

Density and Viscosity of Some Partially Carbonated Aqueous Alkanolamine Solutions and Their Blends

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The densities and viscosities of partially carbonated monoethanolamine (MEA), diethanolamine (DEA), and *N*-methyldiethanolamine (MDEA) solutions were measured at 298 K. With increasing carbon dioxide loadings, significant increases in both density and viscosity were observed. These results were combined with literature data to produce correlations for alkanolamine solution density and viscosity as a function of amine concentration, carbon dioxide loading, and temperature. The resulting single-amine correlations were used to predict the densities and viscosities of DEA + MDEA and MEA + MDEA blends. Predictions are compared with data measured for these blends.

Introduction

Very little information is available concerning the effect of acid gas loading on the physical properties of amine-treating solutions flowing through the absorption and regeneration columns used in gas processing, despite the fact that the small amount of data currently available suggests there is a large effect on some properties. Solution density and viscosity are important in the mass-transfer-rate modeling of absorbers and regenerators because these properties affect the liquid–film coefficient for mass transfer. Design of related operations such as pumps and heat exchangers would also benefit from better knowledge of the physical properties of process solutions.

This work was undertaken to determine the effect of carbon dioxide loading on the density and viscosity of industrial strength alkanolamine solutions as well as certain MDEA-based amine blends. Measurements were made at 25 °C, with carbon dioxide loadings from essentially zero to 0.5 moles of CO₂ per mole of amine. Data were taken on aqueous solutions containing the single amines monoethanolamine (MEA), diethanolamine (DEA), and *N*-methyldiethanolamine (MDEA), as well as binary blends of MDEA with MEA and DEA.

The results of the measurements for single amines were combined with available literature data, and standard regression techniques were employed to produce empirical equations correlating solution densities and viscosities to amine composition, CO₂ loading, and temperature. Amine blend data were used to check our ability to predict binary-amine-mixture properties from the single-amine correlations.

Experimental Section

Density (more precisely, specific gravity) was measured using hydrometers calibrated against distilled water, pure amines, and sodium chloride solutions at 25 °C. Unloaded solution densities were checked against literature data and found to be within ±0.1% of reported values.

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Kinematic viscosity was measured using standard Cannon-Fenske viscometers. The viscometers were calibrated against water and unloaded solutions of MDEA for which there are considerable data (Al-Ghawas et al., 1989; Snijder et al., 1993). Repeat measurements were made with a precision of ±0.2%. The dynamic viscosity, η , was calculated from the product of the kinetic viscosity and the measured density. All measurements were made at 25 °C.

A series of samples having a particular amine concentration but with a varying loading were prepared as follows: A large batch of amine of known concentration was made up. Part of this solution was reserved for later use, and the remainder was loaded to saturation by bubbling carbon dioxide at 1 atm pressure through a sintered glass Dreschel head. Varying proportions of the unloaded and loaded solutions were then mixed together to produce a set of samples having a fixed amine-to-water ratio but with varying loading. The amine concentrations and carbon dioxide loadings were also checked titrimetrically using the method of Weiland and Trass (1969) for the single-amine solutions. For amine mixtures, only the total loading could be checked titrimetrically. Therefore, we relied on being very careful in making up the original solutions to fix accurately the concentration of *each* amine in the mixture.

Results and Discussion

The density and viscosity data for partially loaded MDEA, DEA, and MEA solutions are presented in Tables 1, 2, and 3, respectively. Similar data for DEA + MDEA and MEA + MDEA blends are shown in Tables 4 and 5, respectively. These results, along with available literature data as summarized by Ho et al. (1993), were used in the development of the correlations. The summary of Ho et al. includes their own unpublished data as well as results published by Union Carbide Chemical Company (1957), Dow Chemical Company (1962), Jefferson Chemical Company (1970), Pennwalt Corporation (1980), Khalil (1984), Leibush and Shorina (1947), Dingman (1963), Kohl and Riesenfeld (1985), Otto et al. (1986), and Al-Harbi (1982).

