	1136	1126	1116
140	1183	1172	1160	1150	1139	1129	1119
150	1186	1175	1164	1153	1143	1132	1123
160	1189	1178	1166	1156	1146	1136	1126
170	1192	1180	1169	1159	1149	1139	1130
180	1195	1183	1172	1162	1152	1142	1133
190	1197	1186	1175	1165	1155	1145	1136
200	1200	1189	1178	1168	1158	1148	1139

[a] Measured with a vibrating tube instrument.

Table 3. Experimental Densities (ρ) for 2-Hydroxyethyltrimethylammonium L-Lactate, [$(C_2H_4OH)(CH_3)_3N$][Lactate]

	T/K				
	312.4	332.3	352.2	372.3	392.4
p/MPa	ρ/kg·m^{-3}				
0.1	1130[a]				
10	1133	1121	1109	1098	1086
20	1137	1124	1113	1101	1090
30	1140	1128	1116	1105	1094
40	1143	1131	1120	1109	1098
50	1146	1134	1123	1112	1101
60	1150	1137	1126	1115	1105
70	1152	1140	1129	1118	1108
80	1155	1143	1132	1122	1111
90	1157	1146	1135	1125	1114
100	1160	1149	1138	1128	1117
110	1163	1152	1141	1130	1121
120	1166	1154	1144	1133	1124
130	1168	1157	1146	1136	1126
140	1171	1160	1149	1139	1129
150	1173	1162	1152	1142	1132
160	1176	1165	1154	1144	1135
170	1178	1168	1157	1147	1138
180	1181	1170	1159	1150	1140
190	1183	1172	1162	1152	1143
200	1185	1175	1165	1155	1146

[a] Measured with a vibrating tube instrument.

below 100 MPa and ± 0.25 MPa from (100 to 200) MPa. The temperature uncertainty is estimated to be ± 0.1 K. The relative uncertainty in the density measurements is estimated to be ± 0.1 %.

CORRELATION

Tait Equation. The Tait equation is an empirical equation for representing liquid densities that has the following form in terms of the specific volume, V_s:

$$V_s(P,T) = \frac{1}{\rho(p,T)} = \frac{1}{\rho(p_0,T)}(1 - C\ln(1 + p/B(T))) \tag{1}$$

Table 4. Experimental Densities (ρ) for 1-Butyl-3-methylimidazolium Chloride, [bmim][Cl]

	T/K					
	352.1	372.4	392.3	412.3	432.2	452.0
p/MPa	ρ/kg·m^{-3}					
0.1	1054[a]					
10	1058	1046	1034	1022	1011	1000
20	1062	1050	1038	1027	1016	1005
30	1067	1055	1043	1032	1021	1010
40	1071	1059	1047	1036	1026	1015
50	1074	1063	1052	1041	1030	1020
60	1078	1067	1056	1045	1035	1024
70	1082	1071	1059	1049	1039	1028
80	1085	1074	1063	1053	1043	1032
90	1089	1078	1067	1057	1047	1037
100	1093	1081	1071	1061	1051	1041
110	1096	1085	1075	1064	1055	1045
120	1099	1088	1078	1068	1059	1048
130	1102	1092	1081	1072	1062	1052
140	1106	1095	1085	1076	1066	1056
150	1109	1099	1088	1079	1069	1060
160	1112	1102	1092	1082	1073	1063
170	1115	1105	1095	1086	1076	1067
180	1118	1108	1099	1089	1080	1070
190	1121	1111	1102	1092	1083	1074
200	1124	1115	1105	1096	1087	1077

[a] Measured with a vibrating tube instrument.

$$\frac{1}{\rho(P_0,T)} = a_0 + a_1 \cdot T + a_2 \cdot T^2 \tag{2}$$

$$B(T) = b_0 \exp(-b_1 \cdot T) \tag{3}$$

Derivatives of the Tait equation were used to estimate the thermal expansion coefficient, $\alpha_p = (1/V_s)(\partial V_s/\partial T)_p$ and the isothermal compressibility, $k_T = -(1/V_s)(\partial V_s/\partial p)_T$, as shown in previous works.[16]

Sanchez–Lacombe Equation of State (SL EOS). The SL EOS is based on lattice fluid theory[17] and can be used to predict phase equilibria from the pure component parameters and has the following form:

$$\tilde{\rho}^2 + \tilde{p} + \tilde{T}[\ln(1-\tilde{\rho}) + (1 - 1/r)\tilde{\rho}] = 0 \tag{4}$$

$$\tilde{T} \equiv T/T^*, \quad \tilde{p} \equiv p/p^*, \quad \tilde{\rho} \equiv \rho/\rho^* \quad \varepsilon^* = RT^*,$$
$$v^* = RT^*/p^*, \quad r = Mp^*/RT^*\rho^*$$

where p^*, T^*, and ρ^* represent characteristic parameters of the EOS, M is the molecular weight, and R is the gas constant.

