Distribution of nonionic compounds between the gas phase and sodium dodecyl sulfate micelles

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Regression analysis of the influence of the structure of nonionic organic compounds on their partition coefficients (K_{gm}) between the sodium dodecyl sulfate (SDS) micelles and the gas phase in aqueous micellar SDS solutions was performed using the descriptors characterizing the mechanisms of interaction between these compounds and SDS micelles. For alkanes and compounds with hydroxyl groups, as well as for aromatic and aliphatic compounds without hydroxyl groups, the $\log(K_{gm})$ values are described by separate regression equations, which is due to specific features of intermolecular interactions for these types of organic compounds in the case of their solubilization by SDS micelles.

Key words: sodium dodecyl sulfate, micelles, partition coefficients, organic compounds.

Aqueous micellar solutions of surfactants (Surf) can solubilize various chemical compounds in micelles. Inhomogeneity of the internal structure of micelles favors solvation in the micellar pseudophase of polar and nonpolar molecules. According to modern concepts, a micelle consists of a nonpolar hydrocarbon core, a polar surface layer formed by hydrated head groups and nearest counterions, and a region of intermediate polarity between the core and the surface layer.[1] In actual micelles, these regions have no well-defined boundaries and the gradient of polarity continuously decreases from the surface to the center of the micelles. Different types of microenvironments for different structural types of compounds can result in different mechanisms and intensities of the intermolecular interactions determining their solubilization.[1,2]

The solubilization equilibrium is characterized by the partition coefficients of substances, $K_x = x_m/x_w$ (x_m and x_w are the equilibrium mole fractions of solute in the micellar pseudophase and water, respectively) or $K_{wm} = [C_m]/[C_w]$ ($[C_m]$ and $[C_w]$ are respectively the molar concentrations of the solute in micelles and in water, referred to the volumes of the phases), as well as by the binding constants $K_s = [C_m]/([C_w] \cdot [C_s])$ ($[C_s]$ is the molar concentration of the Surf associated into micelles[2,3]). At low concentrations of Surf in water, the above-mentioned coefficients are related by linear relationships[2]

$$K_{wm} = K_s/v_s \tag{1}$$

$$K_{wm} = K_x/(v_s \cdot n_w), \tag{2}$$

where n_w is the number of moles of water in the standard state and v_S is the molar volume of the Surf associated into micelles.

Solubilization of organic compounds from aqueous solutions by sodium dodecyl sulfate (SDS) micelles was studied in most detail. The K_{wm}, K_x, and K_s values for many organic compounds were analyzed using linear dependences between the free solvation energies of the substances and the empirical parameters (descriptors) characterizing the solvation mechanisms.[4–6] The following regression equation (RE) characterizing different contributions to the $\log(K_{wm})$, $\log(K_x)$, and $\log(K_s)$ values for different compounds was used in the case of micellar SDS solutions[7]

$$\log(\mathrm{SP}) = c + r \cdot R_2 + s \cdot \pi_2^{\mathrm{H}} + a \cdot \Sigma\alpha_2^{\mathrm{H}} + b \cdot \Sigma\beta_2^{\mathrm{H}} + v \cdot V_x, \tag{3}$$

where SP is the analyzed variable (K_{wm}, K_x, K_s); R_2, π_2^{H}, $\Sigma\alpha_2^{\mathrm{H}}$, and $\Sigma\beta_2^{\mathrm{H}}$ are the descriptors[7,8] characterizing the dispersion (expressed through the excess molar refraction), dipole-dipole, proton-donor, and proton-acceptor interactions of the solute with the medium (the last two interactions are due to the formation of H-bonds); V_x is the characteristic volume of the solute (10^{-2} cm^3 mol^{-1}), which is calculated from the molecular structure[7]; r, s, a, b, and v are coefficients at the descriptors, which account for the intensity of the effect of corresponding interactions on the log(SP) values; and c is a constant dependent on the choice of the standard state for SP.

In this work the capability of SDS micelles to solubilize organic compounds from the gas phase is assessed. To this end, we calculated the gas phase—micelle partition coefficients $K_{gm} = [C_m]/[C_g]$ ($[C_m]$ and $[C_g]$ are the equilibrium molar concentrations of

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the solute in the micellar pseudophase and in saturated vapor under standard conditions[4]) for nonionic compounds. To determine the factors affecting the distribution of various compounds between the SDS micelles and the gas phase, regression analysis was performed of the $\log(K_{gm})$ values calculated using both Eq. (3) and the following equation

$$\log(K_{gm}) = c + w \cdot \log(K_{gw}) + h \cdot \log(K_{gh}), \qquad (4)$$

where the K_{gm} values are expressed through the partition coefficients $K_{gw} = [C_w]/[C_g]$ and $K_{gh} = [C_h]/[C_g]$ ($[C_h]$ is the equilibrium molar concentration of the solute in hexadecane). Using Eq. (4), it is possible to simulate the solvation ability of the micellar microenvironment of the solutes using hydrophilic and hydrophobic factors assessed by $\log(K_{gw})$ and $\log(K_{gh})$ values, respectively. The w and h coefficients characterize the intensity of hydrophilic and hydrophobic interactions between the solute and micelles.