On the basis of pure-component molar volumes together with excess molar volumes (due to interactions of various species), an expression to correlate the density of single-amine solutions was developed. The density of a solution

Table 1. Density and Viscosity of MDEA

CO ₂ loading (mol/mol)	CO ₂ loading		CO ₂ loading		
	$\rho/\text{g}\cdot\text{mL}^{-1}$	$\eta/\text{mPa}\cdot\text{s}$	(mol/mol)	$\rho/\text{g}\cdot\text{mL}^{-1}$	$\eta/\text{mPa}\cdot\text{s}$
30 mass % MDEA					
0.00	1.027	3.27	0.30	1.058	3.36
0.05	1.031	3.27	0.35	1.062	3.36
0.10	1.037	3.31	0.40	1.067	3.38
0.15	1.043	3.32	0.45	1.072	3.38
0.20	1.049	3.34	0.50	1.078	3.40
0.25	1.052	3.36			
40 mass % MDEA					
0.00	1.035	5.40	0.30	1.077	6.00
0.05	1.043	5.49	0.35	1.083	6.13
0.10	1.050	5.61	0.40	1.091	6.30
0.15	1.054	5.72	0.45	1.098	6.37
0.20	1.062	5.73	0.50	1.105	6.48
0.25	1.070	5.94			
50 mass % MDEA					
0.00	1.044	9.66	0.30	1.092	11.36
0.05	1.049	9.81	0.35	1.100	11.97
0.10	1.059	10.19	0.40	1.108	12.38
0.15	1.066	10.33	0.45	1.116	12.74
0.20	1.076	10.70	0.50	1.124	12.88
0.25	1.084	11.13			
60 mass % MDEA					
0.00	1.053	17.44	0.38	1.112	25.36
0.06	1.061	17.75	0.44	1.119	27.00
0.13	1.071	19.22	0.51	1.128	
0.19	1.080	20.73	0.57	1.136	
0.25	1.088	21.97	0.64	1.148	
0.32	1.100	23.95			

Table 2. Density and Viscosity of DEA

CO ₂ loading (mol/mol)	CO ₂ loading		CO ₂ loading		
	$\rho/\text{g}\cdot\text{mL}^{-1}$	$\eta/\text{mPa}\cdot\text{s}$	(mol/mol)	$\rho/\text{g}\cdot\text{mL}^{-1}$	$\eta/\text{mPa}\cdot\text{s}$
10 mass % DEA					
0.00	1.009	1.80	0.30	1.023	1.84
0.05	1.012		0.35	1.025	
0.10	1.014	1.83	0.40	1.027	1.85
0.15	1.017		0.45	1.030	
0.20	1.019	1.83	0.50	1.032	1.86
0.25	1.021				
20 mass % DEA					
0.00	1.021	1.97	0.30	1.049	
0.05	1.025	1.97	0.35	1.052	2.09
0.10	1.030		0.40	1.057	
0.15	1.035	2.00	0.45	1.062	2.16
0.20	1.039		0.50	1.167	
0.25	1.043	2.04			
30 mass % DEA					
0.00	1.035	3.10	0.30	1.071	3.48
0.05	1.039		0.35	1.076	
0.10	1.047	3.21	0.40	1.083	3.55
0.15	1.050		0.45	1.090	
0.20	1.059	3.32	0.50	1.094	3.66
0.25	1.064				
40 mass % DEA					
0.00	1.045	5.29	0.30	1.094	6.16
0.05	1.053		0.35	1.102	
0.10	1.062	5.53	0.40	1.111	6.51
0.15	1.069		0.45	1.119	
0.20	1.078	5.88	0.50	1.127	6.90
0.25	1.086				

is given by its average molecular weight divided by its total molar volume

$$\rho = \frac{x_{\text{Am}}M_{\text{Am}} + x_{\text{H}_2\text{O}}M_{\text{H}_2\text{O}} + x_{\text{CO}_2}M_{\text{CO}_2}}{V} \quad (1)$$

