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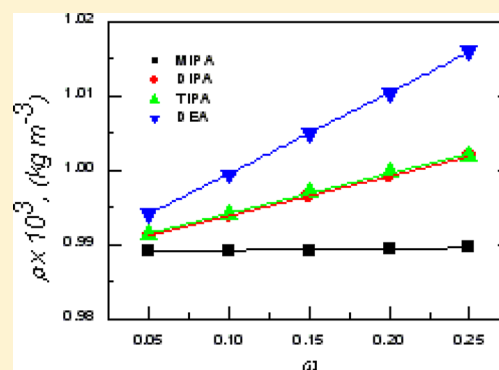
Densities, Viscosities, and Refractive Indices of Aqueous Alkanolamine Solutions as Potential Carbon Dioxide Removal Reagents

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ABSTRACT: To determine the possibility of improvement of the process of CO₂ removal by absorption with alkanolamines, densities, viscosities, and refractive indices of four aqueous alkanolamine solutions (water + monoisopropanolamine (MIPA), diisopropanolamine (DIPA), triisopropanolamine (TIPA), or diethanolamine (DEA)) were measured in the temperature range (298.15 to 343.15) K and at atmospheric pressure. Due to the corrosive effect of increased concentration of alkanolamine solutions, the mass fractions of alkanolamines were selected as $w = 0.05, 0.1, 0.15, 0.20,$ and 0.25 . The measured data were regressed with polynomial equations. Obtained results provide necessary data for industrial equipment design and process optimization regarding greenhouse gases (GHG), but also it is a foundation for further investigation of the potential replacement of usually used monoethanolamine with other mixtures of alkanolamine solutions presented within this work.



INTRODUCTION

Scientific investigations nowadays provide clear evidence that human activities have caused the significant concentration rise of so-called greenhouse gases (GHGs) over the last 200 years.¹ Climate shift changes are affecting weather systems causing negative effects on human health, agriculture, and the global economy which resulted in serious environmental concerns deriving from the need to reduce GHG emissions from industrial resources. Since carbon dioxide (CO₂) accounts for the largest portion of the world's annual emissions of GHGs, its emissions from industrial waste gases have become a major target for reduction, especially flue gases from coal power stations as main emitters of carbon dioxide. Acid gas absorption is a pretty complex technology which has been developed within many various separation techniques. For the purpose of carbon dioxide removal, there are different separation solutions which are under development such as absorption into a liquid solvent, cryogenic separation, adsorption onto a solid, chemical conversion and permeation through membranes. Among these techniques, the majority of scientific attention is being set toward absorption into liquid solvents. These techniques are suitable for treating low carbon dioxide concentration within high volume flue gases streams. These conditions are typical for coal-based power plants. Commonly used solvents for purpose of carbon dioxide removal are aqueous solutions of alkanolamines. One of the most widely adopted liquid absorption agent is monoethanolamine (MEA), due to high carbon dioxide loading capacity. Nowadays MEA is being gradually replaced by other more efficient alkanolamine solutions due to negative properties of MEA such as significant amount of energy required for solvent regeneration, operational problems with process equipment caused by increased corrosion,²

degradation of solvent, and so forth.³ Beside classic alkanolamine-based solutions, some researches^{4–7} show potential use of sterically hindered amines which are characterized by high carbon dioxide loading capacity and easy regeneration of solvent. However, it should be noted that sterically hindered amines do not show a higher carbon dioxide loading capacity than conventional alkanolamines under all conditions. This is particularly a case with the reduction of carbon dioxide partial pressure.^{5,6} A third option for potential carbon dioxide removal is solvent formation, achieved by blending of variety of single alkanolamines. This approach combines the favorable characteristics of different solvents while suppressing their unfavorable characteristics. A small addition of secondary alkanolamines to primary alkanolamines can shift 2–3 times the mass transfer coefficient of carbon dioxide with no change of operation properties of blended constituents.⁷ Since the majority of coal based power plants are operating at atmospheric pressure, and new power plants designs with increased operating pressure are still under development, all measurements were carried out at atmosphere pressure. Equilibrium concentrations of carbon dioxide and other acid gases present in flue gases in solution are proportional to their partial pressures in the gas phase (Henry's law application) so a conclusion can be made that the increase of partial pressures of acid gases will cause all chemical reactions to be driven to the right. Although this conclusion is valid in most cases, primary alkanolamines show no changes in loading capacity with the increase of acid gases partial pressures, due to limiting factors which will be

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discussed later. The reaction equilibrium between alkanolamines and carbon dioxide is also sensitive to temperature, since the rise of temperature causes the rise of partial vapor pressure of acid gases. This property offers the possibility that acid gases can be easily stripped from amine solution by the application of heat with simultaneous regeneration of alkanolamine solvent. The basic concept of acid gases removal includes an absorber and stripping column with the operating temperature around 323.15 K and 393.15 K, respectively, to cover potential low-temperature operating conditions, but also higher temperatures that may occur (due to increase heat of reaction). Due to the wide use of alkanolamines in flue gas treatment, metal processing, and chemical industries, investigations of thermodynamic and transport properties of different alkanolamine solutions have been studied.^{8–28} Although there are many other parameters which affect the absorption of acid gases, the main goal of this work is to determine thermodynamic properties of aqueous solutions of alkanolamines, including effects of temperature and concentration change within primary, secondary, and tertiary aqueous solutions of alkanolamines. These results will provide precise and reliable data regarding thermodynamic and transport properties of aqueous solutions of alkanolamines as a starting point for further investigation of the potential use of blended alkanolamines in future work.

EXPERIMENTAL SECTION

Chemicals. For the purpose of the investigation of properties of alkanolamine solutions, the following chemicals have been used: monoisopropanolamine (MIPA, $x = 0.98$), diethanolamine (DEA, $x = 0.995$), diisopropanolamine (DIPA, $x = 0.98$), and triisopropanolamine (TIPA, $x = 0.95$) were supplied by Merck and Sigma Aldrich (the sample description is given in Table 1).

Table 1. Sample Description

chemical name	source	initial mass fraction purity	purification method
MIPA ^a	Merck	≥0.98	none
DIPA ^b	Merck	≥0.98	none
TIPA ^c	Sigma Aldrich	≥0.95	none
DEA ^d	Merck	≥0.995	none
water		0.999	ultra pure deionized

^aMIPA = monoisopropanolamine. ^bDIPA = diisopropanolamine.

^cTIPA = triisopropanolamine. ^dDEA = diethanolamine.

All chemicals were kept in dark bottles in an inert atmosphere and degassed just before a sample preparation. Aqueous solutions of alkanolamines were prepared with 4D ultra pure deionized water ($x = 0.999$). To prevent the eventual solubility of ambient air, water was degassed by ultrasonic bath at elevated temperature of 323.15 K for 30 min.