Calculation procedure

The partition coefficients K_{gm} of nonionic compounds in micellar SDS solutions[1] were calculated using the K_{wm},[3] K_x,[5,9] and K_s[6] values, as well as the free energies of transfer of the compounds from water to micelles $\Delta G_{tr} = -RT\ln(K_x)$.[10,11] The values of the partition coefficients K_x[5] and the binding constants K_s[6] were averaged before calculating the K_{wm} coefficients using formulas (1) and (2) with $n_w = 55.5$ mol L^{-1} and $v_s = 0.25$ L mol^{-1} (see Ref. 3). The ΔG_{tr} values were converted into K_x values using the formula $K_x = \exp(-\Delta G_{tr}/RT)$ and then recalculated to obtain the K_{wm} values using formula (2). The $\log(K_{gm})$ values were calculated by the formula $\log(K_{gm}) = \log(K_{wm}) + \log(K_{gw})$ using the $\log(K_{gw}) = \log L^w$ values.[8]

The values of descriptors for calculations using Eqs. (3) and (4) were taken from the literature.[7,8] The regression analysis was carried out by the least squares method. The coefficients at the descriptors and their standard deviations, multiple correlation coefficients R corrected for the number of degrees of freedom, and the standard deviations for the samples (sd) were calculated.

Results and Discussion

The capability of SDS micelles to solvate nonionic compounds more efficiently than water follows from the validity of the inequality $\log(K_{gm}) > \log(K_{gw})$ for all compounds considered and from the positive $\log(K_{wm})$ values (Table 1). For many compounds including alcohols, phenols, naphthols, nitriles, nitro compounds, anilines, lower ketones (up to hexanone), benzamide, and dioxane the $\log(K_{gm})$ values also appreciably exceed (by unity and more) the corresponding $\log(K_{gh})$ values, which indicates their higher solvation by SDS micelles compared to hexadecane. The reverse relationship, $\log(K_{gm}) < \log(K_{gh})$, is valid for the alkanes solubilized in the micellar core,[2] as well as for alkylbenzenes, polycyclic aromatic compounds, and some other compounds (see Table 1).

Regression analysis of $\log(K_{gm})$ values for different types of compounds (a total of 104 substances, see Table 1) using Eq. (3) showed that the regression is characterized by the values $sd = 0.310$ and $R = 0.990$ (Table 2). Assuming that solubilization of different types of compounds is specific toward the mechanism of their interactions with micelles or toward the location sites in the SDS micelles, it is possible to improve statistical characteristics of the regressions for a series comprised of compounds of the same type. Therefore, the entire set of $\log(K_{gm})$ values was divided into four series (see Table 1) comprised of alkanes and nonpolar gases (Ar, O_2) solubilized in the micellar core[2] (series I, compounds **1—12**); proton-donor compounds including alcohols, phenols, and naphthols (series II, compounds **13—52**); aromatic compounds without OH groups (series III, compounds **53—78**); and aliphatic compounds without OH groups but with polar bonds in the molecules (series IV, compounds **79—104**).

The results of the regression analysis of the $\log(K_{gm})$ values for series I—IV are listed in Table 2. The essential feature of these regressions is the fact that the coefficients at all descriptors are positive, which distinguishes them from the reported regressions for $\log(K_x)$,[5] $\log(K_s)$,[6] and $\log(K_{wm})$[4] values, for which the s, a, and b coefficients are negative. The regression equation for series I has only the V_x descriptor because of zero values of other descriptors for this group of compounds.[7] For series III and IV, we obtained regression equations with three (π_2^H, $\Sigma\alpha_2^H$, V_x) and four (R_2, π_2^H, $\Sigma\beta_2^H$, V_x) descriptors, respectively, since the coefficients at other descriptors appeared to be statistically insignificant. Appreciable decrease in the standard deviations sd of the regression equations for each of the series I—IV compared to that of the regression equation for the entire set of compounds (see Table 2) confirms the advisability of using separate regression equations for these series and indicates that solubilization by SDS micelles is specific toward the type of compounds. For series II and IV the free terms of the regression equations c have low statistical significance ($sd_c > c$).

