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Density, Speed of Sound, Viscosity, Refractive Index, and Excess Volume of *N*-Methyl-2-pyrrolidone + Ethanol (or Water or Ethanolamine) from T = (293.15 to 323.15) K

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ABSTRACT: Experimental density, viscosity, speed of sound, and refractive index for binary mixtures of *N*-methyl-2-pyrrolidone with water, ethanol, and ethanolamine were measured over the entire composition range at different temperatures of (293.15 to 323.15) K. The effect of composition and temperature upon different physical properties has been analyzed. Excess volumes have been calculated, and their dependence on composition was mathematically represented by a Redlich–Kister-type equation.

INTRODUCTION

Cyclic amides have shown important characteristics such as high density, high boiling point, and high polarity solvents, which allow the usability at an industrial level. Also the high solubility in water allows the use of this kind of substance in a wide range of industrial and laboratory operations. More specifically, N-methyl-2-pyrrolidone (NMP) has shown selectivity with regard to unsaturated and aromatic hydrocarbons and sulfur gases. The low reactivity and high solubility are important characteristics that allow use of the NMP as an extraction agent in lubricant oil processing and the scrubbing treatment of natural gas.¹ On the other hand, the excellent thermal and chemical stability is another interesting characteristic for the use of NMP as solvent in different reaction systems. Also, the NMP could be used as cosolvent with water, hydrocarbons, alcohols, glycol ether, and ketones.² NMP in aqueous solution is used in the carbon dioxide capture process by means of physical absorption.³ The knowledge of different physical properties of systems that involve NMP is an important starting point to optimize different mass transfer operations.

The present work analyzes different physical properties in NMP-based binary liquid systems. More specifically, the liquid systems employed in this study include NMP + water (or ethanol or ethanolamine). Previous studies have analyzed certain physical properties for the NMP + water system at other temperatures.⁴ This kind of system has been chosen due to the important characteristics and uses as a solvent, in the separation process, and in acid gas capture.⁴

EXPERIMENTAL SECTION

N-Methyl-2-pyrrolidone (CAS number 872-50-4) with a mass purity of \geq 0.99 was supplied by Fluka. Ethanolamine (CAS number 141-43-5) and ethanol (CAS number 64-17-5) were supplied by Sigma-Aldrich with a mass purity of \geq 0.99 and \geq 0.995, respectively. Bidistilled water was used to prepare aqueous systems. All solutions were prepared by mass using an analytical balance (Kern 770) with a precision of 10^{-4} g. The uncertainty in the mole fraction for prepared sample solutions was found to be \pm 0.0005.

Density and Speed of Sound. The density and speed of sound of water and aqueous solutions of different solutes were measured with an Anton Paar DSA 5000 vibrating tube densimeter and sound analyzer. The uncertainty in the density and speed of sound measurements was $\pm 1.7 \cdot 10^{-4} \text{ g} \cdot \text{cm}^{-3}$ and $\pm 0.22 \text{ m} \cdot \text{s}^{-1}$, respectively. In general, each value came from an average of three measurements.

Also, the adiabatic compressibility, κ_s , was calculated from speed of sound values and density values using the Laplace equation

$$\kappa_s = \frac{1}{u^2 \cdot \rho} \tag{1}$$

where *u* is the speed of sound and ρ is the density of the solution. **Viscosity.** The kinematic viscosity (ν) was determined from the transit time of the liquid meniscus through different capillary

the transit time of the liquid meniscus through different capillary viscosimeters supplied by Schott, capillary nos. 0c, I, Ic, and Ia, using eq 2.

$$\nu = K \cdot (t - \theta) \tag{2}$$

where t is the efflux time; K is the characteristic constant of the capillary viscometer; and θ is a correction value to correct end effects. Both parameters were obtained from the capillaries supplier (Schott). An electronic stopwatch with an accuracy of \pm 0.01 s was used to measure efflux times. In the measurements, a Schott-Geräte AVS 350 Ubbelohde viscometer was used. Each measurement was repeated at least 5 times, and the uncertainty of this measurement is \pm 0.0017 mm² · s⁻¹. The dynamic viscosity (η) was obtained from the product of the kinematic viscosity (ν)

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and the corresponding density (ρ) of the mixture, in terms of eq 3 for each mixture composition.

$$\eta = \nu \cdot \rho \tag{3}$$

Refractive Index. Refractive index was determined using an Atago RX-5000 refractometer. Before measurements, the refractometer was calibrated using distilled—deionized water in accordance with the instrument instructions. The mixtures were directly injected from the stock solution stored at working temperature to avoid evaporation. The refractive index measurements were done after the liquid mixtures attained the constant temperature of the refractometer. This procedure was repeated at least three times, and the uncertainty of the measurement was $\pm 6.7 \cdot 10^{-5}$. The average of these readings was taken for the refractive index values.

RESULTS AND DISCUSSION

The systems analyzed in the present work are based on N-methyl-2-pyrrolidone (NMP), and the other components are water, ethanol, and ethanolamine (MEA). Tables 1 to 3 show the experimental data determined for density, speed of sound, viscosity, and refractive index for the system NMP + water (or ethanol, or ethanolamine). These tables allow us to analyze the influence of composition and temperature upon the physical property values.

In relation to the first of the physical properties (density), the behavior obtained for the system NMP + water is different than the corresponding one obtained for the other mixtures (with ethanol or MEA). The observed behavior for the systems with ethanol and MEA is a monotonic increase in the value of density when NMP concentration increases in the mixture. With regard to the NMP + water system, the behavior is different because at low NMP concentrations an increase in the value of density is observed, but a maximum value of this physical property at 0.3 mole fraction is reached. When the NMP concentration increases, a decrease in the density is produced but with a minor slope compared to the previous behavior. This behavior is in agreement with previous studies that use this system at different temperatures.⁴

The speed of sound shows different behavior for each system. For the NMP + ethanol system, a monotonic increase is observed when NMP concentration increases in the mixture. The opposite behavior is observed for the NMP + MEA system. In both cases, the temperature produces a decrease in the speed of sound value.

