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Measurement, correlation, and prediction of vapor pressure for binary and ternary systems containing an alkylsulfate-based ionic liquid



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ABSTRACT

Vapor pressure data for water, 1-propanol, 2-propanol, as well as the mixtures of {water + 1-propanol} and {water + 2-propanol}, were measured by a quasi-static ebulliometric method, in the presence of an alkylsulfate-based IL, namely, 1-ethyl-3-methylimidazolium methylsulfate ([EMIM][MS]) or 1-ethyl-3methylimidazolium ethylsulfate ([EMIM][ES]). The experimental vapor pressure data for binary systems containing IL were correlated by NRTL model with an overall relative root mean square deviation (rRMSD) of 0.0053, and the obtained binary NRTL parameters were employed to predict the vapor pressure for two ternary systems with an overall rRMSD of 0.0196. In addition, isobaric vapor-liquid equilibria were predicted for the ternary systems containing [EMIM][MS], [EMIM][ES], 1,3-dimethylimidazolium methylsulfate ([MMIM][MS]), 1-ethyl-3-methylimidazolium tetrafluoroborate ([EMIM] [BF₄]), and 1-ethyl-3-methylimidazolium trifluoromethanesulfonate [EMIM][OTF], respectively, with IL mole fraction of 0.05, 0.15, and 0.25 at 101.325 kPa. It was found that the addition of IL can enhance the relative volatility of propanol to water, and the separation ability follows the order of [MMIM] [MS] > [EMIM][MS] > [EMIM][ES] > [EMIM][BF₄] > [EMIM][OTf], which was further explained at electronic level with quantum chemical calculation. Therefore, the azeotropic mixtures of {water + 1-propanol}, and {water+2-propanol} might be separated effectively by the addition of the alkylsulfate-based ILs in extractive distillation.

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1. Introduction

In chemical industry, the separation of azeotropic or close boiling mixtures is usually realized with special distillation, such as extractive distillation and salt distillation, in which an entrainer is added into the mixtures for increasing relative volatility. However, the volatile organic solvents used in extractive distillation are likely to harm workers, pollute environment, and consume excessive energy in solvent recycle process, while the inorganic salts used in salt distillation may cause the problems of pipeline blockage and equipment corrosion. Ionic liquids (ILs) are prospective alternatives to conventional solvents in special distillation [1], since several ILs can combine the advantages of both organic solvents and inorganic salts, showing the favorable properties of negligible volatility, good stability, easy recyclability, non-flammability, and structural tenability [2].

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Vapor-liquid equilibria (VLE) data are the foundation of process design and equipment selection in distillation. Up to now, many researchers have studied the VLE behavior for systems containing ILs, aiming to screen suitable ILs as entrainers in extractive distillation to enhance separation efficiency [3]. Among those ILs, imidazoliumbased ILs are widely investigated, including the ILs with waterunstable fluorinated anions like tetrafluoroborate $([BF_4]^-)$ and hexafluorophosphate ($[PF_6]^-$), which can release toxic hydrogen fluoride under aqueous environment [4], and some expensive ILs with the anion of bis(trifluromethanesulfonyl)imide $([NTf_2]^-)$ or trifluoromethanesulfonate ([OTf]⁻), which seriously limited their technical application in large scale. Compared with those flawed ILs as entrainers, the hydrophilic ILs with alkylsulfate and dialkylphosphate anions have shown great potential in extractive distillation, owing to their low-viscosity, non-toxicity, and reasonable price [5,6]. Several works have been done to determine the VLE data for systems containing alkylsulfate- and dialkylphosphate-based ILs. Li et al. measured the vapor pressure for binary and ternary systems containing dialkylphosphate-based ILs, including 1-methyl-3methylimidazolium dimethylphosphate ([MMIM][DMP]),

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lable I		
Specifications	of chemical	samples.

