Solubility of Hydrogen Sulfide in Water + Monoethanolamine + 2-Amino-2-methyl-1-propanol

Meng-Hiu Li* and Bei-Chia Chang

Department of Chemical Energineering, Chung Yuan Christian University, Chung Li, Taiwan 32023, Republic of China

The solubilities of hydrogen sulfide in water (1) + monoethanolamine (2) + 2-amino-2-methyl-1-propanol (3) have been measured at 40, 60, 80, and 100 °C and at partial pressures of hydrogen sulfide ranging from 1.0 to 180 kPa. The ternary mixtures studied were $\omega_2 = 0$, $\omega_3 = 0.3$; $\omega_2 = 0.06$, $\omega_3 = 0.24$; $\omega_2 = 0.12$, $\omega_3 = 0.18$; $\omega_2 = 0.18$, $\omega_3 = 0.12$; and $\omega_2 = 0.24$, $\omega_3 = 0.06$ where ω is the mass fraction. The model of Kent and Eisenberg has been extended to represent the solubility of H_2S in the ternary mixtures. The model reasonably reproduces the equilibrium partial pressure of H_2S above the ternary mixtures, not only over a temperature range from 40 to 100 °C, but also at various compositions of the components in the ternary mixture.

Introduction

Alkanolamine aqueous solutions are widely used in gas treating processes to remove acid gases, such as CO_2 and H_2S, from natural, refinery, and synthesis gas streams. Industrially important alkanolamines are monoethanolamine (MEA), diethanolamine (DEA), di-2-propanolamine (DIPA), and *N*-methyldiethanol amine (MDEA). Aqueous MEA solutions have been widely used due to their high reactivity, low solvent cost, ease of reclamation, and low absorption of hydrocarbons (*1*). When the primary (or secondary) amine reacts with CO_2, stable carbamates are usually formed (*2*). The maximum CO_2 loading is limited by stoichiometry to 0.5 mol of CO_2/mol of amine when carbamate formation is the only reaction. At high CO_2 partial pressures, however, carbamates may hydrolyze and generate free amine which can react with additional CO_2; thus, the CO_2 loading of MEA may exceed 0.5. For a gas stream that contains both CO_2 and H_2S, the aqueous MDEA solution is found to be an appropriate solution for the selective removal of H_2S from the gas stream (*3, 4*). Advantages of the use of MDEA solutions include their high loading capacity (about 1.0 mol of CO_2/mol of amine) and low enthalpy of reaction with the acid gases (*5*). The lower enthalpy of reaction leads to lower energy requirements for regeneration. However, a slow reaction rate of CO_2 with tertiary amines limits the use of MDEA solutions (*2*).

Recently the use of blended amines, a solution of two or more amines in varying compositions, finds considerable improvement in absorption and savings in energy requirements (*6*). Blended amines combine the absorption characteristics of the constituent amines such as a higher loading capacity and faster reaction rates. Blends of primary and tertiary amines, such as mixtures of MEA and MDEA, have been suggested for CO_2 removal (*6*).

Sterically hindered alkanolamines have also been proposed as commerically attractive solvents for removal of acid gases from gaseous streams (*2*). An example of the sterically hindered alkanolamines in 2-amino-2-methyl-1-propanol (AMP), which is the hindered form of MEA. Due to the bulkiness of the group attached to a tertiary carbon atom of AMP, the formation of carbamates is inhibited when AMP reacts with CO_2 (*2, 7*). Thus, the reaction of CO_2 with AMP is mainly the formation of the bicarbonate ion as follows:

$$CO_2 + RR'NH_2 + H_2O \rightleftharpoons HCO_3^- + RR'NH_3^+ \quad (1)$$

where $RR'NH_2$ denotes AMP. Therefore, the CO_2 loading of AMP can approach 1 mol of CO_2/mol of amine. The kinetics of the reaction of CO_2 with AMP have been shown to be first order with respect to both CO_2 and AMP, and the rate constant has the value of 1270 $m^3/(kmol\cdot s)$ at 40 °C (*7*). Compared to MDEA, AMP has the same high CO_2 loading capacity (about 1 mol of CO_2/mol of amine) but has a higher reaction rate constant for the reaction with CO_2. The reaction rate constant of CO_2 with MDEA is 3.5 $m^3/(kmol\cdot s)$ at 25 °C (*8*). Therefore, H_2O + MEA + AMP may be an attractive new solvent, in addition to H_2O + MEA + MDEA solutions, for the acid gas treating process.

