bounds of the smoothed data are within this average deviation.

The experimental data were cross-plotted to obtain equilibrium isotherms, as shown in Figure 5. The cross plot of the data of this work did not show maximum and minimum pressures, as reported by Berecz and Balla-Achs. The cross-plotted data reported by Unruh and Katz compare well to the data of this work up to 281 K. At temperatures above 283 K, there are significant discrepancies in the isotherms. We note that Unruh and Katz had only 5 data points compared to our 14 data points above 283 K.

There may be several reasons to describe the discrepancies reported by Berecz and Balla-Achs. They did not report the exact procedures they used to measure carbon dioxide concentrations. Apparently, they reported concentrations at loading (before hydrate formation) and they did not measure the vapor composition at the equilibria. If that is the case, the concentration differences due to carbon dioxide solubility may have been neglected. Discrepancies may also have occurred due to the impurities they had either in their carbon dioxide or in their methane. It may be noted that the impurities become very significant if they are hydrate-formers.

Conclusions

Hydrate equilibrium data of carbon dioxide and methane binary mixtures are presented and compared to the previous data published in the literature. The data reported by Unruh and Katz compare well to those of this work up to 281 K, although they had calculated the carbon dioxide concentration in the vapor phase. This work does not show unusual hydrate phenomena as reported by Berecz and Balla-Achs, whose pure-components impurities may have caused the discrepancies.

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Physical Properties of Aqueous AMP Solutions

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The solubility and diffusivity of N_2O in 2 and 3 M AMP (2-amino-2-methyl-1-propanol) solutions were measured and correlated with temperature. The N_2O analogy was used to estimate the solubility and diffusivity of CO_2 in AMP solutions. The density of aqueous AMP solutions was determined over a wide range of temperatures and compositions. The viscosity of 2 and 3 M AMP solutions was measured over the temperature range 23-77 °C.

1. Introduction

There are many advantages in the use of aqueous AMP (2-amino-2-methyl-1-propanol) solutions for the removal of the acid gas compounds H_2S and CO_2 from various types of gases (*1*, *2*). For the determination of reaction kinetics and in the design of suitable gas-liquid absorbers, it is necessary to use the physicochemical properties of aqueous AMP solutions, such as the density and viscosity of the solutions, as well as the solubility and diffusivity of the acid gas in the solutions. The literature data are insufficient or unavailable, and it is necessary to determine these physicochemical properties of AMP solutions.

Due to the chemical reaction that occurs in the solution when it mixes with the acid gas, it is impossible to obtain these transfer properties directly. In view of the similarities with regard to configuration, molecular volume, and electronic structure, N_2O is often used as a nonreacting gas to estimate the properties of CO_2. It has been proved that "N_2O analogy" may be applied to estimate the solubility of CO_2 in aqueous alkanolamine solutions according to the following equation (*3*, *5*):

$$(\text{solubility of } CO_2) = C_1(\text{solubility of } N_2O) \tag{1}$$

with

$$C_1 = \frac{(\text{solubility of } CO_2 \text{ in water})}{(\text{solubility of } N_2O \text{ in water})} \tag{2}$$

The estimation of the diffusivity of CO_2 in alkanolamine solutions may be done in different ways. Sada et al. (*6*) and Haimour and Sandall (*7*) considered that the N_2O analogy can be used to estimate the diffusivity of CO_2. Diaz et al. (*4*) proved this analogy by using aqueous alcohol solutions. However, Versteeg et al. (*8*) thought that the analogue was not a general relation and suggested a modified Stokes-Einstein relation to calculate the diffusivity of CO_2:

$$(D_{N_2O}\eta^{0.8})_{\text{am soln}} = \text{constant} = (D_{N_2O}\eta^{0.8})_{\text{water}} \tag{3}$$

For convenience, Versteeg et al. (*8*) correlated the data of solubility and diffusivity of N_2O and CO_2 in water based on their own and other experimental results. These correlations are as follows:

$$H_{N_2O} = 8.55 \times 10^6 \exp(-2284/T) \tag{4}$$

$$H_{CO_2} = 2.82 \times 10^6 \exp(-2044/T) \tag{5}$$

$$D_{N_2O} = 5.07 \times 10^{-6} \exp(-2371/T) \tag{6}$$

$$D_{CO_2} = 2.35 \times 10^{-6} \exp(-2119/T) \tag{7}$$

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Table I. Solubility of N_2O in Water[a]

