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Dielectric Constants of Aqueous Diisopropanolamine, Diethanolamine, *N*-Methyldiethanolamine, Triethanolamine, and 2-Amino-2-methyl-1-propanol Solutions

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Dielectric constants of aqueous diisopropanolamine, diethanolamine, *N*-methyldiethanolamine, triethanolamine, and 2-amino-2-methyl-1-propanol solutions for temperatures of (303.15 to 328.15) K were measured using a dielectric analyzer. The dielectric constants of the corresponding pure alkanolamines were also measured for the same temperature range. The measured dielectric constants of diethanolamine and propylene glycol were found to be in good agreement with literature values. The relation between the dielectric constant of alkanolamines and the concentration of the hydroxyl groups per unit volume was discussed. The Kirkwood relation was also applied to represent the temperature dependence of dielectric constants of alkanolamines, and the apparent dipole moments were obtained from the measured data. The dielectric constants of aqueous alkanolamine solutions were correlated satisfactorily by using a Redlich−Kister type equation, and the positions of the relative minimum in the plot of $\Delta\epsilon$ versus x_1 for aqueous alkanolamine solutions were presented. The compositions of the potential complexes formed from alkanolamines and water molecules were discussed.

Introduction

For the removal of CO_2 and H_2S from gas streams, aqueous alkanolamine solutions are industrially important solvents used in the natural gas, petroleum chemical, and ammonia industries. A wide variety of alkanolamines such as monoethanolamine (MEA), diglycolamine (DGA), diethanolamine (DEA), diisopropanolamine (DIPA), *N*-methyldiethanolamine (MDEA), triethanolamine (TEA), 2-amino-2-methyl-l-propanol (AMP), and 2-piperidineethanol (2-PE) can be used.[1] Accurate estimations of the thermophysical properties are essential in process design calculations involving these solutions. The dielectric constant is one of the basic physical properties and provides useful information about the intermolecular interactions in liquids.[2]

In general, the dielectric constants of aqueous alkanolamine solutions cannot be predicted from the dielectric constants of the corresponding individual components. For alkanolamines, however, only few dielectric constant data are available in the literature. The dielectric constants of diethanolamine solution for temperatures of (273.15 to 323.15) K were reported by Ikada et al.[3] Thus, the purpose of this study was to measure the dielectric constants of aqueous DEA, DIPA, MDEA, TEA, and AMP solutions for temperatures of (303.15 to 328.15) K using a dielectric analyzer. The dielectric constants of the corresponding alkanolamines were also measured.

Experimental Section

Chemicals. DEA was Riedel-deHaën reagent grade with purity ≥ mass fraction 0.99; DIPA was Acros reagent grade with purity of mass fraction 0.99; MDEA (purity ≥ mass fraction 0.985), TEA (purity ≥ mass fraction 0.9991), and AMP (purity ≥ mass fraction 0.99) were Acros reagent grade; propylene glycol (PG) was J. T. Baker reagent grade with purity ≥ mass fraction 0.99.

Apparatus and Procedure. Deionic water, with a resistivity of 18.3 MΩ·cm and with a total organic carbon content of less than 15 ppb, produced by Barnstead Thermodyne, model Easy Pure 1052, was used. The aqueous solutions were prepared by mass, with weighting accurate to $\pm 1 \times 10^{-7}$ kg. The estimated uncertainty of the mole fraction of aqueous solutions is $\pm 1.5 \times 10^{-4}$. The prepared aqueous solutions have been degassed by using ultrasonic cleaner (Branson, model 3510).

