

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.

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 (CO₂) 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 CO₂ 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 CO₂ 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 CO₂ removal process.

The objective of this work is to measure density and viscosity, at temperatures from (313.15 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).

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 $\leq 2 \cdot 10^{-6} \text{ ohm}^{-1} \cdot \text{cm}^{-1}$		None
Dodecane	Sigma-Aldrich	≥ 0.99	$\leq 0.01^a$	None

^a as stated by the supplier by gas chromatography

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

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 $\pm 0.7 \text{ kg} \cdot \text{m}^{-3}$.

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 to 140) MPa, and (253.15 to 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 TERMOCAL 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 TERMOCAL laboratory and described in [9,10].

Viscometer calibration was performed at $p = (0.1 \text{ to } 100) \text{ MPa}$ and $T = (293.15 \text{ to } 393.15) \text{ K}$ 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.

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

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

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

Table 3. Uncertainty calculation of dynamic viscosity, η , for water at $p = 5$ MPa and $T = 393.15$ K.

Amount		Estimate	Units	Probability Distribution	Standard Uncertainty $u(x_i)$	Sensitivity Coefficient c_i	Uncertainty Contribution $u(y)$
X_i		x_i					
Reference							
Fluid	Viscosity	0.239	mPa·s	Normal	0.0012	1	0.0012
	Calibration		s	Normal	0.005	0.17	0.00084
Time	Resolution	1.61	s	Rectangular	0.0029	0.17	0.00048
	Repeatability		s	Normal	0.0072	0.17	0.0012
	Calibration		K	Normal	0.010	0.0023	0.00002
Temperature	Resolution	393.15	K	Rectangular	0.0029	0.0023	0.000007
	Uniformity		K	Rectangular	0.029	0.0023	0.00007
	Stability		K	Rectangular	0.014	0.0023	0.00003
	Calibration		MPa	Normal	0.0005	0.00022	0.0000001
Pressure	Resolution	5	MPa	Rectangular	0.0029	0.00022	0.0000006
	Stability		MPa	Rectangular	0.014	0.00022	0.000006
Density	Solid	7673	kg·m ⁻³	Normal	17	0.00004	0.00075
	Fluid	945.5	kg·m ⁻³	Normal	0.005	0.00044	0.00000021
Calibration							
function			mPa·s	Normal	0.0033	1	0.0033
coefficients							
Standard Uncertainty			mPa·s			$u(y)$	± 0.0038
Expanded Uncertainty ($k=2$)			mPa·s			$U(y)$	± 0.0076
Relative Expanded Uncertainty ($k=2$)			(mPa·s/mPa·s)			$U_r(y)$	$\pm \mathbf{0.032}$

Table 4. Uncertainty calculation of dynamic viscosity, η , for DMAE ($w_1 = 0.4$) + H₂O at $p = 60$ MPa and $T = 293.15$ K.

Amount		Estimate	Units	Probability Distribution	Standard Uncertainty	Sensitivity Coefficient	Uncertainty Contribution
X_i		x_i			$u(x_i)$	c_i	$u(y)$
Reference							
Fluid	Viscosity	7.534	mPa·s	Normal	0.037	1	0.038
	Calibration		s	Normal	0.005	0.55	0.0027
Time	Resolution	41.68	s	Rectangular	0.0029	0.55	0.0016
	Repeatability		s	Normal	0.13	0.55	0.071
Temperature	Calibration		K	Normal	0.010	0.018	0.00018
	Resolution	293.15	K	Rectangular	0.0029	0.018	0.000051
	Uniformity		K	Rectangular	0.029	0.018	0.00051
	Stability		K	Rectangular	0.014	0.018	0.00025
Pressure	Calibration		MPa	Normal	0.0060	0.000099	0.0000006
	Resolution	60	MPa	Rectangular	0.0029	0.000099	0.0000003
	Stability		MPa	Rectangular	0.014	0.000099	0.0000014
Density	Solid	7673	kg·m ⁻³	Normal	17	0.0011	0.019
	Fluid	1007.7	kg·m ⁻³	Normal	0.005	0.0011	0.0000057
Calibration							
function			mPa·s	Normal	0.0404	1	0.040
coefficients							
Standard Uncertainty		mPa·s				$u(y)$	± 0.092
Expanded Uncertainty		mPa·s		($k=2$)		$U(y)$	± 0.18
Relative Expanded Uncertainty (mPa·s/mPa·s)					($k=2$)	$U_r(y)$	$\pm \mathbf{0.025}$

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, 6 and 7, respectively.

Table 5. Experimental densities, ρ , for DEA (1) + H₂O (2) mixtures at different conditions of temperature, T , pressure, p , and mass fraction, w_1 .^a

$\rho/\text{kg}\cdot\text{m}^{-3}$						
T/K						
p/MPa	293.15	313.15	333.15	353.15	373.15	393.15
$w_1 = 0.0993$						
0.1	1009.4	1002.9	993.4	981.9	968.1	952.6
0.5	1009.5	1003.1	993.7	982.0	968.3	952.9
1	1009.7	1003.4	993.9	982.2	968.6	953.1
2	1010.2	1003.7	994.3	982.6	969.1	953.6
5	1011.4	1005.0	995.5	983.9	970.4	955.1
10	1013.6	1007.0	997.6	986.1	972.7	957.5
15	1015.6	1009.0	999.6	988.2	974.9	959.9
20	1017.7	1011.0	1001.6	990.3	977.1	962.3
30	1021.8	1015.1	1005.7	994.4	981.4	966.8
40	1025.9	1019.1	1009.7	998.5	985.7	971.3
50	1029.7	1022.8	1013.4	1002.4	989.7	975.4
60	1033.6	1026.6	1017.2	1006.2	993.6	979.7
70	1037.6	1030.4	1020.9	1010.0	997.5	984.0
80	1041.1	1033.9	1024.5	1013.7	1001.4	987.9
90	1044.8	1037.3	1028.0	1017.3	1005.1	991.7

100	1048.5	1040.9	1031.5	1020.9	1008.8	995.5
110	1051.9	1044.4	1034.9	1024.4	1012.4	999.4
120	1055.4	1047.7	1038.4	1027.8	1015.9	1003.1
130	1059.0	1051.1	1041.7	1031.1	1019.4	1006.6
140	1062.2	1054.4	1045.0	1034.4	1022.7	1010.1

$w_1 = 0.2000$

0.1	1021.6	1014.3	1004.2	992.4	978.3	962.8
0.5	1021.7	1014.4	1004.4	992.5	978.6	963.2
1	1021.9	1014.6	1004.6	992.7	978.8	963.3
2	1022.3	1015.0	1005.0	993.1	979.3	963.8
5	1023.5	1016.2	1006.2	994.4	980.6	965.2
10	1025.5	1018.2	1008.2	996.5	982.8	967.7
15	1027.4	1020.1	1010.2	998.6	985.0	969.9
20	1029.4	1022.0	1012.1	1000.6	987.2	972.3
30	1033.3	1025.8	1016.0	1004.7	991.5	976.8
40	1037.0	1029.7	1020.0	1008.5	995.6	981.3
50	1040.7	1033.2	1023.6	1012.4	999.5	985.4
60	1044.4	1036.9	1027.1	1016.3	1003.5	989.6
70	1048.0	1040.4	1030.8	1019.8	1007.3	993.8
80	1051.4	1043.8	1034.2	1023.4	1011.1	997.6
90	1054.9	1047.1	1037.5	1026.9	1014.7	1001.5
100	1058.4	1050.6	1041.0	1030.4	1018.3	1005.2
110	1061.6	1053.9	1044.2	1033.8	1021.9	1008.9
120	1064.9	1057.1	1047.7	1037.1	1025.3	1012.6
130	1068.3	1060.3	1050.9	1040.5	1028.7	1016.1
140	1071.4	1063.5	1054.1	1043.6	1032.0	1019.5