T, K	solubility, (kPa·m³)/kmol	ref
298.2	4171.3	Sada and Kito (*11*)
	4132.2	Sada et al. (*12*)
	4154.1	Joosten and Danckwerts (*13*)
	4173.0	Haimour and Sandall (*7*)
	4211.8	Markham and Kobe (*14*)
	4026.0*	Versteeg and van Swaaij (*8*)
	4176.0	present work
323.2	7282.0*	Versteeg and van Swaaij (*8*)
	7254.2	present work
348.2	12097.0*	Versteeg and van Swaaij (*8*)
	12347.8	present work

[a] Numbers marked with an asterisk come from eq 4 presented by Versteeg and van Swaaij.

In this study, new experimental results on the solubility and diffusivity of N_2O in water and two aqueous AMP solutions at various temperatures and local pressure will be presented. Additionally, the data on the density and viscosity of these solutions will be presented, and these data will be compared with the literature data that are available. Finally, a method that can be used to estimate the diffusivity of CO_2 in AMP solutions will be presented.

2. Experimental Section

2.1. Density and Viscosity. Densities were measured by means of calibrated 25-mL pycnometer bottles, and viscosities were measured by means of Cannon-Fenske routine viscometers. The measurements were performed in a constant-temperature water bath, in which the temperature could be held constant to ±0.05 K. The experimental errors were estimated to be equal to ±0.01 kg/m³ and $\pm 1 \times 10^{-6}$ (N·s)/m², respectively.

Analytical grade AMP with a purity of ≥97%, supplied by BDH Chemicals Co., was used in this experiment. The N_2O used was supplied from Union Carbide Canada Ltd. with minimum purity of 99.8%.

2.2. Solubility. Solubilities were measured in a glass flask with a volume of 250 mL at the local pressure of ~94 kPa. The experimental apparatus and operating method are similar to that of Haimour and Sandall (*7*). The temperatures of the water bath and environment can be kept constant within ±0.05 and ±0.2 K, respectively. About 50 mL of solution was injected into the flask, which was weighed by a balance whose precision is $\pm 1 \times 10^{-6}$ kg. The partial pressures of N_2O in the experiments were corrected for the vapor pressure of the solution. The maximum experimental error in the measured gas solubility is estimated as ±2%.

Before the solubilities in AMP solutions were measured, the solubilities of N_2O in water were measured with this apparatus. Table I compares these data with the values found in the literature. It is seen that the results of this work are in good agreement with the literature values for water.

2.3. Diffusivity. A single-sphere absorber apparatus was used to measure the diffusivities of N_2O in water and in AMP solutions. It included a laminar jet and a sphere of 50.8-mm diameter. This apparatus and operating procedure are similar to that used by Tomcej and Otto (*9*). The gas absorption rate was measured with a calibrated soap-film meter and the liquid flow rate by a rotameter that was calibrated at various experimental temperatures and concentrations of the solutions. The temperature of the experiment was held constant within ±0.1 K, and the total pressure in the absorption chamber was maintained at about 100 kPa for all the experiments. The experimental error was estimated to be ±5%.

Table II. Diffusivity (D) of CO_2 in Water at 25 °C

D, 10⁹ m²/s	ref
1.92	Davidson and Cullen (*15*)
1.92	Ratcliff and Holdcroft (*16*)
2.00	Vivian and King (*17*)
1.95	Thomas and Adams (*18*)
1.93	Khalil (*19*)
1.97	Tomcej and Otto (*9*)
1.93	Versteeg and van Swaaij (*8*)
1.93	present work