where ρ is the solution density ($\text{g}\cdot\text{mL}^{-1}$), V is the molar

Table 3. Density and Viscosity of MEA

CO ₂ loading (mol/mol)	CO ₂ loading		CO ₂ loading		
	$\rho/\text{g}\cdot\text{mL}^{-1}$	$\eta/\text{mPa}\cdot\text{s}$	(mol/mol)	$\rho/\text{g}\cdot\text{mL}^{-1}$	$\eta/\text{mPa}\cdot\text{s}$
10 mass % MEA					
0.00	1.002	1.77	0.30	1.024	1.87
0.05	1.006		0.35	1.029	
0.10	1.011	1.80	0.40	1.034	1.92
0.15	1.016		0.45	1.038	
0.20	1.018	1.83	0.50	1.042	1.93
0.25	1.021				
20 mass % MEA					
0.00	1.007	1.72	0.30	1.053	1.98
0.05	1.015		0.35	1.059	
0.10	1.022	1.83	0.40	1.066	2.12
0.15	1.030		0.45	1.072	
0.20	1.038	1.90	0.50	1.179	2.22
0.25	1.046				
30 mass % MEA					
0.00	1.013	2.52	0.30	1.073	3.21
0.05	1.023		0.35	1.085	
0.10	1.033	2.72	0.40	1.095	3.51
0.15	1.044		0.45	1.106	
0.20	1.054	2.92	0.50	1.117	3.82
0.25	1.065				
40 mass % MEA					
0.00	1.017	3.41	0.30	1.096	4.97
0.05	1.032		0.35	1.114	
0.10	1.043	3.76	0.40	1.126	5.90
0.15	1.056		0.45	1.139	
0.20	1.070	4.30	0.50	1.147	6.73
0.25	1.082				

Table 4. Density and Viscosity of DEA + MDEA Blends

CO ₂ loading (mol/mol)	CO ₂ loading		CO ₂ loading		
	$\rho/\text{g}\cdot\text{mL}^{-1}$	$\eta/\text{mPa}\cdot\text{s}$	(mol/mol)	$\rho/\text{g}\cdot\text{mL}^{-1}$	$\eta/\text{mPa}\cdot\text{s}$
5:45 DEA:MDEA (mass %:mass %)					
0.00	1.045	2.52	0.30	1.090	2.21
0.05	1.050		0.35	1.098	
0.10	1.059	2.72	0.40	1.106	3.51
0.15	1.067		0.45	1.114	
0.20	1.075	2.92	0.50	1.121	3.82
0.25	1.083				
10:40 DEA:MDEA (mass %:mass %)					
0.00	1.048	3.41	0.30	1.094	4.97
0.05	1.054		0.35	1.102	
0.10	1.061	3.76	0.40	1.110	5.90
0.15	1.070		0.45	1.119	
0.20	1.078	4.30	0.50	1.128	6.73
0.25	1.086				
20:30 DEA:MDEA (mass %:mass %)					
0.00	1.050	9.60	0.30	1.098	11.82
0.05	1.057		0.35	1.106	
0.10	1.065	10.25	0.40	1.114	12.69
0.15	1.073		0.45	1.123	
0.20	1.082	11.10	0.50	1.131	13.33
0.25	1.090				

volume of the solution ($\text{mL}\cdot\text{mol}^{-1}$), and x_i and M_i are the mole fractions and molecular weights, respectively, of the amine, water, and carbon dioxide.

