Binary System of Polyethylene Glycol 200 (1) + 3-Dimethylamino-1-propylamine (2) for CO₂ Absorption: Thermophysical Properties and Spectroscopic Study

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ABSTRACT: As the concentration of CO₂ in the atmosphere keeps increasing, the development of a highly efficient CO₂ absorbent is highly desired. In this work, a binary mixture system of polyethylene glycol 200 (PEG 200) (1) + 3-dimethylamino-propylamine (DMAPA) (2) was used for CO₂ absorption. Considering the importance of thermophysical properties to binary solutions, the densities and viscosities of the PEG 200 (1) + DMAPA (2) mixture were measured at T = (298.15, 303.15, 308.15, 313.15, and 318.15) K and atmospheric pressure over the entire composition range. Based on the density and viscosity data, the excess properties and viscous flow thermodynamic parameters were calculated, respectively. To obtain the coefficients and to estimate the standard deviations between the experimental and calculated quantities, the excess molar volume (Vₐₑₓ), the viscosity deviation (Δη), and the excess Gibbs free energies of activation for viscous flow (ΔG*vis) were fitted to the Redlich–Kister equation. Furthermore, based on the results of UV–vis, FTIR, and ¹H NMR spectra, the intermolecular interaction of PEG 200 and DMAPA was discussed. Particularly, a strong intermolecular bonding is formed when the molar ratio of PEG 200 to DMAPA is about 1:2 because of the excess molar volume (Vₐₑₓ). On that account, a mixture of PEG 200 and DMAPA in a ratio of 1:2 was used for studying CO₂ absorption, and a CO₂ absorption of about 0.19 g per gram of absorbent was achieved at room temperature and atmosphere.

1. INTRODUCTION

The extensive use of fossil fuels and the rapid increase of CO₂ content in the atmosphere have caused the greenhouse effect and the negative consequences of global climate changes, and this has attracted widespread attention.12 Experts generally deem CO₂ capture and utilization (CCU) to be one of the most effective methods to reduce global CO₂ emissions.3,4 Various methods have been proposed to capture CO₂, such as solid adsorption,5,6 membrane separation,7,8 and organic solvent scrubbing.9–11 The organic solvent scrubbing method involves either physical or chemical solvents according to the strength of the solvent–solvent interaction. Representative amine scrubbing is the most commonly used commercial technology for CO₂ capture through chemical absorption.12,13 However, the loss of amine by volatilization easily causes secondary pollution in the process of CO₂ removal. Especially, there are also some shortcomings, such as equipment corrosion, relatively high cost, and serious loss of efficiency. To improve the process of amine absorption of CO₂, the selection of a suitable solvent is important. Thus, it is the focus of this study to find an organic solvent that can reduce the volatilization of amine, has better CO₂ uptake, and causes less environmental damage. Previous studies4–17 have shown that addition of certain organic solvents, such as alcohol and ethylene glycol derivatives, to combine with amines, has dramatically reduced the volatilization of amines, and the resulting amine–alcohol binary system showed superior CO₂ absorption capacity. Polyethylene glycol 200 (PEG 200) has been widely studied for CO₂ absorption because it has many good properties.5,6,15 Especially, it has reasonable characteristics to be labeled as a green solvent such as being nonvolatile, noncombustible, nontoxic to humans, animals, and aquatic organisms, and can be biodegraded by bacteria in soil and sewage. These properties make it suitable for replacing the previously used organic auxiliary solvent of alcohol for CO₂ capture.

In the course of our study on the capture of CO₂,20 we found that 3-dimethylamino-1-propylamine (DMAPA) can efficiently absorb CO₂ through intramolecular proton transfer. However, the problem of high volatility of DMAPA is still difficult to solve. Inspired by CO₂ organic binding solutions...
Herein, the present work mainly focuses on investigating the CO$_2$ uptake of the PEG 200 ($\text{1}$) + DMAPA ($\text{2}$) system includes the following three steps: (1) determining the density, viscosity, and excess properties of the PEG 200 (1) + DMAPA (2) mixture, (2) obtaining the gas–liquid equilibrium data for the gas mixture of CO$_2$ + N$_2$ at various concentrations of PEG 200 (1) + DMAPA (2), and (3) determining the spectral properties of the PEG 200 (1) + DMAPA (2) + CO$_2$ system. To the best of our knowledge, studies on the thermodynamic properties of the binary system of PEG 200 (1) + DMAPA (2), such as density ($\rho$), viscosity ($\eta$), and excess properties, over a wide experimental temperature range are very lacking. Herein, the present work mainly focuses on investigating the density and viscosity data at $T = (298.15, 303.15, 308.15, 313.15, \text{and} 318.15) K$ for the whole composition range. Based on the above experimental results, the excess molar volume ($V_m^E$), viscosity deviation ($\Delta\eta$), and the excess Gibbs free energies of activation for viscous flow ($\Delta G^{*E}$) were calculated. According to the results of excess properties, the CO$_2$ uptake of the PEG 200 (1) + DMAPA (2) system was also studied. These results can be used to obtain important basic data for potential industrial applications. Furthermore, the possible intermolecular interaction of PEG 200 with DMAPA was also investigated using UV–vis, FTIR, and $^1$H NMR spectroscopic techniques.

2. RESULTS AND DISCUSSION

2.1. Density and Viscosity. The density of PEG 200 (1) + DMAPA (2) mixtures with different compositions at atmospheric pressure and $T = (298.15, 303.15, 308.15, 313.15, \text{and} 318.15) K$ was determined. The results are shown in Table 1 and Figure 1, where the mole fraction of PEG 200 is expressed by $x_1$.