Table III. Density (ρ) of Aqueous AMP Solutions

t, °C	AMP concn, wt %	ρ, kg/m³	t, °C	AMP concn, wt %	ρ, kg/m³
20.3	9.05	997.5	20.0	73.62	977.8
31.9	9.05	994.0	31.9	73.62	968.5
41.9	9.05	989.3	43.1	73.62	959.3
55.5	9.05	983.0	54.7	73.62	949.6
66.0	9.05	976.9	65.8	73.62	940.3
77.8	9.05	969.6	77.8	73.62	929.9
88.7	9.05	961.5	90.7	73.62	918.1
20.0	27.33	996.7	20.0	88.32	957.9
32.1	27.33	993.1	31.9	88.32	948.3
43.4	27.33	986.6	43.1	88.32	939.3
55.5	27.33	978.9	54.7	88.32	929.5
66.0	27.33	971.6	65.8	88.32	920.1
77.8	27.33	963.2	77.8	88.32	909.6
88.7	27.33	955.0	90.7	88.32	897.8
20.3	50.94	996.0	31.9	100.00	925.4
31.9	50.94	987.9	43.1	100.00	916.0
41.9	50.94	979.7	54.7	100.00	906.2
55.5	50.94	968.8	65.8	100.00	897.0
66.0	50.94	960.6	77.8	100.00	886.4
77.8	50.94	950.7	90.7	100.00	874.8
88.7	50.94	941.5			

Table IV. Viscosity (η) of Aqueous AMP Solutions

	η, 10³ (N·s)/m²			η, 10³ (N·s)/m²	
t, °C	2 M AMP	3 M AMP	t, °C	2 M AMP	3 M AMP
23.6	2.052	3.126	56.5	0.864	1.146
35.4	1.439	2.058	66.9	0.712	0.921
45.9	1.104	1.515	76.7	0.602	0.763

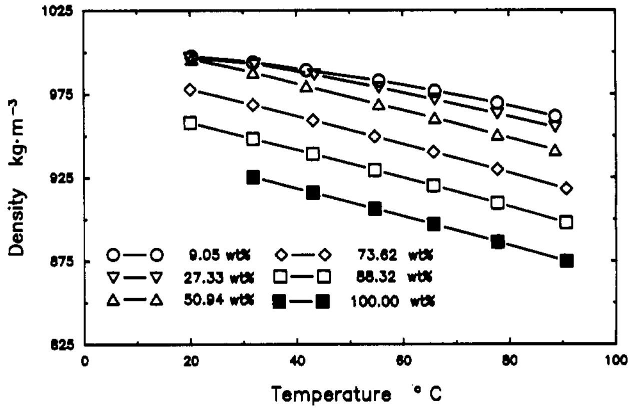

Figure 1. Densities of aqueous AMP solutions.

Sodium lauryl sulfate (Teepol) was added to water or aqueous AMP solution to promote the wetting ability at the sphere surface, especially at high temperatures. It had been proved that the effect of Teepol on the diffusivity is negligible (*9*). Teepol was added to 0.1 wt % for water, 0.01 wt % for 2 M AMP solution, and 0.005 wt % for 3 M AMP solution.

With use of the apparatus, the diffusion coefficient of CO_2 in water at 25 °C was determined to be 1.93×10^{-9} m²/s. As shown in Table II, it is in good agreement with values that were published.

3. Result and Discussion

3.1. Density and Viscosity. The results of the density determination are presented in Table III and plotted in Figure

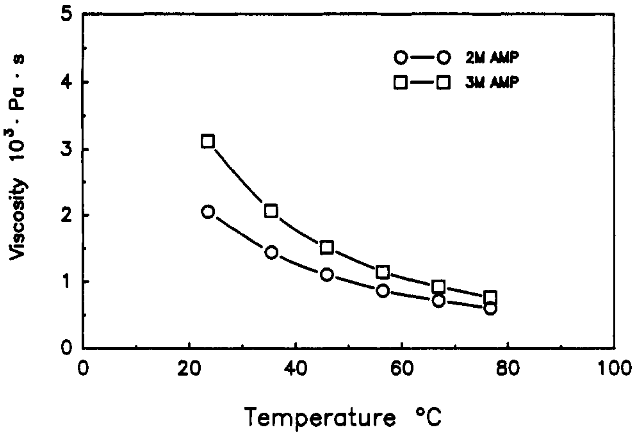

Figure 2. Viscosities of aqueous AMP solutions.