A more complex behavior is observed for the mixtures produced by NMP and water. In the last case, a behavior similar to that previously commented for density is observed (a maximum in the speed of sound value). A similar behavior has been found for other cyclic amide + water systems.^{4,5}

In relation with the influence of temperature upon speed of sound, a different and very characteristic behavior is observed. Depending on the mixture composition, a different type of influence of the temperature can be observed. So, for water-rich composition, an increase in the temperature produces an increase in the value of speed of sound too, whereas for the great part of mixture compositions the behavior is the opposite.

The adiabatic compressibility for these aqueous mixtures could be calculated using density and speed of sound experimental data. For the NMP + water system, the trend observed

Table 1. Density ρ , Speed of Sound <i>u</i> , Viscosity η , and
Refractive Index n_D for NMP (1) + Water (2) from T =
(293.15 to 323.15) K

T/K = 293.15 T/K = 303.15 T/K = 313.15 T/K = 323.15

ρ/g·c·m ⁻³	101	1,10 2,0110	1,10 000110	1,10 010110	1,10 020110		
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0.20001766.51732.01697.01661.60.30011751.61713.01674.31634.90.39941718.41679.11639.31599.40.49971683.71644.51604.61565.50.60011651.61612.71573.21534.10.69971623.81585.31546.81507.60.79821601.21562.61523.91485.40.89901581.01542.61504.11466.01.00001565.31526.51487.81450.2 η/mPa .s0.00000.99330.79740.65350.54650.10003.33162.37161.74951.34810.20005.90283.95762.74872.06490.30016.77394.36533.01012.21470.39945.72353.95882.88982.16110.49974.46943.31782.51011.95890.60013.56612.71032.15321.70650.69972.86532.27881.83421.48530.79822.39391.96951.64091.36110.89902.04051.70521.46001.24251.00001.33301.33191.33061.32900.10001.33301.33191.33631.34810.20001.45731.45331.44251.43870.49971.46481.45651.45241.45810.60011.46181.45761.4425 <t< td=""><td>0.1000</td><td>1713.9</td><td>1694.6</td><td>1674.0</td><td>1651.4</td></t<>	0.1000	1713.9	1694.6	1674.0	1651.4		
0.30011751.61713.01674.31634.90.39941718.41679.11639.31599.40.49971683.71644.51604.61565.50.60011651.61612.71573.21534.10.69971623.81585.31546.81507.60.79821601.21562.61523.91485.40.89901581.01542.61504.11466.01.00001565.31526.51487.81450.2 $\eta/mPa*s$ 0.00000.99330.79740.65350.54650.10003.33162.37161.74951.34810.20005.90283.95762.74872.06490.30016.77394.36533.01012.21470.39945.72353.95882.88982.16110.49974.46943.31782.51011.95890.60013.56612.71032.15321.70650.69972.86532.27881.83421.48530.79822.39391.96951.64091.36110.89902.04051.70521.46001.24251.00001.33301.33191.33061.32900.10001.33301.33191.33631.34810.20001.42211.41871.41651.41140.30011.44021.43631.43311.42830.39941.45071.44671.44251.43670.49971.46181.45761.4534 <t< td=""><td>0.2000</td><td>1766.5</td><td>1732.0</td><td>1697.0</td><td>1661.6</td></t<>	0.2000	1766.5	1732.0	1697.0	1661.6		
0.3994 1718.4 1679.1 1639.3 1599.4 0.4997 1683.7 1644.5 1604.6 1565.5 0.6001 1651.6 1612.7 1573.2 1534.1 0.6997 1623.8 1585.3 1546.8 1507.6 0.7982 1601.2 1562.6 1523.9 1485.4 0.8990 1581.0 1542.6 1504.1 1466.0 1.0000 1565.3 1526.5 1487.8 1450.2 $\eta/mPa \cdot s$ $\eta/mPa \cdot s$ 10000 0.9933 0.7974 0.6535 0.5465 0.1000 0.9933 0.7974 0.6535 0.5465 0.1000 0.9933 0.7974 0.6535 0.5465 0.1000 3.316 2.3716 1.7495 1.3481 0.2000 5.9028 3.9576 2.7487 2.0649 0.3001 6.7739 4.3653 3.0101 2.2147 0.3994 5.7235 3.9588 2.8898 2.1611 0.4997 4.4694 3.3178 2.5101 1.9589 0.6001 3.5661 2.7103 2.1532 1.7065 0.6997 2.8653 2.2788 1.8342 1.4853 0.7982 2.3939 1.9695 1.6409 1.3611 0.8990 1.3300 1.3319 1.3206 1.3290 0.1000 1.3307 1.3882 1.3863 1.3840 0.2000 1.4507 1.4467 1.4425 1.4458 0.3001	0.3001	1751.6	1713.0	1674.3	1634.9		