Table 1. Density Data for the Binary System of PEG 200 (1) + DMAPA (2) as a Function of PEG 200 Mole Fraction ($x_1$) at $T = (298.15, 303.15, 308.15, 313.15, \text{and} 318.15) K$ and Atmospheric Pressure

| $x_1$ | $T = 298.15 K$ | $T = 303.15 K$ | $T = 308.15 K$ | $T = 313.15 K$ | $T = 318.15 K$ |
|-------|----------------|----------------|----------------|----------------|----------------|
| 0.0000 | 0.8098         | 0.8042         | 0.8023         | 0.7949         | 0.7938         |
| 0.0262 | 0.8323         | 0.8296         | 0.8265         | 0.8197         | 0.8216         |
| 0.0537 | 0.8585         | 0.8532         | 0.8476         | 0.8441         | 0.8435         |
| 0.0827 | 0.8807         | 0.8731         | 0.8700         | 0.8677         | 0.8672         |
| 0.1133 | 0.9008         | 0.8973         | 0.8917         | 0.8890         | 0.8879         |
| 0.1455 | 0.9205         | 0.9158         | 0.9119         | 0.9083         | 0.9075         |
| 0.1796 | 0.9394         | 0.9358         | 0.9302         | 0.9278         | 0.9267         |
| 0.2157 | 0.9573         | 0.9522         | 0.9473         | 0.9445         | 0.9434         |
| 0.2541 | 0.9746         | 0.9714         | 0.9664         | 0.9610         | 0.9600         |
| 0.2948 | 0.9907         | 0.9867         | 0.9822         | 0.9784         | 0.9774         |
| 0.3381 | 1.0053         | 1.0012         | 0.9966         | 0.9933         | 0.9922         |
| 0.3844 | 1.0192         | 1.0164         | 1.0116         | 1.0071         | 1.0062         |
| 0.4339 | 1.0328         | 1.0295         | 1.0251         | 1.0215         | 1.0206         |
| 0.4869 | 1.0455         | 1.0417         | 1.0378         | 1.0314         | 1.0308         |
| 0.5438 | 1.0566         | 1.0546         | 1.0497         | 1.0460         | 1.0450         |
| 0.6052 | 1.0690         | 1.0661         | 1.0618         | 1.0574         | 1.0560         |
| 0.6714 | 1.0791         | 1.0767         | 1.0718         | 1.0684         | 1.0659         |
| 0.7433 | 1.0900         | 1.0860         | 1.0811         | 1.0788         | 1.0781         |
| 0.8214 | 1.1001         | 1.0958         | 1.0926         | 1.0851         | 1.0832         |
| 0.9066 | 1.1064         | 1.1043         | 1.1013         | 1.0975         | 1.0964         |
| 1.0000 | 1.1213         | 1.1175         | 1.1139         | 1.1090         | 1.1057         |

*Standard uncertainties $u$ for each variable are $u(T) = 0.01 K$, $u(\rho) = \pm 5\%$, and $u(x_1) = 0.0001$ and the combined expanded uncertainty is $u_c(\rho) = \pm 0.02\%$, with a 0.95 level of confidence ($k \approx 2$).

Figure 1. Changes in experimental densities with molar fraction for PEG 200 (1) + DMAPA (2) at $T = (298.15, 303.15, 308.15, 313.15, \text{and} 318.15) K$.

It can be seen from Figure 1 that the density of mixtures increases with the increase of the mole fraction of PEG 200 at the same temperature. In the same sample, the density of the mixtures decreases with the increase of temperature. The density of the binary solution can be calculated using eq 1)\(^{22}\)

$$
\rho_{cal} = \frac{x_1 M_1 + x_2 M_2}{V_1 + x_1 x_2 \sum_{i=0}^{x_1} A_i(2x_1 - 1)^i}
$$

where $x_1$, $M_1$ and $x_2$, $M_2$ represent the molar fraction and relative molecular weight of pure PEG 200 and pure DMAPA, respectively. $V_1$ and $V_2$ are the molar volumes of pure PEG 200 and DMAPA, respectively. $A_i$ represents the fitting parameters.

The average absolute deviation was calculated using eq 2\(^{22}\):
AAD% = \frac{\sum |(Y_{\text{exp}} - Y_{\text{cal}})|}{n} \times 100 (2)

where \( Y_{\text{exp}} \) is the experimental density value, \( Y_{\text{cal}} \) is the calculated density value, and \( n \) is the number of experiments. The relative deviation of density of the PEG 200 (1) + DMAPA (2) mixture is within \( \pm 0.002 \) and is plotted in Figure 2.

The viscosity of the PEG 200 (1) + DMAPA (2) mixture at \( T = (298.15, 303.15, 308.15, 313.15, \text{and } 318.15) \) K was measured with an Ubbelohde viscometer. The viscosity data are listed in Table 2 and plotted in Figure 3. From Figure 3, one can find that the viscosity decreases gradually with the increase of temperature. This phenomenon indicates that the molecular energy of the binary system solution tends to increase with the increase of temperature. With the increase of the mole fraction of PEG 200, the viscosity of mixtures increases first and then decreases at the same temperature. When the mole fraction of PEG 200 is greater than 0.66, the viscosity increases more slowly. Particularly, when the mole fraction of PEG 200 is 0.8214, the viscosity reaches the maximum value and then begins to decrease.

The viscosity can be calculated using eq 3:

\[ \eta = x_1 \eta_1 + x_2 \eta_2 + x_1 x_2 \sum_{i=0}^{n} A_i (2x_i - 1)^i \] (3)

where \( x_1 \) and \( x_2 \) denote the mole fractions of PEG 200 and DMAPA, respectively. \( \eta \) is the viscosity of mixtures, and \( \eta_1 \) and \( \eta_2 \) represent the viscosity values of pure PEG 200 and DMAPA, respectively. The relative deviation of viscosity between the experimental value and the calculated value is

\[ \frac{\sum |(\rho_{\text{cal}} - \rho_{\text{exp}})|}{n} \times 100 \]

Figure 3. Excess molar volume \( (V_m^E) \) with the mole fraction for the binary system of PEG 200 (1) + DMAPA (2) at \( T = (298.15, 303.15, 308.15, 313.15, \text{and } 318.15) \) K. The symbols represent the experimental values, and the solid curves represent the calculated values.