Measurements. Density measurements were performed on Anton Paar DMA 5000 digital vibrating U-tube densimeter (with automatic viscosity correction) with an instrument accuracy of $\pm 5 \cdot 10^{-3} \text{ kg} \cdot \text{m}^{-3}$. The cell temperature was regulated with built-in solid state thermostat, maintaining a temperature within accuracy of $\pm 0.01 \text{ K}$. The instrument was calibrated before measurements using Milli-Q water and dry air as standards. Mixtures were prepared gravimetrically using a Mettler AG 204 balance with a precision of $1 \cdot 10^{-4} \text{ g}$. The uncertainty of the mass fraction determination was less than $\pm 1 \cdot 10^{-4}$. Refractive index measurements were carried out on Anton Paar RXA 156 refractometer. The repeatability of refractive index was $2 \cdot 10^{-5}$ and temperature within 0.03 K. Viscosities η of aqueous solutions of alkanolamines were measured with a digital Stabinger viscometer (model SVM 3000/G2). The instrument contains two measuring cells. A U-tube is filled with the liquid sample and excited to oscillate via magnetic coils. The other cell which is used for dynamic viscosity measurements contains a straight tube filled with liquid sample. Within this tube, a floating measuring rotor made of low-density material with built-in magnet is placed, centered by centrifugal force. A rotating magnet in SVM 3000 induces an eddy current field with a speed dependent brake torque. After the start of experiment, rotor is reaching a constant speed determined by the equilibrium between the viscosity-dependent driving torque, which is proportional to the speed difference between tube and the rotor, and the brake torque caused by eddy currents. The dynamic viscosity is calculated from the equation:

$$\eta = \frac{ku_2}{(u_1 - u_2)} \quad (1)$$

where u_1 is the speed of tube, u_2 is the rotor speed, and k is the mean adjustment coefficient for the instrument. The instrument measuring ranges were adjusted by the manufacturer. The uncertainty of the dynamic viscosity measurements is below 1.5 % in the temperature and viscosity range used in this work. A detailed description of the above-mentioned apparatus has been given in our previous papers.^{29–32} A comparison of experimental

Table 2. Review of the Literature Data for Densities, Refractive Indices, and Viscosities of 1-Amino-2-propanol (MIPA), Diisopropanolamine (DIPA), Triisopropanolamine (TIPA), and Diethanolamine (DEA) and Water (H₂O) at Atmospheric Pressure^a

T/K	$\rho/\text{kg} \cdot \text{m}^{-3}$		n_D		$\eta/\text{mPa} \cdot \text{s}$	
	this work	lit.	this work	lit.	this work	lit.
298.15	956.644	956.972 ^b	MIPA			
			1.44590	1.44604 ^c	23.259	23.00 ^b
313.15	992.460	991.99 ^b	DIPA			
			1.45517		308.54	
323.15	984.835	984.90 ^d	1.45139		123.21	125.73 ^d
			TIPA			
323.15	990.616		1.45233		132.30	
			DEA			
298.15	1093.221	1093.52 ^e	1.47640	1.4747 ^f	566.57	566.30 ^g

^aStandard uncertainties σ for each variables are $\sigma(T) = 0.01 \text{ K}$; $\sigma_c(\rho) = \pm 1 \cdot 10^{-2} \text{ kg} \cdot \text{m}^{-3}$; $\sigma_c(\eta) = \pm 3 \cdot 10^{-3} \text{ mPa} \cdot \text{s}$ and $\sigma_c(n_D) = 5 \cdot 10^{-5}$, with a 0.95 level of confidence ($k \approx 2$). ^bThe Alkanolamines Handbook.³³ ^cDreisbach.³⁴ ^dHenni et al.³⁵ ^eMurrieta-Guevara and Rodriguez.³⁶ ^fDaubert and Danner.³⁷ ^gTeng et al.³⁸

data with representative literature data of pure components^{27,33–38} is presented in Table 2.

RESULTS AND DISCUSSION

Measurements within this work were carried out in the temperature range from (298.15 to 343.15) K with temperature steps of 5 K, which corresponds to temperature operating conditions within the absorption column. Typical mass concentrations of alkanolamines range from (10 to 35) %.³⁹ Higher alkanolamine mass concentrations can be achieved by implementation of corrosion inhibitors since higher concentrations of alkanolamines have negative effects (corrosion, solvent degradation) on process equipment. Such high concentrations are mainly used in the treatment of natural gas within petrochemical industries.

The density, refractive index, and viscosity data for four aqueous binary mixtures of alkanolamine and water were measured at $w = 0.05, 0.1, 0.15, 0.20$, and 0.25 of alkanolamine. The results for all measured properties over the temperature range (298.15 to 343.15) K are presented in Tables 3 to 5.

Density. The densities of the measured binary systems increase with the increase of mass percentage of alkanolamine. Measured data show a linear dependence over the concentration range with a decrease of density values with the increase of temperature. Received data were fitted using eq 2 with two optimized parameters. The measured densities of all binary systems are presented in Table 3.

Refractive Index. The refractive indices almost linearly decrease with increase in temperature and increase with the rise of mass percentage of alkanolamines. For the purpose of standard deviation reduction, all data used eq 2 with two optimized parameters. The measured values of refractive index for all binary systems of alkanolamines are presented in Table 4.

Viscosity. Viscosities of aqueous solutions of alkanolamines show an increase with the increase of mass percentage of alkanolamines. The increase in temperature causes a decrease of viscosity values. Received results of viscosity values are fitted using eq 2 with three optimized parameters. The polynomial dependence of viscosity from mass percent of alkanolamines is shifting toward linear dependence at higher temperatures. Data regarding temperature and concentration dependence of viscosity of aqueous solutions of alkanolamine are presented in Table 5.

To best of our knowledge, there are no literature data regarding all properties over entire concentration and temperature conditions, as it is presented in this paper. Only data of densities for aqueous solutions of DEA at (313.15 and 333.15) K,^{16,18} and data for density, viscosity, and refractive index for TIPA aqueous solutions (at five temperatures: (303.15, 313.15, 323.15, 333.15, and 343.15) K)²³ are present in literature. A comparison between our and literature data^{16,18} of aqueous solutions of DEA shows excellent agreement (deviation is less than 0.5 %), while TIPA aqueous solutions²³ show very good agreement with maximum absolute average deviations of 0.7 %, 0.3 %, and 1.0 % for density, refractive indices, and viscosities, respectively.

Speaking about the alkanolamine theoretical loading capacity of carbon dioxide, primary alkanolamines tend to favor the reaction of carbamate ion formation, which is stable and has a low rate of hydrolysis to bicarbonate. The carbamate ion can tie up to alkanolammonium ion which prevents 1 mol of alkanolamine to react with carbon dioxide. Thus, theoretical level that can be achieved is 0.5 mol of carbon dioxide per mol of alkanolamine. Tertiary alkanolamine shows a tendency to form bicarbonate with potential theoretical level of 1 mol of carbon dioxide per 1 mol of alkanolamine. Although such a high loading capacity can

Table 3. Densities for Aqueous Solutions of Different Alkanolamines, for the Mass Fraction of Alkanolamines of $w = 0.05, 0.1, 0.15, 0.20$, and 0.25 , at Temperatures of (298.15 to 343.15) K and Atmospheric Pressure^a