0.4997 1683.7 1644.5 1604.6 1565.5 0.6001 1651.6 1612.7 1573.2 1534.1 0.6997 1623.8 1585.3 1546.8 1507.6 0.7982 1601.2 1562.6 1523.9 1485.4 0.8990 1581.0 1542.6 1504.1 1466.0 1.0000 1565.3 1526.5 1487.8 1450.2 η/mPa · η/mPa · 1480.2 11000 0.9933 0.7974 0.6535 0.5465 0.1000 0.9933 0.7974 0.6535 0.5465 0.1000 3.3316 2.3716 1.7495 1.3481 0.2000 5.9028 3.9576 2.7487 2.0649 0.3001 6.7739 4.3653 3.0101 2.2147 0.3994 5.7235 3.9588 2.8898 2.1611 0.4997 4.4694 3.3178 2.5101 1.9589 0.6001 3.5661 2.7103 2.1532 1.7065 0.6997 2.8653 2.2788 1.8342 1.4853 0.7982 2.3939 1.9695 1.6409 1.3611 0.8990 2.0405 1.7052 1.4603 1.3290 0.1000 1.3300 1.3319 1.3306 1.3290 0.1000 1.3907 1.3882 1.3863 1.3840 0.2000 1.4507 1.4467 1.4425 1.4456 0.4001 1.4618 1.4576 1.4455 1.4524 0.79	0.3994	1718.4	1679.1	1639.3	1599.4		
0.6001 1651.6 1612.7 1573.2 1534.1 0.6997 1623.8 1585.3 1546.8 1507.6 0.7982 1601.2 1562.6 1523.9 1485.4 0.8990 1581.0 1542.6 1504.1 1466.0 1.0000 1565.3 1526.5 1487.8 1450.2 $\eta/mPars$ $\eta/mPars$ 1487.8 1450.2 0.0000 0.9933 0.7974 0.65335 0.5465 0.1000 3.3316 2.3716 1.7495 1.3481 0.2000 5.9028 3.9576 2.7487 2.0649 0.3001 6.7739 4.3653 3.0101 2.2147 0.3994 5.7235 3.9588 2.8898 2.1611 0.4997 4.4694 3.3178 2.5101 1.9589 0.6001 3.5661 2.7103 2.1532 1.7065 0.6997 2.8653 2.2788 1.8342 1.4853 0.7982 2.3939 1.9695 1.6409 1.3611 0.8990 2.0405 1.7052 1.4600 1.2425 1.0000 1.3330 1.3319 1.3306 1.3290 0.1000 1.3330 1.3319 1.3306 1.3290 0.1000 1.4507 1.4467 1.4425 1.4387 0.4497 1.4507 1.4467 1.4425 1.4387 0.3011 1.4402 1.4363 1.4331 1.4283 0.3994 1.4507 1.4467 1.4425	0.4997	1683.7	1644.5	1604.6	1565.5		
0.6997 1623.8 1585.3 1546.8 1507.6 0.7982 1601.2 1562.6 1523.9 1485.4 0.8990 1581.0 1542.6 1504.1 1466.0 1.0000 1565.3 1526.5 1487.8 1450.2 $\eta/mPa \cdot s$ $\eta/mPa \cdot s$ 0.0000 0.9933 0.7974 0.6535 0.5465 0.1000 3.3316 2.3716 1.7495 1.3481 0.2000 5.9028 3.9576 2.7487 2.0649 0.3001 6.7739 4.3653 3.0101 2.2147 0.3994 5.7235 3.9588 2.8898 2.1611 0.4997 4.4694 3.3178 2.5101 1.9589 0.6001 3.5661 2.7103 2.1532 1.7065 0.6997 2.8653 2.2788 1.8342 1.4853 0.7982 2.3939 1.9695 1.6409 1.3611 0.8990 2.0405 1.7052 1.4600 1.2425 1.0000 1.3330 1.3319 1.3306 1.3290 0.1000 1.3907 1.3882 1.3863 1.3840 0.2000 1.4221 1.4187 1.4165 1.4114 0.3001 1.4407 1.4467 1.4425 1.4387 0.4997 1.4573 1.4533 1.4492 1.4450 0.6001 1.4618 1.4608 1.4544 1.4546 0.4997 1.4673 1.4628 1.4584 1.4546 0.4997 <t< td=""><td>0.6001</td><td>1651.6</td><td>1612.7</td><td>1573.2</td><td>1534.1</td></t<>	0.6001	1651.6	1612.7	1573.2	1534.1		
0.7982 1601.2 1562.6 1523.9 1485.4 0.8990 1581.0 1542.6 1504.1 1466.0 1.0000 1565.3 1526.5 1487.8 1450.2 $\eta/mPa \cdot s$ $\eta/mPa \cdot s$ $\eta/mPa \cdot s$ 0.0000 0.9933 0.7974 0.6535 0.5465 0.1000 3.3316 2.3716 1.7495 1.3481 0.2000 5.9028 3.9576 2.7487 2.0649 0.3001 6.7739 4.3653 3.0101 2.2147 0.3994 5.7235 3.9588 2.8898 2.1611 0.4997 4.4694 3.3178 2.5101 1.9589 0.6001 3.5661 2.7103 2.1532 1.7065 0.6997 2.8653 2.2788 1.8342 1.4853 0.7982 2.3939 1.9695 1.6409 1.3611 0.8990 2.0405 1.7052 1.4600 1.2425 1.0000 1.3330 1.3319 1.3306 1.3290 0.1000 1.3907 1.3882 1.3863 1.3840 0.2000 1.4221 1.4187 1.4165 1.4114 0.3001 1.4402 1.4363 1.4331 1.4283 0.3994 1.4507 1.4467 1.4425 1.4387 0.4997 1.4573 1.4533 1.4492 1.4450 0.4997 1.4673 1.4528 1.4534 1.4495 0.6001 1.4618 1.4628 1.4584 1.4546 <tr< td=""><td>0.6997</td><td>1623.8</td><td>1585.3</td><td>1546.8</td><td>1507.6</td></tr<>	0.6997	1623.8	1585.3	1546.8	1507.6		
0.8990 1581.0 1542.6 1504.1 1466.0 1.0000 1565.3 1526.5 1487.8 1450.2 $\eta/mPa \cdot s$ $\eta/mPa \cdot s$ $\eta/mPa \cdot s$ $\eta/mPa \cdot s$ 0.0000 0.9933 0.7974 0.6535 0.5465 0.1000 3.3316 2.3716 1.7495 1.3481 0.2000 5.9028 3.9576 2.7487 2.0649 0.3001 6.7739 4.3653 3.0101 2.2147 0.3994 5.7235 3.9588 2.8898 2.1611 0.4997 4.4694 3.3178 2.5101 1.9589 0.6001 3.5661 2.7103 2.1532 1.7065 0.6997 2.8653 2.2788 1.8342 1.4853 0.7982 2.3939 1.9695 1.6409 1.3611 0.8990 2.0405 1.7052 1.4600 1.2425 1.0000 1.3330 1.3319 1.3306 1.3290 0.1000 1.3330 1.3319 1.3306 1.3290 0.1000 1.3907 1.4863 1.4331 1.4283 0.3994 1.4507 1.4467 1.4425 1.4387 0.4997 1.4573 1.4533 1.4492 1.4504 0.6001 1.4618 1.4576 1.4544 1.4546 0.5997 1.4648 1.4608 1.4564 1.4546 0.6997 1.4648 1.4608 1.4564 1.4546 0.6001 1.4618 1.4576 1.4534 <	0.7982	1601.2	1562.6	1523.9	1485.4		