The dielectric constants of aqueous solutions were measured using a Dielectric Analyzer (model DEA 2970, TA Instruments). Based on a coplanar, interdigitated-comb configuration of the electrodes, a ceramic single surface sensor was used in this study. The electrode assembly was confined in a bell jar (which offers protection from humid air) and operated in the presence of a purge gas. In an experiment, a ceramic single surface sensor (without sample) was placed face up on a furnace drip pan at the bottom of the oven. The bell jar was replaced, and nitrogen (with purity 99.9995 mol %) at a flow rate of 500 mL·min^{-1} purged the DEA for 30 min. The sensor calibration procedure was started. The process of sensor calibration was proceeded for about 1 min, and the sensor calibration parameters (geometry and resistance temperature detector) were automatically calculated. The temperature calibration was also performed during a sensor calibration step of each experiment. After sensor calibration, a sample was loaded and distributed evenly over the surface of the sensor, which rests on a furnace. When lowered, ram pressure caused the sensor to embed in the surface of the sample and forced sample to flow into spaces between the electrodes. The ram contains spring-loaded probes to make electrical contact with the pads on the sensor, completing the signal circuits. When voltage was applied, the sample response was measured as a function of time, temperature and frequency (f). A system controller monitored the current in the ceramic single surface

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Table 1. Dielectric Constant ϵ of PG and DEA

T	PG		DEA	
K	ref 8	this study	ref 3	this study
303.15	27.96	27.87 ± 0.03[a]	24.64	24.69 ± 0.09
308.15	27.19	27.13 ± 0.02		24.30 ± 0.06
313.15	26.40	26.43 ± 0.02	23.65	23.86 ± 0.03
318.15	25.67	25.76 ± 0.02		23.28 ± 0.05
323.15	24.97	25.08 ± 0.02	22.65	22.63 ± 0.08
328.15		24.57 ± 0.03		22.13 ± 0.08

[a] Standard deviation determined from (4 to 6) replicated runs.

sensor and the phase angle shift (θ) between the applied voltage (V) and the measured current (I). The capacitance (C) was calculated by

$$C = \frac{I_{\text{measured}}}{V_{\text{applied}}} \frac{\sin \theta}{2\pi f} \tag{1}$$

The channels in the ceramic single surface sensor were precisely 125 μm wide and 12.5 μm deep; electrode spacing and the sample thickness were predetermined, assuming intimate sample/electrode contact. By knowing the electrode area (A), the electrode spacing (d), and the capacitance (C), the dielectric constant was determined using

$$\epsilon = \frac{Cd}{e_o A} \tag{2}$$

where e_o is the absolute permittivity of free space. Besides the sensor and temperature calibration steps, instrument (electronic) calibration was performed by using the DEA module calibration program at least once a week to calibrate the electronics on the analog board. Samples were run from (303.15 to 328.15) K at a heating rate of 5 K/min. The thickness of the sample in ceramic single surface was between (0.125 and 6.0) mm. The static dielectric constant is the dielectric constant measured in static fields or at low frequencies where no relaxation effects occur. In this study, the dielectric constant measurements were carried out in the frequency range of (260 to 300) kHz where no relaxation effects were observed. Raju[2] pointed out that the dielectric constant of water was not appreciably dependent on the frequency up to 100 MHz. In the work of Kinart et al.,[4] the static dielectric constant (relative permittivity) measurements were carried out at the frequency of 3 MHz for two binary mixtures of aqueous glycols solutions. Within the present frequency range, the dielectric constants showed no variation with frequency. Thus, the dielectric constant, measured in this experiment, corresponds to the static dielectric constant. In all the cases, the experiments were generally performed for four to six replicated runs. The DEA gave a dielectric constant sensitivity of 0.01 and an isothermal stability of 0.2 K[5]. The uncertainty[6,7] of the dielectric constant was estimated to be less than ± 1.0 %.

Results and Discussion

The dielectric constants of PG and DEA were measured to reassure the accuracy of the dielectric constant measurements. The measured dielectric constants of PG and DEA are presented in Table 1. The literature values for the dielectric constants for PG of Verbeeck et al.[8] and DEA of Ikada et al.[3] are also listed in Table 1 for comparison. In Figure 1, the relative deviations between the measured and literature dielectric constants for PG and DEA are shown. The overall average absolute relative deviations (in 100 AARD) were 0.24 and 0.48 for PG and DEA, respectively. Thus, the dielectric constants of PG and DEA measured in this study were in good agreement with the literature values.