The values of the v coefficients of the regression equations for series II—IV are close and much smaller than the corresponding value for series I (see Table 2), which is associated with localization of compounds of type I in the micellar core.[1] The smallest of the coefficients (r), which characterizes the dispersion interaction between solutes and the medium, decreases in the series IV > II > III. At the same time, the s coefficient increases, thus indicating that the dipole-dipole interactions begin playing a more important role in the same series IV < II < III. Such changes in the r and s coefficients are likely due to peculiarities of the solute location in the micelles where the compounds of series III are in a more polar microenvironment, whereas the compounds of series IV are in a less polar microenvironment compared to that of the compounds of series II. The coefficient a changes parallel to the s values (IV

Table 1. The $\log(K_{gm})$, $\log(K_{wm})$, $\log(K_{gw})$, and $\log(K_{gh})$ values used in calculations

Compound	$\log(K_{wm})$	Ref.	log K		
			(K_{gm})	(K_{gw})*	(K_{gh})*
Argon (**1**)	0.465	6	−1.002	−1.47	−0.688
Oxygen (**2**)	0.453	6	−1.057	−1.51	−0.723
Methane (**3**)	0.672	5	−0.780	−1.46	−0.323
Ethane (**4**)	1.382	5	0.046	−1.34	0.492
Propane (**5**)	1.962	5	0.526	−1.44	1.050
Butane (**6**)	2.642	5	1.124	−1.52	1.615
Pentane (**7**)	3.324	11	1.620	−1.70	2.162
Hexane (**8**)	3.727	11	1.906	−1.82	2.668
Heptane (**9**)	4.586	11	2.624	−1.96	3.173
Octane (**10**)	5.269	11	3.160	−2.11	3.677
Nonane (**11**)	5.865	11	3.565	−2.30	4.182
Decane (**12**)	6.425	11	4.105	−2.32	4.680
Methanol (**13**)	0.079	3	3.819	3.74	0.922
Ethanol (**14**)	0.380	3	4.050	3.67	1.485
Propanol (**15**)	0.881	3	4.441	3.56	2.031
Propan-2-ol (**16**)	0.747	5	4.227	3.48	1.764
Butanol (**17**)	1.262	3	4.722	3.46	2.601
2-Methylpropanol (**18**)	1.387	5	4.687	3.30	2.413
Butan-2-ol (**19**)	1.267	5	4.657	3.39	2.338
tert-Butanol (**20**)	1.087	5	4.367	3.28	1.963
Pentanol (**21**)	1.835	3	5.185	3.35	3.106
Pentan-2-ol (**22**)	1.552	5	4.772	3.22	2.840
Pentan-3-ol (**23**)	1.287	5	4.477	3.19	2.860
3-Methylbutanol (**24**)	1.737	5	4.977	3.24	3.011
2-Methylbutan-2-ol (**25**)	1.447	5	4.697	3.25	2.630
Hexanol (**26**)	2.288	3	5.518	3.23	3.610
Hexan-2-ol (**27**)	2.187	5	5.067	2.88	3.081
Heptanol (**28**)	2.597	5	5.687	3.09	4.115
Octanol (**29**)	3.077	5	6.077	3.00	4.619
2-Butoxyethanol (**30**)	1.897	5	6.487	4.59	3.806
Benzyl alcohol (**31**)	1.717	5	6.577	4.86	4.221
2-Phenylethanol (**32**)	1.977	5	6.957	4.98	4.628
3-Phenylpropanol (**33**)	2.262	5	7.342	5.08	5.180
Phenol (**34**)	1.817	6	6.456	4.85	3.766
2-Methylphenol (**35**)	1.937	5	6.247	4.31	4.218
4-Methylphenol (**36**)	1.997	5	6.497	4.50	4.312
2,3-Dimethylphenol (**37**)	2.377	5	6.897	4.52	4.952
2,4-Dimethylphenol (**38**)	2.417	5	6.827	4.41	4.770
2,5-Dimethylphenol (**39**)	2.417	5	6.757	4.34	4.774
2,6-Dimethylphenol (**40**)	2.377	5	6.237	3.86	4.680
3,5-Dimethylphenol (**41**)	2.417	5	7.017	4.60	4.856