Chemicals	Source	Purity	Purification method	Analysis method
Water	Distilled in the laboratory	Electrical conductivity < 5 µS cm ⁻¹ (298.15 K)	None	Electrical conductivity measurement ^a
1-Propanol 2-Propanol [EMIM] [MS]	Guangfu Chemical Regent Co., Tianjin, China Guangfu Chemical Regent Co., Tianjin, China Synthesized in the laboratory	Mass fraction > 0.997 Mass fraction > 0.997 Mole fraction > 0.990	4A molecular sieves 4A molecular sieves Distillation	GC ^b GC ^{b[EMIM][MS]} ¹ H NMR, ¹³ C NMR ^c
[EMIM][ES]	Synthesized in the laboratory	Mole fraction > 0.990	Distillation	¹ H NMR, ¹³ C NMR ^c

^a Measured at 298.15 K with a conductivity meter (type EC-215, Hanna Co., Italy).

^b Checked with a gas chromatography (type GC2010, SHIMADZU Co., Japan) equipped with a FID detector and FFAP capillary column.

^c Recorded on a 400.0 MHz NMR spectrometer (Bruker Co., Germany) at T = 300 K using deuterated water (D₂O) as the external reference solvent.

1-ethyl-3-methylimidazolium dimethylphosphate ([EMIM][DMP]), 1-ethyl-3-ethylimidazolium diethylphosphate ([EEIM][DEP]), 1ethyl-3-methylimidazolium diethylphosphate ([EMIM][DEP]), and 1-butyl-3-emthylimidazolium dibutylphosphate ([EMIM][DEP]), and 1-butyl-3-emthylimidazolium dibutylphosphate ([BMIM][DBP]). Their results indicated that the addition of ILs can change the relative volatility of a solvent, and that ILs can exhibit "ionic" or "molecule" character in different solvents [7–9]. On the other hand, 1-ethyl-3-methylimidazolium ethylsulfate ([EMIM][ES]), one of the first commercial "bulk" ionic liquids (available on a ton-scale), was reported to have the ability to break the azeotrope of water–ethanol mixture with sufficient IL-content [10,11].

In our previous work, the effects of 1-methyl-3-methylimidazolium methylsulfate [MMIM][MS] on the vapor pressure of water,

Table 2

Experimental and calculated vapor pressure data for the binary system {water (1)+[EMIM][MS] (2)}.^a

T/K	P ^{exp} /kPa	P ^{cal} /kPa	γ_1^{exp}	γ_1^{cal}
$x_1 = 0.9911$				
325.47	13.58	13.68	0.9910	0.9987
332.85	19.31	19.45	0.9915	0.9988
339.46	26.10	26.25	0.9929	0.9989
347.77	37.35	37.57	0.9932	0.9990
353.71	47.73	47.97	0.9940	0.9990
360.69	62.89	63.18	0.9946	0.9991
367.95	82.80	83.09	0.9957	0.9991
373.32	100.79	100.94	0.9977	0.9992
$x_1 = 0.9801$				
324.84	12.91	13.06	0.9831	0.9939
332.78	18.88	19.09	0.9837	0.9944
341.06	27.44	27.75	0.9837	0.9948
347.98	36.97	37.35	0.9853	0.9952
355.55	50.42	50.91	0.9861	0.9955
362.67	66.52	67.20	0.9857	0.9957
368.80	83.60	84.49	0.9854	0.9960
373.91	100.75	101.65	0.9873	0.9961
$x_1 = 0.9659$				
324.90	12.61	12.77	0.9713	0.9834
334.35	19.81	20.03	0.9739	0.9849
341.79	27.62	27.98	0.9732	0.9859
350.10	39.34	39.87	0.9739	0.9869
355.56	49.12	49.78	0.9744	0.9875
363.58	67.17	68.05	0.9756	0.9883
369.47	83.67	84.75	0.9762	0.9888
374.66	100.80	102.14	0.9763	0.9893
$x_1 = 0.9182$				
332.13	16.38	16.21	0.9390	0.9291
339.27	22.75	22.53	0.9421	0.9332
344.26	28.31	28.10	0.9428	0.9358
349.55	35.37	35.22	0.9425	0.9384
357.67	49.22	49.08	0.9449	0.9421
364.82	64.72	64.82	0.9437	0.9451
371.71	83.58	83.73	0.9461	0.9478
376.98	100.94	101.12	0.9480	0.9497