Some solubility measurements of CO_2 and H_2S in blended alkanolamine aqueous solutions are available in the literature. The solubilities of CO_2 and H_2S in H_2O + MEA + MDEA have been studied (*9–12*). However, the solubility of acid gases in aqueous mixtures of MEA with AMP has not been reported in the literature. Therefore, it is the purpose of this research to measure the solubility of H_2S in aqueous MEA + AMP solutions.

Experimental Section

Alkanolamine aqueous solutions were prepared from distilled water. MEA is Riedel-de Haën reagent grade with a 99% (mol) purity, and AMP is Riedel-de Haën reagent grade with 98% (mol) purity.

The solubility of H_2S in aqueous MEA + AMP solutions was measured in a 1.0-L stainless steel vapor-recirculation equilibrium cell. For the low partial pressures of H_2S, nitrogen was introduced and mixed with H_2S, and the partial pressure of H_2S was determined by on-line chromatography. The solubility (loading capacity) of H_2S in amine solutions was determined by the titration method. When both the system pressure and the gas-phase concentrations, determined by the gas chromatograph, do not vary for 2 h, equilibrium is assumed to have been reached; it usually takes 4–5 h for the system to reach equilibrium. The partial pressure of H_2S was obtained from the pressure of the system and the gas-phase analysis. At equilibrium, the liquid sample was withdrawn from the cell into a vessel containing 1.0 M NaOH, thus converting free dissolved acid gas into the involatile ionic species. The H_2S content of the sample was determined by reacting the liquid with a solution of acidified 0.1 N I_2 (I^-).

---

* To whom correspondence should be addressed.

0021-9568/94/1739-0361$04.50/0

Table 1. Partial Pressure P of H_2S in $\omega_2 = 0.0$, $\omega_3 = 0.3$ H_2O (1) + MEA (2) + AMP (3)[a]

40 °C		60 °C		80 °C		100 °C	
P/kPa	α	P/kPa	α	P/kPa	α	P/kPa	α
1.22	0.394	2.01	0.300	2.86	0.243	4.88	0.173
5.49	0.641	2.36	0.347	5.07	0.302	7.26	0.203
9.42	0.703	7.92	0.539	13.2	0.411	10.4	0.268
10.3	0.735	12.3	0.601	25.7	0.508	20.1	0.338
17.2	0.776	13.4	0.609	28.0	0.521	20.8	0.337
41.8	0.886	26.2	0.693	45.1	0.602	42.6	0.469
81.0	0.900	43.2	0.779	60.0	0.664	58.2	0.484
105.2	0.938	60.1	0.808	73.2	0.682	64.8	0.543
133.5	0.936	68.1	0.810	106.2	0.743	96.0	0.599
		104.0	0.841	106.7	0.756	116.1	0.665
		116.2	0.830	149.2	0.812		

[a] H_2S loading α = mol of H_2S/total mol of amine, ω = mass fraction.

Table 2. Partial Pressure P of H_2S in $\omega_2 = 0.06$, $\omega_3 = 0.24$ H_2O (1) + MEA (2) + AMP (3)[a]

40 °C		60 °C		80 °C		100 °C	
P/kPa	α	P/kPa	α	P/kPa	α	P/kPa	α
1.32	0.407	1.08	0.201	1.52	0.156	1.03	0.132
1.96	0.434	1.98	0.292	1.69	0.180	1.30	0.159
2.66	0.476	3.22	0.331	3.49	0.250	2.65	0.164
8.00	0.624	7.08	0.425	6.08	0.266	9.44	0.247
8.37	0.626	16.0	0.550	9.87	0.333	23.9	0.333
15.0	0.714	24.3	0.636	11.5	0.351	52.2	0.430
38.4	0.796	25.1	0.613	19.0	0.422	83.7	0.531
38.5	0.815	55.5	0.749	26.7	0.478	94.4	0.571
100.2	0.853	86.7	0.799	30.4	0.504		
104.7	0.876	123.4	0.811	50.5	0.576		
161.6	0.901	158.7	0.858	69.7	0.614		
				79.2	0.670		
				119.1	0.731		

[a] H_2S loading α = mol of H_2S/total mol of amine, ω = mass fraction.