1.0001565.31526.51487.81450.2 $\eta/mPa \cdot s$ 0.00000.99330.79740.65350.54650.10003.33162.37161.74951.34810.20005.90283.95762.74872.06490.30016.77394.36533.01012.21470.39945.72353.95882.88982.16110.49974.46943.31782.51011.95890.60013.56612.71032.15321.70650.69972.86532.27881.83421.48530.79822.39391.96951.64091.36110.89902.04051.70521.46001.24251.00001.33001.33191.33061.32900.10001.33071.38821.38631.38400.20001.42211.41871.41651.41140.30011.44021.43631.43311.42830.39941.45071.44671.44251.43870.49971.45731.45331.44921.45060.60011.46181.45761.45341.44540.69971.46481.46081.45651.45240.69971.46481.46081.45641.45640.69971.46491.46491.45641.45640.69971.46481.46081.45641.45640.69971.46481.46081.45641.45640.69971.46481.46281.45641.4564 <td>0.8990</td> <td>1581.0</td> <td>1542.6</td> <td>1504.1</td> <td>1466.0</td>	0.8990	1581.0	1542.6	1504.1	1466.0		
η/mPa·s0.00000.99330.79740.65350.54650.10003.33162.37161.74951.34810.20005.90283.95762.74872.06490.30016.77394.36533.01012.21470.39945.72353.95882.88982.16110.49974.46943.31782.51011.95890.60013.56612.71032.15321.70650.69972.86532.27881.83421.48530.79822.39391.96951.64091.36110.89902.04051.70521.46001.24251.00001.85811.55311.32391.14610.10001.33071.33821.38631.32400.20001.42211.41871.41651.41140.30111.44021.43631.43311.42830.39941.45071.44671.44251.43870.49971.45731.45331.44921.45050.60011.46181.45761.45341.44500.60011.46181.45761.45341.45460.69971.46481.46081.45651.45240.69971.46481.46081.45651.45240.69971.46481.46081.45651.45240.69971.46481.46081.45651.45240.69971.46481.46081.45651.45240.69971.46481.46081.45641.4564<	1.0000	1565.3	1526.5	1487.8	1450.2		
0.0000 0.9933 0.7974 0.6535 0.5465 0.1000 3.3316 2.3716 1.7495 1.3481 0.2000 5.9028 3.9576 2.7487 2.0649 0.3001 6.7739 4.3653 3.0101 2.2147 0.3994 5.7235 3.9588 2.8898 2.1611 0.4997 4.4694 3.3178 2.5101 1.9589 0.6001 3.5661 2.7103 2.1532 1.7065 0.6997 2.8653 2.2788 1.8342 1.4853 0.7982 2.3939 1.9695 1.6409 1.3611 0.8990 2.0405 1.7052 1.4600 1.2425 1.0000 1.3330 1.3319 1.3306 1.3290 0.1000 1.3330 1.3319 1.3306 1.3290 0.1000 1.3907 1.3882 1.3863 1.3840 0.2000 1.4221 1.4187 1.4165 1.4114 0.3001 1.4402 1.4363 1.4331 1.4283 0.3994 1.4507 1.4467 1.4425 1.4387 0.4997 1.4573 1.4533 1.4492 1.4450 0.6001 1.4648 1.4508 1.4524 1.4524 0.7982 1.4673 1.4628 1.4584 1.4546 0.8990 1.4690 1.4649 1.4604 1.4564 1.0000 1.4706 1.4664 1.4621 1.4576			n/m				
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0.2000 5.9028 3.9576 2.7487 2.0649 0.3001 6.7739 4.3653 3.0101 2.2147 0.3994 5.7235 3.9588 2.8898 2.1611 0.4997 4.4694 3.3178 2.5101 1.9589 0.6001 3.5661 2.7103 2.1532 1.7065 0.6997 2.8653 2.2788 1.8342 1.4853 0.7982 2.3939 1.9695 1.6409 1.3611 0.8990 2.0405 1.7052 1.4600 1.2425 1.0000 1.8581 1.5531 1.3239 1.1461 0.0000 1.3330 1.3319 1.3306 1.3290 0.1000 1.3907 1.3882 1.3863 1.3840 0.2000 1.4221 1.4187 1.4165 1.4114 0.3001 1.4402 1.4363 1.4331 1.4283 0.3994 1.4507 1.4467 1.4425 1.4387 0.4997 1.4573 1.4533 1.4492 1.4506 0.6001 1.4618 1.4508 1.4524 1.4524 0.7982 1.4673 1.4628 1.4584 1.4546 0.8990 1.4690 1.4649 1.4604 1.4564 1.0000 1.4706 1.4664 1.4621 1.4576	0.1000	3.3316	2.3716	1.7495	1.3481		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0.2000	5.9028	3.9576	2.7487	2.0649		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0.3001	6.7739	4.3653	3.0101	2.2147		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0.3994	5.7235	3.9588 2.8898		2.1611		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0.4997	4.4694	3.3178	2.5101	1.9589		
0.6997 2.8653 2.2788 1.8342 1.4853 0.7982 2.3939 1.9695 1.6409 1.3611 0.8990 2.0405 1.7052 1.4600 1.2425 1.0000 1.8581 1.5531 1.3239 1.1461 np/- np/- 0.0000 1.3330 1.3319 1.3306 1.3290 0.1000 1.3907 1.3882 1.3863 1.3840 0.2000 1.4221 1.4187 1.4165 1.4114 0.3001 1.4402 1.4363 1.4331 1.4283 0.3994 1.4507 1.4467 1.4425 1.4387 0.4997 1.4573 1.4533 1.4492 1.4450 0.6001 1.4618 1.4576 1.4534 1.4450 0.6997 1.4648 1.4608 1.4565 1.4524 0.7982 1.4673 1.4628 1.4584 1.4546 0.8990 1.4690 1.4649 1.4604 1.4564	0.6001	3.5661	2.7103	2.1532	1.7065		
0.7982 2.3939 1.9695 1.6409 1.3611 0.8990 2.0405 1.7052 1.4600 1.2425 1.0000 1.8581 1.5531 1.3239 1.1461 m _D /- 0.0000 1.3330 1.3319 1.3306 1.3290 0.1000 1.3907 1.3882 1.3863 1.3840 0.2000 1.4221 1.4187 1.4165 1.4114 0.3001 1.4402 1.4363 1.4331 1.4283 0.3994 1.4507 1.4467 1.4425 1.4387 0.4997 1.4573 1.4533 1.4492 1.4450 0.6001 1.4618 1.4576 1.4534 1.4455 0.6997 1.4648 1.4608 1.4565 1.4524 0.7982 1.4673 1.4628 1.4584 1.4546 0.8990 1.4690 1.4649 1.4604 1.4564 1.0000 1.4706 1.4664 1.4621 1.4576	0.6997	2.8653	2.2788	1.8342	1.4853		