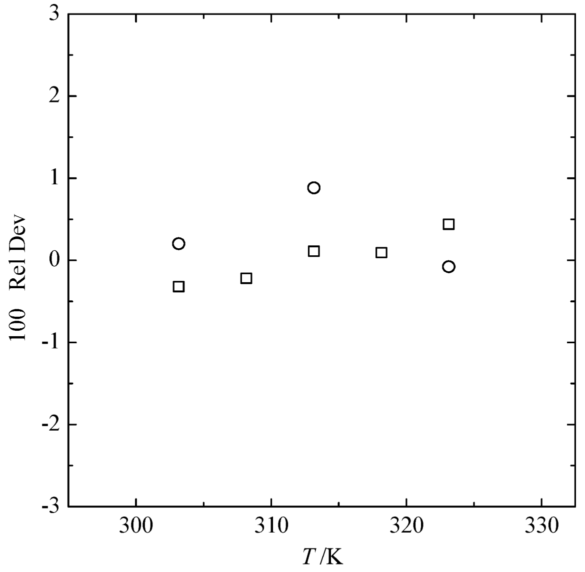

Figure 1. Relative deviation between the measured and literature dielectric constants for PG and DEA. □, PG (using data of ref 8); ○, DEA (using data of ref 3). Relative deviation = $(\epsilon_{\text{exp}} - \epsilon_{\text{lit}})/\epsilon_{\text{lit}}$ where ϵ_{lit} is the literature value.

Table 2. Dielectric Constant ϵ of Pure Alkanolamines

T/K	DIPA	MDEA	TEA	AMP
303.15	13.87 ± 0.04[a]	21.84 ± 0.07	28.11 ± 0.07	20.59 ± 0.06
308.15	13.75 ± 0.04	21.33 ± 0.08	27.41 ± 0.06	20.11 ± 0.03
313.15	13.62 ± 0.04	20.83 ± 0.08	26.73 ± 0.06	19.69 ± 0.01
318.15	13.49 ± 0.04	20.32 ± 0.09	26.07 ± 0.06	19.27 ± 0.10
323.15	13.36 ± 0.04	19.79 ± 0.07	25.41 ± 0.06	18.88 ± 0.09
328.15	13.26 ± 0.05	19.28 ± 0.05	24.80 ± 0.06	18.37 ± 0.10

[a] Standard deviations determined from (4 to 6) replicated runs.

In Table 2, dielectric constants of DIPA, MDEA, TEA, and AMP are presented. The temperature dependence of the dielectric constant of alkanolamines was assumed as follows:

$$\epsilon = a + b(T/\text{K}) + c(T/\text{K})^2 \tag{3}$$

where a, b, and c are parameters. The fitted parameters, by using eq 3, are listed in Table 3. Except for DEA, the fitted results (in 100 AARD) were less than 0.06. For DEA, the data of Ikada et al.[3] were also included in the calculations; the temperature ranged from (273.15 to 328.15) K. The parameters of Wohlfarth[9] for water were also listed in Table 3 for reference. In Figure 2, plots of the dielectric constants versus the reciprocal of temperature for five amines are shown. At a constant temperature, the dielectric constants of alkanolamines were in order as TEA > DEA > MDEA > AMP > DIPA. In their study of the dielectric properties of lower-membered polyethylene glycols, Koizumi and Hanai[10] pointed out that the main contribution to the dielectric polarization of liquid PEGs might be due to the orientation polarization of hydroxyl groups. They found that higher PEGs, having fewer hydroxyl groups per unit volume, showed smaller dielectric constants. In Figure 3, the dielectric constants of alkanolamine versus the concentration of the hydroxyl groups per unit volume (moles of hydroxyl groups/m^3) are shown at 303.15 K. The concentration of the hydroxyl groups per unit volume for each amine was calculated from the product of the density of amine (in mol·m^{-3}) and the number of hydroxyl groups per molecule. The numbers of hydroxyl groups per molecule are 1 for AMP; 3 for TEA; and

