

# Solubility of Ethane in Aqueous Solutions of Monoethanolamine and Diethanolamine

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The solubility of ethane in  $3 \text{ kmol}\cdot\text{m}^{-3}$  aqueous solutions of monoethanolamine and diethanolamine has been determined at temperatures in the range of (298 to 398) K at pressures up to 13.2 MPa. The experimental results were modeled using a Henry's law approach, and the results are summarized in terms of salting-in coefficients and Setchenow coefficients.

## Introduction

Aqueous solutions of alkanolamines are widely used in the natural gas industry to remove the acid gases,  $\text{H}_2\text{S}$  and  $\text{CO}_2$ , from natural gases. Some of the common alkanolamines used in the process are monoethanolamine (MEA), diethanolamine (DEA), and methyldiethanolamine (MDEA). Another alkanolamine, triethanolamine (TEA), is no longer used. The solubility of the light hydrocarbons in alkanolamine solutions is important, as the dissolved hydrocarbons constitute a loss to the process. As such, the estimation of the hydrocarbon content in the alkanolamine is imperative in the design and evaluation phase of these processes. Despite this importance, there are only a limited number of experimental data sets dealing with the solubility of the lighter hydrocarbons in alkanolamines. Lawson and Garst<sup>1</sup> measured the solubility of methane and ethane in MEA and DEA solutions. Jou et al.<sup>2</sup> measured the solubility of ethane in TEA solutions. In that study, several amine concentrations were used, and these data show the effect of amine concentration on solubility. Jou et al.<sup>3</sup> determined the solubility of methane and ethane in MDEA solutions. Carroll and Mather<sup>4</sup> presented a model for the solubility of light hydrocarbons in aqueous alkanolamine solutions. This model will be used for the correlation of the data reported in this paper.

## Experimental Section

The apparatus and experimental technique that were used are similar to those described by Jou et al.<sup>5</sup> The equilibrium cell was mounted in an air bath. The temperature of the contents of the cell was measured by a calibrated iron–constantan thermocouple, and the pressure in the cell was measured by digital Heise gauges (0 to 10, 0 to 35 MPa). These gauges had an accuracy of  $\pm 0.1\%$  of full scale by comparison with a dead-weight gauge. The thermocouple had an accuracy of  $\pm 0.1^\circ\text{C}$  by comparison with a platinum resistance thermometer. The alkanolamines were obtained from Fisher and had a purity of 99.5%, which was determined by titration. The water was double-distilled. Ethane was obtained from Matheson and had a purity of 99%.

Prior to the introduction of the fluids, the cell was evacuated. About  $100 \text{ cm}^3$  of the amine solution was allowed to flow into the cell. The ethane was added to the cell by the cylinder

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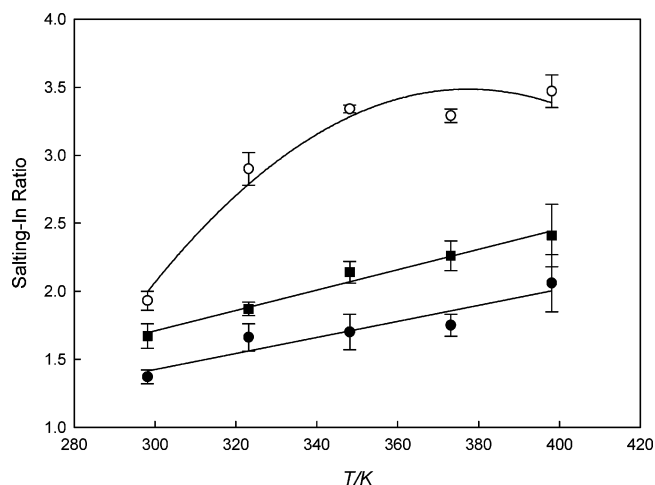