0.8990 2.0405 1.7052 1.4600 1.2425 1.0000 1.8581 1.5531 1.3239 1.1461 n _D /- n _D /- 1.0000 1.3330 1.3319 1.3306 1.3290 0.0000 1.3330 1.3319 1.3306 1.3290 0.1000 1.3907 1.3882 1.3863 1.3840 0.2000 1.4221 1.4187 1.4165 1.4114 0.3001 1.4402 1.4363 1.4331 1.4283 0.3994 1.4507 1.4467 1.4425 1.4387 0.4997 1.4573 1.4533 1.4492 1.4450 0.6001 1.4618 1.4576 1.4534 1.4452 0.6997 1.4648 1.4608 1.4565 1.4524 0.7982 1.4673 1.4628 1.4584 1.4546 0.8990 1.4690 1.4649 1.4604 1.4564 1.0000 1.4706 1.4664 1.4621 1.4576	0.7982	2.3939	1.9695	1.6409	1.3611		
1.0000 1.8581 1.5531 1.3239 1.1461 $n_{\rm D}/ 0.0000$ 1.3330 1.3319 1.3306 1.3290 0.1000 1.3907 1.3882 1.3863 1.3840 0.2000 1.4221 1.4187 1.4165 1.4114 0.3001 1.4402 1.4363 1.4331 1.4283 0.3994 1.4507 1.4467 1.4425 1.4387 0.4997 1.4573 1.4533 1.4492 1.4450 0.6001 1.4618 1.4576 1.4534 1.4595 0.6997 1.4648 1.4608 1.4565 1.4524 0.7982 1.4673 1.4628 1.4584 1.4546 0.8990 1.4690 1.4664 1.4604 1.4564 1.0000 1.4706 1.4664 1.4621 1.4576	0.8990	2.0405	1.7052	1.4600	1.2425		
n _D /- 0.0000 1.3330 1.3319 1.3306 1.3290 0.1000 1.3907 1.3882 1.3863 1.3840 0.2000 1.4221 1.4187 1.4165 1.4114 0.3001 1.4402 1.4363 1.4331 1.4283 0.3994 1.4507 1.4467 1.4425 1.4387 0.4997 1.4573 1.4533 1.4492 1.4450 0.6001 1.4618 1.4576 1.4534 1.4495 0.6997 1.4648 1.4608 1.4565 1.4524 0.7982 1.4673 1.4628 1.4584 1.4546 0.8990 1.4690 1.4649 1.4604 1.4564 1.0000 1.4706 1.4664 1.4621 1.4576	1.0000	1.8581	1.5531	1.3239	1.1461		
0.0000 1.3330 1.3319 1.3306 1.3290 0.1000 1.3907 1.3882 1.3863 1.3840 0.2000 1.4221 1.4187 1.4165 1.4114 0.3001 1.4402 1.4363 1.4331 1.4283 0.3994 1.4507 1.4467 1.4425 1.4387 0.4997 1.4573 1.4533 1.4492 1.4450 0.6001 1.4618 1.4576 1.4534 1.4495 0.6997 1.4648 1.4608 1.4565 1.4524 0.7982 1.4673 1.4628 1.4584 1.4546 0.8990 1.4690 1.4649 1.4604 1.4564 1.0000 1.4706 1.4664 1.4621 1.4576			$n_{\rm D}$	′			
0.10001.39071.38821.38631.38400.20001.42211.41871.41651.41140.30011.44021.43631.43311.42830.39941.45071.44671.44251.43870.49971.45731.45331.44921.44500.60011.46181.45761.45341.44950.69971.46481.46081.45651.45240.79821.46731.46281.45841.45460.89901.46901.46491.46041.45641.00001.47061.46641.46211.4576	0.0000	1.3330	1.3319	1.3306	1.3290		
0.20001.42211.41871.41651.41140.30011.44021.43631.43311.42830.39941.45071.44671.44251.43870.49971.45731.45331.44921.44500.60011.46181.45761.45341.44950.69971.46481.46081.45651.45240.79821.46731.46281.45841.45460.89901.46901.46491.46041.45641.00001.47061.46641.46211.4576	0.1000	1.3907	1.3882	1.3863	1.3840		
0.30011.44021.43631.43311.42830.39941.45071.44671.44251.43870.49971.45731.45331.44921.44500.60011.46181.45761.45341.44950.69971.46481.46081.45651.45240.79821.46731.46281.45841.45660.89901.46901.46491.46041.45641.00001.47061.46641.46211.4576	0.2000	1.4221	1.4187	1.4165	1.4114		
0.39941.45071.44671.44251.43870.49971.45731.45331.44921.44500.60011.46181.45761.45341.44950.69971.46481.46081.45651.45240.79821.46731.46281.45841.45660.89901.46901.46491.46041.45641.00001.47061.46641.46211.4576	0.3001	1.4402	1.4363	1.4331	1.4283		
0.49971.45731.45331.44921.44500.60011.46181.45761.45341.44950.69971.46481.46081.45651.45240.79821.46731.46281.45841.45460.89901.46901.46491.46041.45641.00001.47061.46641.46211.4576	0.3994	1.4507	1.4467	1.4425	1.4387		
0.60011.46181.45761.45341.44950.69971.46481.46081.45651.45240.79821.46731.46281.45841.45460.89901.46901.46491.46041.45641.00001.47061.46641.46211.4576	0.4997	1.4573	1.4533	1.4492	1.4450		
0.69971.46481.46081.45651.45240.79821.46731.46281.45841.45460.89901.46901.46491.46041.45641.00001.47061.46641.46211.4576	0.6001	1.4618	1.4576	1.4534	1.4495		
0.79821.46731.46281.45841.45460.89901.46901.46491.46041.45641.00001.47061.46641.46211.4576	0.6997	1.4648	1.4608	1.4565	1.4524		
0.89901.46901.46491.46041.45641.00001.47061.46641.46211.4576	0.7982	1.4673	1.4628	1.4584	1.4546		
1.0000 1.4706 1.4664 1.4621 1.4576	0.8990	1.4690	1.4649	1.4604	1.4564		
	1.0000	1.4706	1.4664	1.4621	1.4576		

