COMPONENTS:	EVALUATOR:
(1) Methane; CH_4; [74-82-8] (2) Water; H_2O; [7732-18-5]	Rubin Battino Department of Chemistry Wright State University Dayton, OH 45435 USA 1985, January

CRITICAL EVALUATION:

AN EVALUATION OF THE SOLUBILITY OF METHANE IN WATER BETWEEN TEMPERATURES OF 273.15 and 523.15 K AT A METHANE PARTIAL PRESSURE OF 0.101325 MPa.

Normally only one equation and table of smoothed data are given in an evaluation. This evaluation gives three equations and two tables of smoothed data. Equation (1) represents the best evaluation of data prior to 1981. Equation (2) and Table 1 represent the recommended evaluation between temperatures of 273.15 and 328.15 K based on the precise measurements of Rettich *et al*. (22). Equation (3) and Table 2 represent tentative values between temperatures of 273.15 and 523.15 K based on data from Rettich *et al*. (22) and Crovetto *et al*. (9). For applications around room temperature the data represented by Equation (2) and Table 1 are recommended.

The solubility of methane in water is reported in many papers (1-33). Most of these measurements are at *ca*. atmospheric pressure, but there are many measurements reported at larger pressures which will be evaluated separately. Many of the papers contain additional data on the solubility of methane in aqueous electrolyte solutions and in mixed solvents.

The thirty-three papers that were examined for the evaluation are listed alphabetically in the reference list. The recent results reported by Rettich *et al*. (22) are at least an order of magnitude more precise than the data reported in any other papers. Seven papers published prior to 1981 were judged to contain more reliable data (3, 4, 7, 20, 28, 31, 32) than the other papers. A linear regression of the 36 data points in the 274 to 313 K temperature interval from the seven papers gave the equation

$$\ln x_1 = -78.1584 + 104.4791/\tau + 29.7802 \ln \tau \tag{1}$$

with a standard deviation of 0.0050 in $\ln x_1$ or about 0.50 percent in x_1 at the middle of the temperature range. The sixteen data points from Rettich *et al*. (22) give the equation for the 275 to 328 K temperature range of

$$\ln x_1 = -115.647716 + 155.575631/\tau + 65.2552591 \ln \tau - 6.616975729 \, \tau \tag{2}$$

with a standard deviation in x_1 of 0.056 percent. In both equations $\tau = (T/100 \text{ K})$. In Equation (1) the mole fraction solubility, x_1, is for a partial pressure of 0.101325 MPa methane. In Equation (2) the mole fraction is for a fugacity of 0.101325 MPa methane. Rettich *et al*. (22) used a rigorous thermodynamic approach described in their paper to convert their

COMPONENTS:	EVALUATOR:
(1) Methane; CH_4; [74-82-8] (2) Water; H_2O; [7732-18-5]	Rubin Battino Department of Chemistry Wright State University Dayton, OH 45435 USA 1985, January

CRITICAL EVALUATION:

experimental measurements to mole fraction solubility values. The mole fraction solubilities calculated from Equation (2) are 1 - 2 percent larger at 273.15 K and 0.20 percent larger at 313 K. Both equations have been extrapolated to 373 K. At the larger temperatures Equation (2) gives smaller solubility values than Equation (1). At 373 K Equation (2) gives a solubility value that is eleven percent smaller than the value calculated from Equation (1). Table 1 contains the smoothed data calculated from Equation (2). Equation (2) and the data smoothed at 5 K intervals from 273.15 to 328.15 K are *recommended* values of the mole fraction solubility of methane in water at a fugacity of 0.101325 MPa. The extrapolated values between 333.15 and 373.15 K are tentative. Also given in Table 1 are ideal gas Ostwald coefficients and enthalpy, entropy and constant pressure heat capacity changes for the transfer of one mole of methane from the gas phase to the infinitely dilute solution in water. The mole fraction solubility at 0.101325 MPa fugacity methane shows a minimum at 363 K.

Recently Crovetto *et al.* (9) measured the solubility of methane in water from 297 to 518 K over a fugacity range of 1.3 to 6.5 MPa with an estimated precision in the Henry's constant of 1 - 2 percent. The evaluator computed mole fraction methane solubility values in water at a partial pressure of 0.101325 MPa assuming Henry's law is obeyed. The seven values from the work of Crovetto *et al.* (9) were combined with the 16 values of Rettich *et al.* (22) in a linear regression to yield the equation for the mole fraction solubility of methane at a partial pressure of 0.101325 MPa (1 atm) over the 273.15 to 523.15 temperature range of:

$$\ln x_1 = -99.14188 + 132.821/\tau + 51.91445 \ln \tau - 4.25831 \tau \qquad (3)$$

where again $\tau = (T/100$ K$)$. The standard deviation in $\ln x_1$ is 0.015, and the percent error in x_1 at the middle of the temperature range is 1.5 percent. Smoothed tentative values of the mole fraction solubility and the thermodynamic changes on solution at 10 K intervals from 273.15 to 523.15 K are given in Table 2. Equation (3) and the smoothed data in Table 2 are for the use of workers who need values in the 373 to 523 K temperature range at a methane partial pressure of 0.101325 MPa. For values at larger partial pressures see the evaluation of high pressure methane in water solubility data.

COMPONENTS:	EVALUATOR:
(1) Methane; CH_4; [74-82-8] (2) Water; H_2O; [7732-18-5]	Rubin Battino Department of Chemistry Wright State University Dayton, OH 45435 USA 1985, January

CRITICAL EVALUATION:

TABLE 1.   Solubility of methane in water at a methane partial pressure of 0.101325 MPa. Recommended[a] values of the mole fraction and Ostwald solubility, and of the partial molar thermodynamic changes on solution as a function of temperature.

