Acknowledgment

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Appendix: Fugacity Coefficient of the Three-Parameter Pseudocubic Equation of State

The fugacity coefficient of component k in a mixture, $\hat{\phi}_k$, is given as follows:

$$RT \ln \hat{\phi}_k = I_1 + I_2 + I_3 + I_4 + I_5 + I_6 + I_7 \quad \text{(A1)}$$

where

$$I_1 = RT\left[\left(\frac{\theta}{\theta+\lambda}\right)\ln\left(\frac{V}{V-\theta b}\right) + \left(\frac{\lambda}{\theta+\lambda}\right)\ln\left(\frac{V}{V+\lambda b}\right)\right] \quad \text{(A2)}$$

$$I_2 = -RT[3(2\textstyle\sum X_l b_{lk} - b) - 4(\theta_k b + 2\theta(\sum X_l b_{lk} - b))] \times \left[\left(\frac{\theta}{\theta+\lambda}\right)^2 \frac{1}{V-\theta b} + \left(\frac{\lambda}{\theta+\lambda}\right)^2 \frac{1}{V+\lambda b} + \frac{2\theta\lambda}{(\theta+\lambda)^3 b}\ln\left(\frac{V+\lambda b}{V-\theta b}\right)\right] \quad \text{(A3)}$$

$$I_3 = -3RT[\theta b(1-\theta_k) + \theta_k b(1-\theta) + 4\theta(1-\theta) \times (\textstyle\sum X_l b_{lk} - b)]\left[\frac{\lambda}{(\theta+\lambda)^2}\frac{1}{V+\lambda b} - \frac{\theta}{(\theta+\lambda)^2}\frac{1}{V-\theta b} + \frac{\theta-\lambda}{(\theta+\lambda)^3 b}\ln\left(\frac{V+\lambda b}{V-\theta b}\right)\right] \quad \text{(A4)}$$

$$I_4 = -2(\textstyle\sum X_l a_{lk})\left[\left(\frac{\epsilon}{\epsilon+\sigma}\right)^2\frac{1}{V-\epsilon b} + \left(\frac{\epsilon}{\epsilon+\sigma}\right)^2\frac{1}{V+\sigma b} + \frac{2\epsilon\sigma}{(\epsilon+\sigma)^3 b}\ln\left(\frac{V+\sigma b}{V-\epsilon b}\right)\right] \quad \text{(A5)}$$

$$I_5 = 8a[(1-\theta_k)b + 2(1-\theta)(\textstyle\sum X_l b_{lk} - b)]\left[\left(\frac{\epsilon^3}{2(\epsilon+\sigma)^3}\right)\frac{1}{(V-\epsilon b)^2} + \left(\frac{3\epsilon^2\sigma}{(\epsilon+\sigma)^4 b}\right)\frac{1}{(V-\epsilon b)} + \left(\frac{\sigma^3}{2(\epsilon+\sigma)^3}\right)\frac{1}{(V+\sigma b)^2} - \left(\frac{3\epsilon\sigma^2}{(\epsilon+\sigma)^4 b}\right)\frac{1}{(V+\sigma b)} - \frac{3\epsilon\sigma(\epsilon-\sigma)}{(\theta+\lambda)^5 b^2}\ln\left(\frac{V+\sigma b}{V-\epsilon b}\right)\right] \quad \text{(A6)}$$

$$I_6 = -6a[\theta b(1-\theta_k) + \theta_k b(1-\theta) + 4\theta(1-\theta) \times (\textstyle\sum X_l b_{lk} - b)]\left[\left(\frac{\epsilon^2}{2(\epsilon+\sigma)^3}\right)\frac{1}{(V-\epsilon b)^2} + \left(\frac{\epsilon(2\sigma-\epsilon)}{(\epsilon+\sigma)^4 b}\right)\frac{1}{V-\epsilon b} + \left(\frac{\sigma(2\epsilon-\sigma)}{(\epsilon+\sigma)^4 b}\right)\frac{1}{V+\sigma b} - \left(\frac{\sigma^2}{2(\epsilon+\sigma)^3}\right)\frac{1}{(V+\sigma b)^2} + \left(\frac{\epsilon^2 - 4\epsilon\sigma + \sigma^2}{(\epsilon+\sigma)^5 b^2}\right)\ln\left(\frac{V+\sigma b}{V-\epsilon b}\right)\right] \quad \text{(A7)}$$

$$I_7 = -RT \ln Z \quad \text{(A8)}$$

in which

$$\lambda = 3(1-\theta) \quad \text{(A9)}$$

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Solubility of Hydrogen in Aqueous Ethanolamine Solutions at Temperatures between 323 and 423 K