(see Figure 1), as suggested by different authors,⁶ holds over this very narrow interval of concentration where the compressibility

Table 2. Density ρ , Speed of Sound *u*, Viscosity η , and Refractive Index $n_{\rm D}$ for NMP (1) + Ethanol (2) from T = (293.15 to 323.15) K

x_1	T/K = 293.15	T/K = 303.15	T/K = 313.15	T/K = 323.15		x_1	<i>T</i> /K = 293.1	
$ ho/g\cdot cm^{-3}$								
0.0000	0.78954	0.78094	0.77220	0.76329		0.0000	1.01665	
0.0992	0.82976	0.82098	0.81209	0.80305		0.1000	1.01780	
0.1985	0.86484	0.85596	0.84698	0.83787		0.2090	1.01927	
0.2973	0.89436	0.88542	0.87639	0.86727		0.3000	1.02059	
0.3963	0.92089	0.91191	0.90287	0.89376		0.4000	1.02206	
0.4948	0.94441	0.93543	0.92639	0.91730		0.5000	1.02346	
0.5932	0.96500	0.95597	0.94690	0.93779		0.5960	1.02491	
0.6912	0.98396	0.97494	0.96589	0.95680		0.7000	1.02657	
0.7897	1.00086	0.99185	0.98282	0.97377		0.8000	1.02840	
0.8875	1.01679	1.00780	0.99880	0.98978		0.9070	1.03050	
1.0000	1.03287	1.02395	1.01503	1.00610		1.0000	1.03287	
		u/m	$\cdot s^{-1}$					
0.0000	1162.3	1128.9	1095.4	1071.9		0.0000	1735.9	
0.0992	1226.1	1191.3	1157.0	1122.7		0.1000	1701.7	
0.1985	1284.2	1248.9	1214.7	1179.6		0.2090	1673.2	
0.2973	1331.7	1295.9	1260.3	1224.9		0.3000	1649.8	
0.3963	1375.7	1338.9	1302.5	1266.6		0.4000	1630.1	
0.4948	1414.8	1377.6	1341.0	1304.6		0.5000	1613.2	
0.5932	1448.9	1411.4	1374.2	1337.3		0.5960	1599.1	
0.6912	1480.7	1442.9	1405.3	1368.3		0.7000	1587.7	
0.7897	1509.4	1471.4	1433.5	1396.1		0.8000	1578.6	
0.8875	1537.0	1498.5	1460.3	1422.8		0.9070	1571.4	
1.0000	1565.3	1526.5	1487.8	1450.2		1.0000	1565.3	
		$\eta/{ m m}$	Pa•s					
0.0000	1.1160	0.9502	0.7899	0.6304		0.0000	24.614	
0.0992	1.1390	0.9625	0.8030	0.6570		0.1000	17.432	
0.1985	1.1730	0.9860	0.8276	0.6910		0.2090	11.992	
0.2973	1.2180	1.0333	0.8670	0.7310		0.3000	8.496	
0.3963	1.2750	1.0901	0.9234	0.7820		0.4000	6.211	
0.4948	1.3540	1.1500	0.9800	0.8330		0.5000	4.761	
0.5932	1.4361	1.2205	1.0394	0.8950		0.5960	3.622	
0.6912	1.5240	1.2920	1.1094	0.9520		0.7000	2.883	
0.7897	1.6260	1.3710	1.1730	1.0107		0.8000	2.401	
0.8875	1.7264	1.4500	1.2373	1.0710		0.9070	2.089	
1.0000	1.8581	1.5531	1.3239	1.1461		1.0000	1.858	
		n _D	/-					
0.0000	1.3616	1.3574	1.3532	1.3493		0.0000	1.4525	
0.0992	1.3797	1.3760	1.3717	1.3677		0.1000	1.4546	
0.1985	1.3953	1.3916	1.3872	1.3837		0.2090	1.4567	
0.2973	1.4089	1.4051	1.4011	1.3974		0.3000	1.4587	
0.3963	1.4209	1.4168	1.4127	1.4089		0.4000	1.4604	
0.4948	1.4313	1.4273	1.4234	1.4195		0.5000	1.4620	
0.5932	1.4406	1.4366	1.4329	1.4289		0.5960	1.4634	
0.6912	1.4490	1.4449	1.4411	1.4372		0.7000	1.4648	
0.7897	1.4563	1.4523	1.4484	1.4443		0.8000	1.4663	
0.8875	1.4633	1.4593	1.4552	1.4512		0.9070	1.4677	
1.0000	1.4706	1.4664	1.4621	1.4576		1.0000	1.4691	

