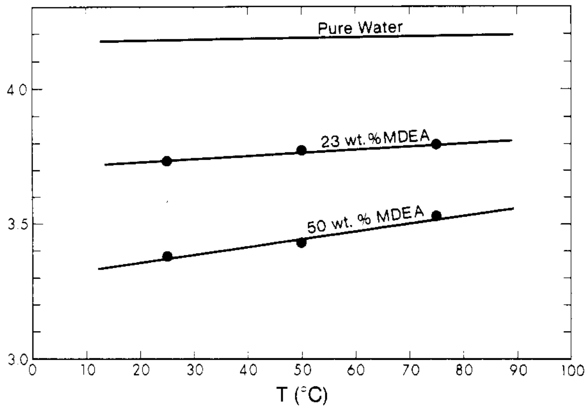

Figure 1. Effect of temperature on the heat capacity of MDEA solutions.

squares, with all points weighted equally. A linear dependence of heat capacity on temperature was found to reproduce the results within the precision of the data and gave the following equations:

23 wt % MDEA solution

$$C_p = 3.7085 + 0.00117t$$

50 wt % MDEA solution

$$C_p = 3.2975 + 0.00295t$$

(t in °C, C_p in kJ/(kg °C)).

These equations are plotted in Figure 1, together with the experimental data and the literature values for pure water. The heat capacities of MDEA solutions are almost the same as those of diethanolamine (DEA) solutions (*5*) at the same temperatures.

Registry No. Methyldiethanolamine, 105-59-9.

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Received for review July 9, 1982. Accepted November 23, 1982.

Solution Thermodynamics of Some Slightly Soluble Hydrocarbons in Water

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This paper summarizes a series of measurements of the solubilities of 14 hydrocarbons in water at temperatures ranging from 275 to 304 K using a generator-column-high-pressure liquid-chromatographic method. The compounds investigated were benzo[*a*]pyrene, benz[*a*]anthracene, anthracene, hexylbenzene, benzene, chrysene, pyrene, triphenylene, fluoranthene, 1-methylphenanthrene, 2-methylanthracene, phenanthrene, fluorene, and naphthalene. We have calculated values of $\Delta G°$, $\Delta H°$, and $\Delta C_p°$ at 298.15 K for the processes compound(liquid or solid) = compound(aqueous) using the model of Clarke and Glew. We have also calculated these same thermodynamic parameters from the information available on these compounds in the literature.

Introduction

For the past few years, this laboratory has been systematically measuring the aqueous solubilities and octanol-water partition coefficients of organic compounds (*1-4*) using a "generator" or "dynamic-coupled-column" liquid-chromatographic method. Because of the environmental importance of polycyclic aromatic hydrocarbons, we wish to summarize the solubility data on 14 of these compounds. Also, while we have already reported measurements on 12 of these compounds, the calculations of the thermodynamic parameters and a comparison with the existing literature data have not been done with an adequate thermodynamic model. The Gibbs energy changes ($\Delta G°$), the enthalpy changes ($\Delta H°$), and the heat capacity changes ($\Delta C_p°$) have been determined from the solubility measurements at different temperatures and compared with values calculated from measurements in the literature.

These measurements are also of interest for a variety of reasons: (1) since many of these compounds are carcinogenic, there is a need to know the degree to which they can enter the environment; (2) the thermodynamic information is of fundamental importance in understanding hydrophobic interactions and in calculating the transfer properties of solutes between various solvents (*5*); and (3) direct calorimetric measurements cannot presently yield meaningful values for substances having very low solubilities. The direct calorimetric measurement is also complicated (*6*) by the volatility problem which exists for many of these compounds.

Experimental Section

The experimental procedures have been described in detail elsewhere (*3*, *4*). All chemicals were obtained from commercial sources and were found to be at least 99 mol % pure by using chromatographic procedures.