$w_1 = 0.3002$

0.1	1034.5	1026.0	1015.4	1003.0	988.7	972.9
0.5	1034.7	1026.2	1015.5	1003.1	988.9	973.2
1	1034.8	1026.4	1015.7	1003.4	989.2	973.4
2	1035.2	1026.7	1016.2	1003.7	989.6	973.9
5	1036.3	1027.9	1017.3	1005.0	990.9	975.3
10	1038.2	1029.7	1019.3	1007.1	993.2	977.7
15	1040.0	1031.6	1021.2	1009.1	995.3	980.0
20	1041.8	1033.4	1023.1	1011.1	997.4	982.4
30	1045.4	1037.1	1026.9	1015.1	1001.6	986.9
40	1049.0	1040.7	1030.6	1018.9	1005.7	991.2
50	1052.5	1044.1	1034.1	1022.6	1009.5	995.3
60	1055.9	1047.6	1037.7	1026.4	1013.4	999.4
70	1059.3	1051.0	1041.1	1029.9	1017.2	1003.6
80	1062.5	1054.3	1044.4	1033.4	1020.9	1007.3
90	1065.8	1057.5	1047.7	1036.8	1024.4	1011.2
100	1069.1	1060.8	1051.1	1040.1	1028.0	1014.8
110	1072.1	1063.9	1054.3	1043.5	1031.5	1018.5
120	1075.3	1067.1	1057.5	1046.8	1034.9	1022.1
130	1078.5	1070.2	1060.6	1050.0	1038.2	1025.5
140	1081.4	1073.3	1063.7	1053.1	1041.5	1029.1

$$w_1 = 0.3997$$

0.1	1047.5	1038.0	1026.6	1013.9	999.2	983.2
0.5	1047.7	1038.2	1026.8	1014.0	999.4	983.5
1	1047.9	1038.4	1027.0	1014.2	999.6	983.7
2	1048.2	1038.7	1027.4	1014.5	1000.1	984.2
5	1049.3	1039.8	1028.5	1015.8	1001.4	985.6
10	1051.0	1041.7	1030.4	1017.8	1003.6	988.0

15	1052.8	1043.4	1032.3	1019.8	1005.7	990.3
20	1054.5	1045.1	1034.2	1021.8	1007.8	992.7
30	1058.0	1048.7	1037.8	1025.7	1012.0	997.1
40	1061.4	1052.3	1041.6	1029.4	1016.0	1001.4
50	1064.7	1055.6	1045.0	1033.0	1019.8	1005.4
60	1068.0	1059.0	1048.4	1036.7	1023.6	1009.6
70	1071.2	1062.2	1051.8	1040.2	1027.3	1013.6
80	1074.3	1065.4	1055.0	1043.6	1031.0	1017.3
90	1077.4	1068.5	1058.2	1047.0	1034.5	1021.2
100	1080.6	1071.6	1061.5	1050.4	1038.0	1024.8
110	1083.5	1074.7	1064.6	1053.6	1041.4	1028.5
120	1086.5	1077.7	1067.8	1056.8	1044.8	1032.0
130	1089.6	1080.7	1070.8	1060.0	1048.1	1035.4
140	1092.4	1083.6	1074.0	1063.0	1051.3	1038.9

^a Standard uncertainties ($k=1$): $u(T) = 0.01$ K; $u_r(p) = 0.0001$; $u(w) = 0.0001$ and $u(\rho) = 0.35$

$\text{kg}\cdot\text{m}^{-3}$

Table 6. Experimental densities, ρ , for TEA (1) + H₂O (2) mixtures at different conditions of temperature, T , pressure, p , and mass fraction, w_1 .^a

$\rho/\text{kg}\cdot\text{m}^{-3}$						
T/K						
p/MPa	293.15	313.15	333.15	353.15	373.15	393.15
$w_1 = 0.0992$						
0.1	1013.3	1006.9	997.2	985.7	971.7	956.1
0.5	1013.5	1007.0	997.5	985.7	971.9	956.4
1	1013.7	1007.2	997.7	985.9	972.2	956.6
2	1014.2	1007.6	998.1	986.4	972.6	957.1
5	1015.5	1008.9	999.3	987.7	973.9	958.6
10	1017.6	1010.9	1001.4	989.9	976.3	961.1
15	1019.6	1013.0	1003.5	992.1	978.5	963.4
20	1021.8	1015.0	1005.5	994.1	980.6	965.9
30	1026.0	1019.1	1009.6	998.2	985.1	970.4
40	1030.0	1023.0	1013.6	1002.4	989.4	974.9
50	1034.0	1026.8	1017.5	1006.2	993.4	979.2
60	1037.9	1030.7	1021.3	1010.2	997.4	983.5
70	1041.8	1034.4	1024.8	1014.0	1001.4	987.7
80	1045.5	1038.0	1028.6	1017.6	1005.3	991.7
90	1049.1	1041.6	1032.0	1021.2	1009.1	995.7
100	1052.9	1045.2	1035.6	1024.8	1012.7	999.4
110	1056.3	1048.6	1039.0	1028.4	1016.4	1003.3
120	1059.8	1052.0	1042.5	1031.8	1019.9	1006.9
130	1063.3	1055.3	1045.8	1035.2	1023.3	1010.4
140	1066.5	1058.5	1049.1	1038.4	1026.8	1014.1
$w_1 = 0.2000$						

0.1	1029.5	1021.9	1011.6	999.5	985.1	969.2
0.5	1029.5	1022.1	1011.8	999.6	985.3	969.5
1	1029.7	1022.3	1012.0	999.8	985.5	969.7
2	1030.2	1022.7	1012.4	1000.1	986.1	970.2
5	1031.4	1023.9	1013.6	1001.5	987.4	971.7
10	1033.4	1025.9	1015.7	1003.7	989.7	974.2
15	1035.4	1027.8	1017.7	1005.8	991.9	976.5
20	1037.4	1029.7	1019.7	1007.8	994.0	978.9
30	1041.4	1033.7	1023.6	1011.9	998.4	983.5
40	1045.2	1037.6	1027.7	1015.9	1002.6	988.0
50	1049.0	1041.2	1031.4	1019.8	1006.7	992.3
60	1052.7	1044.9	1035.0	1023.7	1010.6	996.5
70	1056.4	1048.5	1038.6	1027.4	1014.5	1000.7
80	1059.8	1051.9	1042.1	1031.1	1018.5	1004.6
90	1063.4	1055.3	1045.6	1034.5	1022.1	1008.6
100	1066.9	1058.8	1049.1	1038.1	1025.7	1012.3
110	1070.2	1062.1	1052.3	1041.5	1029.4	1016.1
120	1073.6	1065.6	1055.8	1044.9	1032.9	1019.8
130	1077.0	1068.7	1059.0	1048.2	1036.2	1023.4
140	1080.1	1071.9	1062.4	1051.5	1039.7	1026.9

$$w_1 = 0.2991$$

0.1	1046.1	1037.4	1026.3	1013.6	999.2	982.9
0.5	1046.2	1037.5	1026.5	1013.7	999.3	983.3
1	1046.4	1037.7	1026.7	1013.9	999.6	983.5
2	1046.8	1038.1	1027.2	1014.3	1000.1	984.0
5	1047.9	1039.3	1028.3	1015.6	1001.5	985.5

