

(19). The Muhlbauer and Monaghan data show higher hydrogen sulfide and carbon dioxide partial pressures, especially at low acid gas concentrations. Other mixed acid gas-amine system data are available, but, again, direct comparisons cannot be made because of differing amine concentrations and temperatures.

### Conclusions

The data obtained in this study extend the range of definition of H<sub>2</sub>S and CO<sub>2</sub> solubility in MEA and DEA solutions. The data are accurate and in good agreement with data from other sources.

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## Hydrocarbon Gas Solubility in Sweetening Solutions: Methane and Ethane in Aqueous Monoethanolamine and Diethanolamine

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**The solubility of methane and ethane in monoethanolamine and diethanolamine solutions is measured. The experimental procedures used to collect the data are presented along with selected data and graphs showing effects of temperature, amine concentration, and acid gas content. The solubilities of these hydrocarbons in the amine solutions are about the same as their solubility in an equal weight or volume of water. However, the type and concentration of amine, temperature, and presence of hydrogen sulfide and carbon dioxide affect the methane and ethane solubility.**

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Aqueous solutions of monoethanolamine (MEA) or diethanolamine (DEA) are used as absorbants in natural gas sweetening systems to selectively remove hydrogen sulfide and carbon dioxide from the plant feed gas. In the process some hydrocarbon gas also is absorbed in the amine solution. Knowledge of the amounts of hydrocarbon gas in the amine stream is necessary for design of the sweetening system and associated Claus sulfur recovery unit.

**Previous work.** Published data on hydrocarbon solubility in amine solutions are limited to two curves plotted in Kohl and

Riesenfeld (5) for methane solubility at 80° and 160°F in 25.3 wt % MEA having 0.47 mole of acid gas per mole MEA. The data of this study are in general agreement with the Kohl data.

Published data for hydrocarbon gas solubility in water are more plentiful. These data are given in Culberson and McKetta (2), Davis and McKetta (3), and Kobayashi and Katz (4). A review and presentation of these data are given in the API Technical Data Book (7). Solubilities for hydrocarbon mixtures are given in a publication by Amirjafari and Campbell (1).

### Experimental

Amine solutions were prepared from distilled water and commercial grade MEA and DEA. Both amines checked better than 99 % pure by acid titration. Hydrocarbon gases used were 99 mol % minimum. The gas compositions were checked by mass spectrometer analyses.

The equilibrium cell used for the gas solubility determinations is described in a previous publication by Lawson and Garst (6). The cell was evacuated, and 500-600 g of solution was drawn into the cell. The cell was pressured with hydrocarbon gas and rocked at test temperature for at least 16 hr.

Liquid-phase samples were taken in a 10-ml sample container and weighed. Sample container tare weight established the weight of liquid sample. The volume of gas dissolved was measured volumetrically by water displacement from a sec-

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**Table I. Solubility of Methane in MEA and DEA Solutions**

Amine solution		Vapor-phase partial press of methane, psi	Liquid-phase concn of methane, lb-mol; methane/10 <sup>5</sup> lb soln
Wt % amine	Amine type		
100°F			
5	DEA	512	3.48
		963	6.46
25	DEA	510	3.20
		968	6.00
40	DEA	526	3.24
		933	5.52
15	MEA	954	6.55
40	MEA	500	3.48
		954	6.26
150°F			
5	DEA	516	2.77
		978	5.20
25	DEA	511	2.89
		982	5.40
40	DEA	518	3.06
		937	5.38
15	MEA	498	3.07
40	MEA	993	5.80
		500	3.33
40	MEA	954	6.20
		516	2.58
25	DEA	920	5.02
		530	3.04
40	DEA	963	5.74
		490	9.16
40	MEA	974	16.60
		250°F	
25	DEA	501	2.97
40	DEA	920	5.82
		498	3.56
40	MEA	908	6.30
		508	4.19
40	MEA	950	7.85

ond sample container. Acid gases were scrubbed from the evolving gas with caustic solution placed in the second sample container. The inherent limitations imposed by equipment and sampling procedures probably introduced error in equilibrium pressure observations of 2-3%, which is the magnitude of cell pressure drops observed during sampling.

