Diffusion Coefficients of Substituted Benzenes at High Dilution in Water

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The interdiffusion coefficient D_{aw} of six hydrophobic benzene derivatives (a = chlorobenzene, 1,2-dichlorobenzene, 1,4-dichlorobenzene, bromobenzene, 1,2-dibromobenzene, 1,4-dibromobenzene) in aqueous solution (w) at 25 °C is measured at different concentrations below 10^{-5} solute mole fractions using the Taylor dispersion technique. On the basis of the experimental uncertainty around 4%, a distinct concentration dependence is not recognizable. Analysis of the interdiffusion coefficients with the Stokes−Einstein equation shows greater D_{aw}^{∞} values than expected from the limit of a complete solute−solvent contact ("stick limit"), and in addition substance-specific deviations. Chemical engineering correlations for D_{aw}^{∞} presently available from the literature yield unsatisfactory results for this class of predominantly hydrophobic compounds.

Introduction

The long-term distribution of chemicals in the environment is determined to a large degree by passive transport processes. Among the three basic environmental compartments soil, water, and air, the latter two form the major abiotic dispersion pathways of chemicals, and in unsaturated soil zones it is the pore water that governs vertical leaching of contaminants to the groundwater by advection and diffusion phenomena (cf. Neely and Blau (1985) and Ballschmiter (1992)). The latter is also of particular importance for the water−air transfer of organic compounds. A direct way to study the mobility of chemicals in aqueous systems is the determination of diffusion coefficients. Persistent organic contaminants, however, often have rather low water solubilities, which makes it particularly difficult to perform conventional diffusion measurements due to detection limits.

In addition to the movement through different abiotic compartments, lipophilic organic contaminants may also be taken up by organisms, which then leads to the bioaccumulation of such chemicals with its potential hazardous effects for the receptor species. It is now well known that, with aquatic organisms, uptake and bioconcentration of xenobiotics is most often driven by passive diffusion processes from the water body into living tissues (Gobas et al., 1986; Connell, 1988).

A recent investigation of the bioconcentration of halogenated benzenes in fish has shown that the uptake rates of bromobenzenes are larger than those of the smaller chlorobenzene counterparts (Sijm et al., 1993). These findings are somewhat surprising in view of the fact that the diffusion velocity usually decreases with molecular size. Thus, smaller compounds are expected to be taken up faster by aquatic organisms, if the uptake process is indeed governed by passive diffusion processes. This leads to the question of whether the halogenated benzenes show diffusion coefficients that decrease with increasing molecular size. Another possibility to derive a physicochemical explanation for the relative order of the bioconcentration uptake rates mentioned above would be a detailed analysis of the compound transfer from water to a lipophilic medium.

In the present paper, we analyze the first question by experimentally determining the interdiffusion coefficients of the benzene derivatives in water using the Taylor dispersion method. Due to the low water solubilities of the hydrophobic compounds, a further question is whether this method is applicable for solute mole fractions around and below 10^{-5}. The latter is of particular interest for future practical applications of the Taylor dispersion technique, which is known to have an averge error range of up to 4% (Baldauf and Knapp, 1983), but is much less time-consuming than, e.g., the interferometric method with a typical error around 0.1%. In order to address this latter point, additional compounds (urea, $KClO_3$, KNO_2, benzene, toluene, acetone) are included in the testing scheme, for which reliable reference data are available from the literature. Finally, it will be analyzed whether the experimentally determined diffusion coefficients agree with the Stokes−Einstein relationship, and whether they can be modeled with well-known chemical engineering correlations.