10	1049.8	1041.2	1030.4	1017.8	1003.8	988.0
15	1051.7	1043.1	1032.4	1019.9	1006.0	990.3
20	1053.7	1045.0	1034.3	1021.9	1008.2	992.7
30	1057.4	1048.8	1038.2	1025.9	1012.5	997.3
40	1061.1	1052.6	1042.1	1029.9	1016.8	1001.8
50	1064.7	1056.1	1045.7	1033.8	1020.7	1006.0
60	1068.3	1059.7	1049.4	1037.6	1024.6	1010.2
70	1071.8	1063.2	1052.9	1041.2	1028.4	1014.4
80	1075.1	1066.6	1056.3	1044.8	1032.4	1018.3
90	1078.5	1069.9	1059.7	1048.3	1036.0	1022.3
100	1081.8	1073.3	1063.2	1051.8	1039.5	1026.1
110	1084.9	1076.4	1066.3	1055.2	1043.1	1029.8
120	1088.2	1079.6	1069.8	1058.5	1046.8	1033.4
130	1091.5	1082.8	1072.9	1061.8	1050.1	1036.9
140	1094.5	1085.9	1076.0	1065.0	1053.4	1040.5

$w_1 = 0.4000$

0.1	1065.2	1055.1	1043.1	1029.7	1014.6	998.1
0.5	1065.3	1055.3	1043.3	1029.8	1014.8	998.5
1	1065.4	1055.4	1043.5	1030.0	1015.0	998.6
2	1065.8	1055.8	1043.9	1030.4	1015.5	999.1
5	1066.9	1056.9	1045.1	1031.7	1016.8	1000.6
10	1068.7	1058.8	1047.1	1033.9	1019.1	1003.1
15	1070.5	1060.6	1049.0	1035.8	1021.2	1005.4
20	1072.3	1062.4	1050.9	1037.9	1023.4	1007.8
30	1075.8	1066.0	1054.7	1041.9	1027.7	1012.4
40	1079.3	1069.7	1058.4	1045.8	1032.0	1016.7
50	1082.7	1073.2	1062.0	1049.5	1035.7	1021.0

60	1086.1	1076.6	1065.6	1053.3	1039.7	1025.2
70	1089.4	1079.9	1069.0	1056.9	1043.5	1029.3
80	1092.5	1083.1	1072.3	1060.4	1047.3	1033.1
90	1095.8	1086.3	1075.5	1063.8	1050.8	1037.1
100	1098.9	1089.5	1078.9	1067.2	1054.3	1040.7
110	1101.8	1092.7	1082.0	1070.5	1057.9	1044.5
120	1104.9	1095.7	1085.3	1073.9	1061.3	1048.1
130	1107.9	1098.7	1088.3	1077.1	1064.6	1051.5
140	1110.8	1101.7	1091.4	1080.1	1067.8	1055.0

^a Standard uncertainties ($k=1$): $u(T) = 0.01$ K; $u_r(p) = 0.0001$; $u(w) = 0.0001$ and $u(\rho) = 0.35$

$\text{kg}\cdot\text{m}^{-3}$

Table 7. Experimental densities, ρ , for DMAE (1) + H₂O (2) mixtures at different conditions of temperature, T , pressure, p , and mass fraction, w_1 .^a

$\rho/\text{kg}\cdot\text{m}^{-3}$						
T/K						
p/MPa	293.15	313.15	333.15	353.15	373.15	393.15
$w_1 = 0.1005$						
0.1	994.5	987.4	977.4	965.2	950.7	934.5
0.5	994.7	987.6	977.6	965.3	950.9	934.9
1	994.8	987.8	977.8	965.6	951.2	935.1
2	995.3	988.3	978.2	966.0	951.7	935.6
5	996.5	989.5	979.5	967.3	953.0	937.1
10	998.5	991.5	981.5	969.5	955.4	939.7
15	1000.5	993.4	983.6	971.6	957.7	942.1
20	1002.6	995.4	985.6	973.8	959.9	944.6

30	1006.6	999.4	989.7	978.0	964.4	949.3
40	1010.5	1003.4	993.7	982.0	968.7	953.9
50	1014.3	1007.0	997.3	986.0	972.8	958.2
60	1018.1	1010.8	1001.1	989.9	976.8	962.6
70	1021.8	1014.5	1004.9	993.6	980.9	966.9
80	1025.3	1018.0	1008.4	997.4	984.7	971.0
90	1029.0	1021.4	1012.0	1000.9	988.6	975.0
100	1032.5	1024.9	1015.4	1004.6	992.1	978.8
110	1035.9	1028.3	1018.9	1008.1	995.9	982.7
120	1039.3	1031.6	1022.3	1011.7	999.5	986.5
130	1042.7	1035.0	1025.5	1014.9	1003.0	990.0
140	1045.7	1038.2	1028.8	1018.3	1006.3	993.7

$w_1 = 0.2020$

0.1	992.6	983.2	971.5	958.1	942.4	925.2
0.5	992.7	983.4	971.8	958.1	942.7	925.5
1	992.9	983.6	972.0	958.4	942.9	925.7
2	993.3	984.0	972.4	958.8	943.4	926.3
5	994.4	985.2	973.7	960.1	944.8	927.9
10	996.3	987.2	975.7	962.4	947.2	930.5
15	998.2	989.1	977.7	964.5	949.5	933.0
20	1000.0	990.9	979.7	966.7	951.8	935.6
30	1003.7	994.8	983.7	971.0	956.3	940.5
40	1007.4	998.6	987.7	975.0	960.9	945.2
50	1010.8	1002.1	991.3	978.9	964.9	949.6
60	1014.3	1005.6	995.0	982.7	968.9	954.1
70	1017.9	1009.1	998.6	986.6	973.0	958.5

80	1021.1	1012.5	1002.1	990.1	976.9	962.4
90	1024.4	1015.7	1005.4	993.8	980.6	966.6
100	1027.7	1019.1	1008.9	997.3	984.3	970.5
110	1030.8	1022.3	1012.2	1000.7	988.0	974.3
120	1033.9	1025.6	1015.5	1004.1	991.6	978.2
130	1037.2	1028.6	1018.6	1007.5	995.0	981.7
140	1040.1	1031.7	1021.9	1010.8	998.4	985.3

$w_1 = 0.3005$

0.1	990.6	978.8	965.2	950.2	933.3	915.0
0.5	990.7	979.0	965.4	950.3	933.6	915.5
1	990.9	979.2	965.6	950.6	933.9	915.7
2	991.3	979.6	966.1	951.0	934.4	916.3
5	992.4	980.8	967.3	952.4	935.9	917.9
10	994.2	982.7	969.5	954.7	938.4	920.8
15	996.0	984.6	971.5	956.9	940.8	923.4
20	997.8	986.5	973.5	959.1	943.2	926.1
30	1001.4	990.3	977.5	963.4	947.9	931.2
40	1004.9	994.0	981.5	967.5	952.4	936.2
50	1008.2	997.5	985.2	971.6	956.6	940.8
60	1011.6	1001.0	988.9	975.6	960.9	945.3
70	1014.9	1004.4	992.5	979.3	965.0	949.8
80	1018.0	1007.6	996.0	983.0	969.1	953.9
90	1021.2	1010.9	999.2	986.6	972.8	958.1
100	1024.3	1014.1	1002.7	990.1	976.4	962.2
110	1027.2	1017.3	1006.0	993.6	980.3	966.0
120	1030.2	1020.4	1009.3	997.0	983.8	969.9
130	1033.3	1023.4	1012.4	1000.3	987.3	973.5