1-propanol, 2-propanol, {water+1-propanol}, and {water+2propanol} were studied [12]. In order to discuss the influence of alkylsulfate-based ILs with various anion structures on the VLE behavior of {water+propanol} mixtures, herein, we shifted the study object to 1-ethyl-3-methylimidazolium methylsulfate ([EMIM][MS]) and 1-ethyl-3-methylimidazolium ethylsulfate ([EMIM][ES]). Thus, the vapor pressure data for water, 1-propanol, and 2-propanol, as well as the mixtures of {water+1-propanol} and {water+2-propanol}, were measured in the presence of [EMIM][MS] or [EMIM][ES] at varying IL-content and temperature. The experimental data for binary systems were correlated by NRTL model, and the obtained binary model parameters were used to predict the vapor pressure of the ternary systems of {water+1-

Table 3

Experimental and calculated vapor pressure data for the binary system {water (1)+[EMIM][ES] (2)].^a

$x_1 = 0.9916$ 323.8712.5612.660.99180.9991331.5018.1618.280.99290.9992386.625.2025.350.99340.9992347.3936.8337.010.99420.9993353.9848.3448.540.99520.9993361.9566.1566.380.99480.9944368.2983.9984.210.99680.9944372.7799.0299.040.99920.9994 $x_1 = 0.9813$ $x_1 = 0.9813$ $x_1 = 0.9872$ 0.9959333.9420.0720.200.98850.9962339.0725.3125.480.98800.9964347.5036.4836.700.98840.9967354.1547.9848.250.98810.9968362.5466.7267.030.98940.9972373.2599.0799.490.99130.9973 $x_1 = 0.9684$ $x_1 = 0.9684$ $x_1 = 0.9884$ 0.9970345.5620.2220.370.98220.9889344.5620.92220.370.98220.9893349.3338.5338.870.98240.9910355.4449.4049.870.98210.914362.8466.0266.570.98370.9913373.889.149.930.98430.9223373.889.149.9930.98430.9264373.352.2819.3713.520.94200.9564 <th>T/K</th> <th>P^{exp}/kPa</th> <th>P^{cal}/kPa</th> <th>γ_1^{exp}</th> <th>γ_1^{cal}</th>	T/K	P ^{exp} /kPa	P ^{cal} /kPa	γ_1^{exp}	γ_1^{cal}
323.8712.5612.660.99180.9991331.5018.1618.280.99290.9992386.625.2025.350.99340.9992347.3936.8337.010.99420.9993353.9848.3448.540.99520.9993361.9566.1566.380.99580.9994368.2983.9984.210.99680.9994372.7799.0299.040.99920.9994333.9420.0720.200.98850.9962339.0725.3125.480.98800.9964347.5036.4836.700.98840.9967354.1547.9848.250.98810.9963362.5466.7267.030.98940.9970368.7884.3084.650.99130.9972373.2599.0799.490.99130.9973 $x_1=0.9684$ 25.8426.100.98050.9903349.3338.5338.870.98220.9888340.0425.8426.100.98220.9813355.4449.4049.870.98210.9914362.8466.0266.570.98370.9913373.889.149.930.98430.9923373.889.149.930.98430.9923373.889.149.930.98430.9923373.8519.3713.620.94200.9564347.8026.010.94380.9537 <t< td=""><td>$x_1 = 0.9916$</td><td></td><td></td><td></td><td></td></t<>	$x_1 = 0.9916$				