ε-Modified SL EOS. The SL EOS has been found to have a strong systematic deviation for $p\rho T$ correlation of polar fluids.[12] To improve the correlation for fluids that have strong polar and Coulombic interactions, a functional form of the interaction energy, ε^*, was proposed[12] as follows:

$$\varepsilon^*(T) = \varepsilon_0 \frac{\alpha T}{1 + \alpha T} \tag{5}$$

$$T^* = \frac{\varepsilon^*(T)}{R} = \frac{\varepsilon_0}{R}\frac{\alpha T}{1 + \alpha T} \tag{6}$$

where α is a characteristic parameter that allows for the variation of the interaction energy, ε^*, with temperature.

Objective Function. The Tait equation parameters and the characteristic parameters for the equation of state were determined for each substance by minimizing the average relative deviations (ARDs) between calculated and experimental densities according to the following objective function:

$$\text{ARD} = \sum \left| \frac{\rho^{\text{calc}} - \rho^{\text{exp}}}{\rho^{\text{exp}}} \right| \bigg/ n \tag{7}$$

The deviations in the correlated values of the equations of state are compared.

Table 5. Tait Equation Parameters for the Ionic Liquids Studied and Correlation Deviation from a Least Squares Fit

	V_s (0.1 MPa,T)/m^3·kg^{-1}			$B(T)$/MPa			
	$a_0 \cdot 10^4$	$a_1 \cdot 10^7$	$a_2 \cdot 10^{10}$	b_0	$b_1 \cdot 10^3$		
ionic liquid[a]	m^3·kg^{-1}	m^3·kg^{-1}·K^{-1}	m^3·kg^{-1}·K^{-2}	MPa	K^{-1}	C	100 ARD
[emim][lactate]	7.027	5.938	−0.8187	701.2	3.091	0.09511	0.013
[(C_2H_4OH)(CH_3)$_3$N][lactate]	7.014	6.691	−2.619	688.7	2.489	0.09567	0.009
[bmim][Cl]	7.496	5.706	−0.1097	664.5	2.499	0.1162	0.021
						Average	0.014

[a] 1-Ethyl-3-methylimidazolium L-lactate ([emim][lactate]); 2-hydroxyethyl-trimethylammonium L-lactate ([(C_2H_4OH)(CH_3)$_3$N][lactate]); 1-butyl-3-methylimidazolium chloride ([bmim][Cl]).

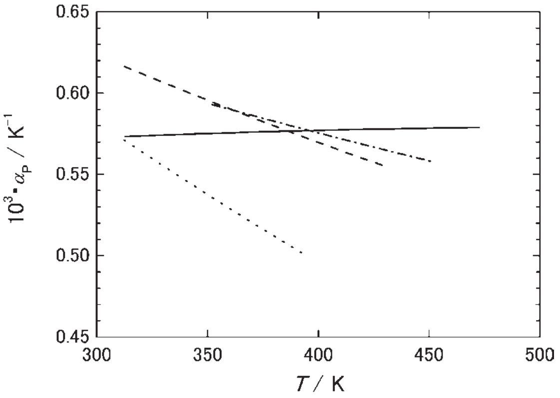

Figure 2. Thermal expansion coefficients of ionic liquids at 0.101 MPa. —, [bmim][PF$_6$] (ref 16); - - -, [emim][lactate]; · · · ·, [(C$_2$H$_4$OH)(CH$_3$)$_3$N][lactate]; -·-·-, [bmim][Cl].