Table 3. Partial Pressure P of H_2S in $\omega_2 = 0.12$, $\omega_3 = 0.18$ H_2O (1) + MEA (2) + AMP (3)[a]

40 °C		60 °C		80 °C		100 °C	
P/kPa	α	P/kPa	α	P/kPa	α	P/kPa	α
0.53	0.259	1.83	0.261	1.65	0.175	0.83	0.133
1.81	0.425	7.51	0.452	8.11	0.302	4.17	0.163
6.07	0.618	20.3	0.610	12.7	0.368	23.3	0.320
16.2	0.762	39.2	0.720	26.6	0.488	74.1	0.492
60.8	0.880	78.2	0.807	63.3	0.624	99.1	0.573
108.4	0.917	114.5	0.850	110.7	0.732		
		159.0	0.885	135.0	0.761		

[a] H_2S loading α = mol of H_2S/total mol of amine, ω = mass fraction.

Table 4. Partial Pressure P of H_2S in $\omega_2 = 0.18$, $\omega_3 = 0.12$ H_2O (1) + MEA (2) + AMP (3)[a]

40 °C		60 °C		80 °C		100 °C	
P/kPa	α	P/kPa	α	P/kPa	α	P/kPa	α
0.67	0.296	1.15	0.213	1.63	0.170	1.24	0.109
1.60	0.401	1.86	0.263	2.62	0.182	8.00	0.219
3.51	0.553	7.00	0.448	5.01	0.249	32.1	0.395
8.44	0.664	15.5	0.579	16.3	0.419	93.0	0.573
21.6	0.796	43.2	0.740	45.8	0.588		
57.9	0.887	100.2	0.847	85.5	0.696		
119.6	0.933	146.4	0.887	138.4	0.773		
181.6	0.962						

[a] H_2S loading α = mol of H_2S/total mol of amine, ω = mass fraction.

Table 5. Partial Pressure P of H_2S in $\omega_2 = 0.24$, $\omega_3 = 0.06$ H_2O (1) + MEA (2) + AMP (3)[a]

40 °C		60 °C		80 °C		100 °C	
P/kPa	α	P/kPa	α	P/kPa	α	P/kPa	α
1.17	0.399	1.16	0.196	1.33	0.123	2.10	0.103
2.91	0.525	3.14	0.291	3.54	0.191	4.57	0.145
7.94	0.676	7.87	0.410	6.20	0.251	6.84	0.174
15.4	0.763	16.5	0.521	14.0	0.344	15.6	0.246
31.1	0.835	30.0	0.603	33.2	0.464	44.6	0.368
74.7	0.905	75.9	0.712	68.9	0.576	95.7	0.499
179.5	0.970	156.2	0.787	123.0	0.667		

[a] H_2S loading α = mol of H_2S/total mol of amine, ω = mass fraction.

The unreacted I_2 was back-titrated with 0.1 N $Na_2S_2O_3$ using starch as the indicator. The materials, vapor–liquid equilibrium apparatus, and methods of analysis are essentially the same as those used in our previous work on the solubility of acid gases in aqueous MEA + MDEA solutions (*10–12*).

Results and Discussion

For the measurements of the solubility of H_2S in water (1) + MEA (2) + AMP (3), the systems studied are $\omega_2 = 0$, $\omega_3 = 0.3$; $\omega_2 = 0.06$, $\omega_3 = 0.24$; $\omega_2 = 0.12$, $\omega_3 = 0.18$; $\omega_2 = 0.18$, $\omega_3 = 0.12$; and $\omega_2 = 0.24$, $\omega_3 = 0.06$ where ω is the mass fraction. The H_2S solubility data in aqueous MEA + AMP solutions at 40, 60, 80, and 100 °C are presented in Tables 1–5.