T/K	$10^{-3} \rho/\text{kg}\cdot\text{m}^{-3}$				
	$w = 0.05$	$w = 0.1$	$w = 0.15$	$w = 0.2$	$w = 0.25$
MIPA					
298.15	0.998574	0.999182	0.999932	1.001027	1.002456
303.15	0.997087	0.997570	0.998142	0.999018	1.000197
308.15	0.995386	0.995747	0.996159	0.996833	0.997780
313.15	0.993483	0.993738	0.994005	0.994489	0.995218
318.15	0.991399	0.991552	0.991681	0.991989	0.992514
323.15	0.989143	0.989197	0.989197	0.989338	0.989679
328.15	0.986722	0.986682	0.986564	0.986546	0.986711
333.15	0.984143	0.984015	0.983779	0.983615	0.983616
338.15	0.981410	0.981198	0.980853	0.980549	0.980398
343.15	0.978537	0.978237	0.977785	0.977354	0.977059
DIPA					
298.15	1.000599	1.003535	1.006988	1.010398	1.014103
303.15	0.999133	1.001970	1.005269	1.008497	1.011982
308.15	0.997454	1.000192	1.003348	1.006408	1.009687
313.15	0.995576	0.998214	1.001240	1.004140	1.007231
318.15	0.993508	0.996055	0.998951	1.001705	1.004617
323.15	0.991263	0.993722	0.996497	0.999106	1.001855
328.15	0.988852	0.991224	0.993882	0.996358	0.998953
333.15	0.986277	0.988560	0.991114	0.993463	0.995910
338.15	0.983349	0.985762	0.988199	0.990404	0.992735
343.15	0.980387	0.982810	0.985138	0.987241	0.989434
TIPA					
298.15	1.000880	1.004303	1.007923	1.011661	1.015037
303.15	0.999387	1.002671	1.006103	1.009612	1.012729
308.15	0.997679	1.000826	1.004079	1.007371	1.010257
313.15	0.995768	0.998785	1.001875	1.004968	1.007640
318.15	0.993671	0.996561	0.999498	1.002408	1.004883
323.15	0.991401	0.994168	0.996957	0.999698	1.001995
328.15	0.988960	0.991613	0.994263	0.996847	0.998980
333.15	0.986362	0.988905	0.991422	0.993857	0.995844
338.15	0.983585	0.985961	0.988426	0.990739	0.992589
343.15	0.980297	0.982830	0.985312	0.987395	0.989222
DEA					
298.15	1.003464	1.009087	1.014940	1.020921	1.027107
303.15	1.001984	1.007530	1.013274	1.019137	1.025187
308.15	1.000305	1.005773	1.011417	1.017178	1.023111
313.15	0.998430	1.003831	1.009390	1.015055	1.020887
318.15	0.996373	1.001713	1.007200	1.012776	1.018512
323.15	0.994141	0.999439	1.004854	1.010350	1.016003
328.15	0.991746	0.996992	1.002355	1.007781	1.013360
333.15	0.989200	0.994401	0.999713	1.005073	1.010552
338.15	0.986501	0.991668	0.996929	1.002231	1.007442
343.15	0.983659	0.988796	0.993878	0.999250	1.004411

^aStandard uncertainties σ for each variables are $\sigma(T) = 0.01$ K; $\sigma(p) = 5$ %; $\sigma(w_i) = 0.0001$, and the combined expanded uncertainty is $\sigma_c(\rho) = \pm 1 \cdot 10^{-2}$ kg·m⁻³; $\sigma_c(\eta) = \pm 3 \cdot 10^{-3}$ mPa·s and $\sigma_c(n_D) = 5 \cdot 10^{-5}$, with a 0.95 level of confidence ($k \approx 2$).

be achieved, bicarbonate is unstable with the limiting factor of very slow rate of reaction of hydrolysis and ionization of dissolved carbon dioxide. This problem can be easily overcome by addition of an activator, typically another amine, which increases the rate of hydration of dissolved carbon dioxide.

The conclusion can be made that by increasing of alkanolamine concentration, acid gas removal will be more efficient with

Table 4. Refractive Indices for Aqueous Solutions of Different Alkanolamines, for the Mass Fraction of Alkanolamines of $w = 0.05, 0.1, 0.15, 0.20$, and 0.25 , at Temperatures of (298.15 to 343.15) K and Atmospheric Pressure^a

T/K	n_D				
	$w = 0.05$	$w = 0.1$	$w = 0.15$	$w = 0.2$	$w = 0.25$
MIPA					
298.15	1.33891	1.34586	1.35281	1.35955	1.36649
303.15	1.33825	1.34506	1.35201	1.35875	1.36560
308.15	1.33749	1.34432	1.35092	1.35767	1.36461
313.15	1.33668	1.34343	1.35012	1.35672	1.36347
318.15	1.33578	1.34259	1.34908	1.35573	1.36228
323.15	1.33483	1.34154	1.34799	1.35454	1.36109
328.15	1.33384	1.34050	1.34705	1.35330	1.35985
333.15	1.33280	1.33936	1.34581	1.35211	1.35861
338.15	1.33173	1.33832	1.34462	1.35092	1.35753
343.15	1.33057	1.33713	1.34343	1.34968	1.35614
DIPA					
298.15	1.34052	1.34766	1.35486	1.36210	1.36925
303.15	1.33988	1.34693	1.35421	1.36130	1.36822
308.15	1.33919	1.34623	1.35332	1.36036	1.36736
313.15	1.33844	1.34533	1.35237	1.35947	1.36641
318.15	1.33755	1.34450	1.35149	1.35838	1.36537
323.15	1.33675	1.34355	1.35044	1.35739	1.36423
328.15	1.33566	1.34256	1.34940	1.35615	1.36294
333.15	1.33467	1.34147	1.34831	1.35501	1.36175
338.15	1.33363	1.34037	1.34702	1.35377	1.36051
343.15	1.33224	1.33899	1.34583	1.35253	1.35922
TIPA					
298.15	1.34069	1.34815	1.35565	1.36317	1.37063
303.15	1.34001	1.34737	1.35487	1.36227	1.36979
308.15	1.33922	1.34656	1.35393	1.36133	1.36869
313.15	1.33853	1.34572	1.35309	1.36039	1.36764
318.15	1.33769	1.34484	1.35209	1.35933	1.36653
323.15	1.33680	1.34387	1.35104	1.35818	1.36532
328.15	1.33565	1.34287	1.34999	1.35718	1.36417
333.15	1.33481	1.34176	1.34883	1.35587	1.36291
338.15	1.33370	1.34063	1.34762	1.35461	1.36154
343.15	1.33265	1.33943	1.34641	1.35319	1.36012
DEA					
298.15	1.33940	1.34654	1.35439	1.36184	1.37017
303.15	1.33875	1.34585	1.35368	1.36108	1.36935
308.15	1.33801	1.34511	1.35288	1.36028	1.36847
313.15	1.33720	1.34427	1.35208	1.35939	1.36756
318.15	1.33633	1.34340	1.35122	1.35862	1.36659
323.15	1.33539	1.34245	1.35028	1.35777	1.36557
328.15	1.33442	1.34148	1.34935	1.35680	1.36453
333.15	1.33340	1.34044	1.34850	1.35582	1.36354
338.15	1.33234	1.33938	1.34755	1.35470	1.36244
343.15	1.33122	1.33820	1.34633	1.35356	1.36140