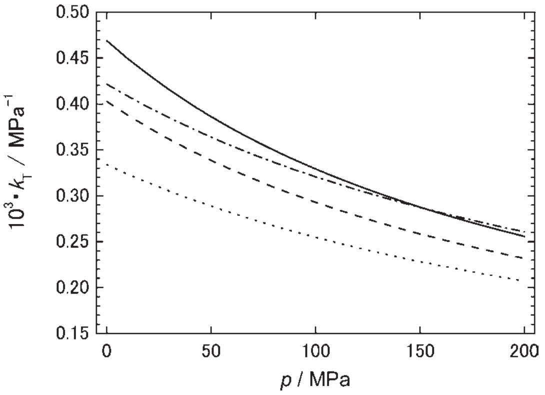

Figure 3. Isothermal compressibilities of ionic liquids at 352.1 K. —, [bmim][PF$_6$] (ref 16); - - -, [emim][lactate]; · · · ·, [(C$_2$H$_4$OH)(CH$_3$)$_3$N][lactate]; -·-·-, [bmim][Cl].

RESULTS AND DISCUSSION

Density data measured at atmospheric pressure are summarized in Table 1, and high-pressure density data measured for each ionic liquid are summarized in Tables 2 to 4. There are no previous $p\rho T$ data reported in the literature for the ionic liquids measured in this work. Comparison data for typical ionic liquid like [bmim][PF$_6$] between our technique and literature data are given in our previous papers.[16,18]

Figure 1 shows $p\rho T$ data measured for [bmim][Cl]. For the case of [bmim][Cl], $p\rho T$ data were measured at temperatures higher than 345 K, since the melting point is about 340 K. However, it should be pointed out that for [bmim][Cl] as well as for other liquids, recrystallization does not readily occur once the ionic liquid is brought to a temperature above its melting point. All ionic liquids exhibited smooth temperature and pressure trends, which means that no phase transitions occurred in the measurement region.

Table 5 shows Tait equation parameters for each ionic liquid. The Tait equation could correlate the density data to within an ARD from 0.013 % to 0.021 % over the entire pressure and temperature ranges studied for all ionic liquids. Figures 2 and 3 show the thermal expansion coefficient and isothermal compressibility for the measured ionic liquids and for one set of values in the literature.[16] Compared with [bmim][PF$_6$], the thermal expansion coefficient of the measured ionic liquids in this work showed temperature variation with $d\alpha_p/dT$ being about $-5.3 \cdot 10^{-7}$ K^{-2} for [emim][lactate], $-8.6 \cdot 10^{-7}$ K^{-2} for [(C$_2$H$_4$OH)(CH$_3$)$_3$N][lactate], and $-3.7 \cdot 10^{-7}$ K^{-2} for [bmim][Cl] at a nominal temperature of 352 K. This might be attributed in some part to the attractive forces of the measured ionic liquids being stronger than the repulsive forces and thus having a greater temperature dependence than that shown for [bmim][PF$_6$]. The negative values of the temperature derivative of the thermal expansion coefficient, $d\alpha_p/dT$, are more typical for metals and minerals than for liquids. The negative $d\alpha_p/dT$ could be evidence for a more structured liquid phase than that for water or organic solvents. For the measured isothermal compressibilities, they were found to be in the order of [(C$_2$H$_4$OH)(CH$_3$)$_3$N][lactate] < [emim][lactate] < [bmim][Cl] < [bmim][PF$_6$] for pressures up to about 100 MPa. The [(C$_2$H$_4$OH)(CH$_3$)$_3$N][lactate] has a large alkyl chain, which probably lowers its free volume, thus causing the isothermal compressibility to be lower than the other ionic liquids shown (Figure 3).

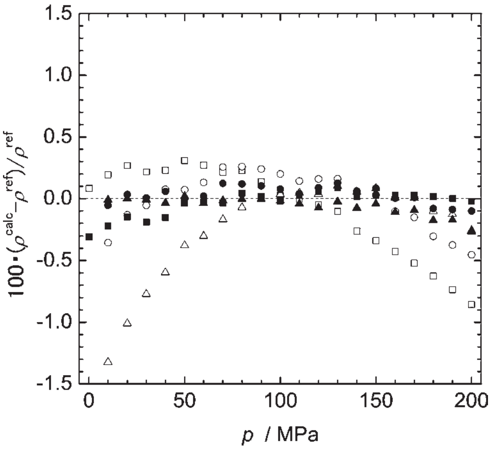

Figure 4. Residual plot between calculated and measured densities for [bmim][Cl] with equations of state (EOS); filled symbols, ε^*-modified Sanchez–Lacombe EOS; ■, 352.1 K; ●, 392.3 K; ▲, 432.2 K; open symbols, Sanchez–Lacombe EOS; □, 352.1 K; ○, 392.3 K; △, 432.2 K.