331.50 18.1618.280.99290.9992 338.66 25.2025.350.99340.9992 347.39 36.8337.010.99420.9993 353.98 48.3448.540.99520.9993 361.95 66.1566.380.99580.9994 368.29 83.9984.210.99680.9994 372.77 99.0299.040.99920.9994 323.35 12.0712.170.98720.9959 339.07 25.3125.480.98850.9962 339.07 25.3125.480.98840.9967 347.50 36.4836.700.98840.9967 354.15 47.9848.250.98810.9968 362.54 66.7267.030.98940.9970 373.25 99.0799.490.99130.9973 $x_1 = 0.9684$ $x_1 = 0.9684$ $x_1 = 0.9822$ 0.9889 34.56 20.2220.370.98220.9889 34.66 20.5266.570.98370.9913 34.56 20.9266.570.98370.9914 362.84 66.0266.570.98370.9913 373.88 9.149.930.98430.9923 373.88 9.149.930.98430.9923 373.88 9.149.930.94200.9567 327.81 13.6313.720.94380.9537 335.28 19.3719.520.94200.9564 341.79	323.87	12.56	12.66	0.9918	0.9991
338.6625.2025.350.99340.9992347.3936.8337.010.99420.9993353.9848.3448.540.99520.9993361.9566.1566.380.99580.9994368.2983.9984.210.99680.9994372.7799.0299.040.99920.9994372.7799.0299.040.99920.999433.9420.0720.200.98850.996233.9420.0720.200.98840.9967354.1547.9848.250.98810.9968362.5466.7267.030.98840.9972373.2599.0799.490.99130.972373.2599.0799.490.99130.9972373.2599.0799.490.99130.9973 $x_1 = 0.9684$ $x_1 = 0.9684$ $x_1 = 0.9824$ 0.9910355.4449.4049.870.98220.9889340.0425.8426.100.98240.9910355.4449.4049.870.98210.9914362.8466.0266.570.98370.9913373.889.149.930.98430.9923373.889.149.930.98430.9923373.889.149.930.94380.9537335.2819.3719.520.94200.9564347.9025.0120.160.94200.9567	331.50	18.16	18.28	0.9929	0.9992
347.39 36.83 37.01 0.9942 0.9993 353.98 48.34 48.54 0.9952 0.9993 361.95 66.15 66.38 0.9958 0.9994 368.29 83.99 84.21 0.9968 0.9994 372.77 99.02 99.04 0.9992 0.9994 372.77 99.02 99.04 0.9992 0.9994 372.77 99.02 99.04 0.9992 0.9994 372.77 99.02 99.04 0.9992 0.9994 372.77 99.02 0.9944 0.9992 0.9994 372.77 99.02 99.04 0.9992 0.9994 372.77 99.02 0.9944 0.9992 0.9994 372.77 99.02 0.9944 0.9992 0.9994 372.77 99.02 0.9044 0.9992 0.9994 333.94 20.07 20.20 0.9885 0.9962 339.07 25.31 25.48 0.9884 0.9967 354.15 47.98 48.25 0.9884 0.9970 368.78 84.30 84.65 0.9913 0.9972 373.25 99.07 99.49 0.9913 0.9973 $x_1 = 0.9684$ $x_1 = 0.9884$ 26.10 0.9865 0.9903 349.33 38.53 38.87 0.9824 0.9910 355.44 49.40 49.87 0.9821 0.9914 362.84 66.02 66.57 0.9837 0.9923 <	338.66	25.20	25.35	0.9934	0.9992
353.98 48.34 48.54 0.9952 0.9993 361.95 66.15 66.38 0.9958 0.9994 368.29 83.99 84.21 0.9968 0.9994 372.77 99.02 99.04 0.9992 0.9994 $x_1=0.9813$ $x_1=0.9813$ $x_1=0.9872$ 0.9959 333.94 20.07 20.20 0.9885 0.9962 339.07 25.31 25.48 0.9880 0.9964 347.50 36.48 36.70 0.9884 0.9967 354.15 47.98 48.25 0.9881 0.9967 354.15 47.98 48.25 0.9881 0.9970 368.78 84.30 84.65 0.9913 0.9972 373.25 99.07 99.49 0.9913 0.9973 $x_1=0.9684$ $x_1=0.9684$ 26.10 0.9805 0.9903 349.33 38.53 38.87 0.9824 0.9910 355.44 49.40 49.87 0.9821 0.9914 46.284 66.02 66.57 0.9837 0.9923 373.88 99.14 99.93 0.9843 0.9923 373.88 99.14 99.93 0.9843 0.9923 373.88 99.14 99.93 0.9438 0.9537 335.28 19.37 19.52 0.9420 0.9564 341.79 25.61 26.16 0.9420 0.9567	347.39	36.83	37.01	0.9942	0.9993