^aStandard uncertainties σ for each variables are $\sigma(T) = 0.01$ K; $\sigma(p) = 5$ %; $\sigma(w_i) = 0.0001$, and the combined expanded uncertainty is $\sigma_c(n_D) = 5 \cdot 10^{-5}$, with a 0.95 level of confidence ($k \approx 2$).

reduction in the required solution circulation rate and overall plant cost. However, this is not the case since more concentrated solutions cause increase of acid gases vapor pressures. Because of exothermic nature of reaction between alkanolamines and carbon dioxide, the increase in concentration of alkanolamine in solution will cause increase in temperature with final increase in vapor pressure of acid gases, thus nullifying positive effects of increase in absorption of carbon dioxide.⁴⁰

Table 5. Viscosities for Aqueous Solutions of Different Alkanolamines, for the Mass Fraction of Alkanolamines of $w = 0.05, 0.1, 0.15, 0.20$, and 0.25 , at Temperatures of (298.15 to 343.15) K and Atmospheric Pressure^a

T/K	$\eta/\text{mPa}\cdot\text{s}$				
	$w = 0.05$	$w = 0.1$	$w = 0.15$	$w = 0.2$	$w = 0.25$
MIPA					
298.15	1.0879	1.2957	1.5686	1.9437	2.4437
303.15	0.97325	1.1478	1.3721	1.6784	2.0830
308.15	0.87612	1.0269	1.2149	1.4681	1.7987
313.15	0.79374	0.92750	1.0842	1.2950	1.5736
318.15	0.72248	0.83624	0.97555	1.1555	1.3844
323.15	0.66173	0.76172	0.88640	1.0391	1.2314
328.15	0.61120	0.69884	0.81131	0.94301	1.1018
333.15	0.56956	0.64702	0.74995	0.86587	0.99336
338.15	0.53810	0.60529	0.69191	0.79996	0.90153
343.15	0.51519	0.57015	0.64780	0.74077	0.82662
DIPA					
298.15	1.1175	1.3939	1.8136	2.2879	3.0264
303.15	0.99681	1.2282	1.5686	1.9521	2.5391
308.15	0.89844	1.0928	1.3801	1.6953	2.1665
313.15	0.81893	0.98142	1.2243	1.4846	1.8711
318.15	0.74422	0.88562	1.0949	1.3128	1.6326
323.15	0.68835	0.80469	0.99239	1.1709	1.4385
328.15	0.64066	0.73572	0.90533	1.0521	1.2785
333.15	0.58329	0.67933	0.83238	0.95118	1.1447
338.15	0.55074	0.63410	0.76927	0.86433	1.0321
343.15	0.52673	0.60056	0.72504	0.81461	0.95787
TIPA					
298.15	1.08305	1.3597	1.7099	2.1728	2.8571
303.15	0.98420	1.1979	1.4862	1.8589	2.3955
308.15	0.88826	1.0682	1.3088	1.6122	2.0507
313.15	0.80339	0.95741	1.1587	1.4177	1.7745
318.15	0.73147	0.86582	1.0376	1.2573	1.5526
323.15	0.66964	0.78737	0.93608	1.1247	1.3727
328.15	0.61637	0.72085	0.84935	1.0130	1.2239
333.15	0.57189	0.66237	0.77545	0.91857	1.1037
338.15	0.53501	0.61337	0.71141	0.83810	0.99910
343.15	0.50643	0.57002	0.65962	0.76939	0.91130
DEA					
298.15	1.0591	1.2698	1.5031	1.8927	2.3760
303.15	0.94983	1.1286	1.3194	1.6487	2.0496
308.15	0.85688	1.0129	1.1742	1.4553	1.7954
313.15	0.77917	0.91418	1.0513	1.2948	1.5847
318.15	0.71370	0.83069	0.94798	1.1615	1.4113
323.15	0.65296	0.75932	0.85985	1.0478	1.2665
328.15	0.60168	0.69689	0.78359	0.95193	1.1410
333.15	0.55965	0.64296	0.71777	0.86732	1.0371
338.15	0.52691	0.59317	0.66193	0.79896	0.95435
343.15	0.50017	0.55339	0.61850	0.74594	0.89222

^aStandard uncertainties σ for each variables are $\sigma(T) = 0.01$ K; $\sigma(p) = 5$ %; $\sigma(w_i) = 0.0001$, and the combined expanded uncertainty is $\sigma_c(\eta) = \pm 3 \cdot 10^{-3}$ mPa·s, with a 0.95 level of confidence ($k \approx 2$).

Experimental and calculated values of density, refractive index, and viscosity for four aqueous alkanolamine solutions are presented in Figures 1 to 3. Calculated values are presented with lines obtained by fittings of experimental values using eq 2:

$$\rho(\text{g}\cdot\text{cm}^{-3}) \text{ or } n_D \text{ or } \eta(\text{mPa}\cdot\text{s}) = A_0 + A_1x + A_2x^2 \quad (2)$$

Parameters A_p of eq 2 for aqueous alkanolamine solutions at different temperatures are presented in Table 6.

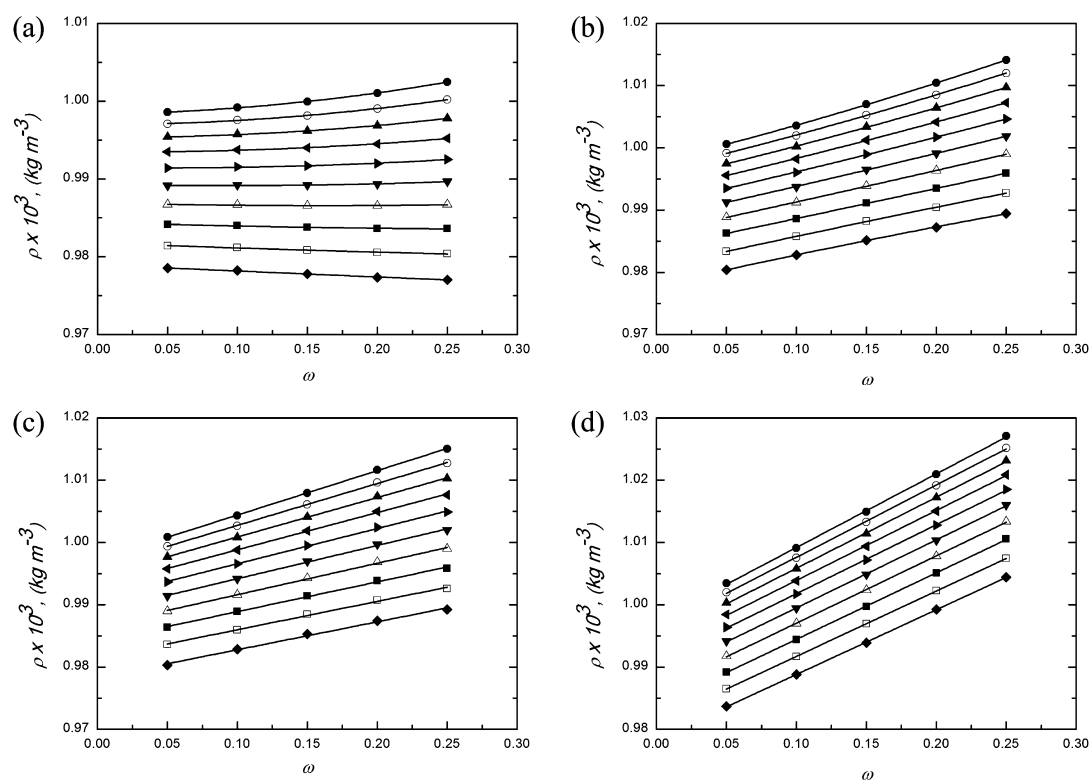


Figure 1. Experimental values of density data for system (a) MIPA (1) + water (2), (b) DIPA (1) + water (2), (c) TIPA (1) + water (2), (d) DEA (1) + water (2). Symbols refer to experimental points at: ●, 298.15 K; ○, 303.15 K; ▲, 308.15 K; ◀, 313.15 K; ▶, 318.15 K; ▼, 323.15 K; △, 328.15 K; ■, 333.15 K; □, 338.15 K and ♦, 343.15 K, while the lines present the results calculated by eq 2.