140	1036.0	1026.4	1015.4	1003.5	990.7	977.2
$w_1 = 0.3995$						
0.1	986.6	972.7	957.4	941.1	923.1	903.9
0.5	986.7	972.9	957.6	941.2	923.4	904.3
1	986.9	973.1	957.9	941.4	923.6	904.6
2	987.3	973.5	958.3	941.9	924.2	905.2
5	988.4	974.7	959.6	943.3	925.8	907.0
10	990.3	976.7	961.8	945.8	928.4	910.0
15	992.1	978.6	963.9	948.1	931.1	912.8
20	994.0	980.6	966.0	950.5	933.6	915.7
30	997.5	984.4	970.2	954.9	938.5	921.0
40	1001.0	988.2	974.2	959.2	943.3	926.3
50	1004.4	991.7	977.9	963.3	947.6	931.0
60	1007.7	995.3	981.7	967.5	952.1	935.9
70	1011.0	998.7	985.4	971.2	956.2	940.5
80	1014.1	1002.0	988.9	975.1	960.4	944.8
90	1017.3	1005.3	992.3	978.7	964.3	949.2
100	1020.4	1008.5	995.8	982.4	968.1	953.2
110	1023.2	1011.7	999.0	985.9	971.9	957.3
120	1026.3	1014.7	1002.4	989.3	975.6	961.2
130	1029.2	1017.7	1005.5	992.7	979.1	964.9
140	1031.9	1020.7	1008.7	996.0	982.6	968.9

^a Standard uncertainties ($k=1$): $u(T) = 0.01$ K; $u_r(p) = 0.0001$; $u(w) = 0.0001$ and $u(\rho) = 0.35$

$\text{kg}\cdot\text{m}^{-3}$

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 0.8% to 3.3% is observed for DMAE solutions, the lowest effect is produced at $T = 293.15$ K and $p = 0.1$ MPa and the highest one at $T = 393.15$ K and $p = 0.1$ MPa.

The experimental values were correlated using a modified Tammann–Tait equation (Eq. (3))

for each composition:

$$\rho(T, p) = \frac{A_0 + A_1T + A_2T^2}{1 - C \ln \left(\frac{B_0 + B_1T + B_2T^2 + p}{B_0 + B_1T + B_2T^2 + 0.1 \text{MPa}} \right)} \quad (3)$$

The fitting results are shown in Table 8, which contains the adjustable parameters and the standard deviation of the adjustment (σ).

Table 8. Fitting parameters of Eq. (3) and standard deviations σ for the density measurements

DEA (1) + H ₂ O (2)	$w_1 = 0.1$	$w_1 = 0.2$	$w_1 = 0.3$	$w_1 = 0.4$
$A_0/\text{kg}\cdot\text{m}^{-3}$	857.266	902.241	952.792	1004.967
$A_1/\text{kg}\cdot\text{m}^{-3}\cdot\text{K}^{-1}$	1.3308	1.1496	0.94477	0.7320
$A_2/\text{kg}\cdot\text{m}^{-3}\cdot\text{K}^{-2}$	-0.00278	-0.00253	-0.00227	-0.00200
B_0/MPa	-547.316	-339.567	-86.929	124.817
$B_1/\text{MPa}\cdot\text{K}^{-1}$	5.4058	4.3792	3.0910	2.0025
$B_2/\text{MPa}\cdot\text{K}^{-2}$	-0.00857	-0.00732	-0.00568	-0.00431
C	0.12973	0.12623	0.12268	0.11924
$\sigma/\text{kg}\cdot\text{m}^{-3}$	0.170	0.141	0.0952	0.0855
TEA (1) + H ₂ O (2)	$w_1 = 0.1$	$w_1 = 0.2$	$w_1 = 0.3$	$w_1 = 0.4$
$A_0/\text{kg}\cdot\text{m}^{-3}$	861.437	908.717	970.071	1033.299
$A_1/\text{kg}\cdot\text{m}^{-3}\cdot\text{K}^{-1}$	1.3335	1.1689	0.92200	0.68858
$A_2/\text{kg}\cdot\text{m}^{-3}\cdot\text{K}^{-2}$	-0.00278	-0.00258	-0.002261	-0.001979
B_0/MPa	-557.630	-357.625	-104.118	169.334
$B_1/\text{MPa}\cdot\text{K}^{-1}$	5.4148	4.4421	3.0708	1.5930
$B_2/\text{MPa}\cdot\text{K}^{-2}$	-0.00857	-0.00739	-0.00559	-0.00366

C	0.12746	0.12533	0.12033	0.11368
$\sigma/\text{kg}\cdot\text{m}^{-3}$	0.164	0.146	0.130	0.0915
DMAE (1) + H ₂ O (2)	$w_1 = 0.1$	$w_1 = 0.2$	$w_1 = 0.3$	$w_1 = 0.4$
$A_0/\text{kg}\cdot\text{m}^{-3}$	845.842	905.967	978.647	1041.673
$A_1/\text{kg}\cdot\text{m}^{-3}\cdot\text{K}^{-1}$	1.3335	1.0172	0.63224	0.28630
$A_2/\text{kg}\cdot\text{m}^{-3}\cdot\text{K}^{-2}$	-0.00282	-0.00246	-0.00202	-0.00162
B_0/MPa	-376.874	83.307	427.581	589.461
$B_1/\text{MPa}\cdot\text{K}^{-1}$	4.5051	1.9517	-0.06778	-1.1043
$B_2/\text{MPa}\cdot\text{K}^{-2}$	-0.00751	-0.00417	-0.00145	-0.00000733
C	0.12745	0.11819	0.10912	0.10348
$\sigma/\text{kg}\cdot\text{m}^{-3}$	0.162	0.119	0.0849	0.0881

Experimental densities were compared with literature data available for us and in the same conditions that our measurements (mainly at atmospheric pressure). For aqueous DEA solutions, Rinker et al. [12] measured at $w_1 = (0.1, 0.2, 0.3)$ and $T = (293.15, 313.15, 333.15, 353.15, 373.15)$ K, Hsu et al. [13] at $w_1 = (0.2, 0.3)$ and $T = (313.15, 333.15, 353.15)$ K, Tseng et al. [14] at $w_1 = (0.1 \text{ to } 0.9)$ and $T = (293.15, 303.15)$ K, Yang et al. [15] at $w_1 = (0.1 \text{ to } 0.9)$ and $T = (283.15 \text{ to } 353.15)$ K, Spasojevic et al. [16] measured at $w_1 = (0.05, 0.1, 0.15, 0.2, 0.25)$ and $T = (298.15 \text{ to } 343.15)$ K, Han et al. [17] measured at $w_1 = (0.3 \text{ to } 1)$ and $T = (298.15 \text{ to } 423.15)$ K. For aqueous TEA solutions, Tseng et al. [14] measured at $w_1 = (0.1 \text{ to } 0.9)$ and $T = (293.15, 303.15)$ K, Zhao et al. [18] measured at $w_1 = 0.3$ and $T = (303.15 \text{ to } 343.15)$ K. For aqueous DMAE solutions, Zhang et al. [19] measured at $w_1 = (0.11, 0.21, 0.31)$ and $T = (293.15, 303.15, 313.15)$ K, Maham et al. [20] measured at $x_1 = (0.1 \text{ to } 0.9)$ and $T = (278.15 \text{ to } 353.15)$ K, Zhang et al. [21] at $w_1 = (0.15, 0.3, 0.45)$ and $T = (298.15 \text{ to } 353.15)$ K.

353.15) K, Chowdhury et al. [22] at $x_1 = (0.1 \text{ to } 0.9)$ and $T = (303.15 \text{ to } 323.15)$ K, Bernal-García et al. [23] measured at $x_1 = (0 \text{ to } 1)$ and $T = (293.15 \text{ to } 363.15)$ K.