361.95 66.15 66.38 0.9958 0.9994 368.29 83.99 84.21 0.9968 0.9994 372.77 99.02 99.04 0.9992 0.9994 $x_1=0.9813$ 323.35 12.07 12.17 0.9872 0.9959 333.94 20.07 20.20 0.9885 0.9962 39.07 25.31 25.48 0.9880 0.9964 347.50 36.48 36.70 0.9884 0.9970 362.54 66.72 67.03 0.9894 0.9970 368.78 84.30 84.65 0.9913 0.9972 373.25 99.07 99.49 0.9913 0.9973 $x_1=0.9684$ $x_1=0.9684$ $x_1=0.9805$ 0.9903 34.56 20.22 20.37 0.9822 0.9889 34.66 20.22 20.37 0.9824 0.9910 355.44 49.40 49.87 0.9821 0.9914 $46.84.53$ 85.21 0.9836 0.9923 373.88 99.14 99.93 0.9843 0.9926 $x_1=0.9292$ $x_1=0.9438$ 0.9537 335.28 19.37 19.52 0.9420 0.9564 37.81 13.63 13.72 0.9438 0.9537 0.9567 0.9567 327.81 13.63 13.72 0.9420 0.9567 327.81 13.63 13.72 0.9420 0.9567 347.92 10.37 19.52 0.9420 0.9567 <td>353.98</td> <td>48.34</td> <td>48.54</td> <td>0.9952</td> <td>0.9993</td>	353.98	48.34	48.54	0.9952	0.9993
368.29 83.99 84.21 0.9968 0.9994 372.77 99.02 99.04 0.9992 0.9994 $x_1 = 0.9813$ 223.35 12.07 12.17 0.9872 0.9959 333.94 20.07 20.20 0.9885 0.9962 339.07 25.31 25.48 0.9880 0.9964 347.50 36.48 36.70 0.9884 0.9967 354.15 47.98 48.25 0.9881 0.9963 362.54 66.72 67.03 0.9894 0.9972 373.25 99.07 99.49 0.9913 0.9973 $x_1 = 0.9684$ $x_1 = 0.9684$ 20.22 20.37 0.9822 0.9889 34.56 20.22 20.37 0.9822 0.9898 340.04 25.84 26.10 0.9805 0.9903 349.33 38.53 38.87 0.9821 0.9914 362.84 66.02 66.57 0.9837 0.9913 373.88 99.14 99.93 0.9843 0.9923 373.88 99.14 99.93 0.9843 0.9923 373.88 19.37 13.62 0.9420 0.9567 327.81 13.63 13.72 0.9438 0.9537 335.28 19.37 19.52 0.9420 0.9564 341.79 25.61 26.16 0.9420 0.9567	361.95	66.15	66.38	0.9958	0.9994
372.77 99.02 99.04 0.9992 0.9994 $x_1 = 0.9813$ $x_1 = 0.9813$ 323.35 12.07 12.17 0.9872 0.9959 333.94 20.07 20.20 0.9885 0.9962 339.07 25.31 25.48 0.9880 0.9964 347.50 36.48 36.70 0.9884 0.9967 354.15 47.98 48.25 0.9881 0.9968 362.54 66.72 67.03 0.9894 0.9970 368.78 84.30 84.65 0.9913 0.9973 $x_1 = 0.9684$ $x_1 = 0.9684$ $x_1 = 0.9684$ $x_1 = 0.9805$ 0.9903 34.56 20.22 20.37 0.9822 0.9898 340.04 25.84 26.10 0.9805 0.9903 349.33 38.53 38.87 0.9821 0.9914 362.84 66.02 66.57 0.9837 0.9919 355.44 49.40 49.87 0.9843 0.9923 373.88 99.14 99.93 0.9843 0.9926 $x_1 = 0.9292$ $x_1 = 0.9242$ $x_1 = 0.9438$ 0.9537 335.28 19.37 13.52 0.9420 0.9564 327.81 13.63 13.72 0.9438 0.9537 335.28 19.37 19.52 0.9420 0.9564 327.81 13.65 13.616 0.9616 0.9567	368.29	83.99	84.21	0.9968	0.9994