Table 3. Parameters of Dielectric Constant Equation for Pure Alkanolamines

	T	parameters				
substance	K	a	b	$10^5\,c$	npts[a]	110 AARD[b]
DEA	273.15−328.15	61.7172	−0.136743	4.94303	12[c]	0.36
DIPA	303.15−328.15	28.8247	−0.07191	7.45260	6	0.04
MDEA	303.15−328.15	44.1399	−0.046824	−8.82158	6	0.02
TEA	303.15−328.15	106.221	−0.37311	38.0795	6	0.01
AMP	303.15−328.15	51.2299	−0.113191	3.98317	6	0.06
H_2O	273.15−373.15	249.21[d]	−0.79069	72.997		

[a] Number of data points. [b] AARD = $(1/n)\sum_{i=1}^{n}((|\epsilon_{\mathrm{exp}} - \epsilon_{\mathrm{calcd}}|)/\epsilon_{\mathrm{exp}})$, where n is the number of data points. [c] Data of ref 3 were included. [d] Parameters of Wohlfarth.[9]

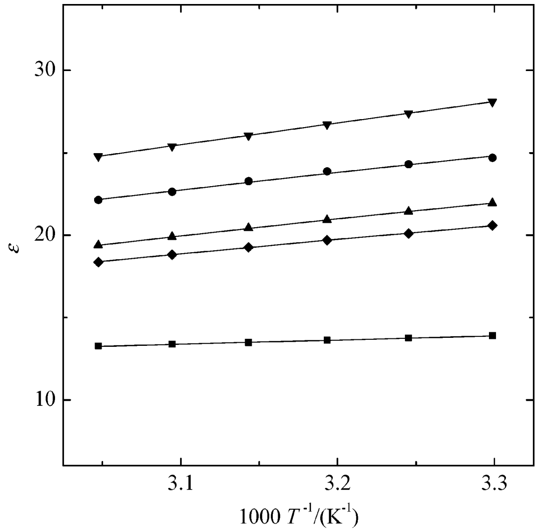

Figure 2. Dielectric constant ϵ of pure alkanolamines as functions of temperature: ▼, TEA; ●, DEA; ▲, MDEA; ◆, AMP; ■, DIPA; lines, calculated using eq 3.

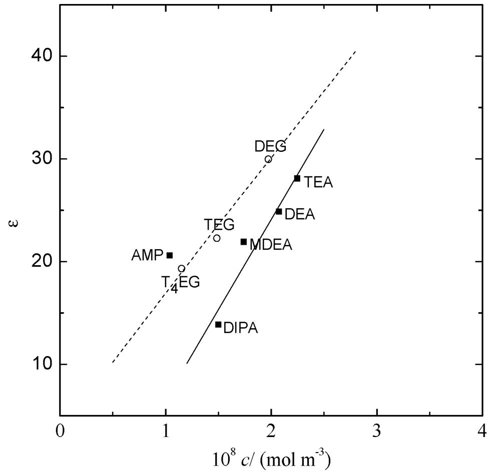

Figure 3. Dielectric constant ϵ vs the concentration of hydroxyl group per unit volume at 303.15 K: ○, polyethylene glycols; ■, alkanolamines; solid line, determined from alkanolamines (exclusion of AMP); dashed line, determined from polyethylene glycols.