4-Ethylphenol (**42**)	2.460	5	6.960	4.50	4.737
4-Propylphenol (**43**)	3.037	5	7.367	4.33	5.185
4-*tert*-Butylphenol (**44**)	3.177	5	7.517	4.34	5.264
4-Fluorophenol (**45**)	1.847	5	6.387	4.54	3.844
4-Chlorophenol (**46**)	2.097	5	7.257	5.16	4.775
4-Bromophenol (**47**)	2.397	5	7.627	5.23	5.135
4-Formylphenol (**48**)	1.707	5	8.187	6.48	5.533
4-Cyanophenol (**49**)	1.787	5	9.247	7.46	5.420
4-Nitrophenol (**50**)	1.817	5	9.627	7.81	5.876
1-Naphthol (**51**)	2.867	5	8.497	5.63	6.130
2-Naphthol (**52**)	2.835	6	8.785	5.95	6.200
Benzene (**53**)	2.014	3	2.644	0.63	2.786
Toluene (**54**)	2.421	3	3.071	0.65	3.325
Ethylbenzene (**55**)	2.778	3	3.358	0.58	3.778
Propylbenzene (**56**)	3.209	3	3.599	0.39	4.230
Butylbenzene (**57**)	3.628	3	3.918	0.29	4.730
Pentylbenzene (**58**)	3.960	3	4.130	0.17	5.230
p-Xylene (**59**)	3.027	5	3.617	0.59	3.839
Anisole (**60**)	2.064	6	3.864	1.80	3.890
Nitrobenzene (**61**)	1.944	6	4.964	3.02	4.557
Fluorobenzene (**62**)	2.501	10	3.091	0.59	2.788
Chlorobenzene (**63**)	2.553	10	3.373	0.82	3.657
Bromobenzene (**64**)	2.904	10	3.974	1.07	4.041
Iodobenzene (**65**)	3.132	10	4.412	1.28	4.502
Benzonitrile (**66**)	2.121	6	5.211	3.09	4.039
1,2-Dichlorobenzene (**67**)	3.271	10	4.271	1.00	4.518
1,3-Dichlorobenzene (**68**)	3.324	10	4.044	0.72	4.410
Naphthalene (**69**)	3.168	6	4.928	1.76	5.161
1-Methylnaphthalene (**70**)	3.666	6	5.456	1.79	5.789
Biphenyl (**71**)	3.699	6	5.649	1.95	6.014
Anthracene (**72**)	4.427	5	7.327	2.90	7.568
Phenanthrene (**73**)	4.457	5	7.307	2.85	7.632
Pyrene (**74**)	5.057	5	8.377	3.32	8.833
Aniline (**75**)	1.777	5	5.807	4.03	3.993
4-Methylaniline (**76**)	1.897	5	5.987	4.09	4.452
Benzamide (**77**)	1.483	6	9.553	8.07	5.767
Acetophenone (**78**)	2.197	5	5.557	3.36	4.501
Acetone (**79**)	0.387	5	3.177	2.79	1.696
Butanone (**80**)	1.121	3	3.841	2.72	2.287
Pentan-2-one (**81**)	1.560	3	4.140	2.58	2.755
Pentan-3-one (**82**)	1.287	5	3.787	2.50	2.811
4-Methylpentan-2-one (**83**)	1.707	5	3.947	2.24	3.089
Hexan-2-one (**84**)	1.977	3	4.387	2.41	3.262
Heptan-2-one (**85**)	2.438	3	4.668	2.23	3.760
Octan-2-one (**86**)	2.817	9	4.927	2.11	4.257
Nonan-2-one (**87**)	3.334	3	5.164	1.83	4.735
Decanone-2 (**88**)	3.977	9	5.697	1.72	5.245
Dichloromethane (**89**)	1.147	5	2.107	0.96	2.019
Trichloromethane (**90**)	1.657	5	2.447	0.79	2.480
Iodoethane (**91**)	1.944	6	2.484	0.54	2.573
1-Iodobutane (**92**)	2.659	6	2.839	0.18	3.628
Halothane** (**93**)	1.643	6	1.723	0.08	2.177
Tetrachloromethane (**94**)	2.297	5	2.237	−0.06	2.823
Dioxane (**95**)	0.597	5	4.307	3.71	2.892
1-Nitrobutane (**96**)	1.810	3	4.080	2.27	3.415
1-Nitropentane (**97**)	2.228	3	4.298	2.07	3.938
Bromoethane (**98**)	1.607	9	2.147	0.54	2.120
1-Bromopropane (**99**)	2.297	9	2.707	0.41	2.620
1-Bromobutane (**100**)	2.297	9	2.977	0.29	3.105
Acetonitrile (**101**)	0.237	9	3.087	2.85	1.739
Propionitrile (**102**)	0.687	9	3.507	2.82	2.082
Butyronitrile (**103**)	1.167	9	3.837	2.67	2.548
Valeronitrile (**104**)	1.667	9	4.247	2.58	3.108

* According to Ref. 8.