Table V. Density (ρ) of Aqueous AMP Solutions at 40 °C

AMP concn, kmol/m^3	ρ, kg/m^3 values of eq 12	values of ref 10
0.000	992.2	992.3
0.258	991.6	990.4
0.462	991.2	989.7
0.900	990.5	987.9
3.000	987.9	985.7

1. The results of the viscosity of 2 and 3 M AMP solutions are listed in Table IV and plotted in Figure 2.

In the experimental temperature region, the density of AMP can be correlated by a polynomial:

$$\rho_{AMP} = 950.65 - 0.71015t - 7.1246 \times 10^{-4}t^2 \quad (8)$$

However, the linear combination

$$\rho_m = y\rho_{AMP} + (1 - y)\rho_{water} \quad (9)$$

is not suitable for calculating the density of aqueous AMP solutions, since the AMP is a polar material. The mixing process of AMP with water results in a nonideal solution. According to the experimental data, the excess density, $\Delta\rho_m$, of aqueous AMP solutions can be empirically regressed by

$$\Delta\rho_m = y(1 - y)(1 + a_1y - a_2y^2)Tg \quad (10)$$

$$Tg = b_1 - b_2t + b_3t^2 - b_4t^3 \quad (11)$$

where

$$a_1 = 2.6362$$

$$a_2 = 0.4453$$

$$b_1 = 11.0048$$

$$b_2 = 5.3281 \times 10^{-3}$$

$$b_3 = -1.1854 \times 10^{-3}$$

$$b_4 = 3.2895 \times 10^{-7}$$

So, the density of aqueous AMP solution can be calculated by eq 12 conveniently. The average deviation of the regression is 0.1% within the experimental region.

$$\rho_{sol.} = \rho_m + \Delta\rho_m \quad (12)$$

Table V shows that densities of AMP solution calculated by eq 12 are in good agreement with the values found in the literature (*10*). Figure 3 compares the viscosities of this work with the values found in the literature (*1*, *10*). The data at 40 °C are in good agreement with the work of Yih and Shen (*10*), but the data at 25 °C differ somewhat from the data of Bosch (*1*).

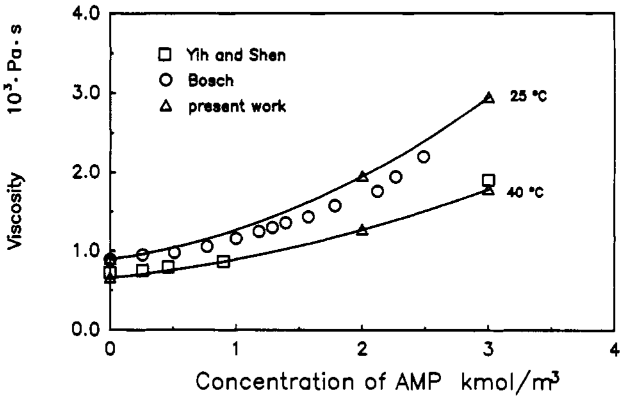

Figure 3. Comparison of viscosity.

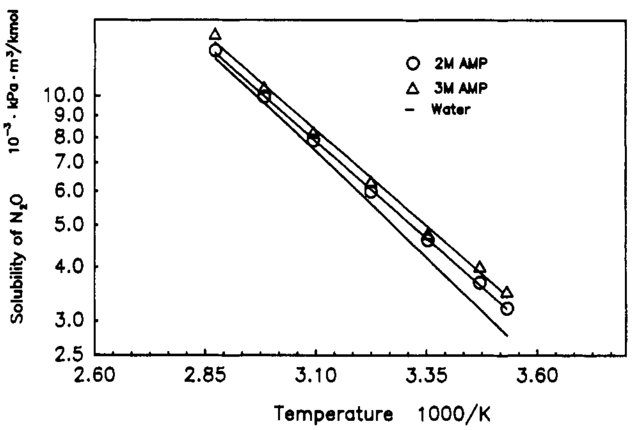

Figure 4. Solubility of N_2O in aqueous AMP solutions.