T/K	Mol Fraction $10^5 x_1$	Ostwald[b] Coefficient L/cm^3 cm^{-3}	$\Delta H_1^0/$ kJ mol^{-1}	$\Delta S_1^0/$ J K^{-1} mol^{-1}	$\Delta C_{p_1}^0/$ J K^{-1} mol^{-1}
Recommended					
273.15	4.6666	0.058055	-19.426	-154.0	262.3
278.15	4.0221	0.050959	-18.127	-149.3	257.2
283.15	3.5192	0.045377	-16.854	-144.8	252.1
288.15	3.1224	0.040946	-15.606	-140.4	246.9
293.15	2.8062	0.037405	-14.384	-136.2	241.8
298.15	2.5523	0.034559	-13.188	-132.2	236.7
303.15	2.3469	0.032267	-12.018	-128.3	231.5
308.15	2.1802	0.030420	-10.873	-124.5	226.4
313.15	2.0445	0.028936	- 9.754	-120.9	221.3
318.15	1.9340	0.027752	- 8.660	-117.5	216.1
323.15	1.8442	0.026821	- 7.592	-114.1	211.0
328.15	1.7717	0.026103	- 6.550	-110.9	205.9
Tentative					
333.15	1.7138	0.025569	- 5.533	-107.9	200.8
338.15	1.6683	0.025196	- 4.542	-104.9	195.6
343.15	1.6336	0.024966	- 3.577	-102.1	190.5
348.15	1.6082	0.024862	- 2.637	- 99.3	185.4
353.15	1.5911	0.024873	- 1.723	- 96.7	180.2
358.15	1.5815	0.024990	- 0.835	- 94.2	175.1
363.15	1.5785	0.025206	+ 0.028	- 91.9	170.0
368.15	1.5817	0.025513	0.865	- 89.6	164.9
373.15	1.5905	0.025909	1.676	- 87.4	159.7

[a] The data are classed as recommended over the 273.15 to 328.15 K temperature interval of the experimental data. The extrapolated data over the 333.15 to 373.15 K temperature interval are classed as tentative.

[b] The Ostwald coefficients were not corrected for non-ideal behavior.

COMPONENTS:	EVALUATOR:
(1) Methane; CH_4; [74-82-8] (2) Water; H_2O; [7732-18-5]	Rubin Battino Department of Chemistry Wright State University Dayton, OH 45435 USA 1985, January

CRITICAL EVALUATION:

TABLE 2.   The solubility of methane in water at unit fugacity. Tentative
values of the mole fraction solubility, and thermodynamic changes
on solution as a function of temperature between 273.15 and
523.15 K.   Values based on the experimental data of Rettich et $al.$
(22) and Crovetto et $al.$ (9).   The Crovetto et $al.$ data were ex-
trapolated to a pressure of 0.101325 MPa assuming Henry's law is
obeyed.

T/K	Mol Fraction $10^5 x_1$	ΔG_1^0 / kJ mol^{-1}	ΔH_1^0 / kJ mol^{-1}	ΔS_1^0 / J K^{-1} mol^{-1}	$\Delta C_{p_1}^0$ / J K^{-1} mol^{-1}
273.15	4.625	22.669	-18.947	-152.4	238.2
283.15	3.507	24.149	-16.600	-143.9	231.1
293.15	2.803	25.549	-14.324	-136.0	224.1
298.15	2.550	26.219	-13.213	-132.3	220.5
303.15	2.344	26.871	-12.119	-128.6	217.0
313.15	2.037	28.132	- 9.985	-121.7	209.9
323.15	1.832	29.307	- 7.922	-115.2	202.8
333.15	1.695	30.428	- 5.929	-109.1	195.7
343.15	1.609	31.491	- 4.007	-103.4	188.6
353.15	1.560	32.498	- 2.156	- 98.1	181.6
363.15	1.542	33.454	- 0.375	- 93.2	174.5
373.15	1.548	34.362	+ 1.334	- 88.5	167.4
383.15	1.577	35.226	2.973	- 84.2	160.3
393.15	1.625	36.047	4.541	- 80.1	153.2
403.15	1.691	36.829	6.038	- 76.4	146.2
413.15	1.776	37.575	7.464	- 72.9	139.1
423.15	1.878	38.288	8.819	- 69.6	132.0
433.15	1.998	38.969	10.104	- 66.6	124.9
443.15	2.137	39.621	11.318	- 63.9	117.8
453.15	2.295	40.247	12.461	- 61.3	110.8
463.15	2.472	40.848	13.533	- 59.0	103.7
473.15	2.670	41.427	14.534	- 56.8	96.6
483.15	2.889	41.986	15.465	- 54.9	89.5
493.15	3.131	42.526	16.325	- 53.1	82.4
503.15	3.395	43.049	17.113	- 51.5	75.4
513.15	3.683	43.557	17.832	- 50.1	68.3
523.15	3.995	44.052	18.479	- 48.9	61.2

COMPONENTS:	EVALUATOR:
(1) Methane; CH_4; [74-82-8] (2) Water; H_2O; [7732-18-5]	Rubin Battino· Department of Chemistry Wright State University Dayton, OH 45435 USA 1985, January

CRITICAL EVALUATION:

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COMPONENTS:	EVALUATOR:
(1) Methane; CH_4; [74-82-8] (2) Water; H_2O; [7732-18-5]	Rubin Battino Department of Chemistry Wright State University Dayton, OH 45435 USA 1985, January

CRITICAL EVALUATION:

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COMPONENTS:	ORIGINAL MEASUREMENTS:
(1) Methane; CH_4; [74-82-8] (2) Water; H_2O; [7732-18-5]	Bunsen, R. W. *Ann. Chem. Pharm.* 1855, *93*, 1-50. [The Journal's title later changed to *J. Liebigs Ann. Chem.*]

VARIABLES:
   T/K = 279.35 - 298.75
	PREPARED BY:
	        H. L. Clever

EXPERIMENTAL VALUES:

Temperature		Mol Fraction	Bunsen Coefficient
$t/^0C$	T/K	$10^5 x_1$	$10^2 \alpha / cm^3 (STP) cm^{-3} atm^{-1}$
6.2	279.35	3.821	4.742
9.4	282.55	3.587	4.451
12.5	285.65	3.326	4.126
18.7	291.85	2.894	3.586
25.6	298.75	2.522	3.121

The compiler calculated the Kelvin temperature and the mole fraction solubility values.