Results and Discussion

The experimental results are summarized in Table I. For each compound, the experimentally determined solubilities as a function of temperature are reported. The molar masses used to calculate the mole fraction solubilities are given in

Table I. Experimental Results for Mole Fraction Solubilities x as a Function of Temperature T[a]

T/K	x	T/K	x
	Benzo[*a*]pyrene (252.32 g mol^{-1})		
283.15	0.3998×10^{-10}	298.15	0.1157×10^{-9}
288.15	0.5712×10^{-10}	303.15	0.1635×10^{-9}
293.15	0.8139×10^{-10}		
	Benz[*a*]anthracene (228.30 g mol^{-1})		
280.05	0.2359×10^{-9}	296.75	0.6313×10^{-9}
283.85	0.2983×10^{-9}	296.25	0.6605×10^{-9}
284.25	0.2849×10^{-9}	298.15	0.6794×10^{-9}
287.45	0.3780×10^{-9}	302.65	0.9785×10^{-9}
291.25	0.4403×10^{-9}	302.85	0.1002×10^{-8}
292.45	0.4995×10^{-9}		
	Anthracene (178.24 g mol^{-1})		
302.45	0.5781×10^{-8}	295.55	0.3760×10^{-8}
296.35	0.3821×10^{-8}	291.45	0.2941×10^{-8}
289.75	0.2537×10^{-8}	287.25	0.2245×10^{-8}
282.85	0.1637×10^{-8}	283.15	0.1769×10^{-8}
301.85	0.5630×10^{-8}	278.35	0.1284×10^{-8}
297.75	0.4387×10^{-8}		
	Hexylbenzene or 1-Phenylhexane (162.28 g mol^{-1})		
278.15	0.1022×10^{-6}	291.15	0.1022×10^{-6}
279.15	0.1022×10^{-6}	292.15	0.1046×10^{-6}
281.15	0.1020×10^{-6}	293.15	0.1053×10^{-6}
282.15	0.1005×10^{-6}	294.15	0.1060×10^{-6}
283.15	0.1003×10^{-6}	295.15	0.1057×10^{-6}
284.15	0.1029×10^{-6}	296.15	0.1090×10^{-6}
285.15	0.1032×10^{-6}	297.15	0.1104×10^{-6}
286.15	0.1020×10^{-6}	298.15	0.1108×10^{-6}
287.15	0.1026×10^{-6}	299.15	0.1108×10^{-6}
288.15	0.1007×10^{-6}	300.15	0.1123×10^{-6}
289.15	0.1019×10^{-6}	302.15	0.1127×10^{-6}
290.15	0.1009×10^{-6}		
	Benzene (78.12 g mol^{-1})		
273.35	0.4232×10^{-3}	290.05	0.4062×10^{-3}
279.35	0.4159×10^{-3}	291.75	0.4073×10^{-3}
284.15	0.4147×10^{-3}	298.15	0.4129×10^{-3}
287.15	0.4080×10^{-3}	298.95	0.4193×10^{-3}
	Triphenylene (228.30 g mol^{-1})		
301.35	0.6400×10^{-9}	387.95	0.2675×10^{-9}
300.45	0.6037×10^{-9}	285.15	0.2391×10^{-9}
293.65	0.3859×10^{-9}	281.15	0.2359×10^{-9}