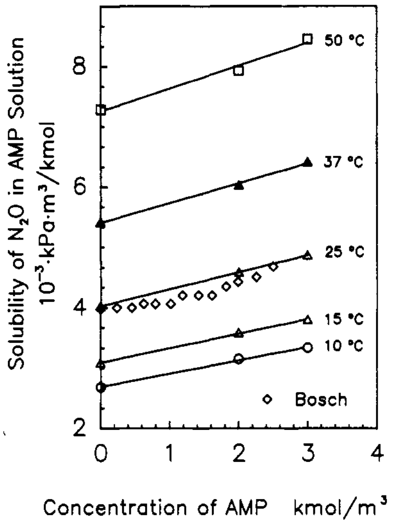

Figure 5. Solubility of N_2O in AMP solutions.

Table VI. Solubility of N_2O in Aqueous AMP Solutions

	solubility, (kPa·m^3)/kmol			solubility, (kPa·m^3)/kmol	
T, K	2 M AMP	3 M AMP	*T*, K	2 M AMP	3 M AMP
283.2	3212.6	3518.0	323.2	7876.6	8266.6
288.2	3683.4	4020.3	335.2	9949.3	10450.6
298.2	4617.1	4842.2	348.2	12754.8	13912.9
310.2	5985.5	6312.9			

3.2. Solubility. The data for the solubility of N_2O in AMP solutions are presented in Table VI and plotted in Figure 4, for

Table VII. Diffusivity (D) of N_2O in Aqueous AMP Solutions

AMP concn, kmol/m^3	t, °C	D, 10^9 m^2/s	AMP concn, kmol/m^3	t, °C	D, 10^9 m^2/s
2.0	25.2	0.81	3.0	21.4	0.59
2.0	26.7	0.91	3.0	23.4	0.62
2.0	28.1	0.97	3.0	34.9	1.06
2.0	31.4	1.02	3.0	45.6	1.57
2.0	40.3	1.47	3.0	55.0	2.27
2.0	50.4	2.09	3.0	64.8	3.00
2.0	60.7	2.82	3.0	74.9	4.07
2.0	70.5	3.88	3.0	75.5	4.17
2.0	74.8	4.44			

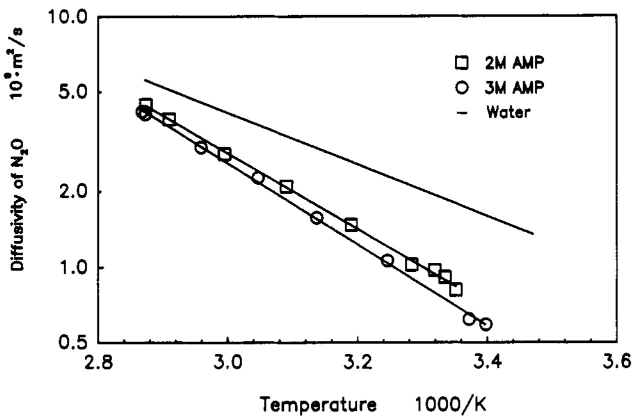

Figure 6. Diffusivity of N_2O in aqueous AMP solutions.

comparison with the solubility of N_2O in water calculated from eq 4. These data may be correlated by eqs 13 and 14 for 2

$$H_{N_2O\text{-}2\,M\,AMP} = 5.446 \times 10^6 \exp(-2110.7/T) \quad (13)$$

$$H_{N_2O\text{-}3\,M\,AMP} = 6.085 \times 10^6 \exp(-2125.9/T) \quad (14)$$

and 3 M AMP solutions, respectively. These data show that the Henry's coefficient increases with an increase in concentration of the AMP solution. The solubility data are linear with the concentration of AMP, as shown by Figure 5. The data are not in agreement with the results of Bosch (*1*) at low concentrations of AMP. Based on these solubility data of N_2O in aqueous AMP solutions, the data can be correlated as a function of temperature and concentration of AMP, viz.

$$H_{N_2O\text{-}AMP} = (6.03 + 0.29M) \times 10^6 \exp(-2170/T) \quad (15)$$

This average deviation of the regression is 2.75% within the experimental region.