The mole fraction of PEG 200, the viscosity of mixtures increases first and then decreases at the same temperature. When the mole fraction of PEG 200 is greater than 0.66, the viscosity increases more slowly. Particularly, when the mole fraction of PEG 200 is 0.8214, the viscosity reaches the maximum value and then begins to decrease.

The viscosity can be calculated using eq 3:

The standard uncertainties \( u \) for each variables are \( u(T) = \pm 0.01 \) K, \( u(p) = 5\% \), and \( u(x_1) = 0.0001 \), and the combined expanded uncertainty is \( u(\eta) = \pm 3\% \), with a 0.95 level of confidence \( (k \approx 2) \).
plotted in Figure 4. It was found that the relative deviation of viscosity data is within ±0.65.

Figure 4. Changes in experimental viscosities with the molar fraction for PEG 200 (1) + DMAPA (2) at T = (298.15, 303.15, 308.15, 313.15, and 318.15) K.

2.2. Excess Property. The excess molar volume \( V_m^E \) of the binary system can be calculated using eq 4\textsuperscript{23–25}

\[
V_m^E = \frac{x_1M_1 + x_2M_2}{\rho} - \left(\frac{x_1M_1}{\rho_1} + \frac{x_2M_2}{\rho_2}\right)
\]

where \( \rho \) represents the density of the mixtures; \( x_1, \rho_1, \) and \( M_1 \) and \( x_2, \rho_2, \) and \( M_2 \) are the mole fraction, density, and relative molecular weight of pure PEG 200 and DMAPA, respectively.

Figure 5 and Table 3 show that the \( V_m^E \) in all composition ranges is negative at the experimental temperatures. That is, the volume of the mixed solution is less than the sum of the volumes before the two liquids are mixed. With the same composition, \( V_m^E \) increases with the increase of temperature. The \( V_m^E \) of the binary solution mixture is mainly related to the chemical action, physical properties, and structural properties of the components. The negative \( V_m^E \) values of mixtures indicate that there is a certain interaction force between PEG 200 and DMAPA, which makes the intermolecular distance smaller and the volume to shrink. Particularly, when the molar ratio of DMAPA and PEG 200 is about 2:1, the \( V_m^E \) is the lowest, meaning that the intermolecular bonding is the strongest at this concentration.

\[
\Delta\eta = \eta - (x_1\eta_1 + x_2\eta_2)
\]

where \( \eta \) represents the viscosity of the PEG 200 (1) + DMAPA (2) system. \( \eta_1 \) and \( \eta_2 \) represent the viscosity of pure PEG 200 and DMAPA, respectively. \( x_1 \) and \( x_2 \) represent the mole fraction of PEG 200 and DMAPA, respectively. Figure 6 and Table 4 show the dependence of viscosity deviations on composition and temperature. It can be seen from each \( \Delta\eta \) curve of the PEG 200 (1) + DMAPA (2) mixture that the value of \( \Delta\eta \) is not always negative but also shows a positive maximum and negative minimum at around \( x_1 \approx 0.6067 \) and \( x_1 \approx 0.1158 \), respectively. The curves become more and more flat, and the absolute values decrease at elevated temperatures. The excess Gibbs free energies of activation for viscous flow (\( \Delta G^{*E} \)) values were calculated from the viscosity data using eq \( 6^26 \)

\[
\Delta G^{*E} = RT[\ln(\eta V) - x_1\ln(\eta_1 V_1) - x_2\ln(\eta_2 V_2)]
\]

where \( R \) is the general constant of the gas, \( T \) is the absolute temperature, \( V, V_1, \) and \( V_2 \) are the molar volumes of the binary mixtures, pure PEG 200, and DMAPA, respectively. \( \eta, \eta_1, \) and \( \eta_2 \) are the absolute viscosity of the binary mixtures, pure PEG 200, and DMAPA, respectively. The values of \( \Delta G^{*E} \) are given in Table 5 and are plotted in Figure 7.

As shown in Figure 7, the values of \( \Delta G^{*E} \) are all positive in the whole concentration range. \( \Delta G^{*E} \) can be used as a reliable standard to detect the interaction between different molecules.\textsuperscript{27} Here, the results of \( \Delta G^{*E} \) show that there is an interaction between PEG 200 and DMAPA.

The polynomial regression values of \( V_m^E, \Delta\eta, \) and \( \Delta G^{*E} \) were calculated using the Redlich Kister eq \( 7^28 \)

\[
Q = x_1(1 - x_1) \sum_{i=0} A_i(2x_1 - 1)^i
\]

where \( Q \) represents \( V_m^E, \Delta\eta, \) and \( \Delta G^{*E} \); \( x_1 \) represents the mole fraction of PEG 200 and \( A_i \) is the polynomial regression coefficient. In order to study the fitting efficiency of \( V_m^E, \Delta\eta, \) and \( \Delta G^{*E} \), the standard deviations between the calculated value and the experimental value were acquired from eq \( 8^26 \)

\[
s = \left[ \frac{\sum_{i=1}^n (Z_{\text{cal},i} - Z_{\text{exp},i})^2}{(N - m)} \right]^{1/2}
\]

where \( Z \) represents \( V_m^E, \Delta\eta, \) or \( \Delta G^{*E} \); \( n \) and \( m \) are the number of experimental points and the number of parameters retained in the respective equations. The calculated values \( A_i \) and \( s \) and the fitting degree \( R^2 \) are shown in Table 6.