T/K	x	T/K	x
	Chrysene (228.30 g mol^{-1})		
301.85	0.1744×10^{-9}	293.55	0.1105×10^{-9}
298.45	0.1491×10^{-9}	284.15	0.6313×10^{-10}
297.15	0.1326×10^{-9}	279.65	0.5603×10^{-10}
	Pyrene (202.26 g mol^{-1})		
303.05	0.1514×10^{-7}	287.45	0.6413×10^{-8}
298.65	0.1211×10^{-7}	282.65	0.5211×10^{-8}
294.35	0.9709×10^{-8}	277.85	0.4382×10^{-8}
291.85	0.8310×10^{-8}		
	Fluoranthene (202.26 g mol^{-1})		
303.05	0.2488×10^{-7}	286.35	0.9531×10^{-8}
297.75	0.1805×10^{-7}	281.25	0.7304×10^{-8}
292.85	0.1321×10^{-7}		
	1-Methylphenanthrene (192.26 g mol^{-1})		
303.05	0.3326×10^{-7}	287.15	0.1377×10^{-7}
300.05	0.2849×10^{-7}	282.05	0.1068×10^{-7}
297.25	0.2389×10^{-7}	279.75	0.8921×10^{-8}
292.35	0.1808×10^{-7}		
	2-Methylanthracene (192.26 g mol^{-1})		
304.25	0.3008×10^{-8}	287.05	0.1040×10^{-8}
300.15	0.2268×10^{-8}	283.95	0.8836×10^{-9}
296.25	0.1790×10^{-8}	282.25	0.7946×10^{-9}
291.45	0.1359×10^{-8}	279.45	0.6615×10^{-9}
	Phenanthrene (178.24 g mol^{-1})		
303.05	0.1240×10^{-6}	285.65	0.5175×10^{-7}
297.45	0.9653×10^{-7}	283.15	0.4730×10^{-7}
294.15	0.8248×10^{-7}	281.65	0.4275×10^{-7}
293.15	0.7955×10^{-7}	277.15	0.3649×10^{-7}
288.15	0.6075×10^{-7}		
	Fluorene (166.23 g mol^{-1})		
304.25	0.2436×10^{-6}	291.15	0.1304×10^{-6}
300.15	0.2000×10^{-6}	286.35	0.1048×10^{-6}
297.15	0.1751×10^{-6}	279.75	0.7786×10^{-7}
	Naphthalene (128.19 g mol^{-1})		
300.15	0.4799×10^{-5}	288.25	0.3019×10^{-5}
298.15	0.4485×10^{-5}	286.55	0.2863×10^{-5}
296.55	0.4142×10^{-5}	284.65	0.2703×10^{-5}
292.45	0.3624×10^{-5}	281.35	0.2376×10^{-5}

[a] Molar masses (in parentheses) are based upon the relative atomic masses given in ref *23*.

parentheses after each compound name in this table.

For the process

$$\text{compound(liquid or solid)} = \text{compound(aqueous)} \qquad \text{(A)}$$

the standard-state Gibbs energy change is given by

$$\Delta G^\circ = -RT \ln fx \qquad (1)$$

where R is the gas constant (8.31441 J K^{-1} mol^{-1}), T is the absolute temperature, x is the solubility on the mole fraction scale, and f is the mole fraction activity coefficient. The activity of the solute is given by the quantity fx and the standard state is chosen such that both f and the activity of water approach unity as the mole fraction of the solvent (water) approaches unity. In calculating values of ΔG°, we have assumed that the activity coefficients of the solutes are unity at these very low experimental concentrations.

In order to represent the temperature dependency of the solubility, we have used the expression recommended by Clarke and Glew (*7*) which is based upon a Taylor series expansion of the heat capacity change at a reference temperature θ:

$$R \ln x = -\Delta G^\circ_\theta/\theta + \Delta H^\circ_\theta[1/\theta - 1/T] + \Delta C_p{}^\circ_\theta[\theta/T - 1 + \ln(T/\theta)] \qquad (2)$$

If the accuracy of the measurements justifies it, additional terms may be added to eq 2 which allow for $\Delta C_p{}^\circ$ to vary with the temperature. The first of these additional terms is $(\theta/2)\cdot(\mathrm{d}\Delta C_p{}^\circ/\mathrm{d}T)_\theta[(T/\theta) - (\theta/T) - 2\ln(T/\theta)]$. This model has the advantage that the adjustable parameters in it are the desired thermodynamic properties. The results of fitting (*8*) the experimental data to this model are given in Table II using a reference temperature θ of 298.15 K. We have found that additional terms which include $\mathrm{d}\Delta C_p{}^\circ_\theta/\mathrm{d}T$, etc., were not needed to represent any of the measurements. For benzo[*a*]pyrene, a meaningful value of $\Delta C_p{}^\circ_\theta$ could not be calculated; therefore, only ΔG°_θ and ΔH°_θ are reported for that compound.