Owing to its simplicity, the model of Kent of Eisenberg (*13*) has often been used to represent the solubility of acid gases in aqueous alkanolamine systems (*14–18*). With the exception of the equilibrium constants for reactions involving amines, the literatures values for all the ionization constants and Henry's law constants were used in the model of Kent and Eisenberg. Jou et al. (*15*) pointed out that the equilibrium constants involving amines are essentially functions of temperature, acid gas loading, and amine concentration, rather than a function of temperature alone, as used in the model of Kent of Eisenberg. On the basis of the model of Kent and Eisenberg, Hu and Chakma (*17, 18*) proposed a modified expression for the equilibrium constants, governing the main amine reactions, as functions not only of temperature but also of acid gas partial pressure and amine concentration. In a similar manner, Li and Shen (*19*) modified the chemical equilibrium constants involving alkanolamines as functions of temperature, amine concentration, and carbon dioxide loading for the calculation of CO_2 solubilities in H_2O + MEA + MDEA. In this study, the method of Li and Shen (*19*) will be applied to correlate the solubility data of H_2S in aqueous MEA + AMP solutions. The constants in the model will be determined by fitting to the H_2S solubility data in H_2O + MEA + AMP.

Equations describing the equilibrium in the H_2S + H_2O + MEA + AMP mixture, with RNH_2 and $RR'NH_2$ representing MEA and AMP, respectively, are as follows:

$$RNH_3^+ \overset{K_{11}}{\rightleftharpoons} H^+ + RNH_2 \tag{2}$$

$$RR'NH_3^+ \overset{K_{12}}{\rightleftharpoons} H^+ + RR'NH_2 \tag{3}$$

$$H_2O \overset{K_3}{\rightleftharpoons} H^+ + OH^- \tag{4}$$

$$H_2S \overset{K_4}{\rightleftharpoons} H^+ + HS^- \tag{5}$$

$$HS^- \overset{K_5}{\rightleftharpoons} H^+ + S^{2-} \tag{6}$$

The expressions for the apparent equilibrium constants are

$$K_{11} = [H^+][RNH_2]/[RNH_3^+] \tag{7}$$

$$K_{12} = [H^+][RR'NH_2]/[RR'NH_3^+] \tag{8}$$

$$K_3 = [\mathrm{H^+}][\mathrm{OH^-}] \tag{9}$$

$$K_4 = [\mathrm{H^+}][\mathrm{HS^-}]/[\mathrm{H_2S}] \tag{10}$$

$$K_5 = [\mathrm{H^+}][\mathrm{S^{2-}}]/[\mathrm{HS^-}] \tag{11}$$

Henry's law relates the H_2S partial pressure to the concentration of the physically dissolved H_2S in the solvent according to

$$P_{\mathrm{H_2S}} = H_{\mathrm{H_2S}}[\mathrm{H_2S}] \tag{12}$$

The following mass balance equations for the reacting species hold:

$$m_1 = [\mathrm{RNH_2}] + [\mathrm{RNH_3^+}] \tag{13}$$

$$m_2 = [\mathrm{RR'NH_2}] + [\mathrm{RR'NH_3^+}] \tag{14}$$

$$(m_1 + m_2)\alpha = [\mathrm{H_2S}] + [\mathrm{HS^-}] + [\mathrm{S^{2-}}] \tag{15}$$

$$[\mathrm{RNH_3^+}] + [\mathrm{RR'NH_3^+}] + [\mathrm{H^+}] = [\mathrm{OH^-}] + [\mathrm{HS^-}] + [\mathrm{S^{2-}}] \tag{16}$$

where m_1 and m_2 are the concentrations of MEA and AMP, respectively, and α is the loading capacity of H_2S of the solvent. For an amine solution with known concentrations m_1, m_2, and α, eqs 7–16 (10 equations overall) can be solved for 10 unknowns: the partial pressure of H_2S and concentrations of the species $[\mathrm{RNH_2}]$, $[\mathrm{RR'NH_2}]$, $[\mathrm{RNH_3^+}]$, $[\mathrm{RR'NH_3^+}]$, $[\mathrm{H_2S}]$, $[\mathrm{HS^-}]$, $[\mathrm{S^{2-}}]$, $[\mathrm{H^+}]$, and $[\mathrm{OH^-}]$.

