



Density and viscosity measurements of aqueous amines at high pressures: DEA-water, DMAE-water and TEA-water mixtures



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ABSTRACT

In this paper, density and viscosity measurements at pressures up to 140 MPa are presented in a temperature range from (293.15 to 393.15) K for diethanolamine (DEA) + water, triethanolamine (TEA) + water and 2-dimethylaminoethanol (DMAE) + water in amine weight concentrations from 10% to 40%. Densities were measured using a vibrating tube densimeter (Anton Paar DMA HPM) with an expanded uncertainty ($k = 2$) less than $\pm 0.7 \text{ kg}\cdot\text{m}^{-3}$. Viscosity measurements were obtained using a falling body viscometer which was calibrated with water and dodecane. The viscosity expanded uncertainty ($k = 2$) ranges from $\pm 2.5\%$ for the highest viscosity to $\pm 3.2\%$ for the lowest.

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1. Introduction

Global climate change, energy efficiency and switching from fossil fuels to biofuels are the most important issues related to environment, energy and economy. Global climate is changing due to greenhouse gas emissions. These GHG emissions include carbon dioxide, methane, NO_x , SO_x etc. Carbon dioxide (CO_2) is one of the most important greenhouse gases (GHG) which is responsible for about 70% of the enhanced greenhouse effect and global warming [1]. Many solutions are focused on removing carbon dioxide from exhaust gases. Post-combustion CO_2 capture technology is the most mature solution which is based on absorption through chemical absorbents. Absorption with amine-based solvents has been extensively studied and regarded as the most effective technology for CO_2 capture [2].

From the industrial point of view, alkanolamines such as diethanolamine (DEA), triethanolamine (TEA), and 2-(dimethylamino)ethanol (DMAE) have been widely used to remove acid gases in industrial processes [3]. The process to capture carbon dioxide using a chemical absorbent consists of an absorber and a stripper, and solubility of carbon dioxide in aqueous alkanolamine solutions, density and viscosity are key data for the design of CO_2 removal process.

The objective of this work is to measure density and viscosity, at temperatures from (293.15 to 393.15) K and pressures from (0.1 to 100) MPa, of aqueous solutions of DEA, TEA and DMAE at mass fractions from 0.1 to 0.4. Both properties are correlated as a function of temperature and pressure using empirical equations.

2. Experimental

2.1. Materials

The sources and chemical purities of the compounds used in this work are detailed in Table 1. Purities were specified by the supplier and no further purification was carried out. Liquid mixtures were prepared by weighting with a standard uncertainty ($k = 1$) in mass fractions less than $\pm 1 \cdot 10^{-4}$ (taking into account the negligible water content of the amines).

2.2. Apparatus and procedure

Densities were obtained using a vibrating tube densimeter (Anton Paar DMAHPM) calibrated with toluene and vacuum which was previously described in [4]. This model is able to measure density in a range from (0 to 3000) $\text{kg}\cdot\text{m}^{-3}$ with a resolution of $10^{-2} - \text{kg}\cdot\text{m}^{-3}$. The uncertainty calculations were performed following the guide to the expression of uncertainty in measurement JCGM100:2008 [5] and the procedure was explained in [4], obtaining an expanded uncertainty ($k = 2$) less than $\pm 0.7 \text{ kg}\cdot\text{m}^{-3}$.

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Table 1
Material description.

Compound	Source	Mass fraction purity ^a	Mass water content (%)	Purification method
DEA	Sigma-Aldrich	≥0.99	<0.002 ^b	None
TEA	Sigma-Aldrich	≥0.99	<0.006 ^b	None
DMAE	Sigma-Aldrich	≥0.995	<0.01 ^b	None
Water	Sigma-Aldrich	Conductivity ≤ 2·10 ⁻⁶ ohm ⁻¹ cm ⁻¹		None
Dodecane	Sigma-Aldrich	≥0.99	≤0.01 ^a	None

^a As stated by the supplier by gas chromatography.

^b Measured by Karl Fisher titration (Mitsubishi CA-200).

Viscosities were obtained using a falling body viscometer. Its operation is based on the falling body measurement through a vertical tube containing the fluid whose viscosity is sought. This equipment is able to measure viscosity in wide pressure and temperature ranges of (0.1–140) MPa, and (253.15–523.15) K. The cell was designed by Groupe de Haute Pression, Laboratoire des Fluides Complexes of the University of Pau [6] and implemented in TER-MOCAL laboratory.

The operation principle of the equipment is based on the combination of Stokes' law of a falling body through a fluid, and Newton's second law. Such behaviour is theoretically described by Eq. (1):

$$\eta = K(p, T)\Delta\rho\Delta t \quad (1)$$

where η is the viscosity, $K(p, T)$ is a calibration constant which depends on the geometry of the equipment and is considered a function of pressure and temperature, $\Delta\rho$ is the difference between body density and the liquid density, and Δt is the fall time.