In addition to the above model, we also considered the equation

$$\ln x = A + B/T + C \ln T \qquad (3)$$

where A, B, and C are now the adjustable parameters. ΔG°_θ, ΔH°_θ, $\Delta C_p{}^\circ_\theta$ may be calculated from eq 3 by calculating $\Delta S^\circ = -(\partial \Delta G^\circ/\partial T)_p$ and from that ΔH° and $\Delta C_p{}^\circ$. We have found that this equation was not as well suited to the problem as eq 2, as evidenced by the calculations which showed A, B, and C to be highly correlated, and a meaningful variance-covariance matrix could not be obtained for the calculation of the standard deviations of ΔG°, ΔH°, and $\Delta C_p{}^\circ$. This was not the case when eq 2 was used. Values of ΔG°_θ, ΔH°_θ, and $\Delta C_p{}^\circ_\theta$ calculated by using eq 3 were found, with the sole exception of naphthalene, to be very close to the values calculated by using eq 2. Parenthetically we note that, for naphthalene, the difference (eq 2 minus eq 3) in ΔH°_θ was 2.2 kJ mol^{-1}. In view of the difficulty of obtaining a meaningful vari-

Table II. Thermodynamic Parameters with their 95% Confidence Limits for the Process Compound(liquid or solid) = Compound(aqueous) at 298.15 K[a]

compd	ΔG°/(kJ mol^{-1})	ΔH°/(kJ mol^{-1})	ΔC_p°/(kJ mol^{-1} K^{-1})	$s(\Delta G^\circ)$/(kJ mol^{-1})	$s(\Delta H^\circ)$/(kJ mol^{-1})	$s(\Delta C_p^\circ)$/(kJ mol^{-1} K^{-1})
benzo[*a*]pyrene	56.71 ± 0.041	50.3 ± 1.3		0.013	0.42	
benz[*a*]anthracene	52.21 ± 0.008	50.0 ± 5.9	0.82 ± 0.81	0.038	2.6	0.35
anthracene	47.69 ± 0.051	47.2 ± 3.5	0.41 ± 0.42	0.022	1.5	0.18
hexylbenzene	39.70 ± 0.020	7.6 ± 1.4	0.51 ± 0.16	0.010	0.67	0.075
benzene	19.31 ± 0.034	2.1 ± 1.9	0.22 ± 0.15	0.013	0.73	0.058
chrysene	56.21 ± 0.17	45.4 ± 16.	1.0 ± 1.9	0.054	4.9	0.58
pyrene	45.29 ± 0.095	41.6 ± 5.8	0.77 ± 0.64	0.034	2.1	0.23
triphenylene	53.09 ± 0.20	56.9 ± 15.	2.9 ± 2.1	0.063	4.6	0.65
fluoranthene	44.16 ± 0.096	45.0 ± 6.0	0.84 ± 0.85	0.022	1.4	0.20
1-methylphenanthrene	43.36 ± 0.075	41.2 ± 5.0	0.30 ± 0.64	0.027	1.8	0.23
2-methylanthracene	49.62 ± 0.047	45.5 ± 2.6	0.47 ± 0.34	0.018	1.0	0.13
phenanthrene	39.98 ± 0.083	36.7 ± 4.7	0.29 ± 0.50	0.034	1.9	0.21
fluorene	38.47 ± 0.024	35.1 ± 1.3	0.33 ± 0.18	0.007	0.41	0.056
naphthalene	30.55 ± 0.037	28.7 ± 3.4	0.31 ± 0.44	0.014	1.3	0.17

[a] Standard deviations *s* of the parameters; ΔC_p° is assumed to be a constant over the temperature range of the measurements. Calculations are based on the experimental results given in Table I.