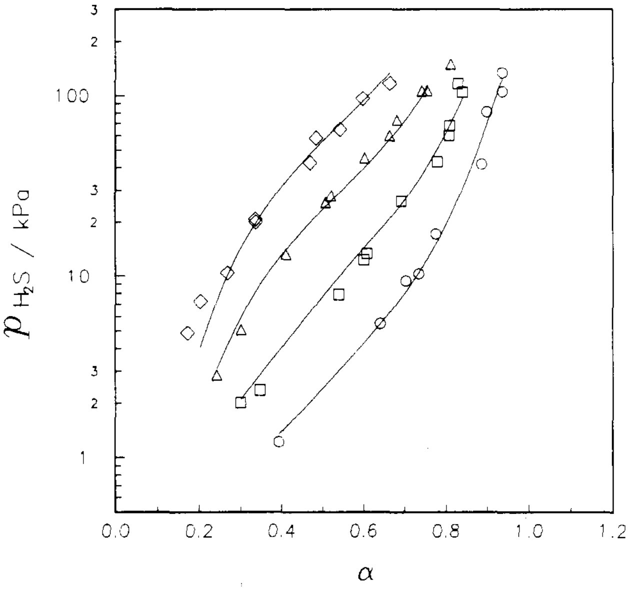

Figure 1. Partial pressure P of H_2S in $\omega_2 = 0$, $\omega_3 = 0.3$ H_2O (1) + MEA (2) + AMP (3) at various temperatures for various loadings α = mol of H_2S/total mol of amine. Lines are calculated by the method of Kent and Eisenberg: ○, 40 °C; □, 60 °C; △, 80 °C, ◇, 100 °C.

Table 6. Equilibrium Constants and Henry's Law Constant Used in This Study[a]

equilibrium constant	A	$B \times 10^{-4}$	$C \times 10^{-8}$	$D \times 10^{-11}$	$E \times 10^{-13}$
$K_3/[(\mathrm{kmol\ m^{-3}})^2]$	39.5554	−9.879	0.568827	−0.146451	0.136145
$K_4/(\mathrm{kmol\ m^{-3}})$	−304.689	38.721	−1.94755	0.438117	−0.37318
$K_5/(\mathrm{kmol\ m^{-3}})$	−657.965	91.631	−4.906296	1.153073	−1.01016
$H_{\mathrm{CO_2}}/[\mathrm{kPa\ (kmol\ m^{-3})^{-1}}]$	102.503	−13.6808	0.737744	−0.17472	0.152162

[a] $K_i = \exp[A + B/T + C/T^2 + D/T^3 + E/T^4]$ for T in kelvin.

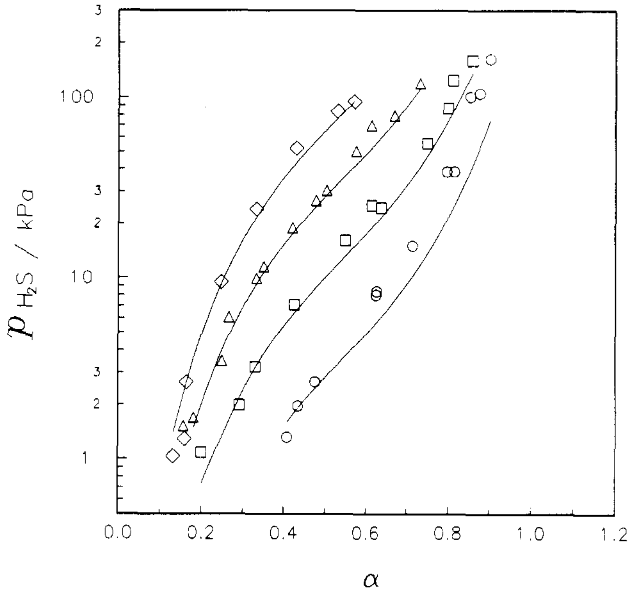

Figure 2. Partial pressure P of H_2S in $\omega_2 = 0.06$, $\omega_3 = 0.24$ H_2O (1) + MEA (2) + AMP (3) at various temperatures for various loadings α = mol of H_2S/total mol of amine. Lines are calculated by the method of Kent and Eisenberg: ○, 40 °C; □, 60 °C; △, 80 °C, ◇, 100 °C.