Experiment and Procedure

The *Taylor dispersion apparatus* used for the diffusivity measurements consists of the following components: a HPLC double piston pump (Merck-Hitachi Model L-6000A) which allows a nearly pulsation-free flow of carrier liquid, a mechanical pulse damping (ProMinent Model 92.00.40.3), a six-way injection valve (Knauer Model A 0258) with a 20 μL sample loop (which corresponds to a length of the injected sample pulse, h = 10.2 cm), a dispersion capillary of stainless steel (length L = 11.30 m; i.d. = 0.50 mm, coiled in a 0.5 m diameter helix), a flow-through differential refractometer (Knauer Model A 0298), a digital voltmeter (Prema Model DMM 6001), and a computer coupled online via an IEEE interface to the voltmeter. A constant temperature of (25.0 ± 0.05) °C around the dispersion capillary and the refractometer cuvettes was maintained with a water thermostat (Julabo Model FP 40). The effective radius of the apparatus (exceeding the technical radius because of the volume of the detector tubing, cuvettes, and fittings) was determined to be R = 0.252 mm in calibration experiments by using an interpolation equation for interdiffusion coefficients in aqueous KCl solutions

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(Woolf and Tilley, 1951) recommended by Kestin and Wakeham (1988). The volumetric flow rate of the carrier liquid was typically 4 mL·h^{-1} (which corresponds to a linear velocity of $u = 33.6$ cm·min^{-1}). The parameters of the apparatus and flow regime meet the requirements (see, for example, Baldauf and Knapp (1983)) to fulfill the boundary conditions under which Taylor (1953) solved the dispersion equation. A Reynolds number of 2.8 indicates laminar-flow conditions in our experiments. The radial concentration gradient created by convection is eliminated by radial diffusion at the same rate because the squared length-to-radius ratio of the capillary ($L^2/R^2 = 2.04 \times 10^9$) is 20 times greater than the Peclet number ($Pe \leq 0.97 \times 10^8$). The effect of axial diffusion can be neglected because $Pe \gg 6.93(L/R) = 0.31 \times 10^6$, and finally, the error in assuming a δ injection instead of the real square-pulse injection is less than 1% due to a ratio of $h/L = 0.009$. Adsorption effects of the organics at the inner wall (cf. Loh et al. (1993)) can be excluded in our measurements because of our nearly symmetric dispersion peaks and reproducible retention times over a long time period (with several cleaning operations for the flow system).

The numerical analysis of recorded dispersion peaks was done by using the same procedure described earlier (Thiel et al., 1993) apart from the use of another Marquardt routine (Press et al., 1990) in our TurboPascal program package.

The *substances* under investigation were used in good commercial quality (purchased from Merck, Darmstadt) without further purification. Water was deionized before use (the water conductivity did not exceed 1 μS·cm^{-1}.) Water had to pass a 3 μm pore filter before it entered the dispersion capillary.

Aqueous solutions of benzene and the halogenated derivatives were prepared using the following procedure: An amount of substance sufficient to yield a saturated solution was added to 600 mL of water, and the mixture was stirred for 2 h at 50 °C (or at a temperature slightly above the melting point of the substance), cooled to room temperature, and stirred once again for 24 h. After separation the aqueous phase was filtrated two times. The stock solution was then used to prepare lower concentrated solutions by dilution with water in volumetric flasks. Potential formation of microdispersion can be excluded, as we did not observe any disturbance in the operation of the differential refractometer due to light scattering throughout all experiments. The solutions of acetone, urea, and potassium salts in water were prepared gravimetrically.

The *dispersion measurements* were carried out with pure water as the carrier liquid. Starting with the nearly saturated aqueous solution, at least two different, more diluted solutions of each substance under investigation were used as samples. The solute mole fractions in these sample solutions are typically between 10^{-4} and 10^{-6} depending on the individual water solubility (see Table 1). For each solution (concentration level) the measurements (mostly three sample injections which were averaged) were repeated two to five times (at different days). Statistical tests confirm the correctness of overall averaging of related experimental data.

The determined interdiffusion coefficients refer to solute concentrations C in the region between the injected sample and the carrier solution that are calculated by means of the following equation (Baldauf 1981):

$$C_{\text{average}} = C_{\text{carrier}} + (C_{\text{sample}} - C_{\text{carrier}})\,\frac{h}{2}\left(\frac{\pi u R^2 L}{48D}\right)^{-1/2} \qquad (1)$$

The relevant parameter values (pulse length h, linear velocity u, capillary radius R, capillary length L) are specified above.