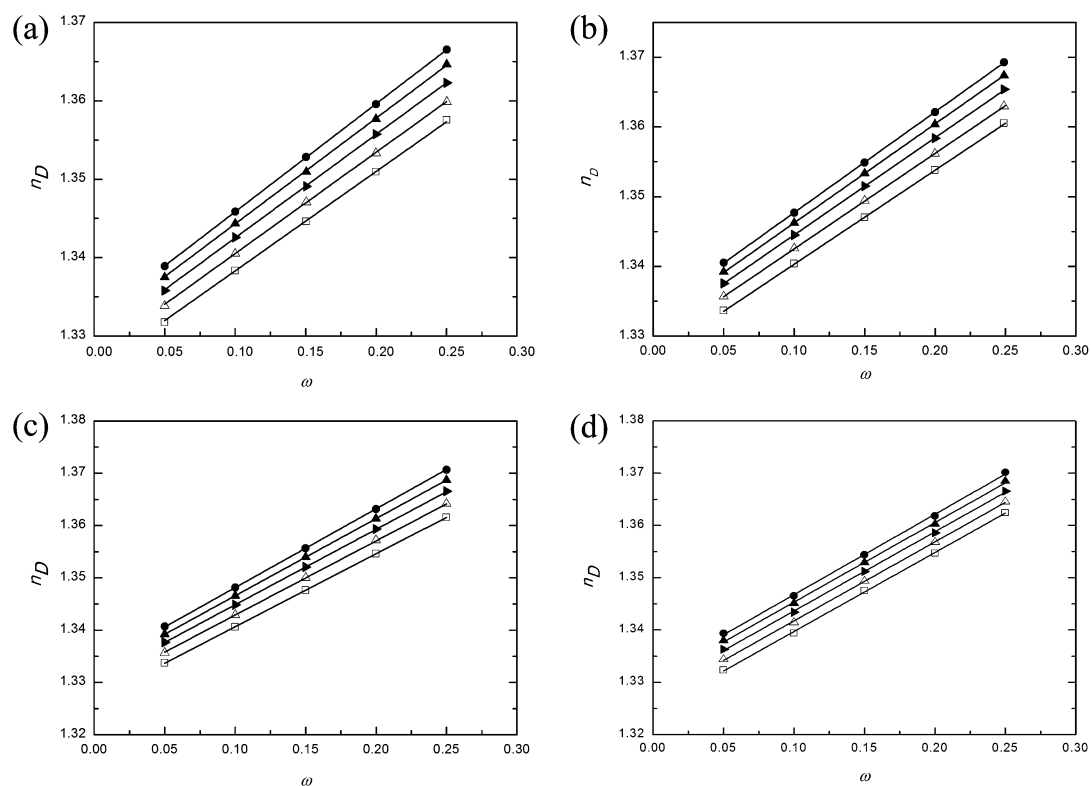


Figure 2. Experimental values of refractive indices data for system (a) MIPA (1) + water (2), (b) DIPA (1) + water (2), (c) TIPA (1) + water (2), (d) DEA (1) + water (2). Symbols refer to experimental points at: ●, 298.15 K; ▲, 308.15 K; ▶, 318.15 K; △, 328.15 K and □, 338.15 K, while the lines present the results calculated by eq 2.

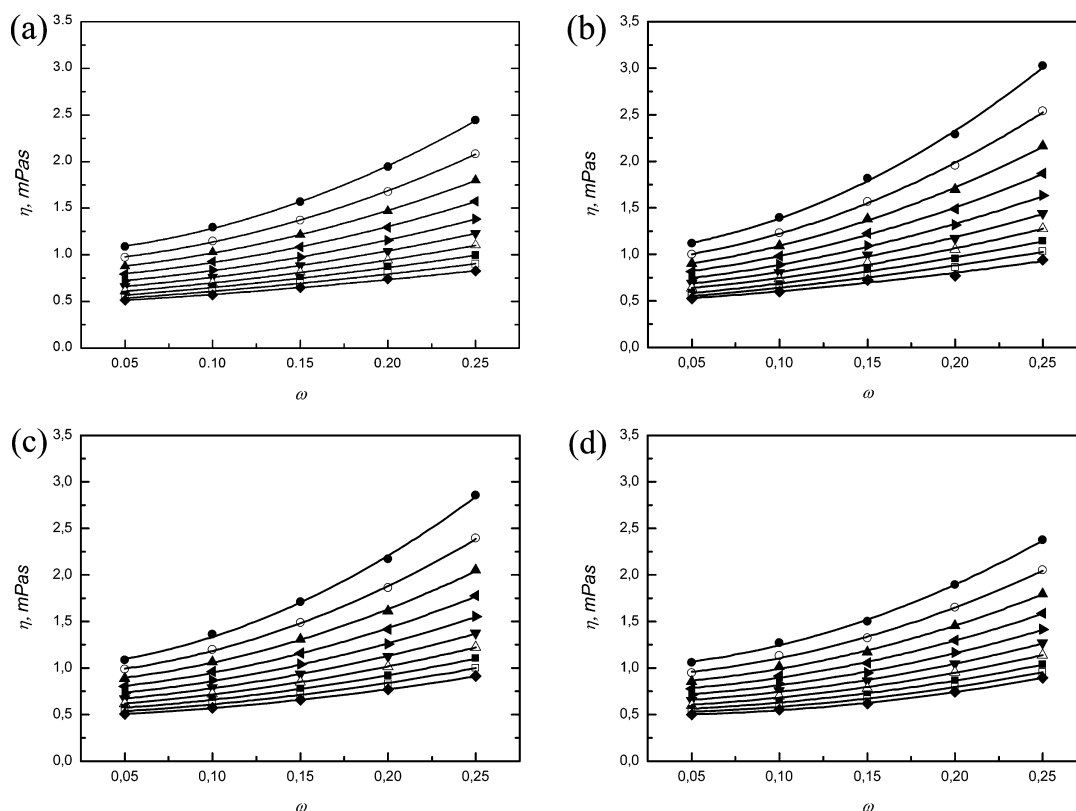


Figure 3. Experimental values of viscosities data for system (a) MIPA (1) + water (2), (b) DIPA (1) + water (2), (c) TIPA (1) + water (2), and (d) DEA (1) + water (2). Symbols refer to experimental points at: ●, 298.15 K; ○, 303.15 K; ▲, 308.15 K; ◀, 313.15 K; ▶, 318.15 K; ▼, 323.15 K; △, 328.15 K; ■, 333.15 K; □, 338.15 K and ♦, 343.15 K, while the lines present the results calculated by eq 2.