In this study, the equilibrium constants for eqs 4–6 and the Henry's law constant used in the calculation are the same as those used in the model of Kent and Eisenberg (*13*) and are presented in Table 6. The equilibrium constants which govern the main amine reactions, i.e., eqs 2 and 3, are assumed to be functions of temperature, amine concentration, and H_2S loading. Using the least-squares fit to the equilibrium partial pressure data of H_2S in H_2O + MEA + AMP solutions, the equilibrium constants were determined as follows:

$$K_{11} = \exp[-27.8641 + 6547.5/(T/\mathrm{K}) - 4.9306 \times 10^8/(T/\mathrm{K})^3 + 3.96352\alpha - 7.46277\alpha^2 + 5.06267\alpha^3 - 0.137338 \ln(m_1/(\mathrm{kmol\ m^{-3}}))] \tag{17}$$

$$K_{12} = \exp[-14.2832 - 1496.15/(T/\mathrm{K}) - 1.70523 \times 10^8/(T/\mathrm{K})^3 + 12.9880\alpha - 26.4005\alpha^2 + 17.0492\alpha^3 + 0.149982 \ln(m_2/(\mathrm{kmol\ m^{-3}}))] \tag{18}$$

where α is the moles of H_2S per total moles of amine.

Figures 1–5 show comparisons of the results between calculated and experimental values for five blended amine aqueous systems. The calculated partial pressures of H_2S over H_2O + MEA + AMP are in good agreement with experimental data for temperatures ranging from 40 to 100 °C and for systems of various concentrations, as shown in Figures 1–5. The model reasonably reproduces the equilibrium partial pressure of H_2S over H_2O + MEA + AMP systems. To test the applicability of the model, calculations of the solubility for data not included in the database have also been performed. Figure 6 gives a comparison between the results of calculated and experimental H_2S solubilities in 2.0 kmol m^{-3} H_2O + AMP solution at 40 and 100 °C (data of Roberts and Mather (*20*)). In Figure 6, the model predicts the solubility of H_2S in 2.0 kmol m^{-3} H_2O + AMP reasonably well at both 40 and 100 °C.

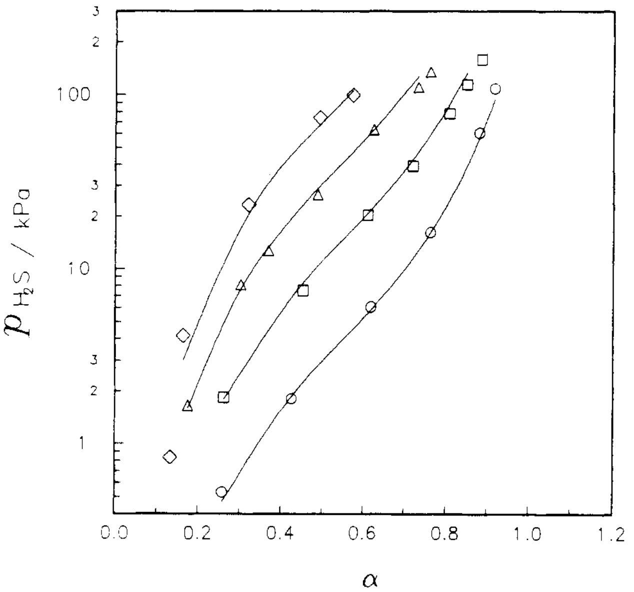

Figure 3. Partial pressure P of H_2S in $\omega_2 = 0.12$, $\omega_3 = 0.18$ H_2O (1) + MEA (2) + AMP (3) at various temperatures for various loadings α = mol of H_2S/total mol of amine. Lines are calculated by the method of Kent and Eisenberg: ○, 40 °C; □, 60 °C; △, 80 °C, ◇, 100 °C.