The complete paper was translated into English by a Mr. Roscoe and published by Bunsen (1). Two long abstracts of the paper were published (2, 3). One was translated into French by M. Verdet (2). The two abstracts presented only the interpolation formula and a table of smoothed values at one degree intervals from 0 to 20 ^0C. The complete set of experimental values, the interpolation equation and smoothed values are also given in Bunsen's book on gasometric methods (4).

Bunsen's interpolation equation for the data is

$$\alpha / cm^3 (STP) cm^{-3} atm^{-1} = 0.05449 - 0.0011807 \ (t/^0C) + 0.000012078 \ (t/^0C)^2.$$

The paper reports solubility measurements of nitrogen, hydrogen, methane, ethane, ethene, carbon monoxide, oxygen and air in water made chiefly by a Dr. Pauli. Only the methane data agree well with modern values.

AUXILIARY INFORMATION

METHOD/APPARATUS/PROCEDURE:	SOURCE AND PURITY OF MATERIALS:
Bunsen's original apparatus and procedure were used. They are described in detail in the paper. Many of the data reported in this paper are of only historical interest. The methane solubility values have stood the test of time fairly well. They are within 2 to 5 percent of modern values.	(1) Methane. Natural gas sample from the mud-volcanoes of Bulganack in the Crimea. Treated with K to remove CO_2. (2) Water. Boiled briskly under vacuum to remove dissolved air. Transferred to the apparatus without contact with air.

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COMPONENTS:	ORIGINAL MEASUREMENTS:
(1) Methane; CH_4; [74-82-8]	Winkler, L. W.
(2) Water; H_2O; [7732-18-5]	*Chem. Ber.* 1901, *34*, 1408-22.

EXPERIMENTAL VALUES:

Temperature		Pressure	Water Volume	Methane Volume (STP)	Bunsen Coefficient
$t/^\circ C$	T/K	p_1/mmHg	v_2/cm^3	v_1/cm^3	$10^2\alpha$/cm^3(STP)cm^{-1}atm^{-1}
0.25	273.40	431.81	2066.43	64.90	5.528
0.30	273.45	431.90	2066.43	64.86	5.523
0.23	273.38	444.58	2098.12	67.69	5.515
0.27	273.42	444.55	2098.12	67.69	5.516
0.28	273.43	444.62	2098.12	67.65	5.511
9.98	283.13	469.59	2066.71	53.35	4.178
10.00	283.15	469.42	2066.71	53.43	4.186
10.00	283.15	469.71	2066.71	53.28	4.171
20.08	293.23	505.46	2069.78	45.52	3.307
20.00	293.15	505.26	2069.75	45.58	3.312
20.00	293.15	505.09	2069.75	45.68	3.321
19.98	293.13	504.99	2069.75	45.54	3.311
20.05	293.20	505.19	2069.77	45.49	3.305
20.02	293.17	505.34	2069.76	45.39	3.298
20.00	293.15	523.86	2101.49	47.83	3.302
20.00	293.15	523.66	2101.49	47.88	3.307
20.00	293.15	523.86	2101.49	47.79	3.299
29.95	303.10	538.90	2074.92	40.67	2.764
30.10	303.25	539.25	2075.00	40.65	2.761
30.00	202.15	539.10	2074.95	40.61	2.759
40.02	313.17	573.54	2082.09	37.31	2.375
40.03	313.18	573.95	2082.10	37.15	2.363
40.00	313.15	573.77	2082.08	37.21	2.367
50.08	323.23	608.54	2090.93	35.70	2.132
50.00	323,15	608.24	2090.86	35.77	2.137
50.00	323.15	608.54	2090.86	35.65	2.129
60.03	333.18	644.69	2101.14	35.09	1.969
59.93	333.08	645.77	2101.04	34.61	1.939
60.00	333.15	644.45	2101.11	35.20	1.977
59.95	333.10	676.49	2133.29	36.93	1.945
60.02	333.17	676.23	2133.47	37.07	1.953
59.95	333.10	676.65	2133.29	36.87	1.941
70.00	343.15	682.54	2112.78	35.03	1.846
70.00	343.15	683.94	2112.78	34.45	1.816
70.05	343.20	684.12	2112.84	34.43	1.811
80.00	353.15	725.54	2125.82	35.78	1.763
80.02	353.17	722.20	2125.84	35.69	1.767
79.97	353.12	721.44	2125.79	35.91	1.779

COMPONENTS:	ORIGINAL MEASUREMENTS:
(1) Methane; CH_4; [74-82-8] (2) Water; H_2O; [7732-18-5]	Winkler, L. W. *Chem. Ber.* <u>1901</u>, *34*, 1408-22.
VARIABLES: T/K = 273.38 - 353.17 p_1/kPa = 57.570 - 96.731	PREPARED BY: H. L. Clever

EXPERIMENTAL VALUES:

The temperatures and Bunsen coefficients below are the average values given by Winkler from the experimental data on the preceeding page.

Temperature		Mol Fraction	Bunsen Coefficient	Ostwald Coefficient
$t/^0C$	T/K	$10^5 x_1$	$10^2\alpha/cm^3$(STP)$cm^{-3}atm^{-1}$	$10^2 L/cm^3$ cm^{-3}
0.27	273.42	4.451	5.519	5.524
9.99	283.14	3.490	4.178	4.331
20.01	293.16	2.864	3.307	3.549
30.02	303.17	2.479	2.761	3.064
40.02	313.17	2.205	2.368	2.715
50.03	323.18	2.058	2.133	2.524
59.98	333.13	1.953	1.954	2.383
70.02	343.17	1.889	1.825	2.293
80.00	353.15	1.897	1.770	2.288

The mole fraction solubility at 101.325 kPa methane partial pressure was calculated by the compiler using a methane molar volume of 22,360.4 cm^3 (STP) mol^{-1}.

The Ostwald coefficients were calculated by the compiler. The Kelvin temperatures were added by the compiler.