The molar volume of an ideal solution is the sum of the partial molar volumes of the components multiplied by their respective mole fractions. Obviously, for ideal solutions the assumption of no reaction or ionization is implicit. In the present context, this would mean that for the purposes of the mole fraction weighting calculation the CO₂ would have to exist solely as *free* CO₂ and not as its carbamate or bicarbonate reaction product (which are the dominant forms of CO₂ in solution). Loaded amine solutions are not ideal, and they certainly require additional terms to account for amine + water and amine + carbon

Table 5. Density and Viscosity of MEA + MDEA Blends

CO ₂ loading (mol/mol)	CO ₂ loading		CO ₂ loading		$\eta/\text{mPa}\cdot\text{s}$
	$\rho/\text{g}\cdot\text{mL}^{-1}$	$\eta/\text{mPa}\cdot\text{s}$	(mol/mol)	$\rho/\text{g}\cdot\text{mL}^{-1}$	
	5:45 MEA:MDEA (mass %:mass %)				
0.00	1.042	8.85	0.30	1.096	10.85
0.05	1.049		0.35	1.106	
0.10	1.059	9.63	0.40	1.115	11.71
0.15	1.068		0.45	1.124	
0.20	1.077	10.11	0.50	1.132	12.65
0.25	1.087				
	10:40 MEA:MDEA (mass %:mass %)				
0.00	1.040	7.36	0.30	1.105	11.30
0.05	1.048		0.35	1.112	
0.10	1.058	8.63	0.40	1.122	12.20
0.15	1.069		0.45	1.132	
0.20	1.079	9.79	0.50	1.140	13.41
0.25	1.090				
	20:30 MEA:MDEA (mass %:mass %)				
0.00	1.035	9.60	0.30	1.103	11.82
0.05	1.047		0.35	1.117	
0.10	1.057	10.25	0.40	1.129	12.69
0.15	1.068		0.45	1.140	
0.20	1.080	11.10	0.50	1.149	13.33
0.25	1.092				

dioxide interactions, in addition to the use of a molar volume for dissolved CO₂, which is unrelated to its pure-component value:

$$V = x_{\text{Am}} V_{\text{Am}} + x_{\text{H}_2\text{O}} V_{\text{H}_2\text{O}} + x_{\text{CO}_2} V_{\text{CO}_2} + x_{\text{Am}} x_{\text{H}_2\text{O}} V^{**} + x_{\text{Am}} x_{\text{CO}_2} V^{**} \quad (2)$$

An expression for the molar volume of pure amine, V_{Am} , was developed using the pure component density data of Al-Ghawas et al. (1989) and DeGuillo et al. (1992) and is given as eq 3

$$V_{\text{Am}} = \frac{M_{\text{Am}}}{aT^2 + bT + c} \quad (3)$$

where T is temperature in kelvin.

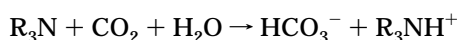
The molar volume associated with the interaction between carbon dioxide and the amine, V^{**} , is given by eq 4:

$$V^{**} = d + ex_{\text{Am}} \quad (4)$$

Coefficients for use in eqs 2, 3, and 4 are given in Table 6. It should be noted that there is a dramatic difference in the partial molar volumes of CO₂ in MEA and DEA solutions compared to that in MDEA solutions. This can be explained in terms of the reaction products formed. When CO₂ reacts with a primary or secondary amine such as MEA or DEA a protonated amine and a carbamate species are formed.



In the reaction between CO₂ and a tertiary amine, such as MDEA, the anion formed is the bicarbonate ion.



Bicarbonate, being a much smaller ion than carbamate, will form a tighter hydration sheath with the water in solution, thus reducing the partial molar volume attributable to the CO₂ in solution.

Equation 1 can be confidently used to calculate MDEA, DEA, and MEA solution densities with CO₂ loadings up to 0.6 mol of CO₂/mol of amine for MEA and DEA, and up to 0.8 mol of CO₂/mol of amine for MDEA, and to a maximum

Table 6. Parameters for Density Correlations

	MEA	DEA	MDEA
a	-5.351 62(-7)	-6.912 9(-7)	-4.860 99(-7)
b	-4.514 17(-4)	-2.066 3(-4)	-4.249 35(-4)
c	1.194 51	1.217 08	1.205 28
d	0	0	12.983
e	0	0	397.72
M	61.09	105.14	119.17
V_{CO_2}	0.047 47	0.044 31	-2.855 8
V^{**}	-1.821 8	-2.965 7	-6.65
std. dev.	0.002 21	0.006 28	0.007 4