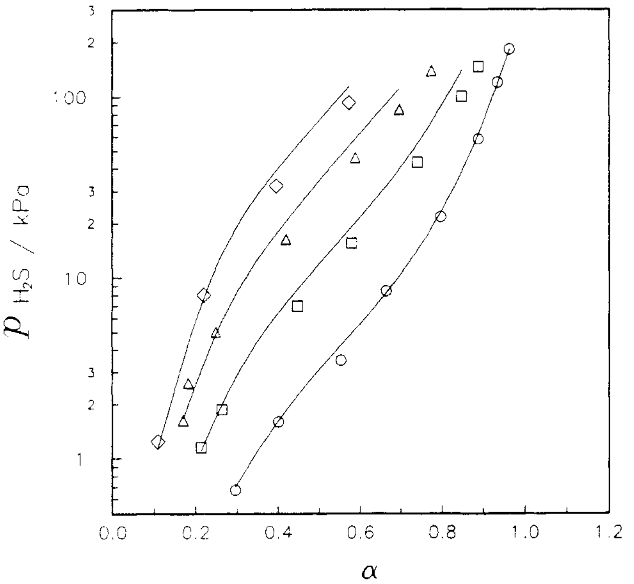

Figure 4. Partial pressure P of H_2S in $\omega_2 = 0.18$, $\omega_3 = 0.12$ H_2O (1) + MEA (2) + AMP (3) at various temperatures for various loadings α = mol of H_2S/total mol of amine. Lines are calculated by the method of Kent and Eisenberg: ○, 40 °C; □, 60 °C; △, 80 °C; ◇, 100 °C.

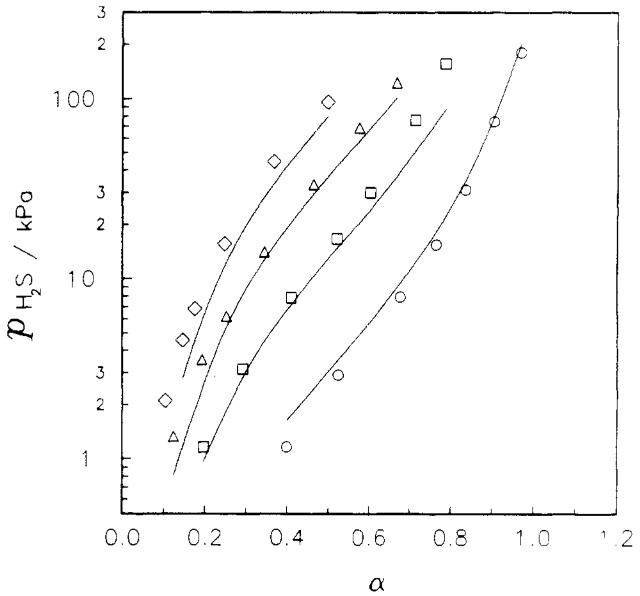

Figure 5. Partial pressure P of H_2S in $\omega_2 = 0.24$, $\omega_3 = 0.06$ H_2O (1) + MEA (2) + AMP (3) at various temperatures for various loadings α = mol of H_2S/total mol of amine. Lines are calculated by the method of Kent and Eisenberg: ○, 40 °C; □, 60 °C; △, 80 °C; ◇, 100 °C.

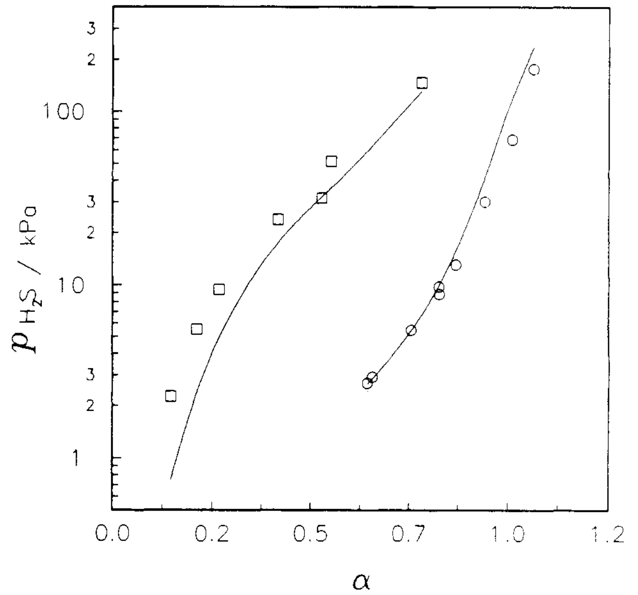

Figure 6. Comparison of calculated and experimental results for the solubility of H_2S in 2.0 kmol m^{-3} H_2O + AMP solution at 40 and 100 °C. Data were reported by Roberts and Mather (*20*). Lines are calculated by the method of Kent and Eisenberg: ○, 40 °C; □, 100 °C.