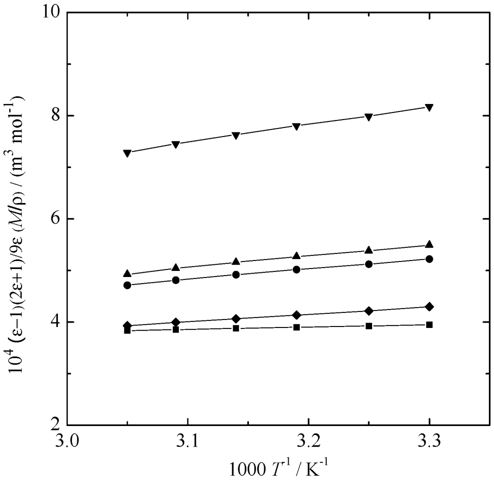

Figure 4. Kirkwood functions vs 1/T for alkanolamines: ▼, TEA; ●, DEA; ▲, MDEA; ◆, AMP; ■, DIPA; lines, calculated using eq 4.

2 for DEA, DIPA, MDEA, diethylene glycol (DEG), triethylene glycol (TEG), and tetraethylene glycol (T_4EG). In the calculations, densities of DEA, MDEA, and AMP were calculated from the correlation of Hsu and Li,[11] density of DIPA was from Henni et al.,[12] density of TEA was from Maham et al.,[13] densities of DEG and TEG were from Valtz et al.,[14] and density of T_4EG was from Moumen et al.[15] For easy observation in Figure 3, a dashed line was determined from DEG, TEG, and T_4EG; a solid line was determined from DEA, DIPA, MDEA, and TEA (exclusion of AMP). As shown in Figure 3, the dielectric constants of PEGs (i.e., DEG, TEG, T_4EG) are normally proportional to the concentrations of the hydroxyl groups per unit volume. Except for AMP, one of the sterically hindered amines, the dielectric constants of the other four amines (i.e., DEA, DIPA, MDEA, and TEA) are also generally proportional to the concentration the hydroxyl groups per unit volume, as shown in Figure 3. As a result, the dielectric constants of alkanolamines also depend on the concentrations of the hydroxyl groups per unit volume, which conforms to the results of Koizumi and Hanai.[10]

For polar fluids, the relation between the dielectric constant and the dipole moment may be considered quantitatively in terms of the Kirkwood relationship[16] after the definition of Onsager's local field.[17] In the theory, an apparent dipole moment of the liquid μ^* can be calculated from the following relation: [18]

$$\frac{(\epsilon - 1)(2\epsilon + 1)}{9\epsilon}\left(\frac{M}{\rho}\right) = \frac{N_0}{3}\left[\alpha + \frac{(\mu^*)^2}{3\epsilon_0 k_B T}\right] \tag{4}$$

where M is the molar mass of the fluid, ρ is the density of fluid, N_0 is the Avogadro number, α is the molecular polarizability of the molecule, ϵ_0 is the electric permittivity in a vacuum, k_B is the Boltzmann constant, T is the absolute temperature, μ^* is the apparent dipole moment ($\mu^* = g^{1/2}\mu$, where μ is the dipole moment in the ideal gas state), and g is the Kirkwood correlation parameter. The Kirkwood parameter measures the restriction to rotation imposed by a cage of molecules surrounding a given one. Plots of the values of the left-hand side of eq 4 versus the reciprocal of temperature for amines are shown in Figure 4. In the calculation, densities of DEA, MDEA, and AMP were calculated using the correlation of Hsu and Li,[11] densities of DIPA were from Henni et al.,[12] and densities of TEA from Maham et al.[13] The slope of the linear regression of the left-

Table 4. Parameters of Kirkwood Equation (i.e., eq 4)

substance	slope	intercept 10^4 m^3·mol^{-1}	μ^* 10^{30} C·m	μ 10^{30} C·m	g
DEA	0.20216	−1.442	19.20	9.34[a]	4.23
DIPA	0.04435	2.484	8.993		
MDEA	0.22564	−1.943	20.28		
TEA	0.3541	−3.501	25.41	12.0[a]	4.48
AMP	0.14707	−0.558	16.37		

[a] Values adopted from ref 19.