The apparent molar volumes \( V_{\psi,1}^E \) and \( V_{\psi,2}^E \) of the binary mixture were obtained from eqs \( 9^26 \) and \( 10^26 \) and the results are listed in Table 7.

\[
V_{\psi,1}^E = \frac{x_2M_2}{x_1} \times \frac{\rho_2 - \rho_m}{\rho_2} + \frac{M_1}{\rho_m}
\]

\[
V_{\psi,2}^E = \frac{x_1M_1}{x_2} \times \frac{\rho_1 - \rho_m}{\rho_1} + \frac{M_2}{\rho_m}
\]

where \( \rho_m \) is the density of the mixture, and \( x_1, \rho_1, \) and \( M_1 \) and \( x_2, \rho_2, \) and \( M_2 \) are the mole fraction, density value, and relative molecular weight of pure PEG 200 and DMAPA, respectively. Table 9 shows that the apparent molar volumes of binary solutions \( V_{\psi,1}^E \) and \( V_{\psi,2}^E \) increase with the increase of temperature at the same composition. At a certain temperature, the apparent molar volume of the binary solution \( V_{\psi,1}^E \)
increases and that of \( V_{g,2} \) decreases with the increase of the mole fraction of PEG 200.

The partial molar volume of the mixed solution was calculated using equation 11 and 12:

\[
\bar{V}_1 = V_m^E + V_1 + (1 - x_1) \left( \frac{\partial V_m^E}{\partial x_1} \right)_{P,T}
\]

\[
\bar{V}_2 = V_m^E + V_2 - x_1 \left( \frac{\partial V_m^E}{\partial x_1} \right)_{P,T}
\]

where \( V_1 \) and \( V_2 \) are the molar volumes of the pure components and \( V_m^E \) is the excess molar volume of the mixture. The calculated data are listed in Table 8. The partial molar volumes of \( \bar{V}_1 \) and \( \bar{V}_2 \) increase with the increase of the mole fraction of PEG 200, while \( V_1 \) and \( V_2 \) do not change significantly with temperatures.

### 2.3. Thermodynamic Parameters.

The \( V_m^E \) of the PEG 200 (1) + DMAPA (2) mixture is negative, which proves that there is an interaction between PEG 200 and DMAPA. This intermolecular interaction can also be explained by the thermodynamics of viscous fluids. To obtain the kinematic viscosity of the PEG 200 (1) + DMAPA (2) system, the thermodynamic parameters of the viscous fluid of the PEG 200 (1) + DMAPA (2) system were calculated, including the activation enthalpy \( \Delta H^* \), activation entropy \( \Delta S^* \), and activation Gibbs free energy \( \Delta G^* \) using eqs 13 and 14:

\[
\Delta G^* = \Delta H^* - T \Delta S^*
\]

\[

\nu = \frac{hN_A}{M} \exp \left( \frac{\Delta G^*}{RT} \right)
\]

where \( \nu, h, N_A, M, \) and \( R \), respectively, represent the kinematic viscosity, Planck constant, Avogadro constant, average molar mass, and molar gas constant of the binary system. From 13 and 14, the following formula can be obtained:

\[
R \ln \left( \frac{M}{hN_A} \right) = \frac{\Delta H^*}{T} - \Delta S^*
\]

\( R \ln(\nu M/hN_A) \) on the left side of eq 15 was found to be in a linear relationship with \( 1/T \) on the right side, when a plot was constructed by taking \( 1/T \) as the abscissa and \( R \ln(\nu M/hN_A) \) as the ordinate. The fitting results are shown in Figure 8. The slope is the \( \Delta H^* \) value and the intercept is the \( -\Delta S^* \) value, and the results are shown in Table 9. The activation Gibbs free energy \( \Delta G^* \) can also be calculated using eq 13, and the results obtained are shown in Table 9 and plotted in Figure 9.

It was found from Table 9 that the value of \( \Delta G^* \) over the range of components is positive at measured temperatures and \( \Delta H^* \) increases from 1.51 to 27.63 kJ mol\(^{-1}\) K\(^{-1}\). The results...
indicate that the viscous flow in pure DMAPA is easier than that in pure PEG 200 or the binary solution. At the same time, $\Delta S^*$ increases from $-215.52$ to $-163.61$ J·mol$^{-1}$·K$^{-1}$ with the increase of the mole fraction of PEG 200. The negative $\Delta S^*$ indicates that the self-association binding force of PEG 200 and DMAPA is stronger than that of DMAPA alone.

### Table 4. Viscosity Deviation ($\Delta \eta$) for the Binary System of PEG 200 (1) + DMAPA (2) at $T = (298.15, 303.15, 308.15, 313.15, \text{and} 318.15)$ K

| $x_1$ | $T = 298.15$ K | $T = 303.15$ K | $T = 308.15$ K | $T = 313.15$ K | $T = 318.15$ K |
|-------|----------------|----------------|----------------|----------------|----------------|
| 0.0000 | 0.00           | 0.00           | 0.00           | 0.00           | 0.00           |
| 0.0262 | $-0.80$        | $-0.62$        | $-0.44$        | $-0.28$        | $-0.15$        |
| 0.0537 | $-1.65$        | $-1.30$        | $-0.97$        | $-0.63$        | $-0.43$        |
| 0.0827 | $-2.18$        | $-1.70$        | $-1.21$        | $-0.72$        | $-0.51$        |
| 0.1133 | $-2.45$        | $-1.86$        | $-1.28$        | $-0.67$        | $-0.41$        |
| 0.1455 | $-2.26$        | $-1.60$        | $-1.00$        | $-0.35$        | $-0.12$        |
| 0.1796 | $-1.52$        | $-1.04$        | $-0.48$        | 0.22           | 0.43           |
| 0.2157 | $-0.26$        | 0.00           | 0.33           | 1.03           | 1.13           |
| 0.2541 | 1.57           | 1.48           | 1.57           | 2.26           | 2.12           |
| 0.2948 | 3.95           | 3.49           | 3.49           | 3.54           | 3.41           |
| 0.3381 | 6.59           | 5.40           | 4.59           | 4.92           | 4.46           |
| 0.3844 | 9.29           | 7.53           | 6.26           | 6.41           | 5.55           |
| 0.4339 | 11.85          | 9.70           | 7.80           | 7.85           | 6.79           |
| 0.4869 | 13.80          | 11.02          | 9.04           | 8.95           | 7.58           |
| 0.5438 | 15.10          | 12.02          | 9.85           | 9.58           | 8.14           |
| 0.6052 | 15.62          | 12.16          | 9.89           | 10.04          | 8.42           |
| 0.6714 | 14.44          | 11.46          | 9.97           | 9.63           | 8.08           |
| 0.7433 | 12.78          | 9.98           | 8.17           | 8.81           | 7.26           |
| 0.8214 | 10.52          | 7.95           | 6.19           | 7.43           | 6.16           |
| 0.9066 | 5.95           | 4.56           | 3.66           | 5.42           | 4.84           |
| 1.0000 | 0.00           | 0.00           | 0.00           | 0.00           | 0.00           |