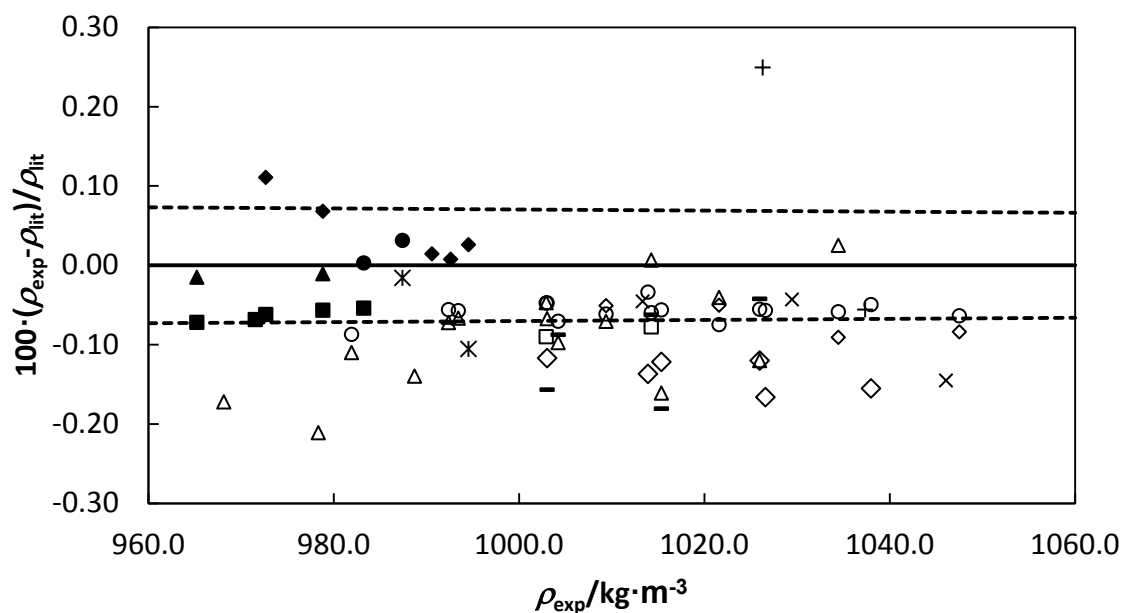


Figure 1. Relative deviations of density measurements (ρ_{exp}) in comparison with literature values (ρ_{lit}): (Δ) Rinker et al. [12], ($-$)Hsu et al. [13], (\diamond) Tseng et al. [14], (\circ)Yang et al. [15] (\square) Spasojevic et al. [16] and (\diamond) Han et al. [17] for DEA aqueous solutions; (\times) Tseng et al. [14] and ($+$) Zhao et al. [18] for TEA aqueous solutions; (\blacklozenge) Zhang et al. [1947], (\blacksquare) Maham et al. [20], (\blacktriangle) Zhang et al. [21], (\bullet) Chowdhury et al. [22] and ($*$) Bernal–García et al. [23] for DMAE aqueous solutions. Dotted lines represent the relative expanded uncertainty of our measurements.

The relative deviations of our measurements and literature values are plotted in Figure 1. Average absolute deviations for DEA-water mixtures are 0.09% from [12], 0.10% from [13], 0.07% from [14], 0.06% from [15], 0.7% from [16] (the disagreement occurs at 80°C) and 0.29% from [17]. As regards TEA-water mixtures, average absolute deviation is 0.09% from [14] and 0.15% from [18]. Finally for DMEA-water mixtures, it is 0.05% from [1947], 0.07%

from [2018], 0.06% from [2149], 0.02% from [2220] and 0.06% from [23]. These average values are close to the uncertainty of our measurements.

Viscosity measurements of DEA-water, DMAE-water and TEA-water mixtures were carried out at amine weight fractions $w_1 = 0.10, 0.20, 0.30$ and 0.40 , $p = (0.1 \text{ to } 100)$ MPa and $T = (293.15, 313.15, 333.15, 353.15, 393.15)$ K, using the falling body viscometer. These measurements are listed in Tables 9-11.

Table 9. Experimental viscosities, η , for DEA (1) + H₂O (2) mixtures at different conditions of temperature, T , pressure, p , and mass fraction, w_1 .^a

$\eta/\text{mPa}\cdot\text{s}$						
T/K						
p/MPa	293.15	313.15	333.15	353.15	373.15	393.15
$w_1 = 0.0993$						
0.1	1.4930	0.9237	0.6268	0.4595		
5	1.4899	0.9150	0.6258	0.4583	0.3586	0.2926
10	1.4858	0.9172	0.6274	0.4614	0.3611	0.2943
15	1.4900	0.9188	0.6269	0.4631	0.3635	0.2964
20	1.4870	0.9192	0.6303	0.4640	0.3652	0.2989
25	1.4875	0.9215	0.6312	0.4671	0.3672	0.3010
30	1.4871	0.9226	0.6339	0.4692	0.3689	0.3024
40	1.4862	0.9253	0.6383	0.4733	0.3724	0.3047
60	1.4873	0.9334	0.6452	0.4815	0.3807	0.3113
80	1.4899	0.9414	0.6532	0.4886	0.3871	0.3178
100	1.4944	0.9509	0.6623	0.4971	0.3943	0.3233
$w_1 = 0.2000$						
0.1	2.2185	1.3042	0.8581	0.6088		

5	2.2149	1.2920	0.8575	0.6083	0.4622	0.3644
10	2.2135	1.2960	0.8602	0.6136	0.4646	0.3676
15	2.2172	1.3011	0.8632	0.6167	0.4670	0.3700
20	2.2169	1.3033	0.8673	0.6198	0.4698	0.3724
25	2.2178	1.3087	0.8717	0.6239	0.4729	0.3748
30	2.2199	1.3120	0.8763	0.6271	0.4761	0.3769
40	2.2212	1.3178	0.8845	0.6333	0.4820	0.3818
60	2.2424	1.3346	0.8989	0.6460	0.4920	0.3919
80	2.2596	1.3525	0.9136	0.6588	0.5046	0.4017
100	2.2773	1.3707	0.9264	0.6731	0.5153	0.4114

$w_1 = 0.3002$

0.1	3.6330	1.9799	1.2470	0.8528		
5	3.6166	1.9760	1.2433	0.8459	0.6145	0.4675
10	3.6346	1.9831	1.2484	0.8544	0.6194	0.4721
15	3.6339	1.9928	1.2539	0.8578	0.6247	0.4778
20	3.6607	2.0039	1.2576	0.8642	0.6304	0.4809
25	3.6764	2.0117	1.2655	0.8704	0.6353	0.4848
30	3.6878	2.0225	1.2707	0.8768	0.6417	0.4908
40	3.7139	2.0393	1.2827	0.8887	0.6515	0.4967
60	3.7657	2.0801	1.3120	0.9096	0.6676	0.5122
80	3.8176	2.1192	1.3373	0.9309	0.6851	0.5270
100	3.8744	2.1585	1.3650	0.9514	0.7019	0.5417

$w_1 = 0.3997$

0.1	6.2479	3.1677	1.8849	1.2260		
5	6.3506	3.1745	1.8723	1.2308	0.8545	0.6194
10	6.4164	3.2222	1.8864	1.2388	0.8630	0.6265
15	6.4632	3.2480	1.9001	1.2468	0.8727	0.6495

20	6.5335	3.2391	1.9116	1.2549	0.8808	0.6555
25	6.5982	3.2797	1.8964	1.2643	0.8890	0.6626
30	6.5996	3.3033	1.9434	1.2750	0.9000	0.6682
40	6.6832	3.3606	1.9733	1.2925	0.9123	0.6810
60	6.8533	3.4383	2.0288	1.3349	0.9445	0.7072
80	6.9727	3.5379	2.0858	1.3714	0.9763	0.7305
100	7.1686	3.6251	2.1361	1.4090	1.0033	0.7538

^a Standard uncertainties ($k=1$): $u(T) = 0.01$ K; $u_r(p) = 0.0001$; $u(w) = 0.0001$ and $u_r(\eta) = 0.016$