Table 1. Water solubilities S_w, Mole Fracitons X_a at Saturated Concentration, and Calculated van der Waals Radii r_a of Hydrophobic Solutes a (cf. Experiment and Procedure Section)

substance	S_w/(mmol·L^{-1})	X_a	r_a/(10^{-10} m)
benzene	22.9[a]	4×10^{-4}	2.55
toluene	6.28[a]	1×10^{-4}	2.71
chlorobenzene	4.43[a]	8×10^{-5}	2.69
1,2-dichlorobenzene	0.612[b]	1×10^{-5}	2.82
1,4-dichlorobenzene	0.204[c]	4×10^{-6}	2.82
bromobenzene	2.62[a]	5×10^{-5}	2.75
1,2-dibromobenzene	0.316[b]	6×10^{-6}	2.92
1,4-dibromobenzene	0.085[c]	2×10^{-6}	2.93

[a] Wasik et al. (1983). [b] Miller et al. (1984). [c] Horvath and Getzen (1985).

Comparison of diffusion coefficients with the Stokes–Einstein relationship requires knowledge of molecular van der Waals radii r_a of solute a in aqueous solution, which can be derived from molecular volumes MV according to

$$r_a = ((3/4\pi)(\text{MV}))^{1/3} \qquad (2)$$

under the simplifying assumption of spherical shape. Estimation of MV through experimental molar volumes divided by the Loschmidt number would imply the unlikely assumption that the solute molar density is identical for the pure organic phase and the diluted aqueous solution. Thus, *a quantum chemical approach* was selected to calculate molecular van der Waals radii in aqueous solution with the COSMO modification (Klamt and Schüürmann, 1993) of the semiempirical AM1 Hamiltonian (Dewar et al., 1985) using MOPAC93 (1993), after initial structure generation with the SYBYL modeling package (1992). Corresponding MV values using standard atomic van der Waals radii were calculated with MOLSV (Smith, 1985).

Results and Discussion

Experimentally determined interdiffusion coefficients D_{aw} for urea, KClO$_3$, KNO$_2$, benzene, toluene, and acetone are listed in Table 2 together with reference values D_{aw}^{ref} from the literature and corresponding relative differences ΔD. Except for acetone and benzene, ΔD is below the above-mentioned typical error range of ±4%.

According to theory, interdiffusion coefficients should converge to intradiffusion coefficients at the limit of very low solute concentrations (Tyrell and Harris, 1984), as is the case with our present results for urea. However, our interdiffusion coefficient of acetone at 10^{-5} mole fraction deviates by −9.3% from the intradiffusion coefficient using the ^{14}C technique in water (Mills and Hertz, 1980). On the other hand, our experimental value is close to a D_{aw}^{∞} value of 1.16×10^{-9} m^2·s^{-1} reported by Cussler (1984), but unfortunately there are no details given about the concentration series from which this value was derived. The apparent discrepancy cannot be resolved at this moment.

With benzene, a deviation of −6.2% is observed from one reference value at 4×10^{-5} mole fraction: However, this reference value was generated by interpolation using an Arrhenius plot of data at other temperatures, and our measured value is indeed covered by the 95% confidence interval of this Arrhenius plot. The overall relatively good agreement with reference data suggests that application

Table 2. Measured Interdiffusion Coefficients D_{aw} and Their Standard Deviations at the Effective Mole Fraction X_a in Water at 25 °C and the Respective Number n of Individual Experiments in Comparison to Reference data D_{aw}^{ref}

substance	X_a	n	$D_{aw} \times 10^9$/ $(m^2 \cdot s^{-1})$	$D_{aw}^{ref} \times 10^9$/ $(m^2 \cdot s^{-1})$	ΔD^g/%
urea	3×10^{-6}	10	1.36 ± 0.01	1.38^a	−1.4
	3×10^{-5}	10	1.33 ± 0.01	$1.38^{a,b}$	−3.6
$KClO_3$	9×10^{-4}	12	1.74 ± 0.02	1.68^c	+3.6
KNO_2	9×10^{-4}	12	1.83 ± 0.02	1.82^c	+0.5
benzene	1×10^{-5}	15	1.13 ± 0.06	1.10^d	+2.7
	4×10^{-5}	15	1.06 ± 0.03	1.13^e	−6.2
toluene	1×10^{-5}	15	0.93 ± 0.04	0.93^e	±0.0
acetone	1×10^{-5}	8	1.17 ± 0.01	1.29^f	−9.3