Table III. Thermodynamic Parameters with Their 95% Confidence Limits for the Process Compound(liquid or solid) = Compound(aqueous) at 298.15 K[a]

ΔG°/(kJ mol^{-1})	ΔH°/(kJ mol^{-1})	ΔC_p°/(kJ mol^{-1} K^{-1})	$s(\Delta G^\circ)$/(kJ mol^{-1})	$s(\Delta H^\circ)$/(kJ mol^{-1})	$s(\Delta C_p^\circ)$/(kJ mol^{-1} K^{-1})	ref
		Anthracene				
47.57 ± 0.10	41.9 ± 6.8	1.6 ± 1.1	0.47	3.0	0.48	*10*
46.89 ± 0.53	51.1 ± 12.	−0.011 ± 0.41	0.24	5.6	0.18	*11*
		Benzene				
19.391 ± 0.012	2.27 ± 0.21	0.253 ± 0.016	0.0056	0.097	0.0076	*12*
19.291 ± 0.018	2.49 ± 0.34	0.266 ± 0.057	0.074	0.13	0.022	*13*
19.397 ± 0.012	2.46 ± 0.44	0.235 ± 0.027	0.0053	0.20	0.012	*14*
	0.46[b]					*15*
	0.80[b]					*16*
	2.08[b]					*17*
		0.220 ± 0.005[b]				*17*
		0.225 ± 0.005[b]				*18*
		0.224 ± 0.020[b]				*19*
		0.416[b]				*20*
		0.238[b]				*21*
		Pyrene				
45.28 ± 0.042	46.9 ± 2.9	−0.029 ± 0.63	0.018	1.2	0.27	*10*
45.03 ± 0.12	36.69 ± 3.2	0.44 ± 0.13	0.051	1.4	0.056	*11*
		Phenanthrene				
39.60 ± 0.071	37.8 ± 3.8	0.25 ± 0.60	0.029	1.5	0.25	*10*
39.62 ± 0.18	39.1 ± 5.4	0.28 ± 0.22	0.083	2.5	0.10	*11*
		Fluorene				
38.26 ± 0.15	37.0 ± 4.5	0.28 ± 0.18	0.066	2.0	0.083	*11*
		Naphthalene				
30.28 ± 0.030	29.9 ± 0.63	0.35 ± 0.071	0.014	0.29	0.033	*13*
30.65 ± 0.030	21.8 ± 1.7	−0.12 ± 0.28	0.013	0.73	0.12	*10*
30.55 ± 0.60	23.8 ± 2.1	0.54 ± 0.096	0.025	0.93	0.042	*11*

[a] Standard devistions *s* of the parameters; ΔC_p° is assumed to be a constant over the temperature range of the measurements. Calculations are based on the experimental data in the cited references. [b] Calorimetric results.

ance-covariance matrix using eq 3, we have exclusively used eq 2 to calculate the values of the thermodynamic parameters and their standard deviations given in Tables II and III.

It is noteworthy that the statistical uncertainties given in Table I, which correspond to the 95% confidence limit, show that, while the ΔG° values are very precise, the values of ΔH° are much less so, and the values of ΔC_p° have very little real meaning. These latter values are included in this table largely for the sake of completeness of presentation of the available information. The ΔC_p° value for hexylbenzene is the most precise of these values reported and we conclude that, if the precision of this method could be improved by a factor of 2 or 3, reasonably meaningful values ($\approx \pm 10\%$) of ΔC_p° could be obtained by taking a sufficient number of closely spaced measurements over an appropriate temperature interval (*9*). King (*9*) has emphasized that both good precision and closely spaced data are required to obtain very precise values of ΔH° and, particularly, ΔC_p°.

We have also examined the information in the literature on these compounds (see Table III). The results fall into two general categories: (i) solubilities measured as a function of temperature and (ii) calorimetric measurements yielding either ΔH° and/or ΔC_p°. The values of ΔG°, ΔH°, and ΔC_p°, their uncertainties, and the standard derivations given in Table III were calculated from the experimental solubility data given in the references cited using eq 2 and the same procedures used for the calculations performed earlier which yielded the quantities given in Table II. The quantities and uncertainties given in Table III for the calorimetric results are the values given by the authors of the respective papers.