Table 10. Experimental viscosities, η , for DMAE (1) + H₂O (2) mixtures at different conditions of temperature, T , pressure, p , and mass fraction, w_1 .^a

$\eta/\text{mPa}\cdot\text{s}$						
T/K						
p/MPa	293.15	313.15	333.15	353.15	373.15	393.15
$w_1 = 0.1005$						
0.1	1.5589	0.9401	0.6336	0.4624		
5	1.5639	0.9415	0.6341	0.4644	0.3590	0.2917
10	1.5638	0.9437	0.6372	0.4671	0.3614	0.2952
15	1.5626	0.9453	0.6403	0.4699	0.3638	0.2973
20	1.5621	0.9476	0.6431	0.4720	0.3659	0.2998
25	1.5615	0.9506	0.6461	0.4744	0.3691	0.3019
30	1.5640	0.9525	0.6477	0.4772	0.3711	0.3036
40	1.5642	0.9570	0.6532	0.4824	0.3746	0.3078
60	1.5623	0.9672	0.6631	0.4910	0.3833	0.3147
80	1.5701	0.9775	0.6729	0.4999	0.3916	0.3220
100	1.5767	0.9881	0.6850	0.5096	0.3984	0.3308

$w_1 = 0.2020$

0.1	2.5334	1.3682	0.8725	0.6055		
5	2.5466	1.3743	0.8770	0.6064	0.4516	0.3533
10	2.5572	1.3819	0.8862	0.6135	0.4543	0.3557
15	2.5659	1.3921	0.8907	0.6195	0.4578	0.3585
20	2.5739	1.3950	0.8970	0.6229	0.4624	0.3624
25	2.5826	1.4004	0.9026	0.6275	0.4659	0.3660
30	2.5893	1.4117	0.9063	0.6338	0.4702	0.3687
40	2.6049	1.4232	0.9178	0.6411	0.4771	0.3736
60	2.6374	1.4505	0.9397	0.6590	0.4923	0.3862
80	2.6717	1.4795	0.9613	0.6779	0.5073	0.3985
100	2.7064	1.5078	0.9835	0.6942	0.5213	0.4097

$w_1 = 0.3005$

0.1	4.2109	2.0424	1.2166	0.8063		
5	4.2230	2.0627	1.2232	0.8120	0.5788	0.4325
10	4.2582	2.0835	1.2391	0.8213	0.5834	0.4367
15	4.2946	2.1028	1.2486	0.8321	0.5895	0.4425
20	4.3298	2.1206	1.2597	0.8407	0.5971	0.4483
25	4.3737	2.1420	1.2736	0.8485	0.6020	0.4536
30	4.4046	2.1613	1.2832	0.8560	0.6099	0.4597
40	4.4643	2.1972	1.3049	0.8750	0.6239	0.4697
60	4.5882	2.2735	1.3527	0.9078	0.6493	0.4890
80	4.7300	2.3479	1.3961	0.9395	0.6735	0.5076
100	4.8605	2.4244	1.4424	0.9726	0.6978	0.5265

$w_1 = 0.3995$

0.1	6.5871	2.9897	1.6512	1.0503		
5	6.5345	3.0311	1.6691	1.0589	0.7177	0.5209

10	6.5643	3.0745	1.6948	1.0740	0.7267	0.5259
15	6.8013	3.1208	1.7186	1.0899	0.7398	0.5368
20	6.8629	3.1641	1.7425	1.1054	0.7525	0.5458
25	6.8217	3.2088	1.7655	1.1184	0.7633	0.5538
30	7.0272	3.2532	1.7893	1.1346	0.7742	0.5613
40	6.8831	3.3419	1.8347	1.1671	0.7998	0.5780
60	7.5912	3.5189	1.9265	1.2254	0.8409	0.6086
80		3.6983	2.0190	1.2811	0.8827	0.6407
100		3.8810	2.1128	1.3375	0.9306	0.6716

^a Standard uncertainties ($k=1$): $u(T) = 0.01$ K; $u_r(p) = 0.0001$; $u(w) = 0.0001$ and $u_r(\eta) = 0.016$

Table 11. Experimental viscosities, η , for TEA (1) + H₂O (2) mixtures at different conditions of temperature, T , pressure, p , and mass fraction, w_1 .^a

$\eta/\text{mPa}\cdot\text{s}$						
T/K						
p/MPa	293.15	313.15	333.15	353.15	373.15	393.15
$w_1 = 0.0992$						
0.1	1.4285	0.9090	0.6124	0.4585		
5	1.4588	0.8937	0.6114	0.4510	0.3584	0.2913
10	1.4550	0.8956	0.6149	0.4541	0.3560	0.2912
15	1.4538	0.8964	0.6154	0.4547	0.3581	0.2933
20	1.4497	0.8965	0.6178	0.4578	0.3595	0.2950
25	1.4473	0.8977	0.6198	0.4595	0.3618	0.2963
30	1.4458	0.8992	0.6221	0.4619	0.3632	0.2988
40	1.4434	0.9027	0.6250	0.4660	0.3659	0.3022
60	1.4398	0.9100	0.6327	0.4738	0.3738	0.3080

80	1.4465	0.9177	0.6418	0.4813	0.3807	0.3138
100	1.4477	0.9261	0.6513	0.4891	0.3871	0.3200

$w_1 = 0.2000$

0.1	2.0375	1.2029	0.8083	0.5805		
5	2.0357	1.2084	0.8084	0.5825	0.4440	0.3551
10	2.0351	1.2102	0.8125	0.5878	0.4475	0.3579
15	2.0400	1.2135	0.8177	0.5898	0.4510	0.3633
20	2.0415	1.2201	0.8200	0.5937	0.4534	0.3620
25	2.0445	1.2248	0.8238	0.5975	0.4562	0.3674
30	2.0461	1.2277	0.8283	0.6006	0.4601	0.3659
40	2.0515	1.2310	0.8329	0.6068	0.4638	0.3704
60	2.0640	1.2507	0.8473	0.6185	0.4753	0.3798
80	2.0799	1.2763	0.8631	0.6313	0.4864	0.3888
100	2.0976	1.2819	0.8789	0.6459	0.4953	0.3982

$w_1 = 0.2991$

0.1	3.2075	1.7864	1.1472	0.8052		
5	3.2125	1.7972	1.1545	0.7994	0.5867	0.4515
10	3.2294	1.8079	1.1644	0.8046	0.5894	0.4583
15	3.1800	1.8184	1.1676	0.8110	0.5951	0.4629
20	3.2337	1.8321	1.1779	0.8144	0.5996	0.4675
25	3.2421	1.8348	1.1815	0.8210	0.6046	0.4703
30	3.2453	1.8463	1.1888	0.8266	0.6109	0.4738
40	3.2875	1.8609	1.2027	0.8374	0.6193	0.4822
60	3.3246	1.8927	1.2254	0.8576	0.6379	0.4973
80	3.3727	1.9329	1.2529	0.8785	0.6550	0.5121
100	3.4457	1.9796	1.2929	0.9019	0.6721	0.5378

$w_1 = 0.4000$

0.1	5.3166	2.7881	1.6702	1.1242		
5	5.3105	2.7938	1.6802	1.1253	0.7909	0.5873
10	5.3312	2.7970	1.6976	1.1358	0.8001	0.5948
15	5.3763	2.8283	1.7037	1.1465	0.8090	0.6019
20	5.4173	2.8522	1.7203	1.1537	0.8190	0.6090
25	5.4451	2.8656	1.7380	1.1636	0.8242	0.6136
30	5.4997	2.8892	1.7477	1.1735	0.8363	0.6246
40	5.5494	2.9279	1.7714	1.1932	0.8525	0.6362
60	5.6746	3.0276	1.8255	1.2293	0.8789	0.6588
80	5.7919	3.1153	1.8777	1.2658	0.9114	0.6835
100	5.9875	3.1926	1.9640	1.3055	0.9403	0.7054

^a Standard uncertainties ($k=1$): $u(T) = 0.01$ K; $u_r(p) = 0.0001$; $u(w) = 0.0001$ and $u_r(\eta) = 0.016$

Viscosities of aqueous DEA solutions are always higher than viscosities of aqueous TEA solutions at the same temperature, pressure and composition in all the studied ranges. On the other hand, viscosities of DMEA + water mixtures are greater than DEA solutions' viscosities at the same temperature and pressure for composition $w_1 = 0.1$, and in the whole composition and pressure ranges at $T = 293.15$ K. However, viscosity behavior is reversed, viscosities of DEA solutions are higher than DMAE solutions, as the amine concentration and temperature are increased in such a way that this occurs at $w_1 = 0.4$ in all the pressure range and temperatures from 313.15 K to 393.15 K.