[a] Calculated interdiffusion data from interferometric measurements (Gosting and Akeley, 1952) and intradiffusion coefficient (Albright and Mills, 1965). [b] Experimental interdiffusion coefficient from the Taylor dispersion measurement at the same concentration (Harris, 1991). [c] Experimental data from interferometric measurements at the same concentrations (Daniel and Albright, 1995). [d] Experimental interdiffusion coefficient from the Taylor dispersion measurement (Tominaga et al., 1984) at molar fraction $X_a \approx 5 \times 10^{-7}$ and 2×10^{-6}. [e] Interpolated interdiffusion data from temperature dependent data at comparable concentration measured using the capillary method (Bonoli and Witherspoon, 1968). [f] Intradiffusion coefficient from ^{14}C tracer measurements (Mills and Hertz, 1980) at infinite dilution. [g] The difference between the experimental and reference data ΔD was calculated by $\Delta D = (D_{aw} - D_{aw}^{ref})/D_{aw}^{ref} \times 100\%$.

Table 3. Interdiffusion coefficients D_{aw} of Halogenated Benzenes Measured at Different Effective Mole Fractions X_a in Aqueous Solution (Where X_a Was Calculated According to Eq 1) at 25 °C[a]

substance	X_a	$D_{aw} \times 10^9/(m^2\ s^{-1})$	n
chlorobenzene	2×10^{-6}	1.04 ± 0.04	11
	4×10^{-6}	1.00 ± 0.02	11
	8×10^{-6}	0.98 ± 0.03	15
1,2-dichlorobenzene	2×10^{-7}	0.94 ± 0.08	15
	5×10^{-7}	0.89 ± 0.06	15
	9×10^{-7}	0.87 ± 0.02	15
1,4-dichlorobenzene	2×10^{-7}	0.99 ± 0.09	22
	4×10^{-7}	0.93 ± 0.09	20
bromobenzene	1×10^{-6}	1.03 ± 0.09	15
	2×10^{-6}	0.99 ± 0.03	15
	5×10^{-6}	0.96 ± 0.02	15
1,2-dibromobenzene	1×10^{-7}	0.87 ± 0.13	15
	3×10^{-7}	0.78 ± 0.04	15
	5×10^{-7}	0.76 ± 0.03	15
1,4-dibromobenzene	2×10^{-7}	0.83 ± 0.11	29

[a] The values are given together with experimental standard deviations and the respective number n of individual experiments.

of the Taylor dispersion technique leads to reliable interdiffusion coefficients in concentration ranges typical for environmental exposure.

The experimentally determined interdiffusion coefficients D_{aw} for the halogenated benzenes (a) in water (w) at all effective concentrations are listed in Table 3. The solute concentration ranges analyzed are substance-specific and go for each compound from the solubility limit to the detection limit of the refractometer. The smallest measurable solute concentration was observed for 1,2-dibromobenzene (effective mole fraction of 1×10^{-7}). As can be seen from Table 3, variations in the mole fraction of 1 order of magnitude correspond to changes in the diffusion coefficient of up to $0.1 \times 10^{-9}\ m^2 \cdot s^{-1}$, which is in the same range as the experimental standard deviations at the lowest solute concentrations. On the basis of the uncertainty of ca. 4% and the reproducibility as indicated in Table 3, the minor variations among the D_{aw} values for different solute concentrations are statistically not significant.