Figure 1. Salting-in ratios for ethane in  $3 \text{ kmol}\cdot\text{m}^{-3}$  aqueous solutions of alkanolamines: ●, MEA; ■, DEA; ○, TEA.<sup>2</sup>

pressure or by means of a spindle press. The circulation pump was started and the vapor bubbled through the solvent for at least 8 h to ensure that equilibrium was reached. A sample of the liquid phase, (2 to 20) g, depending on the solubility, was withdrawn from the cell into a  $50 \text{ cm}^3$  sample bomb that had previously been evacuated and weighed. The bomb contained a magnetic stirring bar to help in degassing the sample. The sample bomb was reweighed to determine the mass of the sample and then attached to a vacuum rack. The rack consisted of 6.35 mm o.d. stainless steel tubing connected to a calibrated Digigauge (0 to 1.0 MPa) and a  $50 \text{ cm}^3$  buret. The rack was evacuated, and the gas was allowed to evolve from the sample bomb into the buret. The moles collected were calculated from the  $P-V-T$  data, assuming ideal gas behavior. A correction was made for the residual ethane left in the sample at atmospheric pressure. The uncertainty in the liquid-phase analyses is estimated to be  $\pm 3\%$ .

## Results and Discussion

The solubility of ethane in  $3 \text{ kmol}\cdot\text{m}^{-3}$  solutions of monoethanolamine and diethanolamine was determined at the temperatures of (298.15, 323.15, 348.15, 373.15, and 398.15) K at pressures up to 13.2 MPa. The experimental data are presented in Tables 1 and 2. The data were correlated using the model of Carroll and Mather.<sup>4</sup> The model used here is identical to that

**Table 1. Solubility of Ethane (2) in a 3 kmol·m<sup>-3</sup> Monoethanolamine Solution**

298.15 K		323.15 K		348.15 K		373.15 K		398.15 K	
<i>P</i> /MPa	<i>x</i> <sub>2</sub> ·10 <sup>3</sup>	<i>P</i> /MPa	<i>x</i> <sub>2</sub> ·10 <sup>3</sup>	<i>P</i> /MPa	<i>x</i> <sub>2</sub> ·10 <sup>3</sup>	<i>P</i> /MPa	<i>x</i> <sub>2</sub> ·10 <sup>3</sup>	<i>P</i> /MPa	<i>x</i> <sub>2</sub> ·10 <sup>3</sup>
4.215	1.11	11.31	1.34	13.10	1.26	12.61	1.42	12.62	1.97
2.34	0.829	9.14	1.22	10.03	1.20	9.97	1.34	8.82	1.60
0.907	0.346	6.37	1.13	6.99	1.09	6.31	0.982	6.29	1.23
0.370	0.162	4.15	0.874	5.76	0.931	5.11	0.926	4.20	0.943
0.092	0.039	2.40	0.574	4.09	0.726	2.99	0.597	2.37	0.618
		0.991	0.274	2.49	0.493	0.854	0.191	0.838	0.213
		0.267	0.086	0.822	0.195	0.279	0.068	0.294	0.0719
				0.276	0.07				

**Table 2. Solubility of Ethane (2) in a 3 kmol·m<sup>-3</sup> Diethanolamine Solution**

298.15 K		323.15 K		348.15 K		373.15 K		398.15 K	
<i>P</i> /MPa	<i>x</i> <sub>2</sub> ·10 <sup>3</sup>	<i>P</i> /MPa	<i>x</i> <sub>2</sub> ·10 <sup>3</sup>	<i>P</i> /MPa	<i>x</i> <sub>2</sub> ·10 <sup>3</sup>	<i>P</i> /MPa	<i>x</i> <sub>2</sub> ·10 <sup>3</sup>	<i>P</i> /MPa	<i>x</i> <sub>2</sub> ·10 <sup>3</sup>
4.218	1.35	12.14	1.43	13.20	1.63	12.78	1.92	12.11	2.31
2.53	1.09	10.61	1.45	10.12	1.55	10.51	1.81	9.57	2.01
1.51	0.659	8.42	1.32	8.61	1.44	6.79	1.39	7.46	1.68
0.507	0.248	6.21	1.25	6.17	1.25	4.06	0.973	4.24	1.14
0.092	0.050	4.07	0.976	4.19	0.926	2.44	0.608	2.98	0.807
		2.56	0.722	2.53	0.664	0.896	0.261	1.01	0.318
		0.783	0.257	0.812	0.243	0.200	0.065	0.279	0.088
		0.244	0.091	0.248	0.072				