Viscosity of the studied mixtures decreases when temperature is increased or pressure is decreased and these effects are similar for all amines. The viscosity decrease is around (79-92)% when temperature increases from 293.15 to 393.15 K and the viscosity increase ranges from 0.1% to 30% when pressure increases from (0.1 to 100) MPa, both effects are higher as amine weight fraction increases.

In addition, viscosities of these mixtures increase with the amine weight fraction and the effect is more significant at lowest temperature and highest pressure for all amines. For example, when the composition is increased from $w_1 = 0.1$ to $w_1 = 0.4$, viscosity of amine-water mixtures is tripled at $T = 293.15$ K and is nearly doubled at $T = 393.15$ K.

Viscosity data were correlated using the modified VFT model, Eq(4), which was successfully used by other authors [24].

$$\eta(T, p) = \exp [a + b \cdot p + (c + d \cdot p + e \cdot p^2)/(T - f)] \quad (4)$$

Fitting of the experimental viscosity data was performed applying the least-squares method contained in the MATLAB software [25]

The fitting results are given in Table 12 which contains the parameters and the standard deviation of the adjustment. The standard deviations are less than the uncertainty measurement of the experimental viscosities, which indicates that the model is appropriate for this type of mixtures.

Table 12. Fitting parameters of Eq. (4) and standard deviations σ for the viscosity measurements

DEA (1) + H ₂ O (2)	$w_1 = 0.1$	$w_1 = 0.2$	$w_1 = 0.3$	$w_1 = 0.4$
a	-3.5117	-3.7494	-3.8529	-3.7495
b/MPa ⁻¹	0.002575	0.002639	0.002711	0.002639
c/K	545.33	793.63	771.57	793.65
d/ K·MPa ⁻¹	-0.36956	-0.16544	-0.31100	-0.16546
e/ K·MPa ⁻²	0.000151	-0.000415	0.0000349	-0.000415
f/K	153.74	151.58	143.33	151.58
σ /mPa·s	0.0019	0.039	0.022	0.069
TEA (1) + H ₂ O (2)	$w_1 = 0.1$	$w_1 = 0.2$	$w_1 = 0.3$	$w_1 = 0.4$

a	-3.4895	-3.61800	-3.810981	-3.9315
b/MPa ⁻¹	0.002509	0.002552	0.002968	0.002895
c/K	537.45	639.05	760.99	863.98
d/ K·MPa ⁻¹	-0.37414	-0.32546	-0.36853	-0.25878
e/ K·MPa ⁻²	0.000266	0.000017	0.000238	-0.000011
f/K	153.99	145.33	140.417	139.01
σ /mPa·s	0.0042	0.0035	0.017	0.026
DMAE (1) + H ₂ O (2)	$w_1 = 0.1$	$w_1 = 0.2$	$w_1 = 0.3$	$w_1 = 0.4$
a	-3.5258	-3.6277	-3.6950	-4.0709
b/MPa ⁻¹	0.002863	0.002900	0.0028229	0.003133
c/K	540.25	598.04	642.32	797.10
d/ K·MPa ⁻¹	-0.36867	-0.27713	-0.13558	-0.06493
e/ K·MPa ⁻²	-0.000045	-0.000168	-0.000339	-0.000357
f/K	157.07	162.05	167.96	159.16
σ /mPa·s a	0.0022	0.012	0.015	0.057

In order to check the reliability of viscosity data, a comparison was carried out at atmospheric pressure (due to the lack of these values at high pressures) between viscosities obtained with our falling body viscometer and the ones obtained using a Stabinger SVM 3000 viscometer available in our laboratory. The results of the comparison are summarized in Table 13 and they are plotted in Figure 4.

Table 13. Viscosity comparison between falling body viscometer (η_{FB})^a and Stabinger SVM 3000 viscometer (η_{SV})^b at $p = 0.1$ MPa.

w_1	T/K	$\eta_{FB}/\text{mPa}\cdot\text{s}$	$\eta_{SV}/\text{mPa}\cdot\text{s}$	$\Delta\eta/\eta_{FB}$	w_1	T/K	$\eta_{FB}/\text{mPa}\cdot\text{s}$	$\eta_{SV}/\text{mPa}\cdot\text{s}$	$\Delta\eta/\eta_{FB}$
DEA (1) + H ₂ O (2)									
0.1	293.15	1.493	1.486	0.005	0.3	293.15	3.633	3.621	0.003
0.1	313.15	0.924	0.914	0.010	0.3	313.15	1.980	1.977	0.001
0.1	333.15	0.627	0.640	-0.021	0.3	333.15	1.247	1.248	-0.001
0.1	353.15	0.460	0.474	-0.031	0.3	353.15	0.853	0.866	-0.016
0.2	293.15	2.219	2.287	-0.031	0.4	293.15	6.248	6.311	-0.010
0.2	313.15	1.304	1.343	-0.030	0.4	313.15	3.168	3.197	-0.009
0.2	333.15	0.858	0.871	-0.015	0.4	333.15	1.885	1.864	0.011
0.2	353.15	0.609	0.627	-0.079	0.4	353.15	1.226	1.244	-0.015
TEA (1) + H ₂ O (2)									
0.1	293.15	1.429	1.410	0.013	0.3	293.15	3.208	3.197	0.003
0.1	313.15	0.909	0.881	0.030	0.3	313.15	1.786	1.786	0.000
0.1	333.15	0.612	0.621	-0.014	0.3	333.15	1.147	1.173	-0.022
0.1	353.15	0.459	0.469	-0.023	0.3	353.15	0.805	0.810	-0.006
0.2	293.15	2.038	2.066	-0.014	0.4	293.15	5.317	5.340	-0.004
0.2	313.15	1.203	1.244	-0.034	0.4	313.15	2.788	2.756	0.012
0.2	333.15	0.808	0.821	-0.016	0.4	333.15	1.670	1.669	0.001
0.2	353.15	0.581	0.569	0.019	0.4	353.15	1.124	1.125	-0.001
DMAE (1) + H ₂ O (2)									

0.1	293.15	1.559	1.587	-0.018	0.3	293.15	4.211	4.334	-0.029
0.1	313.15	0.940	0.947	-0.008	0.3	313.15	2.042	2.102	-0.029
0.1	333.15	0.634	0.623	0.017	0.3	333.15	1.217	1.226	-0.008
0.1	353.15	0.462	0.475	-0.027	0.3	353.15	0.806	0.809	-0.003
0.2	293.15	2.533	2.614	-0.032	0.4	293.15	6.587	6.754	-0.025
0.2	313.15	1.368	1.408	-0.029	0.4	313.15	2.990	3.068	-0.026
0.2	333.15	0.873	0.888	-0.018	0.4	333.15	1.651	1.671	-0.012
0.2	353.15	0.606	0.618	-0.020	0.4	353.15	1.050	1.044	0.006