Georg Kling and Gerd Maurer*

Lehrstuhl für Technische Thermodynamik, Universität Kaiserslautern, D-6750 Kaiserslautern, Federal Republic of Germany

The solubility of hydrogen in aqueous ethanolamine solutions was measured at temperatures between 323 and 423 K at pressures up to 20 MPa. Henry's coefficients derived from the experimental results are successfully predicted by the scaled particle theory.

Introduction

The solubility of hydrogen in aqueous amine solutions is of industrial, as well as of scientific, interest. For example, in the synthesis of ethylenediamine from ethanolamine and ammonia, the degree of conversion is controlled by the hydrogen content of the liquid phase. In the thermodynamics of phase equilibria, the prediction of gas solubility in mixed solvents is still an un-

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* To whom correspondence should be addressed.

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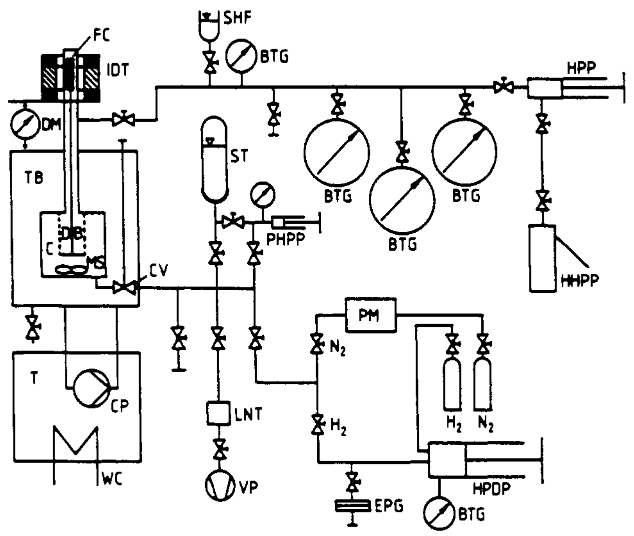

Figure 1. Scheme of the gas solubility apparatus: DB, diaphragm bellows; C, cell; MS, magnetic stirrer; TB, thermostated bath; DM, distance meter; IDT, inductive transducer; FC, ferrite core; SHF, storage of hydraulic fluid; BTG, Bourdon tube gauge; HPP, high-pressure pump; HHPP, high-precision displacement pump; PHPP, precision high-pressure pump; ST, storage tank for liquid; CV, cell valve; T, thermostat; CP, circulation pump; WC, water cooler; LNT, liquid nitrogen trap; VP, vacuum pump; PM, piston gauge; EPG, electronic pressure gauge; HPDP, high precision gas displacement pump.

Table I. Experimental Results for the Solubility of Hydrogen in Aqueous Ethanolamine Solutions with 75, 50, and 25 mol % Water

$\tilde{x}_{H_2O}$	T/K (p^s/MPa)	$10^3 x_{H_2}$	p/MPa
0.75	323.15 (0.008 42)	0.799	4.47
		0.860	4.79
		1.461	8.12
		1.967	10.94
		2.171	12.12
	373.15 (0.072 08)	0.853	3.57
		1.318	5.49
		2.237	9.28
	423.15 (0.3502)	1.048	3.20
		1.342	4.04
		2.225	6.37
		3.476	9.90
0.50	323.15 (0.004 42)	0.618	2.49
		0.782	3.14
		1.019	4.11
		1.525	6.05
		1.538	6.08
		3.468	13.54
	348.15 (0.015 34)	0.712	2.37
		1.037	3.41
		2.160	7.00
		2.855	9.25
		3.748	12.20
	373.15 (0.043 85)	0.743	1.92
		1.640	4.36
		2.443	6.35
		3.091	7.85
		3.757	9.76
		4.238	10.77
	398.15 (0.1077)	2.954	6.30
		3.360	7.26
		4.382	9.36
	423.15 (0.2341)	3.078	5.58
		4.002	7.37
		5.316	9.69
0.25	323.15 (0.002 15)	1.752	4.56
		3.109	8.07
		3.568	9.32
		7.335	19.21
	373.15 (0.022 38)	2.557	4.44
		3.813	6.63
		5.223	9.16
		7.117	12.28
	423.15 (0.1405)	3.053	3.73
		3.638	4.45
		7.326	8.80
		7.892	9.41