AUXILIARY INFORMATION

METHOD/APPARATUS/PROCEDURE:	SOURCE AND PURITY OF MATERIALS:
The original Bunsen absorption method (ref 1) was used. The method and apparatus are described in earlier papers (ref 2).	(1) Methane. Prepared by the decomposition of dimethyl zinc by air free water. (2) Water. Distilled.
	ESTIMATED ERROR: $\delta T/K$ = ± 0.01 $\delta\alpha/\alpha$ ± 0.01 (compiler)
	REFERENCES: 1. Bunsen, R. W. *Gasometrische Methoden*, 2nd. ed., Braunschweig, <u>1858</u>. 2. Winkler, L. W. *Chem. Ber.* <u>1893</u>, *24*, , 3602.

COMPONENTS:	ORIGINAL MEASUREMENTS:
1. Methane; CH_4; [74-82-8] 2. Water; H_2O; [7732-18-5]	Wetlaufer, D. B.; Malik, S. K.; Stoller, L.; Coffin, R. L. *J. Am. Chem. Soc.* <u>1964</u>, *86*, 508-514.

VARIABLES:	PREPARED BY:
Temperature	C. L. Young

EXPERIMENTAL VALUES:

T/K	10^3 Conc. of methane[†] in soln./mol dm^{-3}	Mole fraction[*] of methane x_{CH_4}
278.2	0.00219	0.0000396
298.2	0.00141	0.0000255
318.2	0.00107	0.0000193

[†] at a partial pressure of 101.3 kPa.

[*] calculated by compiler.

AUXILIARY INFORMATION

METHOD/APPARATUS/PROCEDURE:	SOURCE AND PURITY OF MATERIALS:
Modified Van Slyke-Neill apparatus fitted with a magnetic stirrer. Solution was saturated with gas and then sample transferred to the Van Slyke extraction chamber.	1. Matheson c.p. grade, purity 99 mole per cent or better. 2. Distilled.

ESTIMATED ERROR:
$\delta T/K = \pm 0.05$; $\delta x_{CH_4} = \pm 2\%$.

REFERENCES:

COMPONENTS:	ORIGINAL MEASUREMENTS:
(1) Methane; CH_4; [74-82-8] (2) Water; H_2O; [7732-18-5]	Morrison, T. J.; Billett, F. *J. Chem. Soc.* <u>1952</u>, 3819 - 22.

| VARIABLES: T/K = 285.1 - 348.4
p_1/kPa =]0].325 | PREPARED BY:

H. L. Clever |

EXPERIMENTAL VALUES:

Temperature		Mol Fraction	Solubility
$t/^{\circ}C$	T/K	$10^5 x_1$	S/cm^3(STP)kg^{-1}
11.9	285.1	3.215	39.90
14.7	287.9	3.014	37.41
20.5	293.7	2.655	32.96
25.2	298.4	2.422	30.06
35.0	308.2	2.042	25.35
41.0	314.2	1.919	23.82
46.8	320.0	1.808	22.44
51.8	325.0	1.722	21.38
62.3	335.5	1.600	19.86
71.5	344.7	1.539	19.10
75.2	348.4	1.518	18.84

[a] The compiler calculated the mole fraction solubility at 101.325 kPa (1 atm) using the real gas molar volume of 22,360.4 cm^3 at standard conditions of 273.15 K and 101.325 kPa.

[b] The solubility reported by the authors is the same as 10^3 times the Kunen coefficient, or $10^3 s$/cm^3(STP) g^{-1}.

The authors gave the smoothing equation (ref 1).

$$\log_{10}(S/\text{cm}^3(\text{STP})\text{kg}^{-1}) = -77.067 + 4090/(T/K) + 26.20 \log_{10}(T/K)$$

AUXILIARY INFORMATION

METHOD/APPARATUS/PROCEDURE:	SOURCE AND PURITY OF MATERIALS:
The original apparatus was described in references (1, 2). Degassed solvent flows in a thin film through the gas down an absorption helix. The gas absorbed and the solvent volume used are read on burets. NOTE: In the authors earlier paper (ref 2) they report five methane solubility values in water at 298.2 ± 0.1 K of 27.9, 27.8, 27.9, 27.8 and 27.9 cm^3 dm^{-3} (gas volume at STP). The results are about 7 per cent smaller than the results reported in this paper. They are considered preliminary results and no data sheet was prepared for them.	(1) Methane. Prepared by the authors from Grignard reagent. (2) Water. Distilled.

	ESTIMATED ERROR: $\delta T/K$ = ± 0.01 $\delta S/S$ = ± 0.01 (compiler)

REFERENCES:
1. Morrison, T. J.
   J. Chem. Soc. <u>1952</u>, 3814.
2. Morrison, T. J.; Billett, F.
   J. Chem. Soc. <u>1948</u>, 2033.

COMPONENTS:	ORIGINAL MEASUREMENTS:
(1) Methane; CH_4; [74-82-8] (2) Water; H_2O; [7732-18-5]	Claussen, W. F.; Polglase, M. F. *J. Am. Chem. Soc.* 1952, *74*, 4817-9.
VARIABLES: T/K = 274.8 - 312.8 p_1/kPa = 101.325	PREPARED BY: H. L. Clever

EXPERIMENTAL VALUES:

Temperature		Mol Fraction	Bunsen Coefficient	Ostwald Coefficient
$t/^0$C	T/K	$10^5 x_1$	$10^2 \alpha$ cm^3(STP)cm^{-3} atm^{-1}	$10^2 L$/cm^3 cm^{-3}
1.6	274.8		5.52, 5.42	
		4.41	Av. 5.47	5.50
2.0	275.2		5.42, 5.38, 5.32	
		4.33	Av. 5.38	5.42
10.5	283.7		4.25, 4.29, 4.28 4.316, 4.251	
		3.45	Av. 4.28	4.44
19.8	293.0		3.488, 3.251, 3.509	
		2.83	Av. 3.51	3.76
30.4	303.6		2.886, 2.897	
		2.34	Av. 2.89	3.21
39.6	312.8		2.542, 2.563	
		2.07	Av. 2.55	2.92

The authors reported the Bunsen coefficients and their average. The compiler calculated the mole fraction and the Ostwald coefficient values. A methane volume of 22,360.4 cm^{3}STP) mol^{-1} was used to calc- ulate the methane mole fraction at 101.325 kPa.