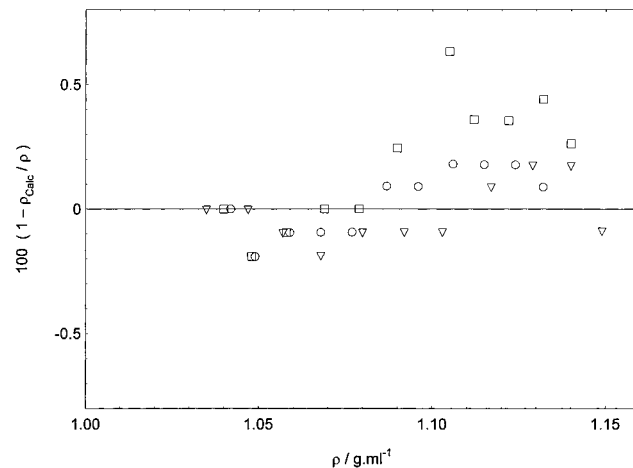


Figure 1. Comparison between predicted and measured density of MEA/MDEA blends: ∇ , 20 mass % MEA + 30 mass % MDEA; \square , 10 mass % MEA + 30 mass % MDEA; \circ , 5 mass % MEA + 45 mass % MDEA.

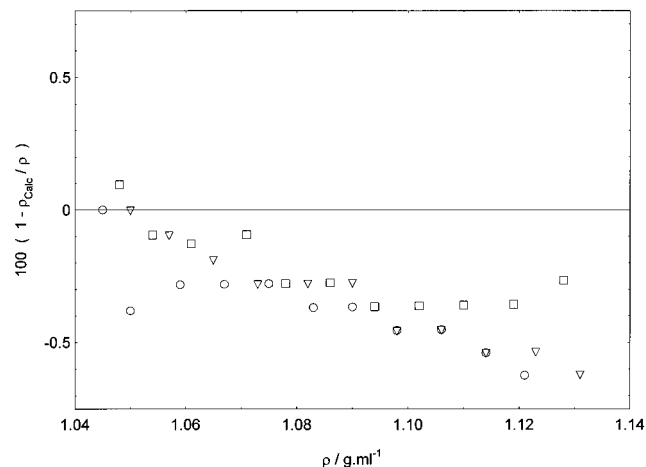


Figure 2. Comparison between predicted and measured density of DEA/MDEA blends: ∇ , 20 mass % DEA + 30 mass % MDEA; \square , 10 mass % DEA + 30 mass % MDEA; \circ , 5 mass % DEA + 45 mass % MDEA.

temperature of 398 K. Table 6 shows the standard deviations of the fits for each of the three amines examined.

For amine blends we used eq 2 modified to include (i) a molar volume term for each amine and (ii) the interactions with CO₂ and water for each amine. In other words, we predicted mixed-amine densities strictly from single-amine data by assuming no amine-amine interaction. The closeness-of-fit is shown in the residual error plots of Figures 1 and 2 for MEA + MDEA and DEA + MDEA blends, respectively. For the MEA + MDEA mixtures, the predicted densities agree with measured data to within the

Table 7. Parameters for Solvent Viscosity

	MEA	DEA	MDEA
<i>a</i>	0	-0.0724	-0.1944
<i>b</i>	0	-3.4363	+0.4315
<i>c</i>	21.186	54.319	80.684
<i>d</i>	2373.	3628.	2889.1
<i>e</i>	+0.01015	-0.0015	+0.0106
<i>f</i>	0.0093	0	0
<i>g</i>	-2.2589	+0.2104	-0.2141
std. dev.	0.0732	0.1355	0.4728

scatter in the data. For the DEA + MDEA mixtures, however, this approach led to an overprediction of density by up to nearly 0.7%.