solved problem. Testing and improving thermodynamic models for gas solubility in mixed solvents require more experimental data. This holds especially for the solubility of quantum gases like hydrogen in liquid mixtures of polar compounds like water and ethanolamine. Therefore the solubility of hydrogen in three water/ethanolamine mixtures was measured and compared with several predictive methods. With the scaled particle theory, a very good prediction is possible.

Experimental Section

The equipment used for the experimental part of this work has been described in detail in previous publications (*1*, *2*). A simplified scheme of the apparatus is shown in Figure 1. A thermostated high-pressure cell of variable volume is charged with known amounts of mixed solvent and gas. The volume of the cell is varied by adding or removing oil from a diaphragm bellows mounted inside the cell. Thus an isothermal pressure–volume diagram of the ternary mixture is taken. In crossing from the one-phase liquid region into the two-phase vapor–liquid region the slope of the p–V isotherm changes drastically. The change of slope occurs at the bubble point pressure of the ternary mixture. Usually about 150 g of solvent together with about 0.006–0.07 g of hydrogen are filled into the cell. The uncertainty of the amounts of solvent and solute is smaller than ±0.26 and ±1.1%, respectively. The temperature is measured by means of a calibrated platinum resistance thermometer within ±0.02 K. The pressure is determined by three high-precision Bourdon tube gauges with ranges of 1, 10, and 25 MPa and an accuracy of ±0.1% of full scale.

Materials

High-grade hydrogen (99.999 mol %) was purchased from Messer Griesheim (Ludwigshafen, FRG). Deionized water was degassed by vacuum distillation. Ethanolamine was kindly provided by BASF AG (Ludwigshafen) with a purity of better than 99.45 mol %. It was degassed by crystallization.

Results

The experimental results for the solubility of hydrogen in three aqueous ethanolamine solutions at temperatures between 323.15 and 423.15 K and pressures up to 20 MPa are given in Table I. The given numbers are averaged values of at least two direct experimental results which agreed within the uncertainties given before with each other. As no data on the solubility of hydrogen in aqueous ethanolamine solutions has been published before, the quality of the new data cannot be checked against literature data. From earlier results on the solubility of hydrogen in pure water determined with the same apparatus (*1*, *2*), it is assumed that the experimental results for the boiling pressure have an accuracy of better than ±1.5%. Some of the experimental data are plotted in Figure 2, showing that in a pressure versus mole fraction plot the isotherms are straight lines.

Henry's Constant

Henry's constant for the solubility of hydrogen in a solvent mixture is defined by

$$H_{H_2,\mathrm{mix}} = \lim_{p \to p^s_{\mathrm{mix}}} \left[\frac{p y_{H_2} \varphi''_{H_2}}{x_{H_2}} \right]_{T=\mathrm{const}} \tag{1}$$

The determination of Henry's constant from gas solubility data therefore requires knowledge of the vapor pressure of the mixed solvent and the fugacity of hydrogen in the vapor phase.

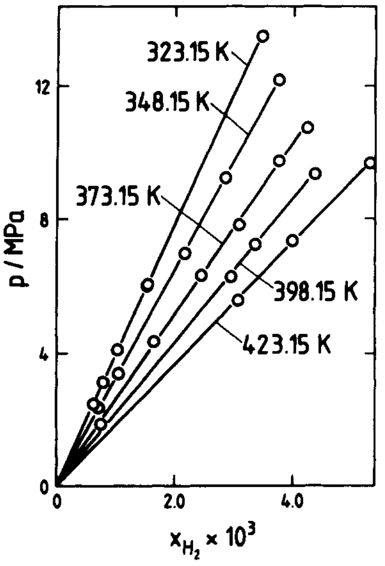

Figure 2. Experimental results for the solubility of hydrogen in an equimolar aqueous ethanolamine solution.