According to the N_2O analogy, with use of eqs 1, 4, 5, and 15, the solubility of CO_2 in an AMP solution can be obtained.

3.3. Diffusivity. The diffusivities of N_2O in 2 and 3 M AMP solutions are presented in Table VII and plotted in Figure 6, together with the diffusivity of N_2O in water calculated from eq 6. These data also show that the diffusivity changes with temperature at the same concentration of AMP in a exponential fashion. The diffusivity decreases with increasing amine concentration. These results are in agreement with the trend of the data reported in the literature (*10*), which are plotted in Figure 7. The data differ by 15% for the 3 M AMP solution; however, the diffusivity in water published by Yih and Shen (*10*) is lower than that calculated from eq 6.

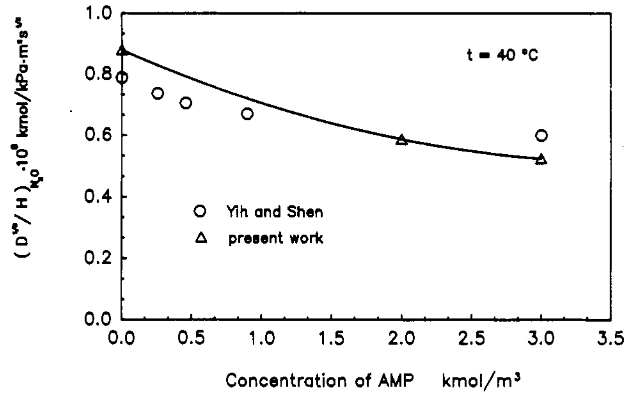

Figure 7. Comparison of diffusivity.

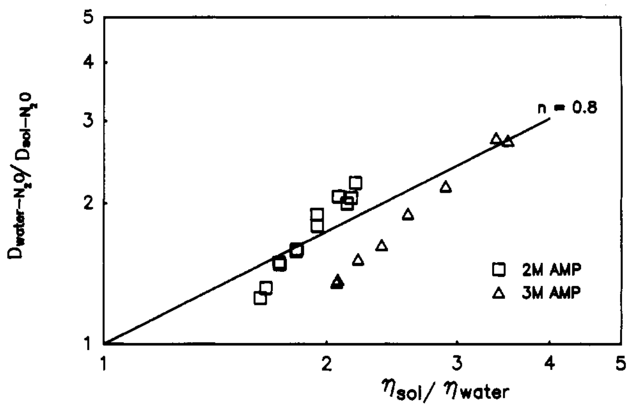

Figure 8. Stokes-Einstein plot for diffusivity.

Table VIII. Diffusivity (D) of CO_2 in Aqueous AMP Solutions

	2 M AMP		3 M AMP	
t, °C	D_1,[a] 10^9 m^2/s	D_2,[b] 10^9 m^2/s	D_1,[a] 10^9 m^2/s	D_2,[b] 10^9 m^2/s
25.0	0.73	1.03	0.78	0.74
35.0	1.25	1.38	1.12	1.04
45.0	1.75	1.81	1.58	1.40
55.0	2.39	2.32	2.18	1.84
65.0	3.20	2.93	2.95	2.37
75.0	4.43	3.62	3.93	2.99

[a] $D_1 = D_{CO_2}(\text{in water})\,[D_{N_2O}(\text{in amine})/D_{N_2O}(\text{in water})]$.
[b] $D_2 = D_{CO_2}(\text{in water})\,(\eta_{water}/\eta_{amine})^{0.8}$.

Figure 8 shows the values of diffusivity measured in the present study divided by the diffusivity of N_2O in water and plotted against the ratio of the viscosity of the AMP solution to that of water. Unfortunately, it indicates that the modified Stokes-Einstein relation of Versteeg et al. (*8*) is not suitable for estimating the diffusivity of CO_2 in AMP solutions. Yih and Shen (*10*) have used a method similar to the N_2O analogy method to estimate the diffusivity of CO_2 in aqueous AMP solutions; the results were used to calculate the rate constants for CO_2 in AMP solutions. The two methods to estimate the diffusivity of CO_2 in AMP solutions from the diffusivity of N_2O in AMP solutions are compared in Table VIII. The results indicate that the N_2O analogy is more reasonable because the calculated diffusivities of CO_2 in AMP solutions estimated by the N_2O analogy are similar to the data of the diffusivity of N_2O in AMP solutions. The N_2O analogy method to estimate the diffusivity of CO_2 in AMP solution is recommended.