**Table 3. Salting-in and Setchenow Coefficients for Ethane in Aqueous MEA Solutions**

<i>T</i> /K	amine concentration		salting-in coefficient	Setchenow coefficient	reference
	mass %	molarity			
298.15	18.1	3.00	1.37 ± 0.05	0.105 ± 0.012	this work
310.95	15.0	2.48	1.38 ± 0.01	0.130 ± 0.003	1
310.95	40.0	6.69	1.93 ± 0.34	0.098 ± 0.026	1
323.15	18.1	3.00	1.66 ± 0.10	0.169 ± 0.020	this work
338.75	15.0	2.48	1.51 ± 0.07	0.166 ± 0.019	1
338.75	40.0	6.69	2.50 ± 0.06	0.137 ± 0.004	1
348.15	18.1	3.00	1.70 ± 0.13	0.177 ± 0.025	this work
373.15	18.1	3.00	1.75 ± 0.08	0.187 ± 0.015	this work
398.15	18.1	3.00	2.06 ± 0.21	0.241 ± 0.034	this work

**Table 4. Salting-in and Setchenow Coefficients for Ethane in Aqueous DEA Solutions**

<i>T</i> /K	amine concentration		salting-in coefficient	Setchenow coefficient	reference
	mass %	molarity			
298.15	30.4	3.00	1.67 ± 0.09	0.171 ± 0.018	this work
310.95	5.0	0.48	1.17 ± 0.03	0.327 ± 0.053	1
310.95	25.0	2.45	1.52 ± 0.04	0.171 ± 0.011	1
323.15	30.4	3.00	1.87 ± 0.05	0.209 ± 0.009	this work
338.75	5.0	0.48	1.18 ± 0.01	0.345 ± 0.018	1
338.75	25.0	2.45	1.68 ± 0.05	0.212 ± 0.012	1
348.15	30.4	3.00	2.14 ± 0.08	0.254 ± 0.012	this work
373.15	30.4	3.00	2.26 ± 0.11	0.272 ± 0.016	this work
398.15	30.4	3.00	2.41 ± 0.23	0.293 ± 0.032	this work

presented in that paper, and the required parameters are taken from that work. Salting-in ratios, (*S*) defined as the mole fraction solubility in the amine solution divided by the mole fraction solubility in pure water, were calculated from the experimental data. The term salting-in is used because the solubility of hydrocarbons in the amine solution is greater than that in pure water. Ionic salts reduce the solubility and hence the term salting-out. Because there are no experimental data at the exact conditions of the data measured here, the model of Carroll and Mather was used to calculate the solubilities of ethane in water. The salting-in ratios for ethane in the 3 kmol·m<sup>-3</sup> solutions are presented in Tables 3 and 4 for MEA and DEA, respectively. The salting-in ratios were calculated point by point. The values given in the tables are the mean for a given temperature, along with the standard deviation. The results are plotted in Figure 1, together with the values for ethane in a 3 kmol·m<sup>-3</sup> TEA

solution. It can be seen that the salting-in ratios are an increasing function of the "size" of the amine. The data were fit by a linear function of temperature for MEA and DEA and a quadratic function of temperature for TEA. The results are as follows:

$$S_{C_2H_6-MEA} = -0.339 + 5.88 \times 10^{-3} T$$

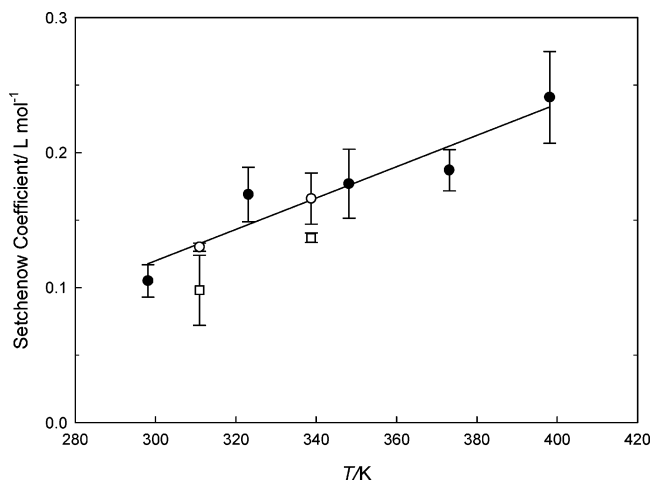
$$S_{C_2H_6-DEA} = -0.534 + 7.48 \times 10^{-3} T$$