AUXILIARY INFORMATION

METHOD/APPARATUS/PROCEDURE:

   The solubility was determined by a micro combustion technique. Methane was bubbled through the water via a sintered glass disc to saturate the water.

   The methane in the saturated solu- tion was removed by bubbling oxygen. The train for analysis was composed of an oxygen tank to sweep out the dissolved gas, pressure regulators, mercury manometer, preheater, absorption U-tube containing ascarite and anhydrone, aerator, combustion tube containing copper oxide at 973 K, weighing tubes containing ascarite and anhydrone, and a Marriotte flask.

SOURCE AND PURITY OF MATERIALS:
(1) Methane. Phillips Petroleum Co. 99.7 % with N_2 the greatest impurity by mass spectrometry.

(2) Water. Doubly distilled.

ESTIMATED ERROR:
    $\delta T/K$ = ± 0.1
    $\delta \alpha / \alpha$ = ± 0.01

REFERENCES:

COMPONENTS:	ORIGINAL MEASUREMENTS:
(1) Methane; CH_4; [74-82-8] (2) Water; H_2O; [7732-18-5]	Lannung, A.; Gjaldbaek, J. C. *Acta Chem. Scand.* 1960, *14*, 1124-8.
VARIABLES: T/K = 291.15 - 310.15 p_1/kPa = 101.325	PREPARED BY: J. Chr. Gjaldbaek

EXPERIMENTAL VALUES:

Temperature		Mol Fraction	Bunsen Coefficient
$t/^0C$	T/K	$10^3 x_1$	α/cm^3 (STP)$cm^{-3}atm^{-1}$
18	291.15	2.82 2.85	0.0350 0.0354
25	298.15	2.55 2.50	0.0315 0.0310
37	310.15	2.10 2.11	0.0259 0.0261

AUXILIARY INFORMATION

METHOD/APPARATUS/PROCEDURE:

The apparatus is a calibrated all-glass combined manometer and bulb enclosed in an air thermostat (ref 1). The entire apparatus is shaken until equilibrium is reached.

The absorbed volume of gas is calculated from the initial and final amounts, both saturated with solvent vapor. The amount of solvent is determined by the weight of displaced mercury. Details in the reference.

SOURCE AND PURITY OF MATERIALS:

(1) Methane. Generated from magnesium methyliodide. Purified by fractional distillation. Specific gravity corresponing with the molecular weight 16.08.

(2) Water. Redistilled. Specific conductivity 2 x 10^{-7} $(\Omega$ cm$)^{-1}$.

ESTIMATED ERROR:
$$\delta T/K = \pm 0.05$$
$$\delta x_1/x_1 = \pm 0.015$$

REFERENCES:
1. Lannung, A. *J. Am. Chem. Soc.* 1930, *52*, 68.

COMPONENTS:	ORIGINAL MEASUREMENTS:
(1) Methane; CH_4; [74-82-8] (2) Water; H_2O; [7732-18-5]	Namiot, A. Yu. *Zh. Strukt. Khim.* 1961, 2, 408-17. *J. Struct. Chem. (Engl. Transl.)* 1961, 2, 381-9.

VARIABLES:	PREPARED BY:
T/K = 273, 283 p_1/kPa = 101.3	H. L. Clever

EXPERIMENTAL VALUES:

Temperature		Henry's Constant K/atm	Mol Fraction at One Atm (compiler) $10^5 x_1$
$t/^0C$	T/K		
0	273.15	22500	4.44
10	283.15	29000	3.45

Henry's constant, K/atm = $(p_1/atm)/x_1$.

AUXILIARY INFORMATION

METHOD/APPARATUS/PROCEDURE:	SOURCE AND PURITY OF MATERIALS:
No experimental details are given. The paper does not make clear whether these are new experimental values or literature values. The paper does contain literature values of the partial molar volume of the gas in water and other thermodynamic information.	No details.
	ESTIMATED ERROR:
	REFERENCES:

COMPONENTS:	ORIGINAL MEASUREMENTS:
(1) Methane; CH_4; [74-82-8] (2) Water; H_2O; [7732-18-5]	Wen, W.-Y.; Hung, J. H. *J. Phys. Chem.* <u>1970</u>, *74*, 170 - 80.

VARIABLES: $T/K = 278.15 - 308.15$ $p_1/kPa = 101.325$	PREPARED BY: H. L. Clever

EXPERIMENTAL VALUES:

T/K	Mol Fraction $10^5 x_1$	Bunsen Coefficient $10^3 \alpha/$ $cm^3(STP)cm^{-3}atm^{-1}$	Ostwald Coefficient $10^3 L/$ $cm^3 \, cm^{-3}$	Kunen Coefficient $10^3 S/$ $cm^3(STP)g^{-1}atm^{-1}$
278.15	3.987	49.48	50.39	49.48 ± 0.08
288.15	3.100	38.46	40.57	38.49 ± 0.11
298.15	2.526	31.26	34.12	31.35 ± 0.10
308.15	2.136	26.35	29.73	26.51 ± 0.08

The authors reported the solubility of methane as $cm^3(STP) \, kg^{-1}$. This is the same as 10^3 time the Kunen coefficient reported above.

The compiler calculated the mole fraction, Bunsen coefficient, and Ostwald coefficients using the real gas molar volume of 22,360.4 $cm^3 \, mol^{-1}$ for methane at standard conditions of 273.15 K and 101.325 kPa (1 atm).

AUXILIARY INFORMATION

METHOD/APPARATUS/PROCEDURE:	SOURCE AND PURITY OF MATERIALS:

The apparatus was similar to that described by Ben-Naim and Baer (ref 1). Teflon needle valves were used in place of stopcocks.
 	
   The apparatus consists of three parts, a dissolution cell of 300 to 600 cm^3 capacity, a gas volume measuring column, and a manometer.
 	
   The solvent is degassed in the dissolution cell, the gas is introduced and dissolved as the gas is stirred by a magnetic stirrer. Dissolution of the gas results in the change in the height of a column of mercury which is measured by a cathetometer.
	(1) Methane. Matheson Co., Inc. Stated to be better than 99.9 percent pure.
	