Our viscosity data for partially loaded MDEA, DEA, and MEA at 298 K shown in Tables 1, 2, and 3 were combined with literature data (Al-Ghawas et al., 1989; DeGuillo et al., 1992; Ho et al., 1993; Snidjer et al., 1993) to produce a correlation that can be used to calculate the viscosity of an amine solution at a given temperature, amine concentration, and CO₂ loading.

The form of the correlating equation is

$$\frac{\eta}{\eta_{\text{H}_2\text{O}}} = \frac{[(a\Omega + b)T + (c\Omega + d)][\alpha(e\Omega + fT + g) + 1]\Omega}{\exp\left(\frac{\quad}{T^2}\right)} \quad (5)$$

where η and $\eta_{\text{H}_2\text{O}}$ are the viscosities of the amine solution and water, respectively (mPa·s), Ω is the mass percent amine, T is the temperature (K), and α is the CO₂ loading (mol of CO₂/mol of amine). Coefficients for use in eq 5 are given in Table 7. The main difficulty in correlating the data was in the selection of a suitable correlating form. We evolved the unlikely looking equation above from the initial supposition that viscosity might follow an Andrade correlation (see Reid et al. (1987), p 439) in which viscosity depends on the exponential of the inverse temperature. (The Andrade correlation has been commonly used with success to express the temperature dependence of pure component viscosities.) We then developed correlating forms for the effect of amine concentration and CO₂ loading assuming that the loading and amine concentration functions appeared as products; i.e., the correlating form used was $\eta/\eta_{\text{H}_2\text{O}} = \exp\{f(w)g(\alpha)/T\}$. The function $f(w)$ was determined from unloaded solution data, and it turned out to be temperature-dependent (except for MEA). The function $g(\alpha)$ turned out to depend on both temperature (although only for MEA and then very weakly) and amine concentration. Thus, disregarding the weak (for MEA) or nonexistent (for DEA and MDEA) dependence of $g(\alpha)$ on T , we have indeed a $1/T$ dependence of the viscosity ratio on temperature. Thus, the Andrade form can be made to fit the data for this rather complex system by allowing what are constants for pure components to become functions of composition for mixtures.

Equation 5 can be used to calculate MDEA, DEA, and MEA solution viscosities up to amine concentrations of 60 mass %, 77.2 mass %, and 40 mass %, respectively, with CO₂ loadings up to 0.5 mol of CO₂/mol of amine for MDEA and DEA and 0.6 mol of CO₂/mol of amine for MEA and to a maximum temperature 398 K. There are minimal experimental data above these limits. The standard deviations of the fits for each of the amines is given in Table 7.

Rather than correlate the viscosity for loaded, mixed-amine systems, we attempted to develop a mixing rule that would allow us to predict the viscosity of a mixed-amine

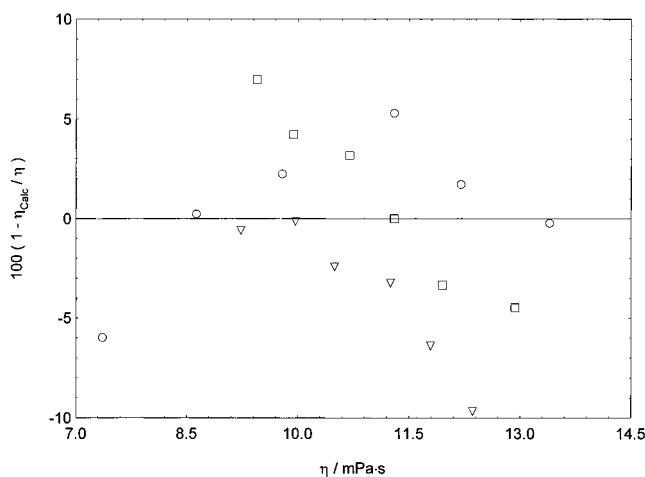


Figure 3. Comparison between predicted and measured viscosity of MEA/MDEA blends: ▽, 20 mass % MEA + 30 mass % MDEA; □, 10 mass % MEA + 30 mass % MDEA; ○, 5 mass % MEA + 45 mass % MDEA.