4. Conclusions

The results of the experimental work in the present study lead to the following conclusions:

1. The solubility of N_2O in aqueous AMP solutions changes with temperature based on an exponential relationship at a constant concentration of AMP; at constant temperature the solubility is a linear function of concentration. Under the experimental conditions, the solubility data of N_2O in aqueous AMP solution can be obtained by eq 15. The solubility of CO_2 in aqueous AMP solutions can be estimated by the N_2O analogy.

2. It is preferable to use the N_2O analogy to estimate the diffusivity of CO_2 in aqueous AMP solutions rather than the Stokes–Einstein relation.

3. An empirical correlation, based on the experimental data, is presented for calculating the density of aqueous AMP solutions over the temperature range 20–90 °C and the complete range of concentration.

Nomenclature

a	parameter of eq 10
b	parameter of eq 11
C_1	constant defined by eq 1
D	diffusivity, m^2/s
H	Henry's coefficient, $(kPa \cdot m^3)/kmol$
M	molarity of amine solution, $kmol/m^3$
t	temperature, °C
T	temperature, K
y	volume fraction
η	viscosity, $(N \cdot s)/m^2$
ρ	density, kg/m^3

Subscripts

am	alkanolamine
AMP	2-amino-2-methyl-1-propanol
CO_2	carbon dioxide
m	mixture
N_2O	nitrous oxide
sol.	amine solution
water	pure water

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Excess Volumes of *n*-Hexane + *n*-Undecane between 288.15 and 308.15 K

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Excess molar volumes of *n*-hexane + *n*-undecane have been determined from density measurements at 288.15, 293.15, 298.15, 303.15, and 308.15 K. The densities were obtained with an Anton Paar densimeter. The minimum values of V_m^E for the systems *n*-hexane, *n*-heptane, *n*-octane, and *n*-nonane + *n*-undecane are plotted against temperature.

Introduction

In previous works (*1–3*) the excess volumes of *n*-heptane + *n*-undecane, *n*-octane + *n*-undecane, and *n*-nonane + *n*-undecane at 288.15, 293.15, 298.15, 303.15, and 308.15 K have been determined. This article reports similar measurements for $(x)n\text{-}C_6H_{14} + (1 - x)n\text{-}C_{11}H_{24}$, together with three-parameter empirical equations fitted to the results obtained at each temperature.

Experimental Section

Densities were measured with an Anton Paar DMA 60/602 densimeter with a resolution of $\pm 1.5 \times 10^{-6}$ g cm^{-3}. The temperature of the water bath containing the oscillator tube was kept constant to within ±0.005 K with a Hetofrig thermostat. The operational technique has been described elsewhere (*4*). The *n*-hexane (Fluka purum grade, >99 mol %) and *n*-undecane (Merk, >99 mol %) were carefully dried with Union Carbide Type 4A molecular sieve (from Fluka) and were used directly without purification. The observed densities of *n*-hexane and *n*-undecane at the various temperatures employed are given in Table I, along with published values (*5–10*) for comparison.

Table I. Experimental Densities (g cm^{-3}) at Various Temperatures and Comparison with Published Values

	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K
			Experimental Values		
n-hexane	0.664 02	0.659 47	0.654 95	0.650 33	0.645 72
n-undecane	0.744 29	0.740 59	0.736 90	0.733 20	0.729 51
			Literature Values		
n-hexane	0.663 45[a]	0.659 33[b]	0.655 09[c]	0.650 18[b]	0.645 78[d]
n-undecane	0.743 55[a]	0.740 17[e]	0.736 50[f]	0.732 84[f]	0.729 07[d]

[a] Reference 5. [b] Reference 6. [c] Reference 7. [d] Reference 8. [e] Reference 9. [f] Reference 10.

Results and Discussion

Table II lists the excess molar volumes V_m^E calculated from the observed densities for each temperature T and each value

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