isotherms cross one another and point to formation of a clathrate-like structure with the stoichiometry corresponding to

the concentration at which the crossing occurs. The structure is relatively temperature resistant, and consequently, the rigidity of

Table 3. Density ρ , Speed of Sound *u*, Viscosity η , and Refractive Index n_D for NMP (1) + MEA (2) from T = (293.15 to 323.15) K

.15	x_1	T/K = 293.15	T/K = 303.15	T/K = 313.15	T/K = 323.15	
			$\rho/g \cdot cm^{-3}$			
29	0.0000	1.01665	1.00874	1.00077	0.99275	
05	0.1000	1.01780	1.00980	1.00160	0.99331	
87	0.2090	1.01927	1.01100	1.00238	0.99400	
27	0.3000	1.02059	1.01210	1.00320	0.99464	
76	0.4000	1.02206	1.01350	1.00420	0.99560	
30	0.5000	1.02346	1.01490	1.00550	0.99670	
79	0.5960	1.02491	1.01630	1.00690	0.99814	
80	0.7000	1.02657	1.01800	1.00869	0.99974	
77	0.8000	1.02840	1.01970	1.01068	1.00173	
78	0.9070	1.03050	1.02172	1.01279	1.00386	
10	1.0000	1.03287	1.02395	1.01503	1.00610	
			<i>u/</i> m	·s ⁻¹		
	0.0000	1735.9	1703.7	1671.2	1638.8	
	0.1000	1701.7	1668.1	1634.1	1600.2	
	0.2090	1673.2	1638.4	1603.1	1568.9	
	0.3000	1649.8	1613.8	1578.6	1544.4	
	0.4000	1630.1	1593.6	1557 3	1521.5	
	0.5000	1613.2	1576.0	1538.9	1502.0	
	0.5960	1599.1	1561.2	1523.9	1486.7	
	0.7000	1587.7	1549.8	1512.1	1474.5	
	0.8000	1578.6	1540.5	1502.6	1464.8	
	0.9070	1571.4	1532.7	1494.4	1456.8	
	1,0000	1565.3	1526.5	1487.8	1450.2	
	10000	100010	102010	Date	110012	
4	0.0000	24 (14	η/ III 14.056	Pa-8	6 002	
+	0.0000	24.014	14.950	9.839	5.296	
	0.1000	17.432	10.928	7.429	5.380	
0	0.2090	11.992	7.985	5.559	4.159	
0	0.3000	8.496	5.834	4.278	3.289	
	0.4000	6.211	4.433	3.358	2.625	
	0.5000	4./01	3.752	2.097	2.1/3	
0	0.5960	3.622	2.822	2.245	1.883	
-	0.7000	2.883	2.296	1.866	1.5/4	
/	0.8000	2.401	1.950	1.620	1.356	
1	0.9070	2.089	1.725	1.462	1.240	
1	1.0000	1.858	1.553	1.324	1.146	
	0.0000	1.4505	n _D ,	/-	1 4 4 5	
3	0.0000	1.4525	1.4488	1.4449	1.4417	
7	0.1000	1.4546	1.4507	1.4467	1.4434	
/	0.2090	1.4567	1.452/	1.4486	1.4451	
+	0.3000	1.4587	1.4546	1.4505	1.4466	
7	0.4000	1.4604	1.4563	1.4520	1.4479	
>	0.5000	1.4620	1.4578	1.4534	1.4493	
9	0.5960	1.4634	1.4592	1.4548	1.4506	
2	0.7000	1.4648	1.4605	1.4561	1.4519	
5	0.8000	1.4663	1.4619	1.4575	1.4533	
2	0.9070	1.4677	1.4632	1.4588	1.4545	
D	1.0000	1.4691	1.4647	1.4603	1.4559	



Figure 1. Influence of composition and temperature upon adiabatic compressibility κ_s for systems used in the present work. \bigcirc , T = 293.15 K; \bigcirc , T = 303.15 K; \square , T = 313.15 K; \blacksquare , T = 323.15 K.