Table 6. Parameters A_p of eq 2 for Aqueous Alkanolamine Solutions at Temperature T

	T/K	A_0	A_1	A_2		T/K	A_0	A_1	A_2
Monoisopropanolamine (MIPA) + Water					Monoisopropanolamine (MIPA) + Water				
$10^{-3} \rho/\text{kg}\cdot\text{m}^{-3}$	298.15	0.99834	$2.00\cdot 10^{-5}$		328.15	0.54639	$1.07\cdot 10^{-2}$	$4.60\cdot 10^{-4}$	
	303.15	0.99695	$-1.00\cdot 10^{-5}$		333.15	0.50174	$1.16\cdot 10^{-2}$	$3.20\cdot 10^{-4}$	
	308.15	0.99534	$-1.00\cdot 10^{-5}$		338.15	0.47599	$1.07\cdot 10^{-2}$	$2.60\cdot 10^{-4}$	
	313.15	0.99350	$-2.00\cdot 10^{-5}$		343.15	0.46061	$9.26\cdot 10^{-2}$	$2.20\cdot 10^{-4}$	
	318.15	0.99149	$-3.00\cdot 10^{-5}$		Diisopropanolamine (DIPA) + Water				
	323.15	0.98930	$-4.00\cdot 10^{-5}$		$10^{-3} \rho/\text{kg}\cdot\text{m}^{-3}$	298.15	0.99771	$5.50\cdot 10^{-4}$	
	328.15	0.98695	$-5.00\cdot 10^{-5}$		303.15	0.99632	$5.40\cdot 10^{-4}$		
	333.15	0.98443	$-6.00\cdot 10^{-5}$		308.15	0.99471	$5.30\cdot 10^{-4}$		
	338.15	0.98176	$-7.00\cdot 10^{-5}$		313.15	0.99290	$5.20\cdot 10^{-4}$		
	343.15	0.97896	$-8.00\cdot 10^{-5}$		318.15	0.99090	$5.10\cdot 10^{-4}$		
n_D	298.15	1.33281	$1.25\cdot 10^{-3}$		323.15	0.98873	$5.00\cdot 10^{-4}$		
	303.15	1.33220	$1.24\cdot 10^{-3}$		328.15	0.98638	$4.80\cdot 10^{-4}$		
	308.15	1.33146	$1.24\cdot 10^{-3}$		333.15	0.98388	$4.70\cdot 10^{-4}$		
	313.15	1.33065	$1.24\cdot 10^{-3}$		338.15	0.98087	$5.00\cdot 10^{-4}$		
	318.15	1.32975	$1.25\cdot 10^{-3}$		343.15	0.97790	$5.10\cdot 10^{-4}$		
	323.15	1.32880	$1.25\cdot 10^{-3}$		n_D	298.15	1.33429	$1.27\cdot 10^{-3}$	
	328.15	1.32780	$1.26\cdot 10^{-3}$		303.15	1.33363	$1.27\cdot 10^{-3}$		
	333.15	1.32677	$1.26\cdot 10^{-3}$		308.15	1.33293	$1.26\cdot 10^{-3}$		
	338.15	1.32571	$1.26\cdot 10^{-3}$		313.15	1.33227	$1.24\cdot 10^{-3}$		
	343.15	1.32452	$1.27\cdot 10^{-3}$		318.15	1.33156	$1.22\cdot 10^{-3}$		
$\eta/\text{mPa}\cdot\text{s}$	298.15	1.00334	$8.34\cdot 10^{-3}$	$1.96\cdot 10^{-3}$	323.15	1.33077	$1.20\cdot 10^{-3}$		
	303.15	0.89693	$8.54\cdot 10^{-3}$	$1.55\cdot 10^{-3}$	328.15	1.32986	$1.19\cdot 10^{-3}$		
	308.15	0.80346	$9.31\cdot 10^{-3}$	$1.21\cdot 10^{-3}$	333.15	1.32890	$1.18\cdot 10^{-3}$		
	313.15	0.72853	$9.08\cdot 10^{-3}$	$9.80\cdot 10^{-4}$	338.15	1.32782	$1.18\cdot 10^{-3}$		
	318.15	0.65736	$9.64\cdot 10^{-3}$	$7.70\cdot 10^{-4}$	343.15	1.32674	$1.15\cdot 10^{-3}$		
	323.15	0.59737	$1.01\cdot 10^{-2}$	$6.10\cdot 10^{-4}$	$\eta/\text{mPa}\cdot\text{s}$	298.15	1.00372	$1.03\cdot 10^{-2}$	$2.80\cdot 10^{-3}$