	(2) Water. Distilled from an all glass apparatus. Specific conductivity 1.5×10^{-6} $(ohm \; cm)^{-1}$

ESTIMATED ERROR:

$\delta S/S = \pm 0.003$
$\delta T/K = \pm 0.005$

REFERENCES:

1. Ben-Naim, A.; Baer, S.
Trans. Faraday Soc. <u>1963</u>, *59*, 2735.

COMPONENTS:	ORIGINAL MEASUREMENTS:
(1) Methane; CH_4; [74-82-8]	Ben-Naim, A.; Wilf, J.; Jaacobi, M.
(2) Water; H_2O; [7732-18-5]	*J. Phys. Chem.* 1973, *77*, 95 - 102.

VARIABLES:	PREPARED BY:
$T/K = 278.15 - 298.15$ $p_1/kPa = 101.325$	H. L. Clever

EXPERIMENTAL VALUES:

Temperature		Mol Fraction	Ostwald Coefficient
$t/^0C$	T/K	$10^5 x_1$	$L/cm^3 \; cm^{-3}$
5	278.15	3.97	0.0502
10	283.15	3.48	0.0448
15	288.15	3.10	0.0405
20	293.15	2.78	0.0370
25	298.15	2.53	0.0342

The compiler added the Kelvin temperatures.

The compiler calculated the mole fraction solubilities for 1 atm (101.325 kPa) using real methane molar volumes. The real molar volumes increased the mole fraction solubility by about 0.22 percent.

AUXILIARY INFORMATION

METHOD/APPARATUS/PROCEDURE:	SOURCE AND PURITY OF MATERIALS:
The method of Ben-Naim and Baer (ref 1) was used. The apparatus was modified by the addition of Teflon stopcocks. The degassed solvent in a volumetric container is forced by a stirrer created vortex up side arms and through tubes containing solvent vapor saturated gas. The gas uptake is deterimned on a buret at constant pressure.	(1) Methane. Matheson Co., Inc. Purity 99.97 percent. (2) Water. Doubly distilled.
	ESTIMATED ERROR: $\delta L/L = \pm 0.005$ (compiler)
	REFERENCES: 1. Ben-Naim, A.; Baer, S. *Trans. Faraday Soc.* 1963, *59*, 2735.

COMPONENTS:	ORIGINAL MEASUREMENTS:
(1) Methane; CH_4; [74-82-8] (2) Water; H_2O; [7732-18-5]	Ben-Naim, A.; Yaacobi, M. *J. Phys. Chem.* 1974, *78*, 170 - 5.
VARIABLES: T/K = 283.15 - 303.15 p_1/kPa = 101.325	PREPARED BY: H. L. Clever

EXPERIMENTAL VALUES:

Temperature		Mol Fraction	Ostwald Coefficient
$t/°C$	T/K	$10^5 x_1$	$L/cm^3\ cm^{-3}$
10	283.15	3.482	0.04480
15	288.15	3.096	0.04051
20	293.15	2.781	0.03700
25	298.15	2.530	0.03420
30	303.15	2.326	0.03192

a Mole fraction solubility at 101.325 kPa (1 atm) partial pressure methane calculated by the compiler using the second viral coefficient of methane. The values are about one per cent larger than the values obtained when ideal gas behavior is assumed.

b The same Ostwald coefficients appear in the author's paper Yaacobi, M.; Ben-Naim, A. *J. Soln. Chem.* 1973, *2*, 425. They appear to be the same as published by Ben-Naim, Wilf and Yaacobi *J. Phys. Chem.* 1973, *77*, 95, and by Yaacobi and Ben-Naim *J. Phys. Chem.* 1974, *78*,175, but the values are rounded to three digits in the two papers.

AUXILIARY INFORMATION

METHOD/APPARATUS/PROCEDURE:	SOURCE AND PURITY OF MATERIALS:
Used the method of Ben-Naim and Baer (ref 1) as modified by Wen and Hung (ref 2). Degassed liquid in a volumetric container is forced by a stirrer created vortex up side-arms and through tubes containing gas saturated with liquid. Gas absorption is measured on a buret at a constant gas pressure.	(1) Methane. Matheson Co., Inc. Stated to be 99.97 per cent methane. (2) Water. Passed through an ion exchanger and double distilled.
	ESTIMATED ERROR: $\delta L/L$ = ± 0.005 (compiler)

	REFERENCES:
	1. Ben-Naim, A.; Baer, S.
	    Trans. Faraday Soc. 1963,*59*,2735.
	
	2. Wen, W.-Y.; Hung, J. H.
	    J. Phys. Chem. 1970, *74*, 170.

COMPONENTS:	ORIGINAL MEASUREMENTS:
(1) Methane; CH_4; [74-82-8] (2) Water; H_2O; [7732-18-5]	Moudgil, B. M.; Somasundaran, P.; Lin, L. J. *Rev. Sci. Instrum.* <u>1974</u>, *45*, 406-9.

VARIABLES:	PREPARED BY:
T/K = 298.15 p_1/kPa = 101.325	H. L. Clever

EXPERIMENTAL VALUES:

Temperature		Solubility/	Mol Fraction	Ostwald Coefficient
$t/^0C$	T/K	$cm^3(STP)$ kg^{-1}	$10^5 x_1$	L/cm^3 cm^{-3}
25.0	298.15	31.67	2.546	0.03447

The mole fraction and Ostwald coefficient values were calculated
by the compiler assuming ideal gas behavior.

AUXILIARY INFORMATION

METHOD/APPARATUS/PROCEDURE:	SOURCE AND PURITY OF MATERIALS:

The apparatus is based on the design of Ben Naim and Baer (1). The apparatus consists of an absorption cell, a gas measuring column and the pressure control system.
 	
  The pressure control system is automated.
	(1) Methane. Matheson Co., Inc.
	    Stated to be 99.9 percent.
	