Figure 2. Influence of composition upon dynamic viscosity η for systems used in the present work. \bigcirc , T = 293.15 K; \bigcirc , T = 303.15 K; \square , T = 313.15 K; \blacksquare T = 323.15 K; \bigstar , T = 298.15 K.⁴

the mixture is almost independent of temperature. On the basis of experimental data, this structure for water is created at molar fraction near 0.05 (a proportion of approximately 19:1 molecules), similar to the system formed by 1-ethyl-2-pyrrolidone.⁵ It is possible to consider that the change observed for the structure of this kind of mixture produces the behavior observed for the different properties commented on previously, as well as the important deviations obtained for each of them. For the other systems NMP + ethanol (or MEA) the behavior is very different from that previously commented on for the NMP + water system. For the NMP + ethanol system a monotonic increase in the value of adiabatic compressibility is observed when ethanol concentration increases. The opposite behavior is observed of the NMP + MEA system. In the nonaqueous systems, the characteristic behavior previously commented on for the NMP + water system in relation with the influence of temperature is not observed.

In relation with the value of viscosity for the systems analyzed in the present study, the behaviors are shown in Figure 2 for the different systems and for all the temperatures employed. As in the case of the physical properties analyzed previously, the most complex behavior was found for the NMP + water system. In the case of NMP + ethanol (or MEA) monotonic behaviors were obtained. For NMP + ethanol, a decrease in viscosity is observed when ethanol concentration increases in the mixture, and the opposite behavior is observed when MEA is used in the experimental system. This fact is due to that pure MEA takes a viscosity value higher in comparison with NMP. In the case of the NMP + water system, a behavior similar to that for the previously commented effect of composition upon density and speed of sound is observed. A maximum is reached when one of the components is added to the system. The viscosity maximum is obtained near 0.3 mol fraction (similar to the other physical properties). This behavior is in agreement with previous studies,⁴ and bibliographic data have been included in Figure 2. This behavior shows the important interactions between the molecules present in the experimental system.

In relation with the influence of NMP concentration in the systems analyzed in the present study upon the refractive index value, a similar behavior for all mixtures was obtained. An increase in the value of refractive index is observed when NMP composition increases. Also an increase in temperature produces in all cases a decrease in the value of this physical property.

To estimate the nonideality of these systems, the excess molar volume was calculated, using eq 4.

$$V^{\rm E} = \sum_{i=1}^{3} x_i \cdot M_i \cdot (\rho^{-1} - \rho_i^{-1})$$
(4)

where x_i , M_i , and ρ_i are the molar fractions, molecular weights, and densities of pure components, respectively.



Figure 3. Excess volume and Redlich—Kister (solid lines) fit for systems used in the present work. \bigcirc , T = 293.15 K; \bigcirc , T = 303.15 K; \square , T = 313.15 K; \blacksquare , T = 323.15 K; \blacktriangle , T = 298.15 K (ref 4).

Table 4. Fit Parameters Corresponding to the Redlich– Kister Equation for Excess Volume V^{E} from T = (293.15 to 323.15) K

parameter	T/K = 293.15	T/K = 303.15	T/K = 313.15	T/K = 323.15				
NMP (1) + Water (2)								
A_0	-8.334	-7.793	-7.185	-6.575				
A_1	30.840	28.899	26.566	24.203				
A_2	-56.050	-53.196	-49.536	-45.692				
A_3	27.773	26.827	25.325	23.626				
σ	0.02	0.02	0.02	0.02				
NMP (1) + Ethanol (2)								
A_0	-1.780	-1.781	-1.768	-1.805				
A_1	2.970	3.270	3.502	3.925				
A_2	-2.684	-3.435	-4.129	-5.107				
A_3	-1.290	-0.836	-0.390	0.190				
σ	σ 0.01 0.01		0.01	0.01				
NMP $(1) + MEA (2)$								
A_0	1.768	2.288	2.340	2.143				
A_1	-0.577	-3.951	-4.751	-3.008				
A_2	-2.057	4.159	7.961	4.835				
A_3	1.773	-1.633	-4.739	-3.044				
σ	0.05	0.05	0.07	0.07				

The behaviors obtained for excess volume for NMP + water (see Figure 3) are in agreement with previous studies for other alkyl-pyrrolidones^{4,5,7} in water. Also, aqueous solutions of pyrrolidine have shown a similar behavior in relation with the sign and magnitude of the NMP + water system.⁸ Also, negative deviations were found for the system NMP + ethanol, but the opposite behavior was obtained when MEA was employed. The effect caused by ethanol is in agreement with the conclusions reached in a previous work⁹ that analyze the influence of alcohol chain length upon the excess volume sign. Positive values for this parameter are reached when alcohol chain length increases.

The excess volume values shown in Figure 3 indicate that the degree of hydrogen-bonded complex formation between NMP and the other substances employed in the mixtures decreases in the manner: water > ethanol > MEA. The breaking of hydrogen bonds leads to liquid structure breaking and positive excess

volume, while hydrogen-bond formation leads to negative excess volume.

The values calculated for excess molar volumes were fitted using the Redlich—Kister equation (eq 5). The results obtained for fitting parameters are shown in Table 4 for the systems studied in this work.

$$\Delta Y = x_1 \cdot x_2 \cdot \sum_{j=1}^{4} q_j \cdot x_2^{(j-1)/2}$$
 (5)

The Redlich—Kister equation fits satisfactorily the excess molar volume calculated from experimental data in the present work.

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