Table II. Experimental Results for the Vapor Pressure of Aqueous Ethanolamine Solutions with 75, 50, and 25 mol % Water

$\tilde{x}_{H_2O}$	T/K	p^s/kPa	$\tilde{x}_{H_2O}$	T/K	p^s/kPa
0.75	310.66	4.32	0.50	373.16	43.94
	323.18	8.23		385.66	69.88
	335.66	15.76	0.25	323.15	2.03
	348.17	26.85		335.66	4.17
	360.65	44.85		348.15	7.63
	373.16	72.12		360.65	13.51
0.50	323.15	4.52		373.16	22.63
	335.66	8.40		385.66	36.93
	348.15	15.33		398.15	58.14
	360.65	26.45		410.64	92.19

The only literature source for the vapor pressure of aqueous ethanolamine solutions is a data sheet by Dow Chemical Co. (*3*). But, as only a few values were reported and no information on the origin of the data is available, the vapor pressure of the mixed solvent was also measured in this work. The experimental procedure followed a method described by Nath and Bender (*4*). The experimental results are given in Table II. It is assumed that the uncertainty of the new data is about ±0.08 kPa or about ±1%, whichever is larger. The results by Dow are up to 5 kPa and up to 20% larger than the results of the present work. Vapor pressures interpolated to the temperatures of the gas solubility experiments are also given in Table I. As can be seen, the vapor pressure of the mixed solvent rarely exceeds 2% of the measured bubble point pressure of the ternary mixture of water, ethanolamine, and hydrogen. Therefore the fugacity of hydrogen in the vapor phase was approximated by the fugacity of pure hydrogen at $p^* = p - p^s_{mix}$, which again was calculated from the Bender equation of state (*5*). It is assumed that the uncertainty caused by these approximations is smaller than about 1%. As a typical example, Figure 3 shows the extrapolation of the experimental results to determine Henry's constant for the solubility of hydrogen in an equimolar mixture of ethanolamine and water. The Henry's constants determined in this work are given in Table III. As a result of additional uncertainties in calculating the fugacity of hydrogen and the extrapolation procedure, the uncertainty of the numbers given in Table III is estimated to be about ±2%.

Prediction of Henry's Constants and Comparison with Experimental Data

The Henry's constant for the solubility of a gas 1 in a mixed solvent of components j is related to the Henry's constant of the same gas 1 in the pure components j by the following equation:

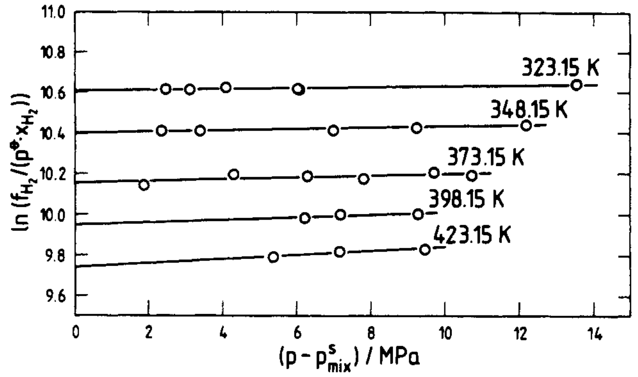

Figure 3. Plot of experimental results to determine Henry's constants for the solubility of hydrogen in an equimolar aqueous ethanolamine solution.