$x_1 = 0.9813$ 323.35 12.0712.170.98720.9959 333.94 20.0720.200.98850.9962 339.07 25.3125.480.98800.9964 347.50 36.4836.700.98840.9967 354.15 47.9848.250.98810.9968 362.54 66.7267.030.98940.9970 368.78 84.3084.650.99130.9972 373.25 99.0799.490.99130.9973 $x_1 = 0.9684$ $x_1 = 0.9684$ $x_1 = 0.9789$ 0.9889 34.56 20.2220.370.98220.9898 340.04 25.8426.100.98050.9903 349.33 38.5338.870.98240.9910 355.44 49.4049.870.98210.9914 362.84 66.0266.570.98370.9913 373.88 99.1499.930.98430.9926 $x_1 = 0.9292$ $x_1 = 0.9292$ $x_1 = 0.9248$ 0.9537 335.28 19.3719.520.94200.9564 341.79 25.6120.1025.057 327.81 13.6313.720.94380.9537 325.28 19.3719.520.94200.9564 341.79 25.6120.1025.057	372.77	99.02	99.04	0.9992	0.9994
323.3512.0712.17 0.9872 0.9959 333.9420.0720.20 0.9885 0.9962 339.0725.3125.48 0.9880 0.9964 347.5036.4836.70 0.9884 0.9967 354.1547.9848.25 0.9884 0.9970 368.7884.3084.65 0.9913 0.9972 373.2599.0799.49 0.9913 0.9973 x1 = 0.9684 </td <td>$x_1 = 0.9813$</td> <td></td> <td></td> <td></td> <td></td>	$x_1 = 0.9813$				
333.94 20.07 20.20 0.9885 0.9962 39.07 25.31 25.48 0.9880 0.9964 347.50 36.48 36.70 0.9884 0.9967 354.15 47.98 48.25 0.9881 0.9968 362.54 66.72 67.03 0.9894 0.9970 368.78 84.30 84.65 0.9913 0.9972 373.25 99.07 99.49 0.9913 0.9973 $x_1 = 0.9684$ $x_1 = 0.9684$ $x_1 = 0.9789$ 0.9889 34.56 20.22 20.37 0.9822 0.9898 340.4 25.84 26.10 0.9805 0.9903 349.33 38.53 38.87 0.9824 0.9910 355.44 49.40 49.87 0.9821 0.9914 462.84 66.02 66.57 0.9837 0.9923 373.88 99.14 99.93 0.9843 0.9926 $x_1 = 0.9292$ $x_1 = 0.9242$ $x_1 = 0.9438$ 0.9537 335.28 19.37 19.52 0.9420 0.9564 341.79 25.61 26.16 0.9420 0.9567	323.35	12.07	12.17	0.9872	0.9959
339.0725.3125.480.98800.9964347.5036.4836.700.98840.9967354.1547.9848.250.98810.9968362.5466.7267.030.98940.9970368.7884.3084.650.99130.9972373.2599.0799.490.99130.9973 $x_1 = 0.9684$ $x_1 = 0.9684$ $x_1 = 0.972$ 20.370.982234.5620.2220.370.98220.9889340.0425.8426.100.98050.9903349.3338.5338.870.98210.9914362.8466.0266.570.98370.9919369.4684.5385.210.98360.9923373.8899.1499.930.98430.9926 $x_1 = 0.9292$ $x_1 = 0.9292$ $x_1 = 0.9242$ 0.9577335.2819.3719.520.94200.9564341.7925.6120.6160.40230.5577	333.94	20.07	20.20	0.9885	0.9962
347.50 36.48 36.70 0.9884 0.9967 354.15 47.98 48.25 0.9881 0.9968 362.54 66.72 67.03 0.9894 0.9970 368.78 84.30 84.65 0.9913 0.9972 373.25 99.07 99.49 0.9913 0.9973 $x_1 = 0.9684$ $x_1 = 0.9684$ $x_1 = 0.9684$ $x_1 = 0.9684$ 34.56 20.22 20.37 0.9822 0.9893 34.04 25.84 26.10 0.9824 0.9910 355.44 49.40 49.87 0.9821 0.9914 362.84 66.02 66.57 0.9837 0.9913 373.88 99.14 99.93 0.9843 0.9923 373.88 99.14 99.93 0.9843 0.9926 $x_1 = 0.9292$ $x_1 = 0.9248$ 13.63 13.72 0.9438 0.9537 335.28 19.37 19.52 0.9420 0.9564	339.07	25.31	25.48	0.9880	0.9964