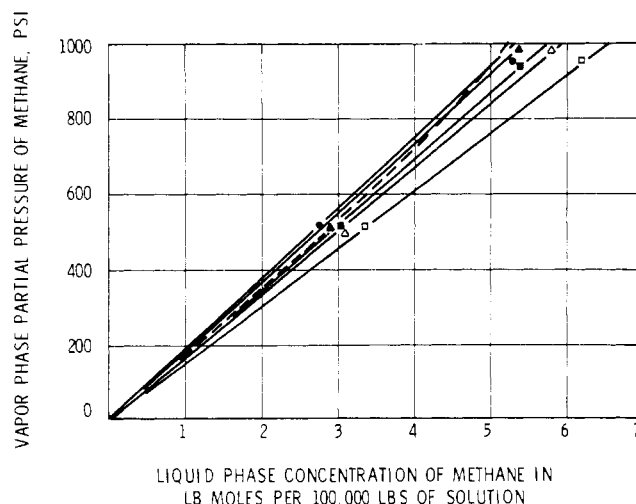
**Results and Discussion**

Tables I and II list selected solubility data for methane and ethane in different concentrations of MEA and DEA solutions. The solubilities are reported as moles of hydrocarbon per unit weight of amine solution rather than the more usual units of mole fraction. Using the chosen units allows for a more direct comparison to data for hydrocarbon solubility in water. Also, since amine solution weights, rather than volumes, were measured, presenting the data on the basis of unit weight of amine solution does not require measuring or estimating solution densities.

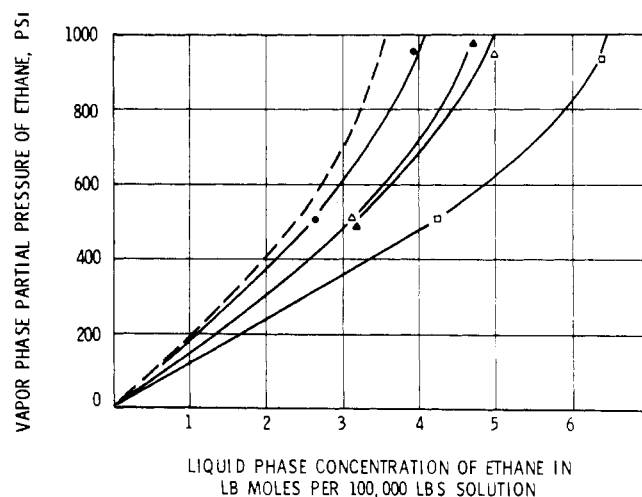
Figure 1 shows the comparison of methane solubility in different amine solutions and water at 150°F. Figure 2 shows the solubilities for ethane at 150°F. The hydrocarbons are more soluble in MEA solutions than in equal weights of corresponding DEA solutions. In general, methane and ethane solubility increases with increasing amine concentration. However, the opposite trend of methane solubility decreasing with increasing amine concentration is shown by the data at 100°F. Methane is somewhat less soluble in dilute solutions of MEA and DEA and somewhat more soluble in concentrated solutions of MEA and DEA than equal weights of water. Eth-

**Table II. Solubility of Ethane in MEA and DEA Solutions**

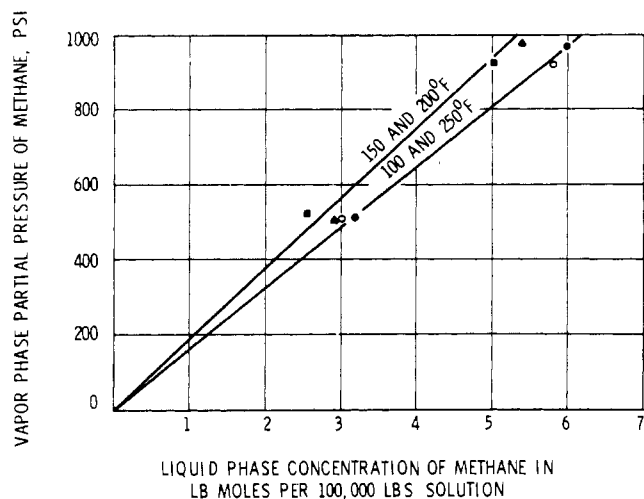
Amine solution		Vapor-phase partial press of ethane, psi	Liquid-phase concn of ethane, lb-mol; ethane/10 <sup>5</sup> lb of soln
Wt % amine	Amine type		
100°F			
5	DEA	501	2.65
		957	3.94
25	DEA	480	3.83
		868	5.02
15	MEA	491	3.90
		868	5.04
150°F			
5	DEA	501	2.65
25	DEA	957	3.94
		498	3.14
15	MEA	972	4.78
		501	3.11
40	MEA	955	4.98
		503	4.25
40	MEA	932	6.38