^a Standard uncertainties ($k=1$): $u(T) = 0.01$ K; $u_r(p) = 0.0001$; $u(w) = 0.0001$; $u_r(\eta) = 0.016$

^b Standard uncertainties ($k=1$): $u(T) = 0.02$ K; $u_r(p) = 0.005$; $u(w) = 0.0001$; $u_r(\eta) = 0.01$

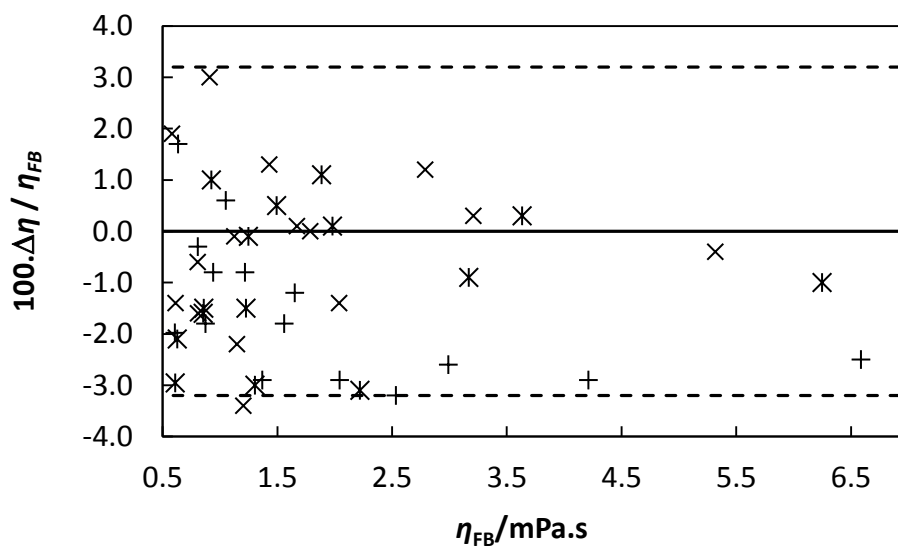


Figure 4. Relative viscosity deviation between falling body viscometer (η_{FB}) and Stabinger SVM 3000 viscometer (η_{SV}) for the mixtures: (x) DEA + water; (x) TEA + water) and (+) DMAE + water. Discontinuous lines represent the relative expanded uncertainty of our measurements.

In addition, our experimental data were also contrasted with the scarce literature data at atmospheric pressure [12,16,18,21,22,26], we have selected those data measured in the same conditions of composition and temperature as our data. For aqueous DEA solutions, Rinker et al. [12] measured at $w_1 = (0.1, 0.2, 0.3)$ and $T = (293.15, 313.15, 333.15, 353.15, 373.15)$ K, Spasojevic et al. [16] measured at $w_1 = (0.05, 0.1, 0.15, 0.2, 0.25)$ and $T = (298.15 \text{ to } 343.15)$ K, Hsu et al. [26] measured at $w_1 = (0.1, 0.2)$ and $T = (313.15, 333.15, 353.15)$ K. For aqueous TEA solutions, Zhao et al. [18] measured at $w_1 = 0.3$ and $T = (303.15 \text{ to } 343.15)$ K. For aqueous DMAE solutions, Zhang et al. [21] at $w_1 = (0.15, 0.3, 0.45)$ and $T = (303.15 \text{ to } 353.15)$ K, and Chowdhury et al. [22] at $x_1 = (0.1 \text{ to } 0.9)$ and $T = (303.15 \text{ to } 323.15)$ K. Relative viscosity deviations are shown in Figure 5.

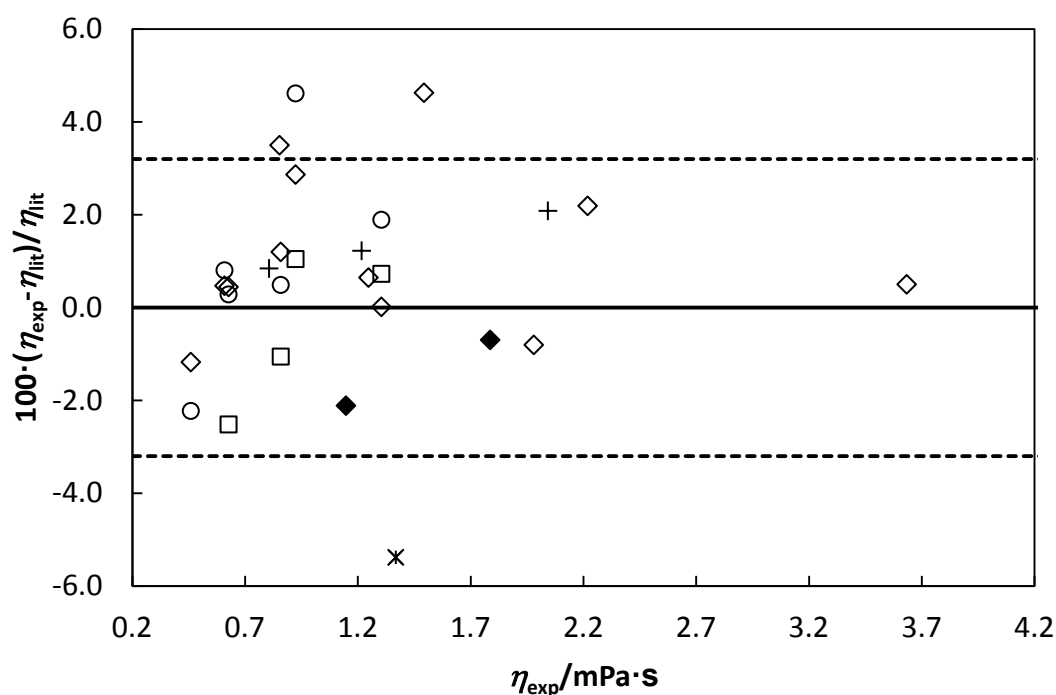


Figure 5. Relative viscosity deviation between our experimental data (η_{exp}) and literature values (η_{lit}): (\diamond) Rinker et al. [12], (\square) Spasojevic et al. [16], and (\circ) Hsu et al. [26] for DEA + water mixtures; (\blacklozenge) Zhao et al. [18] for TEA + water mixtures, and (+) Zhang et al. [21]

and (×) Chowdhury et al. [22] for DMAE + water mixtures. Dotted lines represent the relative expanded uncertainty of our measurements.

Average absolute deviations between our measurements and those reported in the literature are 1.5% in comparison with Rinker et al. [12], 1.3% with Spasojevic et al. [16], 1,7 % with Hsu et al. [26], all of them for DEA aqueous solutions, 2.0% with Trusler et al. [21] and Chowdhury et al. 5.4% [22] for DMEA solutions, this deviation might be due to the difference in composition as the authors measured at $w= 0.22$. Most values are in agreement with our uncertainties. There are other few viscosity measurements in the literature for the systems studied in this paper but they were measured at different compositions and it is not possible to compare the data.

4. Conclusions

Density and viscosity measurements of different amine aqueous solutions (DEA + H₂O, TEA+H₂O and DMAE +H₂O) were measured at amine mass fractions of 10%, 20%, 30% and 40%, and wide ranges of temperature and pressure. Densities increase for richer amine solutions for DEA and TEA but the contrary occurs for DMEA mixtures. A modified Tamman-Tait equation fits quite well the density as a function of pressure and temperature for a given composition.

In addition, viscosities of these mixtures increase with amine weight fraction being this effect more significant at low temperatures for all amines. They were successfully correlated using a modified VFT model.

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