354.15 47.98 48.25 0.9881 0.9968 362.54 66.72 67.03 0.9894 0.9970 368.78 84.30 84.65 0.9913 0.9972 373.25 99.07 99.49 0.9913 0.9973 $x_1 = 0.9684$ $x_1 = 0.9684$ $x_1 = 0.9684$ 325.26 12.97 13.10 0.9789 0.9889 34.56 20.22 20.37 0.9822 0.9898 340.04 25.84 26.10 0.9805 0.9903 349.33 38.53 38.87 0.9824 0.9910 355.44 49.40 49.87 0.9821 0.9914 362.84 66.02 66.57 0.9837 0.9923 373.88 99.14 99.93 0.9843 0.9926 $x_1 = 0.9292$ $x_1 = 0.9242$ $x_1 = 0.9438$ 0.9537 335.28 19.37 19.52 0.9420 0.9564 341.72 25.614 26.16 0.9423 0.5567	347.50	36.48	36.70	0.9884	0.9967
362.54 66.72 67.03 0.9894 0.9970 368.78 84.30 84.65 0.9913 0.9972 373.25 99.07 99.49 0.9913 0.9973 $x_1 = 0.9684$ 25.26 12.97 13.10 0.9789 0.9889 34.56 20.22 20.37 0.9822 0.9898 340.04 25.84 26.10 0.9805 0.9903 349.33 38.53 38.87 0.9824 0.9910 355.44 49.40 49.87 0.9821 0.9914 362.84 66.02 66.57 0.9837 0.9923 373.88 99.14 99.93 0.9843 0.9926 $x_1 = 0.9292$ $x_1 = 0.9292$ $x_1 = 0.9438$ 0.9537 335.28 19.37 19.52 0.9420 0.9564 241.79 25.614 26.616 0.9420 0.5567	354.15	47.98	48.25	0.9881	0.9968
368.78 84.30 84.65 0.9913 0.9972 373.25 99.07 99.49 0.9913 0.9973 $x_1 = 0.9684$ 325.26 12.97 13.10 0.9789 0.9889 34.56 20.22 20.37 0.9822 0.9888 340.04 25.84 26.10 0.9805 0.9903 349.33 38.53 38.87 0.9824 0.9910 355.44 49.40 49.87 0.9821 0.9914 362.84 66.02 66.57 0.9837 0.9923 373.88 99.14 99.93 0.9843 0.9926 $x_1 = 0.9292$ $x_1 = 0.9292$ $x_1 = 0.9438$ 0.9537 335.28 19.37 19.52 0.9420 0.9564 241.78 25.014 26.16 0.9420 0.9564	362.54	66.72	67.03	0.9894	0.9970
373.25 99.07 99.49 0.9913 0.9973 $x_1 = 0.9684$	368.78	84.30	84.65	0.9913	0.9972
$x_1 = 0.9684$ 325.26 12.97 13.10 0.9789 0.9889 334.56 20.22 20.37 0.9822 0.9898 340.04 25.84 26.10 0.9805 0.9903 349.33 38.53 38.87 0.9824 0.9910 355.44 49.40 49.87 0.9821 0.9914 362.84 66.02 66.57 0.9837 0.9923 373.88 99.14 99.93 0.9843 0.9926 $x_1 = 0.9292$ $x_1 = 0.9242$ $x_1 = 0.9438$ 0.9537 327.81 13.63 13.72 0.9438 0.9537 325.28 19.37 19.52 0.9420 0.9564	373.25	99.07	99.49	0.9913	0.9973
325.26 12.97 13.10 0.9789 0.9889 334.56 20.22 20.37 0.9822 0.9898 340.04 25.84 26.10 0.9805 0.9903 349.33 38.53 38.87 0.9824 0.9910 355.44 49.40 49.87 0.9821 0.9914 362.84 66.02 66.57 0.9837 0.9919 369.46 84.53 85.21 0.9836 0.9923 373.88 99.14 99.93 0.9843 0.9926 $x_1 = 0.9292$ $x_1 = 0.9292$ $x_1 = 0.9438$ 0.9537 335.28 19.37 19.52 0.9420 0.9564	$x_1 = 0.9684$				