** 2-Bromo-2-chloro-1,1,1-trifluoroethane.

< II < III), whereas the coefficient *b* decreases in the same order (see Table 2). This likely means that the proton-donor and proton-acceptor interactions between the solute molecules, occurring due to the formation of H-bonds, are also dependent on their microenvironment.

Localization of the solute molecules in micelles should be considered as averaged, since the solute mol-

Table 2. Parameters of regression for log(K_{gm}) values, calculated using Eq. (3)

Series	Compound*	*c*	*r*	*s*	*a*	*b*	*v*	*sd*	*R*	*n*
I—IV	**1—104**	−0.947±0.112	0.392±0.107	2.027±0.140	3.834±0.144	2.242±0.182	2.869±0.123	0.310	0.990	104
I	**1—12**	−1.622±0.074	—	—	—	—	3.833±0.083	0.129	0.998	12
II	**13—52**	−0.210±0.457	0.662±0.185	2.236±0.280	2.710±0.735	2.206±0.510	2.322±0.183	0.212	0.990	40
III	**53—78**	−0.531±0.172	—	3.445±0.144	5.523±0.385	—	2.148±0.190	0.184	0.994	26
IV	**79—104**	−0.528±0.291	0.750±0.355	2.016±0.270	—	2.300±0.289	2.249±0.167	0.192	0.983	26

* See Table 1.

ecules can be distributed inside the micelles within a certain range of the gradient of polarity.[12] This fact is likely responsible for the known contradictions regarding the location sites of aromatic compounds in the micelles of SDS and other Surf.[13,14]

Regression analysis of log(K_{gm}) values using Eq. (4) showed its applicability to the data considered. As previously (calculations using Eq. (3)), the inclusion of all compounds in a combined series (n = 104) deteriorates statistical characteristics (*sd*, *R*) of the corresponding regression equation compared to those of the regression equations for the separate series I—IV (Table 3). The decrease in the hydrophobic coefficient *h* in the series I > IV > II > III is in agreement with weakening of the dispersion interactions characterized by the *r* coefficients in Eq. (3) in the series IV > II > III. An increase in the coefficient of hydrophilic solvation in the same series could also be expected; however, a negative *w* value with low statistical significance ($sd_w > w$) for series I and positive *w* values for series II—IV are observed (see Table 3).

Equation (4) can also be applied to the log(K_{wm}) values used for assessing the distribution of the compounds between water and micelles. The results of analyses show the advantage of using separate regression equations for series I—IV (Table 4). It should be noted that for all the series the *h* and *c* parameters of regression equations for the log(K_{wm}) values coincide with those obtained in the corresponding regressions for the log(K_{gm}) values (see Tables 3 and 4). This is due to the use of the values log(K_{gw}) = log(K_{gm}/K_{wm}) as one of the descriptors in Eq. (4).

Thus, the results of the analysis of log(K_{gm}) and log(K_{wm}) values using Eq. (3) agree with those obtained by using Eq. (4) and indicate specificity of solubilization of organic compounds of types I—IV by SDS micelles. Equation (4) has better statistical characteristics (*sd* and *R*) than Eq. (3); therefore, the former is preferential for prognostication.

Table 3. Regression parameters for log(K_{gm}) values, calculated using Eq. (4)

Series	Compound*	*c*	*w*	*h*	*sd*	*R*	*n*
I—IV	**1—104**	0.630±0.096	0.593±0.019	0.716±0.027	0.393	0.984	104
I	**1—12**	−0.773±0.251	−0.243±0.173	0.912±0.032	0.081	0.999	12
II	**13—52**	1.063±0.098	0.551±0.031	0.708±0.027	0.158	0.994	40
III	**53—78**	0.597±0.117	0.624±0.021	0.647±0.026	0.170	0.995	26
IV	**79—104**	−0.087±0.144	0.619±0.042	0.864±0.042	0.188	0.984	26

* See Table 1.

Table 4. Regression parameters for log(K_{wm}) values, calculated using Eq. (4)

Series	Compound*	*c*	*w*	*h*	*sd*	*R*	*n*
I—IV	**1—104**	0.630±0.095	−0.407±0.019	0.716±0.027	0.393	0.942	104
I	**1—12**	−0.773±0.251	−1.243±0.173	0.912±0.032	0.081	0.999	12
II	**13—52**	1.063±0.098	−0.449±0.031	0.708±0.027	0.158	0.975	40
III	**53—78**	0.597±0.117	−0.376±0.021	0.647±0.026	0.170	0.983	26
IV	**79—104**	−0.087±0.144	−0.381±0.035	0.864±0.042	0.188	0.977	26

* See Table 1.

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