Table III. Henry's Constants $H_{H_2,mix}$/MPa for the Solubility of Hydrogen in Aqueous Ethanolamine Solutions with 75, 50, and 25 mol % Water

	$\tilde{x}_{H_2O}$		
T/K	0.75	0.50	0.25
323.15	5566	4046	2595
348.15		3295	
373.15	4102	2571	1736
398.15		2089	
423.15	2720	1697	1187

$$\ln [H_{1,mix}] = \sum_{j=2}^{m} \tilde{x}_j \ln \left[H_{1,j} \frac{\gamma^\infty_{1,mix}}{\gamma^\infty_{1,j}} \right] \qquad (2)$$

$\gamma^\infty_{1,mix}$ and $\gamma^\infty_{1,j}$ stand for the limiting activity coefficients (normalized according to Raoult's law) of solute 1 in the mixed solvent and the pure components j, respectively. A similar relationship can be derived on the basis of volume fractions, ϕ_j. As information on the activity coefficients is rarely available, the ratio of activity coefficients is usually approximated to unity:

$$\ln [H_{1,mix}] = \sum_{j=2}^{m} \tilde{x}_j \ln [H_{1,j}] \qquad (3)$$

$$\ln [H_{1,mix}] = \sum_{j=2}^{m} \tilde{\phi}_j \ln [H_{1,j}] \qquad (4)$$

Both simplified procedures predict the Henry's constant of hydrogen in the water/ethanolamine mixture with large uncertainties. Typical examples are shown in Figure 4, where predictions—based on the Henry's constant of hydrogen in water and ethanolamine as published recently (*1*, *2*)—are compared with the experimental results of the present work at 373.15 K. Equation 3 overestimates the Henry's constant of hydrogen in an equimolar aqueous ethanolamine solution up to 13%, whereas eq 4 yields a prediction which is too small by about 27%. Much better predictions result from the scaled particle theory of Reiss et al. (*6*). This model starts from the statistical thermodynamics expression for the Henry's coefficient:

$$\ln [H_{1,mix}] = \ln \left[\frac{RT}{V^s_{mix}} \right] + \frac{\Delta\mu'_{1,pot.}}{RT} \qquad (5)$$

The contribution of intermolecular forces to the chemical po-

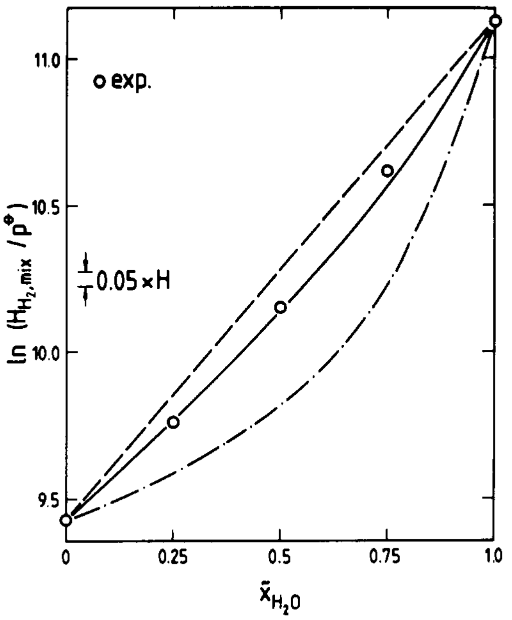

Figure 4. Henry's constants of hydrogen in aqueous ethanolamine solutions at 373.15 K: —, scaled particle theory; --, eq 3 (mole fraction averaging); –·–, eq 4 (volume fraction averaging).

tential of component *i* is a combination of effects caused by repulsive and attractive forces:

$$\Delta\mu_{1,\text{pot.}} = \Delta\mu'_v + \Delta\mu'_c \tag{6}$$

Here the repulsive term of Lebowitz et al. (*7*)

$$\frac{\Delta\mu'_v}{RT} = -[\ln(1-\eta_3)] + \left[\frac{3\eta_2}{1-\eta_3}\right]d_1 + \left[\left(\frac{3\eta_1}{1-\eta_3}\right) + \frac{9\eta_2^2}{2(1-\eta_3)^2}\right]d_1^2 + \frac{\pi}{6}\frac{p^s_{\text{mix}}}{kT}d_1^3 \tag{7}$$

with

$$\eta_k = \frac{\pi}{6}\sum_{j=2}^{m}(\rho^*_j\, d_j^k); \quad k = 1, 2, 3 \tag{7a}$$

and the attractive term of Shoor and Gubbins (*8*), which takes into account van der Waals forces and forces between permanent dipoles, as well as forces between permanent and induced dipoles, are used:

$$\frac{\Delta\mu'_c}{RT} = -\frac{32}{9}\frac{\pi}{kT}\sum_{j=2}^{m}(\rho^*_j\epsilon_{1j}\sigma_{1j}^3) - \frac{4}{3}\frac{\pi}{kT}\sum_{j=2}^{m}\left(\frac{\rho^*_j}{\sigma_{1j}^3}(\mu_1^2\alpha_j + \mu_j^2\alpha_1)\right) - \frac{8}{9}\frac{\pi}{(kT)^2}\sum_{j=2}^{m}\frac{\rho^*_j\mu_1^2\mu_j^2}{\sigma_{1j}^3} \tag{8}$$

Applying the scaled particle theory therefore requires the following information: ν^s_{mix} = saturated liquid molar volume of the mixed solvent; d_1, d_j = hard-sphere diameters of solute 1 and solvent components *j*; p^s_{mix} = vapor pressure of the mixed solvent; ϵ_{1j} = Lennard-Jones energy parameter for interactions between solute 1 and solvent component *j*; σ_{1j} = Lennard-Jones distance parameter for interactions between solute 1 and solvent component *j*; μ_k = dipole moment of molecular species *k*; α_k = polarizability of molecular species *k*. Lennard-Jones parameters for interactions of solute 1 and solvent component *j* were estimated by using the Lorentz–Berthelot mixing rules:

$$\sigma_{1j} = (\sigma_1 + \sigma_j)/2 \tag{9}$$

$$\epsilon_{1j} = (\epsilon_1\epsilon_j)^{1/2} \tag{10}$$

Table IV. Potential Parameters and Constants of Equation 11 Needed To Predict Henry's Coefficients for the Solubility of Hydrogen in Aqueous Ethanolamine (MEA) Solutions

	H_2	H_2O	MEA
a_0	2.87	2.8336	5.0361
$a_1 \times 10^5$	0	−3.3819	−133.894
$a_2 \times 10^7$	0	−7.1048	6.8571
$(\epsilon_i/k)/\text{K}$	29.2	79.3	494.3
μ_i/D	0	1.82	2.59
$\alpha_i/(10^{-30}\ \text{m}^3)$	0.8023		

Table V. Prediction of Henry's Constants for the Solubility of Hydrogen in Aqueous Ethanolamine Solutions with 75, 50, and 25 mol % Water from the Scaled Particle Theory

$\tilde{x}_{H_2O}$	T/K	$H^{\text{spt}}_{H_2,\text{mix}}/\text{MPa}$	$100(H^{\text{spt}}_{H_2,\text{mix}} - H^{\text{exp}}_{H_2,\text{mix}})/H^{\text{exp}}_{H_2,\text{mix}}$
0.75	323.15	5263	−5.4
	373.15	3884	−5.3
	423.15	2707	−0.5
0.50	323.15	3735	−7.7
	348.15	3073	−6.7
	373.15	2528	−1.7
	398.15	2078	−0.5
	423.15	1706	0.5
0.25	323.15	2634	1.5
	373.15	1733	−0.2
	423.15	1168	−1.6

Lennard-Jones distance parameters σ were approximated by the hard-sphere diameters *d*. The Lennard-Jones potential parameters for hydrogen (σ_1 and ϵ_1) and energy parameters for the solvents were taken from Hirschfelder et al. (*9*) and Liabastre (*10*).

Polarizabilities and dipole moments were taken from Liabastre (*10*) and McClellan (*11*). As Henry's constants are very sensitive to hard-sphere diameters, the distance parameters of water and ethanolamine were assumed to depend on temperature

$$\sigma/\text{Å} = a_0 + a_1(T/\text{K}) + a_2(T/\text{K})^2 \tag{11}$$

and fitted to experimental results for Henry's coefficients of hydrogen in the pure solvents (*1*, *2*). For these calculations vapor pressures and liquid densities were taken from Saul and Wagner (*12*), McGarry (*13*), this work, and Aksmanović (*14*). The potential parameters needed to predict Henry's coefficients for the solubility of hydrogen in aqueous ethanolamine solutions are compiled in Table IV. In Table V the predictions are compared with the experimental data. For 373 K the comparison is also shown in Figure 4. The agreement between predicted and measured Henry's coefficients is very good. The deviations exceed the experimental uncertainty only in 4 of 11 data points. The maximum deviation exceeds the estimated experimental error by a factor of 4.