334.56 20.22 20.37 0.9822 0.9898 340.04 25.84 26.10 0.9805 0.9903 349.33 38.53 38.87 0.9824 0.9910 355.44 49.40 49.87 0.9821 0.9914 362.84 66.02 66.57 0.9837 0.9923 373.88 99.14 99.93 0.9843 0.9926 $x_1 = 0.9292$ $x_1 = 0.9292$ $x_1 = 0.9438$ 0.9537 335.28 19.37 19.52 0.9420 0.9564	325.26	12.97	13.10	0.9789	0.9889
340.04 25.84 26.10 0.9805 0.9903 349.33 38.53 38.87 0.9824 0.9910 355.44 49.40 49.87 0.9821 0.9914 362.84 66.02 66.57 0.9837 0.9919 369.46 84.53 85.21 0.9836 0.9923 373.88 99.14 99.93 0.9843 0.9926 $x_1 = 0.9292$ $x_1 = 0.9292$ $x_1 = 0.9438$ 0.9537 327.81 13.63 13.72 0.9438 0.9537 335.28 19.37 19.52 0.9420 0.9564 241.79 256.01 26.16 0.9420 0.9567	334.56	20.22	20.37	0.9822	0.9898
349.33 38.53 38.87 0.9824 0.9910 355.44 49.40 49.87 0.9821 0.9914 362.84 66.02 66.57 0.9837 0.9913 369.46 84.53 85.21 0.9836 0.9923 373.88 99.14 99.93 0.9843 0.9926 $x_1 = 0.9292$ $x_1 = 0.9242$ 0.9438 0.9537 327.81 13.63 13.72 0.9438 0.9537 325.28 19.37 19.52 0.9420 0.9564 241.79 256.01 26.16 0.9420 0.9567	340.04	25.84	26.10	0.9805	0.9903
355.44 49.40 49.87 0.9821 0.9914 362.84 66.02 66.57 0.9837 0.9919 369.46 84.53 85.21 0.9836 0.9923 373.88 99.14 99.93 0.9843 0.9926 $x_1 = 0.9292$ $x_1 = 0.9292$ $x_1 = 0.9242$ $x_1 = 0.9438$ 0.9537 335.28 19.37 19.52 0.9420 0.9564 $x_{0.9577}$	349.33	38.53	38.87	0.9824	0.9910
362.84 66.02 66.57 0.9837 0.9919 369.46 84.53 85.21 0.9836 0.9923 373.88 99.14 99.93 0.9843 0.9926 $x_1 = 0.9292$ 327.81 13.63 13.72 0.9438 0.9537 335.28 19.37 19.52 0.9420 0.9564 241.79 25.614 26.616 0.9420 0.9564	355.44	49.40	49.87	0.9821	0.9914
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	362.84	66.02	66.57	0.9837	0.9919
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	369.46	84.53	85.21	0.9836	0.9923
$x_1 = 0.9292$ 327.81 13.63 13.72 0.9438 0.9537 335.28 19.37 19.52 0.9420 0.9564 241.78 25.01 26.16 0.0422 0.9564	373.88	99.14	99.93	0.9843	0.9926
327.81 13.63 13.72 0.9438 0.9537 335.28 19.37 19.52 0.9420 0.9564 241.79 25.01 26.16 0.0422 0.5565	$x_1 = 0.9292$				
335.28 19.37 19.52 0.9420 0.9564 241.79 25.01 26.10 0.0422 0.9565	327.81	13.63	13.72	0.9438	0.9537
241 70 25 01 20 10 00422 00505	335.28	19.37	19.52	0.9420	0.9564
341.78 25.91 26.16 0.9433 0.9585	341.78	25.91	26.16	0.9433	0.9585
349.88 36.66 37.01 0.9441 0.9610	349.88	36.66	37.01	0.9441	0.9610
356.81 48.60 49.07 0.9447 0.9629	356.81	48.60	49.07	0.9447	0.9629
363.92 64.01 64.72 0.9470 0.9648	363.92	64.01	64.72	0.9470	0.9648
371.26 84.05 85.01 0.9511 0.9665	371.26	84.05	85.01	0.9511	0.9665
375.7098.7099.700.95500.9675	375.70	98.70	99.70	0.9550	0.9675

^a Standard uncertainties *u* are u(T) = 0.02 K, u(x) = 0.0001, and u(P) = 0.04 kPa.

^a Standard uncertainties *u* are u(T) = 0.02 K, u(x) = 0.0001, and u(P) = 0.04 kPa.

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