**Figure 1. Methane solubility in MEA and DEA solutions at 150°F**  
 ● 5 wt % DEA, ▲ 25 wt % DEA, ■ 40 wt % DEA, △ 15 wt % MEA, □ 40 wt % MEA, -- water



**Figure 2. Ethane solubility in MEA and DEA solutions at 150°F**  
 ● 5 wt % DEA, ▲ 25 wt % DEA, △ 15 wt % MEA, □ 40 wt % MEA, -- water



**Figure 3.** Effect of temperature on methane solubility in 25 wt % DEA from 100° to 250°F  
 ● 100°F, ▲ 150°F, ■ 200°F, ○ 250°F

**Table III.** Solubility of Methane in 25 Wt % DEA Containing H<sub>2</sub>S or CO<sub>2</sub>

Temp, °F	Vapor-phase partial press of methane, psi	Liquid-phase concentration		Lb-mol of methane/10 <sup>5</sup> lb of soln
		Mol/mol DEA		
		H <sub>2</sub> S	CO <sub>2</sub>	
100	973	0.20	...	5.37
	862	1.10	...	4.52
150	456	0.19	...	2.01
	495	0.25	...	2.30
100	951	0.20	...	4.36
	790	0.94	...	7.15
	500	...	0.27	2.74
150	956	...	0.27	4.90
	489	...	0.25	2.34
	970	...	0.25	4.10

ane exhibits increasing solubility in more concentrated amine to a greater extent than does methane.

Figure 3 shows that methane solubility exhibits a minimum with temperature between 100–200°F in 25 wt % DEA solutions just as it does with water.

Table III gives the solubility of methane in 25 wt % DEA containing hydrogen sulfide or carbon dioxide. The hydrocar-

bon solubility data obtained with solutions containing hydrogen sulfide and carbon dioxide are not as accurate as the other measurements, and quantitative explanations of the effects of the acid gases are difficult to make with these data. However, the presence of about 0.25 mole of hydrogen sulfide or carbon dioxide per mole of amine in the solution lowered the methane solubility 10–20% from the acid gas-free solubility. It would be expected that increasing acid gas content would decrease hydrocarbon solubility but not necessarily in direct proportion to acid gas concentration.

Additional data were collected using mixtures of methane and ethane. The presence of ethane had little effect on methane solubility in the MEA or DEA solutions used in this study. The total solubility of a gas containing 50 vol % methane and 50 vol % ethane was somewhat greater than if the gas were all methane, but not as much as calculations from individual methane and ethane system data would indicate, assuming no interaction between the two hydrocarbon species. Also, the data indicate that ethane solubility is 10–15% less in the presence of methane, which is consistent with the above observation of total solubility.

The interaction effect of increasing solubilities of methane and ethane in amine solutions is not nearly as large as that for mixtures of methane and ethane in water reported by Amirijafari. In water a 50–50% mixture of methane and ethane has twice the solubility at 160°F that would be predicted by data from separate systems assuming no interaction. The increased mixture solubility would then be expected to occur to a greater extent as amine solution strength is lowered. However, data on 5 wt % DEA did not show a marked increase in total hydrocarbon solubility with methane–ethane mixtures, indicating that the mixture behavior will not be exhibited by amine solutions used in gas sweetening units.

### Conclusions

Data were obtained on methane and ethane solubility in MEA and DEA sweetening solutions. The hydrocarbon gas solubility in the amine solutions is about the same as it is for water. The presence of H<sub>2</sub>S or CO<sub>2</sub> lowers methane solubility in the sweetening solutions.

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