Table 5. Dielectric Constant ϵ of Aqueous Alkanolamine Solutions

T	mole fraction of alkanolamine, x_1			
K	0.2	0.4	0.6	0.8
		DEA + H_2O		
303.15	52.65	39.11	30.57	26.95
308.15	51.53	38.06	29.75	26.41
313.15	49.58	36.95	28.91	25.86
318.15	48.31	35.81	28.07	25.42
323.15	46.78	34.68	27.36	24.81
328.15	45.68	33.73	26.70	24.13
		DIPA + H_2O		
303.15	41.34	24.52	18.46	16.65
308.15	40.27	24.00	18.15	16.45
313.15	39.35	23.48	17.84	16.26
318.15	38.46	23.00	17.53	16.05
323.15	37.74	22.52	17.20	15.83
328.15	37.16	22.19	16.97	15.66
		MDEA + H_2O		
303.15	48.77	33.92	26.68	23.44
308.15	47.54	33.09	26.03	22.86
313.15	46.36	32.16	25.40	22.31
318.15	45.14	31.25	24.69	21.77
323.15	43.98	30.37	23.96	21.15
328.15	42.68	29.41	23.21	20.56
		TEA + H_2O		
303.15	49.25	36.34	31.48	29.67
308.15	48.14	35.44	30.66	28.92
313.15	47.23	34.53	29.85	28.17
318.15	46.33	33.60	29.05	27.43
323.15	45.45	32.73	28.25	26.69
328.15	44.63	31.93	27.51	26.04
		AMP + H_2O		
303.15	48.24	33.65	26.63	23.35
308.15	47.33	32.99	26.17	22.82
313.15	46.43	32.33	25.65	22.28
318.15	45.46	31.80	25.14	21.87
323.15	44.31	31.15	24.95	21.42
328.15	43.13	30.11	24.37	21.03

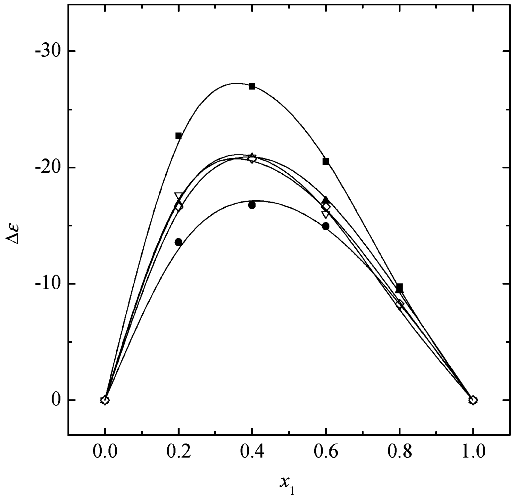

Figure 5. Plot of $\Delta\epsilon$ as a function of composition for aqueous alkanolamine solutions at 303.15 K: ■, DIPA + H_2O; ●, DEA + H_2O; ▲, MDEA + H_2O; ▽, TEA + H_2O; ◇, AMP + H_2O; lines, calculated using eq 6.

hand side of eq 4 versus $1/T$ yields the value of the apparent dipole moment μ^*. Provided that the dipole moment in the ideal gas state μ is available, the Kirkwood correlation parameter g can then be calculated. In Table 4, the determined slopes, the intercepts of the lines in Figure 4, and the apparent dipole moment μ^* were presented. The determined apparent dipole moments vary from 8.993×10^{-30} (C·m) for DIPA to 25.41×10^{-30} (C·m) for TEA. Also, the Kirkwood correlation parameter g for DEA and TEA, determined from the available dipole moment μ, are also listed in Table 4. The calculated Kirkwood correlation parameters were 4.23 and 4.48 for DEA and TEA, respectively. The value of the Kirkwood parameter g was found as a value of 3.01 for 1,1,1,2,3-pentafluoropropane as reported from Gurova et al.[18]