The equipment requires a calibration procedure because in practice the actual behaviour differs from the simplified model expressed in Eq. (1) in several factors [7,8]. Various calibration procedures have been successfully used [8] which are based on the use of a known viscosity reference fluid under (p, T) conditions in which the viscosity is sought, obtaining $K(p, T)$ for each.

In our case, the model described by Eq. (1) is adjusted to viscosities up to 4.9 mPa·s (approximately). However, the addition of an independent term to Eq. (1) allows a better approximation to the behaviour of our viscometer and therefore, it is modified to obtain the expression (2):

$$\eta = a + b\Delta\rho\Delta t \quad (2)$$

This Eq. (2) was successfully used previously in [9], and offers the advantage of providing viscosities at any pressure and temperature condition, within measuring calibration range, through a single adjustment.

The experimental setup was entirely developed in the TERMO-CAL laboratory and described in [9,10].

Viscometer calibration was performed at $p = (0.1–100)$ MPa and $T = (293.15–393.15)$ K with water, which was chosen because it is used in the mixtures, and dodecane. Both have been extensively studied in the pressure and temperature ranges of this study.

Fall time was recorded considering fifteen repetitions for each pressure and temperature. After that, calibration consists of fitting all points using the model expressed by Eq. (2), whose parameters are given in Table 2.

Uncertainty calculation was carried out following the model expressed by Eq. (2) and the procedure described in JCGM

Table 2
Coefficients of Eq. (2) obtained for the falling body viscometer calibration.

	Parameters	Standard deviation
$a/\text{mPa}\cdot\text{s}$	$-5.713\cdot 10^{-2}$	$2.9\cdot 10^{-3}$
$b/\text{mPa}\cdot\text{m}^3\cdot\text{kg}^{-1}$	$2.753\cdot 10^{-5}$	$3.3\cdot 10^{-8}$

100:2008 [5] and its results are presented in Tables 3 and 4 [11]. Uncertainty was evaluated at the limits of the viscosity calibration range for the studied mixtures: the lowest viscosity is 0.260 mPa·s for water at $T = 393.15$ K and $p = 5$ MPa, and the highest viscosity is 7.591 mPa·s for aqueous DMAE solution ($w = 0.4$) at $T = 293.15$ K and $p = 60$ MPa. A normal distribution was considered with a coverage factor $k = 2$ (confidence level of 95.45%), obtaining a relative expanded uncertainty which varies from $\pm 2.5\%$ to $\pm 3.2\%$ for the highest and lowest viscosities, respectively. It is interesting to highlight that the most significant contribution in both cases is the uncertainty associated to calibration function coefficients.

3. Results and discussion

Density measurements of aqueous solutions of diethanolamine (DEA), triethanolamine (TEA) and 2-dimethylaminoethanol (DMAE) were carried out at pressures from (0.1 to 140) MPa and at six temperatures ranging between (293.15 and 393.15) K for amine mass fractions of 0.1, 0.2, 0.3 and 0.4. The experimental results are shown in Tables 5–7, respectively.

Density measurements of the amine solutions studied show $\rho_{\text{TEA}} > \rho_{\text{DEA}} > \rho_{\text{DMAE}}$ at the same composition, temperature and pressure conditions. Density differences between amine solutions are greater as amine weight fraction increases and they are also bigger between DEA and DMAE than TEA and DEA.

As is expected, density increases with pressure and decreases with temperature for all the mixtures, however the effect of amine weight fraction is different.

The density rise due to an increase of pressure from 0.1 MPa to 140 MPa is similar for all the amines and ranges from 4.3% to 6.1% for TEA and DEA solutions, being the lowest increase at $w_1 = 0.4$ and $T = 293.15$ K and the highest increase at $w_1 = 0.1$ and $T = 393.15$ K. For DMAE solutions, the density increase ranges from 4.6% to 7.2% both extreme values at $w_1 = 0.4$ and $T = 293.15$ K and $T = 393.15$ K, respectively.

Furthermore, the density decrease when temperature changes from 293.15 K to 393.15 K ranges from 4.9% to 6.3% for TEA aqueous mixtures and from 4.8% to 6.1% for DEA aqueous mixtures, being higher at lower pressures. In the case of DMAE aqueous mixtures, this density decrease varies from 5.0% to 6.0% at $w_1 = 0.1$ and $p = 140$ MPa and $p = 0.1$ MPa, respectively, and from 6.1% to 8.4% at $w_1 = 0.4$ and $p = 140$ MPa and $p = 0.1$ MPa, respectively. Therefore, the effect of temperature is higher at lower pressures and higher amine weight fraction.

Finally, density increases with increasing amine weight fraction for DEA and TEA aqueous solutions but decreases for DMAE aqueous solutions. Comparing for the different amine solutions how is the relative change of density when the composition changes from $w_1 = 0.1$ to $w_1 = 0.4$, the following percentages are obtained: a decrease ranging from 4.0% to 5.1% for TEA solutions, a decrease ranging from 2.8% to 3.8% for DEA solutions. For both mixtures, the density decrease is nearly constant at 140 MPa at any temperature and the highest change is observed at $T = 293.15$ K and $p = 0.1$ MPa. On the contrary, a density increase ranging from

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