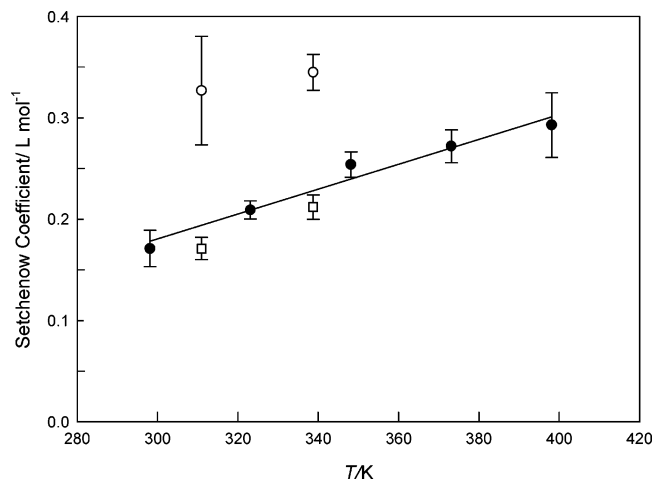
$$S_{C_2H_6-TEA} = -30.2 + 0.179T - 2.37 \times 10^{-4} T^2$$

Also given in Tables 3 and 4 are the data of Lawson and Garst. Their amine concentrations were different from those used here. However, the use of the Setchenow equation:

$$\ln S = kC_a$$

where *S* is the salting-in coefficient, *k* is the Setchenow coefficient, and *C<sub>a</sub>* is the amine concentration in molarity allows for the effect of amine concentration on the salting-in coefficient. The data of Lawson and Garst have Setchenow coefficients that are a function of the amine concentration. This is unlikely to be the case. The data presented for ethane in TEA solutions by

**Figure 2.** Temperature dependence of the Setchenow coefficients of ethane in MEA solutions: ●, this work; ○, 2.48 M, and □, 6.69 M, ref 1.



**Figure 3.** Temperature dependence of the Setchenow coefficients of ethane in DEA solutions: ●, this work; ○, 0.48 M, and □, 2.45 M, ref 1.

Jou et al.<sup>2</sup> were well-correlated by the Setchenow approach. However, there is some agreement between the present data and those of Lawson and Garst. The Setchenow coefficients for ethane in MEA solutions are plotted in Figure 2. There is good agreement for amine solutions with similar concentrations but discrepancies for higher concentrations. Similar results were

obtained for DEA solutions, as shown in Figure 3. The present data were fit by a linear function of temperature:

$$k_{C_2H_6,MEA} = -0.228 + 1.16 \times 10^{-3}T$$

$$k_{C_2H_6,DEA} = -0.188 + 1.23 \times 10^{-3}T$$

#### Literature Cited

- (1) Lawson, J. D.; Garst, A. W. Hydrocarbon gas solubility in sweetening solutions: methane and ethane in aqueous monoethanolamine and diethanolamine. *J. Chem. Eng. Data* **1976**, *21*, 30–32; Errata. *Ibid.* **1996**, *41*, 1210.
- (2) Jou, F.-Y.; Otto, F. D.; Mather, A. E. Solubility of ethane in aqueous solutions of triethanolamine. *J. Chem. Eng. Data* **1996**, *41*, 794–795.
- (3) Jou, F.-Y.; Carroll, J. J.; Mather, A. E.; Otto, F. D. Solubility of methane and ethane in aqueous solutions of methyl-diethanolamine. *J. Chem. Eng. Data* **1998**, *43*, 781–784.
- (4) Carroll, J. J.; Mather, A. E. A model for the solubility of light hydrocarbons in water and aqueous solutions of alkanolamines. *Chem. Eng. Sci.* **1997**, *52*, 545–552; Errata. *Ibid.* **1998**, *53*, 3911.
- (5) Jou, F.-Y.; Otto, F. D.; Mather, A. E. Equilibria of H<sub>2</sub>S and CO<sub>2</sub> in triethanolamine solutions. *Can. J. Chem. Eng.* **1985**, *63*, 122–125.

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