Effects of mixed surfactants on the volatilization of naphthalene from aqueous solutions

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Abstract

The effects of mixed anionic–nonionic surfactants, Tween40–SDS (sodium dodecyl sulfate), Tween40–SDBS (sodium dodecylbenzene sulfonate), Tween20–SDS and Tween20–SDBS, on the solubility and volatilization of naphthalene from static aqueous solutions were investigated. The experiment results indicated that mixed anionic–nonionic surfactants can solubilize naphthalene synergistically, which was resulted from the reduction in critical micelle concentration (CMC) of the mixed surfactant and the increase in micellar partition coefficient (K_{mc}) of naphthalene between micelles and aqueous phase. The synergistic effects of mixed surfactants resulted in further reduction in volatilization of naphthalene than that induced by single surfactant. A positive linear correlation was found between the synergistic solubilization ratio (ΔS) and the synergistic inhibitory capacity on naphthalene volatilization (ΔC) in the presence of mixed surfactants. Results from this study imply that mixed surfactants can be employed in environmental remediation to formulate the needed solubility and volatilization of volatile and semivolatile compounds in aqueous solutions.
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1. Introduction

Surfactants have many domestic, industrial and environmental applications. The widespread uses have made surfactants a ubiquitous type of contaminants in the environment. Consequently, the fate and transport of surfactants in natural waters have been studied extensively [1–4]. As a co-existing compound along with its unique properties, surfactant can markedly change the physicochemical properties of other contaminants as well, therefore the fate and transport processes of these contaminants in the environment can be affected.

The effects of individual surfactant on contaminant solubility have been the subject of extensive studies and the enhanced solubility due to monomeric and micellar surfactant aggregates are well defined [5–8]. Surfactants were also found to increase

both the density and viscosity of the aqueous phase containing tetrachloroethylene, with the net effect of decreasing the conductivity when the surfactant concentration was increased [9]. Several studies have addressed the effects of surfactants on the apparent Henry's law constants (H^*) of volatile organic compounds (VOCs) by the equilibrium tests in closed systems [10–14]. These studies indicated that the presence of surfactants significantly altered equilibrium vapor–liquid partitioning, resulting in substantial reductions in H^*.

The effects of surfactant on the vapor–liquid partitioning of VOCs have also been tested in several non-steady state open container systems. Chern and Chou [15] reported that an anionic surfactant at a sub-critical micelle concentration (CMC) significantly reduced the VOC emission in a diffused aeration system. In our previous studies with static aqueous solutions containing BTEX compounds (benzene, toluene and ethylbenzene) and naphthalene, we have demonstrated that both the volatilization rate and the H^* of these VOCs were decreased in the presence of single surfactant when its concentration

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was above CMC [16,17]. We found that the inhibitory effect of surfactant on VOC volatilization loss correlated well with the decreased ratio (α) of VOC concentration in liquid film at the interface to that in the bulk liquid as a result of micellization. The inhibitory effects were more distinct for surfactants with a higher hydrophile-lipophile balance (HLB) number and for VOCs with a higher hydrophobicity (K_{ow}). In a related study using surfactant to remove polycyclic aromatic hydrocarbons (PAHs) from gaseous phase, Huang and Lee [18] revealed that the mass transfer coefficient from gas to liquid phase decreased with increasing surfactant concentration owing to the increase in interfacial resistance and viscosity. The net absorption rate of PAHs by surfactant solution, however, increased because micellar solubilization effects exceeded the reduction effects of the mass transfer coefficient above the CMC.

However, commercial surfactant products and surfactants in the real natural environment are always present in a mixed form. Mixed surfactants are also of great interest in scientific and industrial applications now, because mixed surfactants can offer certain properties (e.g. synergistic solubilization, surface tension reduction, and inhibitory effect on volatilization) that are desirable in many industrial and environmental applications [19,20]. However, The extent of such desirable effects, varies according to the structural type and composition of mixed surfactant systems, and few quantitative was examined in the literature. In some studies using anionic–nonionic surfactant mixture, a synergistic solubilization of PAHs was noted, particularly at low surfactant concentrations [20–22]. On the contrary, Morgan et al. [23] noted small decreases in the value of solubilization equilibrium constant of pentanol in a binary mixture of two cationic surfactants. Moreover, only a limited number of studies have been reported with the volatilization of organic contaminates from mixed surfactants [14]. It is therefore apparent that further studies are warranted to determine the effect of mixed surfactants on water solubility, volatilization and partitioning characteristic of organic compounds.