Tables 6 and 7 show the characteristic parameters determined for the SL EOS and the ε-modified SL EOS. The values for r and ε_0 of the ε-modified SL EOS are typically larger than the r and ε^* values for the original SL EOS according to previous determinations.[12] The v^* values determined (Table 7) were smaller than the values for v^* of the original SL EOS, and this is also observed in the previously studied systems.[12] As shown by the ARD in Tables 6 and 7, the ε-modified SL EOS gave a better correlation than that of the SL EOS. The differences between the SL EOS and the ε-modified SL EOS can be seen clearly in a residual plot (Figure 4). The systematic deviations in density with pressure for the ε-modified SL EOS were greatly reduced over those of the SL EOS (Figure 4). In the ε-modified SL EOS, $1/\alpha$ has a tendency to increase as Coulombic and van der Waals forces increase.[12] For the group of ILs studied, $1/\alpha$ was in the order of [bmim][Cl] > [(C$_2$H$_4$OH)(CH$_3$)$_3$N][lactate] > [emim][lactate]. Yasaka et al. reported that the small chloride anion attracts water much stronger than the larger hydrophobic anion PF$_6$ with a rotational dynamics study.[19] The [bmim][Cl] ionic liquid has strong Coulombic interactions that make its interactions larger than those of more highly polar molecules. Thus, this trend corresponds with those reported for other ionic liquids.[12]

Table 6. Pure Parameters Determined for the Sanchez−Lacombe Equation of State and the Average Relative Deviation (ARD) of the Correlation

ionic liquid[a]	M	ε^*	v^*	r	100 ARD
		$J \cdot mol^{-1}$	$cm^3 \cdot mol^{-1}$		
[emim][lactate]	200.2349	5490	9.272	17.73	0.28
$[(C_2H_4OH)(CH_3)_3N]$[lactate]	193.2408	5628	8.569	18.71	0.13
[bmim][Cl]	174.6711	5575	9.756	15.50	0.23
average					0.21

[a] 1-Ethyl-3-methylimidazolium L-lactate ([emim][lactate]); 2-hydroxyethyl-trimethylammonium L-lactate ($[(C_2H_4OH)(CH_3)_3N]$[lactate]); 1-butyl-3-methylimidazolium chloride ([bmim][Cl]).

Table 7. Pure Parameters Determined for the ε-Modified Sanchez−Lacombe Equation of State and the Average Relative Deviation (ARD) of the Correlation

ionic liquid[a]	$1/\alpha$	ε_0	v^*	r	100 ARD
	K	$J \cdot mol^{-1}$	$cm^3 \cdot mol^{-1}$		
[emim][lactate]	590	9903	1.759	81.47	0.091
$[(C_2H_4OH)(CH_3)_3N]$[lactate]	641	10680	1.667	84.34	0.048
[bmim][Cl]	713	11050	1.951	67.14	0.061
average					0.067

[a] 1-Ethyl-3-methylimidazolium L-lactate ([emim][lactate]); 2-hydroxyethyl-trimethylammonium L-lactate ($[(C_2H_4OH)(CH_3)_3N]$[lactate]); 1-butyl-3-methylimidazolium chloride ([bmim][Cl]).

CONCLUSIONS

Pressure−density−temperature data for three ionic liquids were measured and correlated. Among the three correlations evaluated, the Tait equation provided the lowest ARDs in density (0.014 %). The ε-modified SL EOS could correlate the density data to within an ARD of 0.067 %, which was much lower than that of the original SL EOS. The ε-modified SL EOS is presently being applied to CO_2−ionic liquid mixtures, and results will be reported in the near future.

ASSOCIATED CONTENT

Supporting Information. Thermal expansion coefficient and isothermal compressibility values for [emim][Lactate], $[(C_2H_4OH)(CH_3)_3N]$[lactate], and [bmim][Cl] determined from the Tait equation parameters. This material is available free of charge via the Internet at http://pubs.acs.org.

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