### Table 5. Excess Gibbs Free Energy of Activation of Viscous Flow ($\Delta G^{*E}$) for the Binary System of PEG (1) + DMAPA (2) at $T = (298.15, 303.15, 308.15, 313.15, \text{and} 318.15)$ K

| $x_1$ | $T = 298.15$ K | $T = 303.15$ K | $T = 308.15$ K | $T = 313.15$ K | $T = 318.15$ K |
|-------|----------------|----------------|----------------|----------------|----------------|
| 0.0000 | 0.00           | 0.00           | 0.00           | 0.00           | 0.00           |
| 0.0262 | 0.84           | 0.78           | 0.78           | 0.77           | 0.86           |
| 0.0537 | 1.31           | 1.24           | 1.19           | 1.19           | 1.27           |
| 0.0827 | 1.92           | 1.83           | 1.77           | 1.76           | 1.77           |
| 0.1133 | 2.47           | 2.35           | 2.27           | 2.25           | 2.26           |
| 0.1455 | 3.00           | 2.88           | 2.77           | 2.73           | 2.69           |
| 0.1796 | 3.47           | 3.30           | 3.18           | 3.13           | 3.09           |
| 0.2157 | 3.85           | 3.68           | 3.52           | 3.47           | 3.40           |
| 0.2541 | 4.15           | 3.96           | 3.80           | 3.77           | 3.67           |
| 0.2948 | 4.37           | 4.19           | 4.07           | 3.94           | 3.88           |
| 0.3381 | 4.48           | 4.28           | 4.09           | 4.04           | 3.94           |
| 0.3844 | 4.49           | 4.29           | 4.11           | 4.07           | 3.94           |
| 0.4339 | 4.40           | 4.23           | 4.03           | 4.01           | 3.89           |
| 0.4869 | 4.20           | 4.03           | 3.87           | 3.86           | 3.73           |
| 0.5438 | 3.91           | 3.75           | 3.61           | 3.60           | 3.48           |
| 0.6052 | 3.52           | 3.36           | 3.24           | 3.28           | 3.17           |
| 0.6714 | 3.01           | 2.89           | 2.83           | 2.85           | 2.75           |
| 0.7433 | 2.43           | 2.32           | 2.25           | 2.33           | 2.24           |
| 0.8214 | 1.76           | 1.68           | 1.60           | 1.74           | 1.68           |
| 0.9066 | 0.95           | 0.91           | 0.87           | 1.03           | 1.02           |
| 1.0000 | 0.00           | 0.00           | 0.00           | 0.00           | 0.00           |

2.4. Spectral Properties. 2.4.1. UV–Vis Spectra. The preceding results may be related to the hydrogen bond and interaction between PEG 200 and DMAPA. The UV–vis spectra of the PEG 200 (1) + DMAPA (2) mixture were measured and are shown in Figure 10. PEG 200 was used as a reference solvent. The absorption peak of DMAPA was found at about 234 nm, which is attributed to N electrons undergoing the $n \rightarrow \sigma^*$ electron transition. The peak red-shifted from 234 to 237 nm with the increase of the DMAPA concentration. This phenomenon is mainly due to the self-association hydrogen bond between PEG 200 molecules was
broken, and the new hydrogen bond was formed between PEG 200 and DMAPA through the formation of (CH₃)₂N···HOCH₂CH₂O− and H₂N···HOCH₂CH₂O−. Particularly, the ability of DMAPA molecules to form a hydrogen bond with PEG 200 molecules was reinforced with the increase of the DMAPA concentration, which will make the nitrogen atom in DMAPA molecules interact more easily with hydroxyl on PEG 200, leading to the red shift of the absorption peak.

2.4.2. FTIR Spectra. To further study the interaction between DMAPA and PEG 200, the FTIR spectra of PEG 200, DMAPA, and the binary system of PEG 200 (1) + DMAPA (2) at various concentrations were obtained. In the FTIR spectra shown in Figure 11, the stretching vibration peak of the O−H bond in PEG 200 molecules appears at 3422 cm⁻¹. Interestingly, the absorption peak moves to a low wavenumber of 3356 cm⁻¹ with an increase of the DMAPA concentration; meanwhile, the bending vibration band of the hydroxyl group in PEG 200 moved from 1104 cm⁻¹ to a lower frequency of 1100 cm⁻¹ with the increase of the DMAPA concentration. These results suggest that the intermolecular hydrogen bonds in PEG 200 are broken gradually, and new hydrogen bonds are formed. The absorption band of hydroxyl groups in PEG 200 moves to a lower frequency, which is due to the vibrational properties of hydroxyl groups in PEG 200, further indicating the hydrogen bonding and interaction of the hydroxyl hydrogen atoms in PEG 200 with nitrogen atoms in DMAPA.

2.4.3. ¹H NMR Spectra. The ¹H NMR spectra of PEG 200, DMAPA, and the binary system of PEG 200 (1) + DMAPA (2) with various concentrations were also compared. Figure 12 shows that the chemical displacement of methyl hydrogen in DMAPA is 2.158 ppm. With the increase of the PEG 200 concentration, the chemical displacement gradually moves to a low field of 2.000 ppm; meanwhile, the chemical shifts of other methylene groups in DMAPA also move to the lower field. The fact is that the electron cloud density of protons on DMAPA becomes thinner due to hydrogen bonding interactions between DMAPA and PEG 200. The most reasonable form of hydrogen bonding was proposed as the formation of (CH₃)₂N···HOCH₂CH₂O− and H₂N···HOCH₂CH₂O−. The results are in well accordance with the FTIR and UV−vis results.