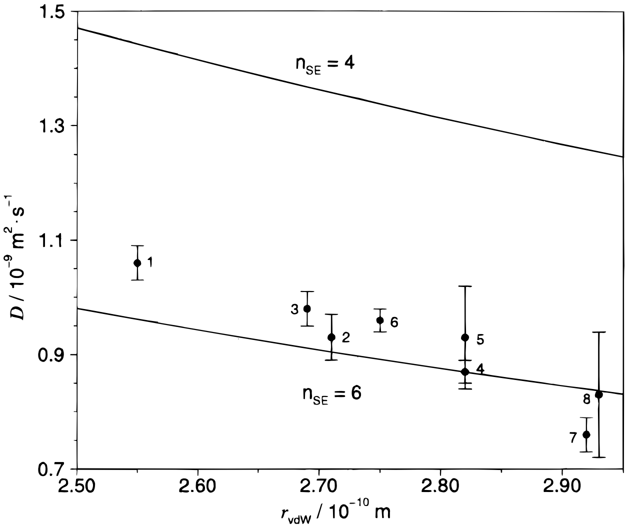

Figure 1. Experimental interdiffusion coefficient D_{aw} plotted against the calculated molecular van der Waals radius r_a for nine benzene compounds (1, benzene; 2, toluene; 3, chlorobenzene; 4, 1,2-dichlorobenzene; 5, 1,4-dichlorobenzene; 6, bromobenzene; 7, 1,2-dibromobenzene; 8, 1,4-dibromobenzene). The solid lines represent the two limiting cases of the Stokes−Einstein equation; cf. eq 3.

Our experimental results can be compared with theoretical values calculated by the Stokes−Einstein equation

$$D_{ab}^{\infty} = k_B T / n_{SE} \pi \eta_b r_a \tag{3}$$

that relates the interdiffusion coefficient D_{ab}^{∞} of compound a in solvent b at infinite dilution to the van der Waals radius r_a of the solute (assumed to be spherical) and the dynamic viscosity η_b of the solvent (k_B is the Boltzmann constant, T the absolute temperature, and n_{SE} the Stokes−Einstein number). Within this hydrodynamic model, the assumption of a complete contact between the solute surface and the solvent, the so-called stick limit, corresponds to $n_{SE} = 6$, and the absence of any direct solvent−solute contact, the slip limit, is represented by $n_{SE} = 4$ (cf. Bird et al. (1960)).

In Figure 1, estimates of D_{aw}^{∞} based on our measurements for all eight benzene compounds are plotted against calculated molecular van der Waals radii (see Table 1) together with lines representing the above-mentioned two limiting cases of eq 3. For the hydrophobic compounds, D_{aw}^{∞} is set equal to the D_{aw} values with the lowest experimental error at the solubility limit (solute concentration). It is seen that with this set of hydrophobic compounds, the solute−solvent interaction is rather close to the stick limit of an (almost) complete contact.

Current chemical engineering correlations derived from modifications of the Stokes−Einstein equation to estimate D_{aw}^{∞} (cf. Reid et al. (1987), Rutten (1992), and Siddiqi and Lucas (1987)) yield unsatisfactory results for this class of predominantly hydrophobic compounds. The best results accounting for 79% of the total variance ($r^2 = 0.79$) are obtained with the Wilke−Chang equation, which makes use of the compounds' molar volumes at the boiling point estimated by the Le Bas method. It follows that application of current group contribution schemes cannot be recommended for more hydrophobic compounds.

Summary and Conclusions

The results indicate that the Taylor dispersion technique offers a promising tool to measure aqueous diffusion coefficients in concentration ranges around and below 10^{-5}

solute mole fraction. It is suggested that replacement of the differential refractometer by a more sensitive detection unit (cf. for example Tominaga et al. (1984)) will allow direct measurements of the diffusion coefficient still closer to D_{aw}^{∞} and for a wider range of compounds.

For the present set of compounds including benzene and more hydrophobic benzene derivatives, the relative order of diffusion coefficients is inversely related to molecular size, and in that respect agrees with expectation. Comparison with the Stokes–Einstein relationship indicates that the solute–water interaction corresponds qualitatively to the stick limit. Current chemical engineering correlations yield substantial deviations from the experimental values and should not be applied to hydrophobic compounds without further analysis.

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