Conclusion

The solubilities of H_2S in H_2O + MEA + AMP have been reported from 40 to 100 °C and at H_2S partial pressures up to 180 kPa. The systems studied are $\omega_2 = 0$, $\omega_3 = 0.3$; $\omega_2 = 0.06$, $\omega_3 = 0.24$; $\omega_2 = 0.12$, $\omega_3 = 0.18$; $\omega_2 = 0.18$, $\omega_3 = 0.12$; and $\omega_2 = 0.24$, $\omega_3 = 0.06$ where ω is the mass fraction. The model of Kent and Eisenberg for calculating the solubilities of acid gases in alkanolamine solutions has been extended to represent the solubility of H_2S in H_2O + MEA + AMP. The equilibrium constants of chemical reactions involving amines are expressed as functions of temperature, amine concentration, and H_2S loading. The literature values for ionization constants and Henry's law constants are adopted directly in the calculations as in the Kent and Eisenberg method. The model reasonably reproduces the equilibrium partial pressure of H_2S over H_2O + MEA + AMP over a temperature range from 40 to 100 °C at various concentrations. The model has also been tested for systems not included in the database. Satisfactory results were obtained for the calculation of the solubility of H_2S in H_2O + AMP.

Literature Cited

(1) Isaacs, E. E.; Otto, F. D.; Mather, A. E. *J. Chem. Eng. Data* **1980**, *25*, 118.
(2) Sartori, G.; Savage, D. W. *Ind. Eng. Chem. Fundam.* **1983**, *22*, 239.
(3) Srinivasan, V.; Aiken, R. C. *Fuel Process. Technol.* **1988**, *19*, 141.
(4) Yu, W. C.; Astarita, G. *Chem. Eng. Sci.* **1987**, *42*, 419.
(5) Riesenfeld, F. C.; Brocoff, J. C. *Technol. Oil Gas J.* **1986**, *84*, 61.
(6) Chakravarty, T.; Phukan, U. K.; Weiland, R. H. *Chem. Eng. Prog.* **1985**, *81*, 32.

(7) Yih, S. M.; Shen, K. P. *Ind. Eng. Chem. Res.* **1988**, *27*, 2237.
(8) Crooks, J. E.; Donnellan, J. P. *J. Org. Chem.* **1990**, *55*, 1372.
(9) Austgen, D. M.; Rochelle, G. T.; Chen, C. C. *Ind. Eng. Chem. Res.* **1991**, *30*, 543.
(10) Shen, K. P.; Li, M. H. *J. Chem. Eng. Data* **1992**, *37*, 96.
(11) Li, M. H.; Shen, K. P. *J. Chem. Eng. Data* **1992**, *37*, 288.
(12) Li, M. H.; Shen, K. P. *J. Chem. Eng. Data* **1993**, *38*, 105.
(13) Kent, R. L.; Eisenberg, B. *Hydrocarbon Process.* **1976**, *55* (2), 87.
(14) Lee, J. I.; Otto, F. D.; Mather, A. E. *Can. J. Chem. Eng.* **1976**, *54*, 214.
(15) Jou, F. Y.; Mather, A. E.; Otto, F. D. *Ind. Eng. Chem. Process Des. Dev.* **1982**, *21*, 539.
(16) Pitsinigos, V. D.; Lygeros, A. I. *Hydrocarbon Process.* **1989**, *68* (4), 43.
(17) Hu, W.; Chakma, A. *Chem. Eng. Commun.* **1990**, *94*, 53.
(18) Hu, W.; Chakma, A. *Can. J. Chem. Eng.* **1990**, *68*, 523.
(19) Li, M. H.; Shen, K. P. *Fluid Phase Equilib.* **1993**, *85*, 129.
(20) Roberts, B. E.; Mather, A. E. *Chem. Eng. Commun.* **1988**, *64*, 105.

Received for review September 7, 1993. Accepted January 19, 1994.[•] This research was supported by a grant, NSC 82-0402-E033-044, of the National Science Council of the Republic of China.

[•] Abstract published in *Advance ACS Abstracts*, March 1, 1994.