The results obtained for benzene are particularly interesting. The measurements of Arnold et al. (*12*), Bohon and Claussen

(*13*), and Franks et al. (*14*) involved the determination of the solubility of benzene in water as a function of temperature. These three very precise investigations are in very good agreement with each other. The calorimetric measurements of Krishnan and Friedman (*15*) were performed in a calorimeter open to the atmosphere and contain a very large systematic error due to vaporization effects (*6*). The results of Reid, Quickenden, and Franks (*16*), even though obtained in a closed system, may also have a large systematic error since it is not clear what corrections were taken into account for the volume change accompanying the mixing of benzene and water. The three values of ΔH° obtained from an analysis of the temperature dependence of the solubility data are in agreement with the calorimetric ΔH° of 2.08 kJ mol^{-1}, albeit they are all higher. The $\Delta C_p{}^\circ$ value obtained from Jolicoeur et al. (*20*) was redetermined by the same investigators (*21*) and found to be in error. Thus, the calorimetric $\Delta C_p{}^\circ$ values appear to be in agreement with each other and with two of the values of $\Delta C_p{}^\circ$ obtained from an analysis of the temperature dependence of the solubilities. We have found, however, that the calculated value of $\Delta C_p{}^\circ$ is sensitive to the inclusion of a $\mathrm{d}\Delta C_p{}^\circ/\mathrm{d}T$ term in the model. For example, the inclusion of the first of the additional terms to eq 2 results in a value of $\Delta C_p{}^\circ = 0.322 \pm 0.090$ J mol^{-1} K^{-1} as compared to a value of 0.235 ± 0.027 J mol^{-1} K^{-1} for the data of Franks et al. (*14*). The results obtained in this laboratory for benzene are seen to be in fortuitously good agreement with the best of the literature results.

A comparison of the results in Tables II and III indicates several interesting items. Our ΔG° value for anthracene is in agreement with the value obtained from Schwarz (*10*), but not with that obtained from Wauchope and Getzen (*11*). Our ΔH° values (Table II) are in agreement with all of the ΔH° values from the literature, with the exception of naphthalene where our ΔH° value is close to the precise value of Bohon and Claussen (*13*), but not to the value obtained from Schwarz (*10*) or from Wauchope and Getzen (*11*). We also note that most of the $\Delta C_p{}^\circ$ values obtained from the temperature dependence of the solubilities in Table III have large uncertainties. Further experiments which resolve these few discrepancies would be useful.

All earlier solubility measurements (*10*–*14*) were obtained by using a "shake-flask" technique where the solute is equilibrated with water and then analyzed in solution. May et al. (*2*, *3*) and DeVoe et al. (*1*) have pointed out the possible systematic errors inherent in this type of procedure: colloidal dispersions, adsorption on surfaces, and possible loss of volatile solutes to the atmosphere. The generator-column method does not suffer from these drawbacks and, if the solute does not absorb in the ultraviolet region, the method can still be used for measurements by using a gas-chromatographic analysis following extraction of the solute.

We have used the mole fraction scale to obtain values of ΔG° and then taken derivatives with respect to temperature. If we had used the same procedure with ΔG° values based on the molarity scale and not corrected for the thermal expansion of the solution, we would have made a systematic error in the calculations (*22*). We have assumed that the mole fraction activity coefficients (f) of the solutes were unity when we calculated values of ΔG° using eq 1 and that the solid or liquid phases in equilibrium with the solution are not hydrated. While both of these assumptions are subject to future investigation, we do feel safe with the first assumption in that it would require a very high degree of association among the solute particles to cause the activity coefficient to deviate any real amount from unity at these very low experimental concentrations.

Acknowledgment

We thank Janet Donaldson of the NBS Boulder Laboratory for her useful discussions on the use of STATLIB.

Registry No. Benzo[*a*]pyrene, 50-32-8; benz[*a*]anthracene, 56-55-3; anthracene, 120-12-7; hexylbenzene, 1077-16-3; benzene, 71-43-2; chrysene, 218-01-9; pyrene, 129-00-0; triphenylene, 217-59-4; fluoranthene, 206-44-0; 1-methylphenanthrene, 832-69-9; 2-methylanthracene, 613-12-7; phenanthrene, 85-01-8; fluorene, 86-73-7; naphthalene, 91-20-3.

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Received for review August 10, 1982. Accepted January 11, 1983.