2.5. CO₂ Absorption. Based on the above experimental results, the PEG 200 plus DMAPA mixture was used to measure CO₂ absorption. Figure 13a shows the relationship between CO₂ uptake and time at room temperature and atmospheric pressure. The absorption equilibrium of PEG 200 + DMAPA (1:1 mole ratio), PEG 200 + DMAPA (1:2), and PEG 200 + DMAPA (2:1) was all achieved within 45 min. In contrast, the absorption capacity of CO₂ of PEG 200 + DMAPA (1:2) is quite large, reaching 0.19 g CO₂ per gram solvent. Interestingly, when the molar ratio of PEG 200 and DMAPA is 1:2, the intermolecular bonding is the most strongest based on the results obtained for Vm of the mixture. This intermolecular hydrogen bonding can reduce the volatility of amines, which will be further discussed later. Considering the importance of CO₂ removal efficiency in practical industrial processes, we investigated low-concentration CO₂ removal under environmental conditions. As shown in Figure 13b, it was found that CO₂ was completely removed in the first 40 min using PEG 200 + DMAPA (1:2) as the absorbent, then the removal efficiency was reduced, and the final absorption reaches saturation in about 60 min.

3. CONCLUSIONS

In summary, the density and viscosity of the PEG 200 (1) + DMAPA (2) binary solution in the whole concentration range

| Table 6. Coefficients of the Redlich–Kister Equation and Standard Deviations for Vm/(cm³·mol⁻¹), Δη/(mPa·s), and ΔGφ/K (kJ·mol⁻¹) for the Binary System of PEG 200 (1) + DMAPA (2) at Different Temperatures; R² is the Degree of Fitting |
|---|---|---|---|---|---|---|---|
| T/(K) | Vm¹/(cm³·mol⁻¹) | A₁ | A₂ | A₃ | A₄ | σ | R² |
| 298.15 | -32.8245 | 21.1379 | -12.4724 | 16.3136 | 0.1487 | 0.9981 |
| 303.15 | 53.57 | 60.38 | -63.77 | 3.84 | 0.72 | 0.9953 |
| 308.15 | 16.52 | -9.70 | 2.45 | 2.28 | 0.06 | 0.9993 |
| 313.15 | -33.7457 | 21.6664 | -11.7893 | 15.0121 | 0.1296 | 0.9987 |
| 318.15 | 42.93 | 45.03 | -52.19 | 4.16 | 0.55 | 0.9958 |
| 323.15 | 15.86 | -9.33 | 2.21 | 2.21 | 0.05 | 0.9994 |
| 328.15 | -33.0189 | 22.2396 | -10.7541 | 9.7849 | 0.0948 | 0.9991 |
| 333.15 | 35.80 | 36.07 | -40.87 | -0.49 | 0.35 | 0.9969 |
| 338.15 | 15.24 | -8.66 | 2.20 | 1.46 | 0.05 | 0.9989 |
| 343.15 | -34.4147 | 21.8592 | -14.3372 | 18.8922 | 0.1476 | 0.9980 |
| 348.15 | 34.45 | 32.45 | -22.57 | 16.16 | 0.45 | 0.9953 |
| 353.15 | 15.07 | -8.00 | 3.17 | 2.27 | 0.06 | 0.9992 |
| 358.15 | -34.6386 | 21.4404 | -14.8585 | 16.5294 | 0.2882 | 0.9963 |
| 363.15 | 29.11 | 24.10 | -17.83 | 18.60 | 0.44 | 0.9931 |
| 368.15 | 14.47 | -7.79 | 3.44 | 1.75 | 0.10 | 0.9983 |
were negative, it also shows a stronger intermolecular bonding is formed. A flow thermodynamics. The properties and viscous flow thermodynamics. Therefore, $T_1 = 298.15$, $303.15$, $308.15$, $313.15$, and $318.15$ K at atmospheric pressure were reported. The density and viscosity data have been used to compute the parameter of excess properties and viscous flow thermodynamics. The $V_m$ values were negative, $\Delta_f$ is not always negative, it also shows a positive maximum and a negative minimum at around $x_1 \approx 0.6067$ and $x_1 \approx 0.1158$; meanwhile, the $\Delta H^\circ$, $\Delta S^\circ$, and $\Delta G^\circ$ values were calculated. Based on the results of $V_m$, it was found that a stronger intermolecular bonding is formed. A mixture of PEG 200 and DMAPA in a ratio of 1:2 was used for investigating CO$_2$ absorption at a pressure of 1 bar, and a CO$_2$ absorption of about 0.19 g per gram of the absorbent at room temperature was achieved. In addition, the UV–vis, FTIR, and $^1$H NMR results indicate that there are hydrogen bonding and interactions of hydroxyl hydrogen atoms in PEG 200 with nitrogen atoms in DMAPA leading to the formation of (CH$_3$)$_2$N···HOCH$_2$CH–O– and H$_2$N···HOCH$_2$CH–O–.