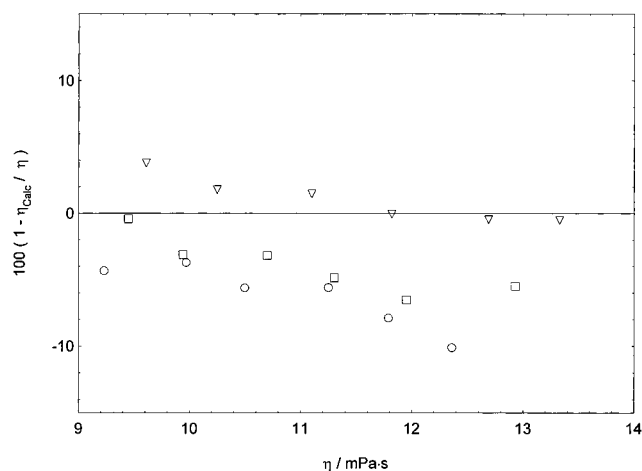


Figure 4. Comparison between predicted and measured viscosity of DEA/MDEA blends: ▽, 20 mass % DEA + 30 mass % MDEA; □, 10 mass % DEA + 30 mass % MDEA; ○, 5 mass % DEA + 45 mass % MDEA.

system from data for single amines. The following formulation involving a mass fraction weighting was found best to represent the mixed amine data:

$$\eta_{\text{mix},\alpha} = \frac{w_1}{w_1 + w_2} \eta_{1,\alpha} + \frac{w_2}{w_1 + w_2} \eta_{2,\alpha} \quad (6)$$

Here $\eta_{\text{mix},\alpha}$ indicates the viscosity of the mixed amines in water and $\eta_{1,\alpha}$ and $\eta_{2,\alpha}$ are the viscosities of the single amines in water (at concentration $w_1 + w_2$), all at loading α , and w_i is the weight fraction of amine i . This approach to the calculation of loaded, aqueous, mixed-amine viscosity correlated the results with the deviations shown in Figures 3 and 4 for MEA + MDEA and DEA + MDEA blends, respectively. Predictions based on single-amine data agree with the mixed-solvent data to within, at worst, about 10%; however, there is a general tendency to underestimate slightly the viscosity of these MDEA-based blends containing small amounts of MEA or DEA and to overestimate the viscosity when there are relatively large amounts of MEA or DEA. Perhaps it is not surprising that the predictions for blends do not give closer agreement with the data. When such a blend is gradually loaded with CO₂, the CO₂ tends to react first with the most reactive amine

(MEA or DEA), and only after the MEA or DEA component has been extensively loaded does the CO₂ react to form predominantly bicarbonate ion. Therefore, a simple mole or mass fraction weighting cannot be expected to represent the data correctly, because such weightings completely ignore the fact that the CO₂ distributes itself between the amines more in proportion to amine reactivity than to amine concentration. An approach based on speciation would be expected to yield a better representation of the data, but that order of complexity is probably not warranted and it would make the correlation difficult to use.

It is significant that existing data on the effect of H₂S loading on solution density and viscosity are not only insufficient to permit their correlation but they are often quite inconsistent. It is not even known, for example, whether increasing H₂S loads cause solution viscosities to increase or decrease, let alone by how much—the few data available often show opposite trends. H₂S being an important acid gas removed in gas-treating processes, this serious lack of data needs to be corrected.

Conclusions

The density and viscosity of aqueous alkanolamine solutions show a significant dependence on amine concentration and acid gas loading. These properties increased with increasing carbon dioxide loading and amine concentration. Correlations were developed to allow the calculation of alkanolamine solution density and viscosity as a function of amine concentration, CO₂ loading, and temperature for single-amine solutions. These correlations can be used for mixed solvents.

Acknowledgment

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