	(2) Water. Triple distilled, specific
	    conductivity 1.5 x 10^{-6} Ω^{-1} cm^{-1}

ESTIMATED ERROR: $\delta T/K$ = ± 0.1
$\delta p/kg$ cm^{-2} = ± 0.1
Maximum error ± 0.4 percent
(authors)

REFERENCES:

1. Ben Naim, A.; Baer, S.
   Trans. Faraday Soc. <u>1963</u>, *59*,
   2735.

COMPONENTS:	ORIGINAL MEASUREMENTS:
(1) Methane; CH_4; [74-82-8] (2) Water; H_2O; [7732-18-5]	Yamamoto, S.; Alcauskas, J. B.; Crozier, T. E. *J. Chem. Eng. Data* 1976, *21*, 78 - 80.

EXPERIMENTAL VALUES:

Temperature		Mol Fraction[1]	Bunsen Coefficient
t ^{0}C	T/K	$10^5 x_1$	$10^2 \alpha$/cm^3(STP) cm^{-3} atm^{-1}
0.76	273.91	4.506	5.592
0.78	273.93	4.522	5.612
0.79	273.94	4.523	5.613
0.80	273.95	4.512	5.600
0.80	273.95	4.505	5.591
0.81	273.96	4.515	5.604
4.92	278.07	3.994	4.957
4.94	278.09	3.999	4.963
4.95	278.10	4.068	4.973
4.96	278.11	3.998	4.963
4.97	278.12	4.003	4.968
10.90	284.05	3.413	4.235
10.93	284.08	3.398	4.216
10.93	284.08	3.419	4.242
10.94	284.09	3.404	4.224
10.94	284.09	3.401	4.220
10.94	284.09	3.416	4.238
10.95	284.10	3.404	4.224
10.95	284.10	3.406	4.226
10.96	284.11	3.427	4.252
17.99	291.14	2.909	3.606
18.00	291.15	2.907	3.603
18.01	291.16	2.920	3.619
18.02	291.17	2.910	3.607
18.02	291.17	2.909	3.606
24.10	297.25	2.582	3.196
24.11	297.26	2.579	3.192
24.15	297.30	2.576	3.189
24.16	297.31	2.594	3.211
24.17	297.32	2.585	3.200
29.52	302.67	2.358	2.915
29.54	302.69	2.357	2.913
29.54	302.69	2.358	2.215
29.54	302.69	2.355	2.911
29.55	302.70	2.347	2.901

[1] The mole fraction values were calulated by the compiler using a methane molar volume of v_1/cm^3 mol^{-1} = 22,360.4 and water density values from the *SMOW* tables.

The Bunsen ceofficients are repeated on the sea water data sheet from the paper.

COMPONENTS:	ORIGINAL MEASUREMENTS:
(1) Methane; CH_4; [74-82-8] (2) Water; H_2O; [7732-18-5]	Yamamoto, S.; Alcauskas, J. B.; Crozier, T. E. *J. Chem. Eng. Data* <u>1976</u>, *21*, 78-80.

VARIABLES:	PREPARED BY:
T/K = 273.91 - 302.70 p_1/kPa = 101.325	H. L. Clever

EXPERIMENTAL VALUES:

The author's equation

$$\ln\ (\alpha/cm^3(STP)cm^{-3}atm^{-1}) = -67.1962 + 99.1624/(T/100\ K)$$

$$+\ 27.9015\ \ln\ (T/100\ K)$$

was obtained by the method of least squares from the solubility data in water and in sea water at the various temperatures. Only the pure water part of the equation is given above.

AUXILIARY INFORMATION

METHOD/APPARATUS/PROCEDURE:	SOURCE AND PURITY OF MATERIALS:
Solubility measurements were made by the Scholander microgasometeric method (ref 1) as modified by Douglas (ref 2). The author's procedure was described in an earlier paper (ref 3). The solubilities were corrected for the effect of dissolved gas on the volume of the aqueous phase by using a value of 37 cm^3 for the partial molal volume of methane in water. The correction increased the Bunsen coefficients by about 0.16 percent. The standard deviations of a single measurement at a constant temperature and pressure ranged from 0.09 to 0.53 percent.	(1) Methane. Linde Specialty Gas. Research grade, 99.99 percent purity. The gas was passed through Ascarite to remove CO_2 prior to use. (2) Water. Distilled.

Estimated Error and References for the table:

ESTIMATED ERROR:
$\delta\alpha/\alpha$ = ± 0.003 (authors)

REFERENCES:
1. Scholander, P. F.
   J. Biol. Chem. <u>1947</u>, *167*, 235.
2. Douglas, E.
   J. Phys. Chem. <u>1964</u>, *68*, 169.
3. Crozier, T. E.; Yamamoto, S.
   J. Chem. Eng. Data <u>1974</u>, *19*, 242.

COMPONENTS:	ORIGINAL MEASUREMENTS:
(1) Methane; CH_4; [74-82-8] (2) Water; H_2O; [7732-18-5]	Muccitelli, J. A.; Wen, W.-Y. *J. Solution Chem.* <u>1980</u>, *9*, 141 - 61.

VARIABLES: $T/K = 278.15 - 298.15$ $p_1/kPa = 101.325$	PREPARED BY: H. L. Clever

EXPERIMENTAL VALUES:

T/K	Mol Fraction $10^5 x_1$	Bunsen Coefficient $10^3 \alpha/cm^3 (STP) cm^{-3} atm^{-1}$	Ostwald Coefficient $10^3 L/cm^3 \, cm^{-3}$
278.15	4.044	50.19	51.12 ± 0.35
283.15	3.501	43.44	45.04 ± 0.17
288.15	3.086	38.27	40.39 ± 0.18
293.15	2.813	34.85	37.42 ± 0.10
298.15	2.535	31.37	34.26 ± 0.15

The compiler calculated the mole fraction and Bunsen coefficient values using the real methane gas volumes in F. Din, *Thermodynamic Functions of Gases*, Butterworths, London, 1961, Vol. 3.

AUXILIARY INFORMATION

METHOD/APPARATUS/PROCEDURE:

   The solubility apparatus and procedure employed were similar to that described by Ben-Naim and Baer (ref 1) with modifications suggested by Wen and Hung (ref 2). The apparatus consists of a mercury manometer, a gas-volume measuring buret, a dissolution cell of about 450 cm^3 capacity, and a mercury reservoir.

   The degassing apparatus and procedure used were similar to that described by Battino *et al.* (ref 3).