Table 6. continued

	T/K	A ₀	A ₁	A ₂		T/K	A ₀	A ₁	A ₂
10 ⁻³ ρ/kg·m ⁻³	Diisopropanolamine (DIPA) + Water				10 ⁻³ ρ/kg·m ⁻³	Triisopropanolamine (TIPA) + Water			
	303.15	0.89158	1.15·10 ⁻²	2.16·10 ⁻³		328.15	0.55653	0.00889	
	308.15	0.79583	1.29·10 ⁻²	1.66·10 ⁻³		333.15	0.52012	0.00760	
	313.15	0.72653	1.23·10 ⁻²	1.33·10 ⁻³		338.15	0.49049	0.00644	
	318.15	0.65556	1.28·10 ⁻²	1.04·10 ⁻³		343.15	0.46902	0.00503	
	323.15	0.60568	1.22·10 ⁻²	8.40·10 ⁻⁴		Diethanolamine (DEA) + Water			
	328.15	0.56476	1.13·10 ⁻²	6.90·10 ⁻⁴		298.15	0.99799	1.07·10 ⁻³	
	333.15	0.50013	1.41·10 ⁻²	4.60·10 ⁻⁴		303.15	0.99658	1.06·10 ⁻³	
	338.15	0.47658	1.28·10 ⁻²	3.70·10 ⁻⁴		308.15	0.99498	1.05·10 ⁻³	
	343.15	0.47048	9.93·10 ⁻³	3.40·10 ⁻⁴		313.15	0.99316	1.04·10 ⁻³	
	Triisopropanolamine (TIPA) + Water					318.15	0.99115	1.03·10 ⁻³	
	298.15	0.99727	7.10·10 ⁻⁴			323.15	0.98896	1.02·10 ⁻³	
	303.15	0.99588	6.90·10 ⁻⁴			328.15	0.98661	1.02·10 ⁻³	
	308.15	0.99429	6.80·10 ⁻⁴			333.15	0.98408	1.02·10 ⁻³	
	313.15	0.99249	6.60·10 ⁻⁴			338.15	0.98129	1.04·10 ⁻³	
	318.15	0.99049	6.40·10 ⁻⁴			343.15	0.97858	1.01·10 ⁻³	
n _D	323.15	0.98833	6.20·10 ⁻⁴		n _D	298.15	1.33240	1.37·10 ⁻³	
	328.15	0.98600	6.00·10 ⁻⁴			303.15	1.33176	1.37·10 ⁻³	
	333.15	0.98351	5.80·10 ⁻⁴			308.15	1.33102	1.37·10 ⁻³	
	338.15	0.98082	5.60·10 ⁻⁴			313.15	1.33020	1.37·10 ⁻³	
	343.15	0.97738	6.00·10 ⁻⁴			318.15	1.32920	1.40·10 ⁻³	
	298.15	1.33130	1.81·10 ⁻³			323.15	1.32816	1.42·10 ⁻³	
	303.15	1.33070	1.80·10 ⁻³			328.15	1.32711	1.43·10 ⁻³	
	308.15	1.32992	1.80·10 ⁻³			333.15	1.32595	1.46·10 ⁻³	
	313.15	1.32914	1.80·10 ⁻³			338.15	1.32482	1.48·10 ⁻³	
	318.15	1.32819	1.83·10 ⁻³			343.15	1.32383	1.44·10 ⁻³	
	323.15	1.32712	1.85·10 ⁻³			η/mPa·s			
	328.15	1.32618	1.85·10 ⁻³			298.15	0.99388	5.01·10 ⁻³	2.00·10 ⁻³
	333.15	1.32523	1.83·10 ⁻³			303.15	0.89471	4.44·10 ⁻³	1.67·10 ⁻³
	338.15	1.32424	1.82·10 ⁻³			308.15	0.80708	4.56·10 ⁻³	1.39·10 ⁻³
	343.15	1.32308	1.82·10 ⁻³			313.15	0.73541	4.16·10 ⁻³	1.19·10 ⁻³
η/mPa·s	298.15	0.91106	0.01721		318.15	0.67616	3.50·10 ⁻³	1.03·10 ⁻³	
	303.15	0.90456	0.00708		323.15	0.61867	3.56·10 ⁻³	8.90·10 ⁻⁴	
	308.15	0.81493	0.00767		328.15	0.56959	3.59·10 ⁻³	7.70·10 ⁻⁴	
	313.15	0.73322	0.00834		333.15	0.53502	2.36·10 ⁻³	7.10·10 ⁻⁴	
	318.15	0.66375	0.00898		338.15	0.51213	8.00·10 ⁻⁵	7.00·10 ⁻⁴	
	323.15	0.60529	0.00912		343.15	0.49932	-2.36·10 ⁻³	7.20·10 ⁻⁴	

To assess calculated and measured data, each set of determined parameters was valuated through average absolute deviation (AADs). Values of AADs for each system and all parameters are given in Table 7. The average absolute deviation is calculated using eq 3:

$$\text{AAD} = \frac{1}{N} \sum_{i=1}^N \left| \frac{Y_{\text{exp},i} - Y_{\text{cal},i}}{Y_{\text{exp},i}} \right| \quad (3)$$

Table 7. Average Absolute Deviations for Aqueous Alkanolamine Solutions (AAD) Obtained by Equation 3

	AAD %		
	density	refractive index	viscosity
MIPA	0.008	0.024	0.386
DIPA	0.016	0.048	1.019
TIPA	0.009	0.017	0.545
DEA	0.009	0.015	0.856

where Y_{exp} are experimental values, Y_{cal} represent calculated values, and N are the number of data points.

CONCLUSION

Thermodynamic and transport properties such as densities, refractive index, and viscosity for four aqueous alkanolamine solutions (MIPA, DIPA, TIPA, and DEA) were measured in the temperature range from (298.15 to 343.15) K and at atmospheric pressure. To investigate the influence of concentration of alkanolamine solution on system properties, mass concentration levels were set at (5, 10, 15, 20, and 25) % of alkanolamines. Received data were correlated using a polynomial equation. Measured and correlated data agreed in almost all cases within 0.01 % for density, 0.025 % for refractive indices, and 0.85 % for viscosities calculated on basis of average absolute deviation, except for values obtained for the system with DIPA, where deviations are slightly higher. Data presented in this work provide not only necessary information for process optimization and design for carbon dioxide removal but also thermodynamic properties essential for precise heat and mass transfer calculations. The obtained data are a necessary contribution for the design of amine-blended solutions which with their positive properties (lower cost of regeneration, reduction of corrosion, etc.) present the future for carbon dioxide removal as a technology which is self-sustained, low-cost, and feasible.