Acknowledgment

We express our gratitude to BASF AG, Ludwigshafen, for supplying ethanolamine samples.

Glossary

a_0, a_1, a_2	coefficients of correlation for Lennard-Jones distance parameter
d	hard sphere diameter
f	fugacity
H_2	hydrogen
$H_{i,j}$	Henry's constant of component *i* in component *j*

H_2O	water
k	Boltzmann's constant
m	number of components in the mixture
MEA	ethanolamine
p	pressure
R	universal gas constant (R = 8.3143 J mol^{-1} K^{-1})
T	temperature
v	molar volume
V	volume
x	mole fraction in liquid phase
y	mole fraction in vapor phase

Greek Symbols

α_i	polarizability of component i
$\gamma_{i,j}$	activity coefficient for component i dissolved in component j
ϵ	Lennard-Jones energy parameter
η_0, η_1, η_2, η_3	reduced densities
μ	chemical potential, dipole moment
ρ	molar density
ρ^*	number density
σ	Lennard-Jones distance parameter
φ	fugacity coefficient
ϕ	volume fraction in liquid phase

Subscripts

c	critical value
ij	binary interactions or attractive property between molecules of species i and j
j	component j
H_2	hydrogen
mix	mixture
pot.	value resulting from intermolecular forces
v	repulsive property

Superscripts

exp	experimental value
s	saturated
spt	value calculated from the scaled particle theory
∞	value at infinite dilution
$'$	liquid phase
$''$	vapor phase
$*$	referred to number of particles, reduced
$\sim$	solute free
$\ominus$	standard value ($p^{\ominus}$ = 0.1 MPa)

Registry No. H_2, 1333-74-0; ethanolamine, 141-43-5.

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Ultrasonic Speed for Liquid Trichlorofluoromethane and 1,1-Dichloro-2,2,2-trifluoroethane at Temperatures from 283 to 373 K and Pressures up to 75 MPa

Toshiharu Takagi
Faculty of Engineering and Design, Kyoto Institute of Technology, Matsugasaki, Sakyoku, Kyoto 606, Japan

The ultrasonic speeds for liquid trichlorofluoromethane (CCl_3F) and 1,1-dichloro-2,2,2-trifluoroethane ($C_2HCl_2F_3$) were measured from 283 to 373 K and from 0.1 MPa or their saturated vapor pressure to about 75 MPa. The measurements were carried out by a sing-around technique operated at a frequency of 2 MHz with the uncertainty of ±0.2%. The temperature and pressure variation of ultrasonic speed and related properties are discussed.

Introduction

Trichlorofluoromethane, CCl_3F (CFC-11), has a high solubility for polymers and is usually used as a solvent in the foam and textile industries. Due to its ability to destroy ozone, its use as a foaming agent and refrigerant is being curtailed. 1,1-Dichloro-2,2,2-trifluoroethane, $C_2HCl_2F_3$ (HCFC-123), is considered a suitable replacement compound of CCl_3F (*1*).

Ultrasonic speeds in fluids, which travel with the compression waves, give a powerful clue for elucidating the thermodyanmic properties. In previous papers (*2-4*), we have been reported the ultrasonic speed of liquid refrigerants at several conditions of temperature and pressure. This paper describes the new experimental results of ultrasonic speeds in the liquid phase of CCl_3F and $C_2HCl_2F_3$. The variation of the ultrasonic speed and related thermodynamic properties for these fluids as a function of temperature and pressure are discussed and compared with related properties of tetrachloromethane, CCl_4.

Experimental Section

Materials. Trichlorofluoromethane, CCl_3F, and 1,1-dichloro-2,2,2-trifluoroethane, $C_2HCL_2F_3$, were supplied by Daikin Industrials Ltd. Their purities were better than 99.8 wt %, as measured by GLC. Tetrachloromethane, CCl_4, was a "spectral grade" reagent supplied by Dojin Chemical Ltd.; its purity, de-

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