SOURCE AND PURITY OF MATERIALS:

(1) Methane. Matheson Co., Inc. Specified to have a purity of 99.95 percent.

(2) Water. Carbon dioxide free.

ESTIMATED ERROR:
$$\delta T/K = \pm 0.005$$
$$\delta p_1/mmHg = \pm 3$$
$$\delta L/L = \pm 0.005$$

REFERENCES:
1. Ben-Naim, A.; Baer, S.
   Trans. Faraday Soc. <u>1964</u>, *60*, 1736.
2. Wen, W.-Y.; Hung, J. H.
   J. Phys. Chem. <u>1970</u>, *74*, 170.
3. Battino, R.; Banzhof, M.; Bogan, M.; Wilhelm, E.
   Anal. Chem. <u>1971</u>, *43*, 806.

COMPONENTS:	ORIGINAL MEASUREMENTS:
(1) Methane; CH_4; [74-82-8] (2) Water; H_2O; [7732-18-5]	Cosgrove, B. A.; Walkley, J. *J. Chromatogr.* <u>1981</u>, *216*, 161 - 7.

VARIABLES:	PREPARED BY:
T/K = 278.15 - 318.15 p_1/kPa = 101.325	H. L. Clever

EXPERIMENTAL VALUES:

Temperature T/K	Mol Fraction $10^4 x_1$
278.15	0.3978
283.15	0.3467
288.15	0.2940
293.15	0.2720
298.15	0.2485
303.15	0.2278
308.15	0.2140
313.15	0.1943
318.15	0.1899

AUXILIARY INFORMATION

METHOD/APPARATUS/PROCEDURE:	SOURCE AND PURITY OF MATERIALS:
A 20 ml volume of degassed solvent (sublimation technique) is transferred to a previously evacuated (10^{-4} mmHg) saturation cell immersed in an insulated controlled (± 0.01 K) water bath. The gas is dispersed through the constantly stirred solution at 1 atm by a course, fritted glass disc. Saturation is obtained within a few hours. Prior to analysis the solution is allowed to sit under 1 atm gas pressure for one hour. A saturated sample is withdrawn from the saturation cell using a greaseless, gas tight (2.500 ± 0.001 ml) Gilmont syringe. A 0.250 ml sample is injected to "wet" the frit. It is stripped and then four 0.500 ml samples are injected sequentially into the cell. The stripped gas is dried before entering the column. The gas is analyzed on a dual filament conductivity detector. Calibrations with pure gas are made dry before and wet after each series of runs.	(1) Methane. No information. (2) Water. No information.
	ESTIMATED ERROR: $\delta x_1/x_1$ = ± 0.015 (compiler)
	REFERENCES:

COMPONENTS:	ORIGINAL MEASUREMENTS:
(1) Methane; CH_4; [74-82-8] (2) Water; H_2O; [7732-18-5]	Rettich, T. R.; Handa, Y. P.; Battino, R.; Wilhelm, E. *J. Phys. Chem.* 1981, *85*, 3230-7.

VARIABLES: T/K = 275.46 - 328.15 p_1/kPa = 50.772 - 118.310	PREPARED BY: H. L. Clever

EXPERIMENTAL VALUES:

T/K	Pressure		Henry's Constant		Mol Fraction
	p_1/atm	p_1/kPa[a]	H/atm[b]	H/GPa[b]	$10^5 x_1$[c]
275.46	0.8063	81.694	22984	2.3288	4.3509
278.14	0.8455	85.673	24877	2.5207	4.0197
283.95	0.8556	86.693	29000	2.9385	3.4482
288.10	0.8491	86.036	31989	3.2413	3.1261
288.15	0.9032	91.520	32010	3.2434	3.1240
293.16	0.9051	91.709	35618	3.6090	2.8076
298.14	0.5079	51.462	39166	3.9685	2.5532
298.16	0.8531	86.442	39214	3.9734	2.5501
298.16	0.9470	95.959	39186	3.9705	2.5520
303.15	0.9701	98.296	42588	4.3152	2.3481
308.15	1.0309	104.458	45876	4.6483	2.1798
313.16	1.0344	104.810	48999	4.9648	2.0409
318.15	0.5011	50.772	51691	5.2376	1.9346
318.16	1.0904	110.482	51678	5.2362	1.9351
323.16	1.1471	116.234	54157	5.4874	1.8465
328.15	1.1676	118.310	56491	5.7239	1.7702

[a] Calculated by the compiler.

[b] Henry's constant evaluated at saturation pressure of the solvent from:

$$H/atm = \lim_{x_1 \to 0}(f_1/x_1) \text{ where } f_1 \text{ is the fugacity.}$$

[c] Mole fraction determined at unit fugacity of 1 atm (101.325 kPa).

AUXILIARY INFORMATION

METHOD/APPARATUS/PROCEDURE:	SOURCE AND PURITY OF MATERIALS:
The apparatus used was modelled after that of Benson, Krause, and Peterson (1). Degassed water is flowed in a thin film over the surface of a one dm^3 sphere to contact the gas. After equilibrium is attained the solution is sealed in a chamber of calibrated volume. The dissolved gas is extracted and its amount determined by a direct PVT measurement. A sample of the gas phase is analyzed in an identical manner. From the results, the saturation pressure of the solvent and Henry's constant are calculated in a thermodynamically rigorous manner, applying all non-ideal corrections.	(1) Methane. Airco. Both chemical pure grade, 99.0 minimum mole percent, and ultrahigh purity grade, 99.99 minimum mole percent were used with no detectable difference in results. (2) Water. Reverse osmosis, "house-distilled". Resistivity greater than 5 x 10^4 Ωm.
	ESTIMATED ERROR: $\delta H/H = \pm 0.0008$ $\delta T/K = \pm 0.01$
The authors smoothing equation, which fits their data to 0.06 %, is: $\ln H = 127.174 - 155.5756/(T/100 \text{ K})$ $- 65.2553 \ln(T/100 \text{ K})$ $+ 6.1698 (T/100 \text{ K})$. For H/Pa.	REFERENCES: 1. Benson, B. B.; Krause, D.; Peterson, M. A. *J. Soln. Chem.* 1979, *8*, 655.

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