Table 7. Apparent Molar Volumes $V_{\text{app}}$ and $V_{\text{app}}$ for the Binary System of PEG 200 and DMAPA at $T$ = (298.15, 303.15, 308.15, 313.15, and 318.15) K

| $x_1$ | $T/K = 298.15$ & $303.15$ & $308.15$ & $313.15$ & $318.15$ |
|-------|------------------|------------------|------------------|------------------|------------------|
| 0.0000 | 113.12 & 128.99 & 147.16 & 187.20 & 175.73 |
| 0.0262 | 106.83 & 121.15 & 136.83 & 146.44 & 148.31 |
| 0.0537 | 114.44 & 127.55 & 132.93 & 137.13 & 137.95 |
| 0.0827 | 122.15 & 126.55 & 133.55 & 136.94 & 138.26 |
| 0.1133 | 128.17 & 132.57 & 136.30 & 139.85 & 140.57 |
| 0.1455 | 133.41 & 136.07 & 140.37 & 142.26 & 143.14 |
| 0.1796 | 138.25 & 141.42 & 144.52 & 146.32 & 147.02 |
| 0.2157 | 142.56 & 144.27 & 146.91 & 149.79 & 150.38 |
| 0.2541 | 146.73 & 148.57 & 150.62 & 152.37 & 152.87 |
| 0.2948 | 150.90 & 152.52 & 154.37 & 155.73 & 156.15 |
| 0.3381 | 154.71 & 155.67 & 157.39 & 159.00 & 159.33 |
| 0.4339 | 158.09 & 159.11 & 160.52 & 161.67 & 161.95 |
| 0.4689 | 161.30 & 162.39 & 163.49 & 165.33 & 165.52 |
| 0.5438 | 164.55 & 165.07 & 166.35 & 167.29 & 167.57 |
| 0.6052 | 167.12 & 167.82 & 168.82 & 169.85 & 170.21 |
| 0.6714 | 169.93 & 170.44 & 171.51 & 172.24 & 172.81 |
| 0.7433 | 172.28 & 173.08 & 174.06 & 174.53 & 174.66 |
| 0.8214 | 174.56 & 175.36 & 175.95 & 177.35 & 177.72 |
| 0.9066 | 177.29 & 177.64 & 178.16 & 178.82 & 179.01 |
| 1.0000 | 178.82 & 179.65 & 180.11 & 180.59 & 180.69 |

Table 8. Partial Molar Volumes $\bar{V}_1$ and $\bar{V}_2$ for the Binary System of PEG 200 and DMAPA at $T$ = (298.15, 303.15, 308.15, 313.15, and 318.15) K

| $x_1$ | $T/K = 298.15$ & $303.15$ & $308.15$ & $313.15$ & $318.15$ |
|-------|------------------|------------------|------------------|------------------|------------------|
| 0.0000 | 107.04 & 106.83 & 110.07 & 104.60 & 106.46 |
| 0.0262 | 116.10 & 115.48 & 118.29 & 114.01 & 115.05 |
| 0.0537 | 124.08 & 123.88 & 126.50 & 122.89 & 124.02 |
| 0.0827 | 132.04 & 132.22 & 133.97 & 131.26 & 132.09 |
| 0.1133 | 139.62 & 139.27 & 140.95 & 139.23 & 139.92 |
| 0.1455 | 146.53 & 146.40 & 147.47 & 146.70 & 147.16 |
| 0.1796 | 152.76 & 152.54 & 153.56 & 153.28 & 153.65 |
| 0.2157 | 158.30 & 158.36 & 159.07 & 159.36 & 159.62 |
| 0.2541 | 163.06 & 162.95 & 163.51 & 164.54 & 164.73 |
| 0.2948 | 167.11 & 167.18 & 167.59 & 168.65 & 168.82 |
| 0.3381 | 170.49 & 170.65 & 171.06 & 172.19 & 172.35 |
| 0.3844 | 173.12 & 173.19 & 173.65 & 174.94 & 175.11 |
| 0.4339 | 175.00 & 175.23 & 175.68 & 176.77 & 176.97 |
| 0.4869 | 176.29 & 176.69 & 177.13 & 178.47 & 178.68 |
| 0.5438 | 177.22 & 177.45 & 178.12 & 178.89 & 179.18 |
| 0.6052 | 177.53 & 177.98 & 178.61 & 179.30 & 179.66 |
| 0.6714 | 177.91 & 178.35 & 179.10 & 179.51 & 180.02 |
| 0.7433 | 178.09 & 178.83 & 179.55 & 179.76 & 179.92 |
| 0.8214 | 178.41 & 179.22 & 179.65 & 180.68 & 180.97 |
| 0.9066 | 179.30 & 179.74 & 180.11 & 180.68 & 180.78 |
| 1.0000 | 178.82 & 179.65 & 180.11 & 180.59 & 180.69 |

Table 9. Apparent Molar Volumes $V_{\text{app}}$ and $V_{\text{app}}$ for the Binary System of PEG 200 and DMAPA at $T$ = (298.15, 303.15, 308.15, 313.15, and 318.15) K
These results are helpful to understand and optimize the CO₂ absorption capacity of the binary solution.

4. EXPERIMENTAL SECTION

4.1. Materials. Analytical grade DMAPA (≥99.0%) and PEG 200 (≥99.0%) were purchased from Shanghai Titan Technology Co., Ltd., China. They were dried over 4 Å molecular sieves and degassed by ultrasonication just before the experiment. The water contents of PEG 200 and DMAPA were determined by the Carl Fischer method to be 0.21 and 0.07 wt %, respectively. Moreover, double-distilled water and HPLC grade ethanol were used to calibrate the pycnometer and the Ubbelohde viscometer, respectively. All specifications of chemical samples are shown in Table S1.

4.2. Instrumentation. All measurements of mass were performed on an electronic balance with an accuracy of ±0.1 mg (Sartorius BS224S). The uncertainty of mole fraction was estimated to be ±0.0001. The UV–vis spectra were recorded on a Shimadzu UV-3600 plus spectrometer. FTIR spectra were recorded on a Bruker Tensor 27 FTIR spectrometer.