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Notes

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REFERENCES

- Hofmann, D. J.; Butler, J. H.; Tans, P. P. A new look at atmospheric carbon dioxide. *Atmos. Environ.* **2009**, *43*, 2084–2086.
- White, L.; Street, D. E. Corrosion control in amine treating units. *Proceedings of corrosion in the oil refining industry conference*, Phoenix, AZ, Sept 17–18, 1998.
- Chi, S.; Rochelle, G. T. Oxidative degradation of monoethanolamine. Presented at the *First National Conference on Carbon Sequestration*, Washington, DC, May 14, 2001.
- Sartori, G.; Savage, D. W. Sterically hindered amines for carbon dioxide removal from gases. *Ind. Eng. Chem. Fundam.* **1983**, *22*, 239–249.
- Maddox, R. N. *Gas conditioning and processing*; Campbell & Co: Norman, OK, 1984.
- Rubin, E. S. *Introduction to Engineering and the Environment*; McGraw-Hill: New York, 2001.
- Park, J. Y.; Yoon, S. J.; Lee, H. Effect of steric hindrance on carbon dioxide absorption into new amine solutions. *Environ. Sci. Technol.* **2003**, *37*, 1670–1675.
- Kuss, E. Die Viskosität von Komprimierten Flüssigkeiten. *Z. Angew. Phys.* **1955**, *7*, 372.
- Riddick, J. A.; Bunger, W. B. *Organic Solvents: Physical Properties and Methods of Purification*, 3rd ed.; Wiley Interscience: New York, 1970.
- Ethanolamines. Product Data; Olin Chemicals: Stamford, CT, 1980.
- Murrieta-Guevara, F.; Trejo, A. Liquid Density as a Function of Temperature of Five Organic Solvents. *J. Chem. Eng. Data* **1984**, *29*, 204–206.
- Weast, R. C.; Astle, M. J. *Handbook of Data on Organic Compounds*; CRC Press, Inc.: Boca Raton, FL, 1985.
- DiGuilio, R. M.; Lee, R. J.; Schaeffer, S. T.; Brasher, L. L.; Teja, A. S. Densities and Viscosities of the Ethanolamines. *J. Chem. Eng. Data* **1992**, *37*, 239–242.
- Wang, Y. W.; Xu, S.; Otto, F. D.; Mather, A. E. Solubility of N₂O in Alkanolamines and in Mixed Solvents. *Chem. Eng. J.* **1992**, *48*, 31–40.
- Maham, Y.; Teng, T. T.; Hepler, L. G.; Mather, A. E. Densities, Excess Molar Volumes, and Partial Molar Volumes for Binary Mixtures of Water with Monoethanolamine, Diethanolamine, and Triethanolamine from 25 to 80 °C. *J. Solution Chem.* **1994**, *23*, 195–205.
- Rinker, B. E.; Oelschlager, W. D.; Colussi, T. A.; Henry, R. K.; Sandall, C. O. Viscosity, Density, and Surface Tension of Binary Mixtures of Water and N-Methyldiethanolamine and Water and Diethanolamine and Tertiary Mixtures of These Amines with Water over the Temperature Range 20–100 °C. *J. Chem. Eng. Data* **1994**, *39*, 392–395.
- Herba, H.; Czechowski, G.; Zywicki, B.; Stockhausen, M.; Jadzyn, J. Molar Excess Volumes of Binary Mixtures of Amino Alcohols with 1,4-Dioxane. *J. Chem. Eng. Data* **1995**, *40*, 214–215.
- Hsu, H. C.; Li, H. M. Densities of aqueous blended amines. *J. Chem. Eng. Data* **1997**, *42*, 502–507.
- Hawrylak, B.; Burke, S. E.; Palepu, R. Partial Molar and Excess Volumes and Adiabatic Compressibilities of Binary Mixtures of Ethanolamines with Water. *J. Solution Chem.* **2000**, *29*, 575–594.
- Ko, J.-J.; Tsai, T.-C.; Lin, C.-Y.; Wang, H.-M.; Li, M.-H. Diffusivity of Nitrous Oxide in Aqueous Alkanolamine Solutions. *J. Chem. Eng. Data* **2001**, *46*, 160–165.
- Aguila-Hernandez, J.; Gomez-Quintana, R.; Murrieta-Guevara, F.; Romero-Martinez, A.; Trejo, A. Liquid Density of Aqueous Blended Alkanolamines and N-Methylpyrrolidone as a Function of Concentration and Temperature. *J. Chem. Eng. Data* **2001**, *46*, 861–867.
- Maham, Y.; Liew, C. N.; Mather, A. E. Viscosities and Excess Properties of Aqueous Solutions of Ethanolamines from 25 to 80 °C. *J. Solution Chem.* **2002**, *31*, 743–756.
- Chauhan, K. R.; Yoon, J. S.; Lee, H.; Kang, M. C.; Min, B.-M. Physical and transport properties of aqueous triisopropanolamine solutions. *J. Chem. Eng. Data* **2003**, *48*, 291–293.
- Mokraoui, S.; Valtz, A.; Coquelet, C.; Richon, D. Volumetric Properties of the Isopropanolamine-Water at Atmospheric Pressure from 283.15 to 353.15 K. *Thermochim. Acta* **2006**, *440*, 122–128.
- Aguila-Hernandez, J.; Trejo, A.; Garcia-Flores, E. B.; Molnar, R. Viscometric and Volumetric Behaviour of Binary Mixtures of Sulfolane and N-methylpyrrolidone with Monoethanolamine and Diethanolamine in the Range 303–373 K. *Fluid Phase Equilib.* **2008**, *267*, 172–180.
- Alvarez, E.; Cerdeira, F.; Gomez-Diaz, D.; Navaza, M. Density, Speed of Sound, Isentropic Compressibility, and Excess Volume of Binary Mixtures of 1-Amino-2-propanol or 3-Amino-1-propanol with 2-Amino-2-methyl-1-propanol, Diethanolamine, or Triethanolamine from (293.15 to 323.15) K. *J. Chem. Eng. Data* **2010**, *55*, 2567–2575.
- Rayer, A. V.; Kadiwala, S.; Narayanaswamy, K.; Henni, A. Volumetric Properties, Viscosities, and Refractive Indices for Aqueous 1-Amino-2-Propanol (Monoisopropanolamine (MIPA)) Solutions from (298.15 to 343.15) K. *J. Chem. Eng. Data* **2010**, *55*, 5562–5568.
- Gomez-Diaz, D.; La Rubia, M. D.; Lopez, A.; Navaza, J.; Pacheco, R.; Sanchez, S. Density, Speed of Sound, Refractive Index, and Viscosity of 1-Amino-2-Propanol {or Bis(2-hydroxypropyl)amine} + Triethanolamine + Water from T = (288.15 to 333.15) K. *J. Chem. Eng. Data* **2012**, *57*, 1104–1111.
- Šerbanović, S. P.; Kijevčanin, M. Lj.; Radović, I. R.; Djordjević, B. D. Effect of temperature on the excess molar volumes of some alcohol + aromatic mixtures and modelling by cubic EOS mixing rules. *Fluid Phase Equilib.* **2006**, *239*, 69–82.
- Kijevčanin, M. Lj.; Djuriš, M. M.; Radović, I. R.; Djordjević, B. D.; Šerbanović, S. P. Volumetric properties of the binary methanol + Chloroform and Ternary Methanol + Chloroform + Benzene Mixtures at (288.15, 293.15, 298.15, 303.15, 308.15, and 313.15) K. *J. Chem. Eng. Data* **2007**, *52*, 1136–1140.
- Kijevčanin, M. Lj.; Radović, I. R.; Živković, E. M.; Djordjević, B. D.; Šerbanović, S. P. Influence of chain length and degree of branching of alcohol + chlorobenzene mixtures on determination and modelling of V^E by CEOS and CEOS/G^E mixing rules. *Fluid Phase Equilib.* **2008**, *263*, 205–213.
- Živković, E. M.; Kijevčanin, M. Lj.; Radović, I. R.; Šerbanović, S. P.; Djordjević, B. D. Viscosity of the binary systems 2-methyl-2-propanol with n-alkanes at T = (303.15, 308.15, 313.15, 318.15 and 323.15) K: Prediction and correlation—New UNIFAC–VISCO interaction parameters. *Fluid Phase Equilib.* **2010**, *299*, 191–197.
- The Alkanolamines Handbook*; Dow Chemical Company: Midland, MI, 1981.
- Dreisbach, R. R. *Physical Properties of Chemical Compounds, III*; American Chemical Society: Washington, DC, 1961.
- Henni, A.; Hromek, J.; Tontiwachwuthikul, P.; Chakma, A. Volumetric Properties and Viscosities for Aqueous Diisopropanolamine Solutions from 25 °C to 70 °C. *J. Chem. Eng. Data* **2003**, *48*, 1062–1067.
- Murrieta-Guevara, F.; Rodriguez, A. T. Liquid Density as a Function of Temperature of Five Organic Solvents. *J. Chem. Eng. Data* **1984**, *29*, 204–209.
- Daubert, T. E.; Danner, R. P. *Technical Data Book-Petroleum Refining*; American Petroleum Institute: Washington, DC, 1992-extant.

(38) Teng, T. T.; Maham, Y.; Hepler, L. G.; Mather, A. E. Viscosity of Aqueous Solutions of N-Methyldiethanolamine and of Diethanolamine. *J. Chem. Eng. Data* **1994**, 39, 290.

(39) Adisorn, A.; Amornvadee, V. Characterization and comparison of the CO₂ absorption performance into single and blended alkanolamines in a packed column. *Ind. Eng. Chem. Res.* **2004**, 43, 2228–2237.

(40) Kohl, A. L.; Nielsen, R. B. *Gas Purification*, 5th ed.; Gulf Publishing Company: Houston, TX, 1997.