The dielectric constants of aqueous alkanolamine solutions for temperatures of (303.15 to 328.15) K are presented in Table 5. To calculate the dielectric constant of binary solutions, a deviation of the dielectric constants from linearity on a mole fraction basis[4] was applied as follows:

$$\Delta\epsilon = \epsilon - (x_1\epsilon_1 + x_2\epsilon_2) \tag{5}$$

where ϵ_i and ϵ are the dielectric constants of the pure component i and the solution, respectively. The parameters of Wohlfarth[9] for water were applied for the calculation of the dielectric constants of water. The values of deviation of dielectric constant $\Delta\epsilon$ were then fitted by a Redlich−Kister type equation as follows:

$$\Delta\epsilon = x_1x_2\sum_{i=0}^{n} A_i(x_1 - x_2)^i \tag{6}$$

where A_i is pair parameter and assumed to have the following temperature dependence:

$$A_i = a_i + b_i(T/\mathrm{K}) + c_i(T/\mathrm{K})^2 \tag{7}$$

where a_i, b_i, and c_i are parameters for binary systems. The

Table 6. Binary Parameters of the Redlich−Kister Equation for Dielectric Constants

	aqueous alkanolamine systems				
parameters	DEA + H_2O	DIPA + H_2O	MDEA + H_2O	TEA + H_2O	AMP + H_2O
a_1	−12.8794	−102.291	−485.334	−193.066	−737.704
b_1	−0.454693	−0.371671	2.35596	0.456797	3.82443
$10^3\ c_1$	0.917211	1.25823	−3.35820	−0.251668	−5.43524
a_2	−654.928	−407.264	214.260	23.2668	1551.39
b_2	4.12102	3.23324	−1.08555	0.44596	−9.41604
$10^3\ c_2$	−6.20097	−5.52077	1.64978	−1.19933	0.146513
npts[a]	24	24	24	24	24
100 AARD[b]	0.9	0.8	0.9	0.7	0.9

[a] Number of data points. [b] AARD = $(1/n)\sum_{i=1}^{n}((|\epsilon_{\text{exp}} - \epsilon_{\text{calcd}}|)/\epsilon_{\text{exp}})$, where n is the number of data points.

determined parameters and the fitted results are presented in Table 6. The overall fitted result (in 100 AARD) for the calculation of the dielectric constants for five aqueous alkanolamine solutions was 0.9 for a total of 120 data points.

As shown in Table 5, the dielectric constant of aqueous alkanolamine solutions decreases as the temperature increases or the mole fraction of alkanolamine increases. In Figure 5, plots of the values of $\Delta\epsilon$ for aqueous alkanolamine solutions versus mole fraction of alkanolamine at 303.15 K are shown. Figure 5 shows that the values $\Delta\epsilon$ are all negative for aqueous alkanolamine systems, and the minimum value of $\Delta\epsilon$ appears near $x_1 = 0.4$. Due to hydrogen bonding interactions, some complexes may form between unlike pairs of molecules at a composition with a minimum value of $\Delta\epsilon$.[4] For aqueous alkanolamine solutions, the formation of alkanolamine−water complexes may occur near $x_1 = 0.4$.

Conclusions

The dielectric constants of aqueous diisopropanolamine, diethanolamine, *N*-methyldiethanolamine, triethanolamine, and 2-amino-2-methyl-1-propanol solutions for temperatures of (303.15 to 328.15) K were reported. The dielectric constants of the corresponding pure alkanolamines were also presented. For pure alkanolamines, the dielectric constants were found proportional to the concentration of the hydroxyl groups per unit volume. Applying the Kirkwood relation, the apparent dipole moments for five alkanolamines were determined. Applying the available ideal gas-state dipole moments, the Kirkwood correlation parameters for DEA and TEA were also determined. For aqueous alkanolamine solutions, the dielectric constants of solutions were found to decrease as the temperature increases or the mole fraction of alkanolamine increases. The deviations of dielectric constants $\Delta\epsilon$ for five aqueous alkanolamine solutions studied were found all negative values and the minimum value of $\Delta\epsilon$ appear near $x_1 = 0.4$.

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