Table 9. Gibbs Free Energy Changes (ΔG°/kJ/mol⁻¹), Enthalpy of Activation (ΔH°), and Entropy of Activation (ΔS°) for the Viscous Flow of the Binary System PEG 200 (1) + DMAPA (2) at Different Concentrations

| x₁   | ΔH°/(kJ·mol⁻¹) | ΔS°/(J·mol⁻¹·K⁻¹) | ΔG°/(kJ·mol⁻¹) |
|------|---------------|-------------------|---------------|
|      |               |                   | T/K = 298.15  |                |
|      |               |                   | 303.15        |                |
|      |               |                   | 308.15        |                |
|      |               |                   | 313.15        |                |
|      |               |                   | 318.15        |                |
| 0.0000| 1.51 ± 0.087  | −215.52 ± 0.28    | 65.77         |
| 0.0262| 2.83 ± 0.99   | −214.73 ± 3.23    | 66.89         |
| 0.0537| 5.07 ± 1.18   | −209.77 ± 3.84    | 67.66         |
| 0.0827| 7.80 ± 0.92   | −203.70 ± 3.00    | 68.58         |
| 0.1133| 10.07 ± 1.08  | −199.00 ± 3.50    | 69.45         |
| 0.1455| 12.86 ± 0.86  | −192.61 ± 2.79    | 70.32         |
| 0.1796| 15.17 ± 1.13  | −187.60 ± 3.66    | 71.15         |
| 0.2157| 17.64 ± 1.22  | −181.89 ± 3.95    | 71.92         |
| 0.2541| 19.26 ± 1.20  | −178.82 ± 3.88    | 72.62         |
| 0.2948| 20.91 ± 0.97  | −175.52 ± 3.15    | 73.28         |
| 0.3381| 22.68 ± 1.35  | −171.45 ± 4.37    | 73.85         |
| 0.3844| 23.95 ± 1.21  | −168.90 ± 3.92    | 74.35         |
| 0.4339| 24.64 ± 1.26  | −168.05 ± 4.10    | 74.79         |
| 0.4869| 25.06 ± 1.16  | −167.88 ± 3.77    | 75.15         |
| 0.5438| 25.60 ± 1.10  | −167.11 ± 3.58    | 75.47         |
| 0.6052| 25.55 ± 1.27  | −168.13 ± 4.13    | 75.73         |
| 0.6714| 25.37 ± 0.82  | −169.45 ± 2.65    | 75.92         |
| 0.7433| 25.49 ± 1.10  | −169.62 ± 3.57    | 76.10         |
| 0.8214| 25.40 ± 1.43  | −170.45 ± 4.63    | 76.27         |
| 0.9066| 24.56 ± 1.45  | −173.60 ± 4.70    | 76.37         |
| 1.0000| 27.63 ± 1.18  | −163.61 ± 3.85    | 76.41         |

![Figure 8](https://doi.org/10.1021/acsomega.1c00667)

Figure 8. Plots of R ln(υM/υNA) against 1/T for PEG 200 (1) + DMAPA (2) at different temperatures, and the molar fractions corresponding to lines are as follows: (A: x₁ = 0.0000 ~ U: x₁ = 1.0000).

![Figure 9](https://doi.org/10.1021/acsomega.1c00667)

Figure 9. Gibbs free energies of activation (ΔG°) with the mole fraction for the viscous flow of PEG 200 (1) + DMAPA (2) at T = (298.15, 303.15, 308.15, 313.15, and 318.15) K and atmospheric pressure. The symbols represent experimental values, and the solid curves represent the values calculated from eq 7.

![Figure 10](https://doi.org/10.1021/acsomega.1c00667)

Figure 10. UV–vis spectral changes with increasing DMAPA concentration in the binary system of PEG 200 (1) + DMAPA (2).
were recorded on a Nicolet 50 FTIR spectrometer. $^1$H NMR spectra were recorded on a JNM-ECZ-400 spectrometer. All spectral experiments of PEG 200 ($^1$) + DMAPA ($^2$) were performed at room temperature and atmospheric pressure.

4.3. Measurements. The density of the sample was measured by the pycnometer method. Briefly, the pycnometer containing the binary solvent solution was placed in a constant temperature water bath at a desired temperature for 30 min, and then weighed at least three times to obtain the average value. The uncertainty of density measurement was estimated to be ±0.02%. The viscosity of the sample was measured using an Ubbelohde viscometer. First, the Ubbelohde viscometer was calibrated with double-distilled water and HPLC grade ethanol, and the viscometer constant was calculated. A stopwatch was used to measure the time when the liquid flows through the capillary of the viscometer and this was repeated at least six times for each sample to obtain the average value. The uncertainty of the viscosity value was ±3%. In order to determine the reliability of the experiment, the experimentally measured values of density and absolute viscosity of DMAPA and PEG 200 were compared with those reported in the literature,$^{30-35}$ as shown in Table S2. The agreement between the experimental and literature values was found to be satisfactory.

4.4. Absorption of CO$_2$. The bubble weight method was used in the experiment of CO$_2$ absorption. In short, CO$_2$ absorption was carried out at room temperature at a pressure of 1 bar. As shown in Figure S1, a known amount of absorbent was placed in a glass tube (inner diameter: 10 mm; length: 200 mm). The gas containing CO$_2$ is bubbled into the absorption solution through a long stainless-steel needle. During gas absorption, the gas solubility was determined by weighing the tube on an electronic analytical balance (Sartorius BS224S). To ensure balance is achieved, the weighing procedure was repeated until the mass remains constant between successive measurements. For diluted CO$_2$ absorption, in a typical procedure, about 2.0 g of absorbent was added to a U-shaped absorption tube with an inner diameter of 8 mm, and then the absorption tube was placed in a 25 °C water bath. The CO$_2$ gas (10 wt %) was controlled by a mass flow meter to control the flow rate to 30 mL·min$^{-1}$. CO$_2$ was absorbed by the absorbent when it passed through the U-shaped tube. Then, the concentration of the CO$_2$ tail gas was detected using the flue gas analyzer (LY800-CO$_2$, Shenzhen Liye Industrial Co., LTD, China) and the change in CO$_2$ concentration was recorded online using a computer.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.1c00667.

Specification of chemical samples; comparison of experimental density and viscosity values of pure PEG and DMAPA with literature values at different temper-
atures; and experimental diagram for CO₂ absorption (PDF)

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Notes
The authors declare no competing financial interest.

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