In the present study, we attempted to determine the solubilization and volatilization of naphthalene in the presence of mixed anionic–nonionic surfactants under static conditions in a system similar to previously described for a single surfactant system. Four types of mixed surfactants were used in this study, including Tween40–SDS (sodium dodecyl sulfate), Tween40–SDBS (sodium dodecylbenzene sulfonate), Tween20–SDS, and Tween20–SDBS. Results from this study are intended to provide some additional insight into the mechanism as well as the practical strategies for the pollution con-

trol of surfactant-laden aqueous wastes containing volatile and semivolatile organic compounds.

2. Materials and methods

2.1. Chemicals

Naphthalene with a purity of greater than 98% was purchased from Shanghai Yuanhang Reagent Plant, China, with the water solubility of 32.05 mg/L and the log K_{ow} of 3.36 [28]. Anionic surfactants, sodium dodecyl sulfate (SDS) and sodium dodecylbenzene sulfonate (SDBS), were purchased from Tokyo Kasei Kogyo Co. Ltd. Two nonionic surfactants used in this study, Tween20 and Tween40, were purchased from Acros Organics. All test surfactants possessed a linear aliphatic chain. These surfactants (all analytical reagents except SDS with a chemical purity of 98%) were used as received without further purifications. The physicochemical properties of compounds used in this study are listed in Table 1. Surfactant solutions were prepared by dissolving the relevant surfactant in deionized water. The stock solutions of naphthalene were also prepared by dissolving it in deionized water.

2.2. Apparatus

The experimental apparatus for the volatilization of naphthalene in a surfactant solution under static condition is shown in Fig. 1. A lidless glass dish of 5.6 cm in diameter and 6.2 cm in height was used to hold the aqueous solution. The solution depth in the dish was maintained at about 4.1 cm, giving a total volume of approximately 100 mL. A temperature-humidity meter (WHM5) was placed in the enclosed test chamber to monitor the relative humidity of the ambient air, which was varied between 75% and 85%. A DHZ-D temperature stabilizer was used as the temperature controller. Ultimately a Plexiglas cuboid without the top and the bottom covers was used to support the static surrounding. In all the processes of volatilization experiment, the glass dishes remain open with the temperature controller closed. A digital reading pipette (Acura 821, Socorex, Switzerland) was used to collect samples from the surfactant solution.

2.3. Batch study on solubilization of naphthalene

The procedures for the preparation of surfactant mixture and solubility determination were essentially the same as described previously [21,22]. Single and mixed surfactant solutions of 5 mL with different concentrations and proportions were placed

Table 1
Properties of the test compounds

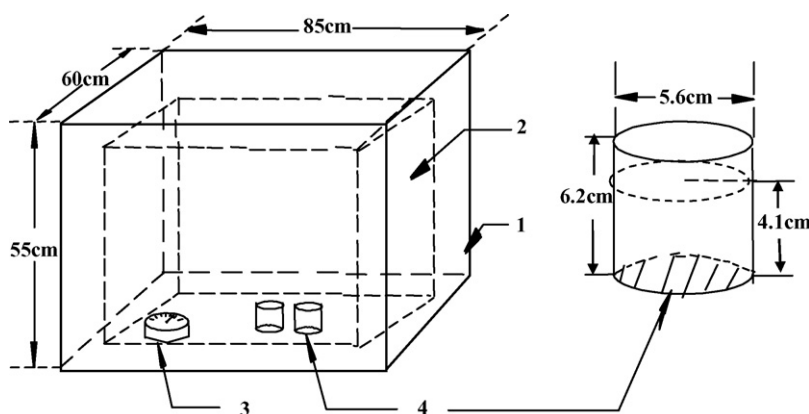| Compound	Formula	MW (g/mol)	CMC (mg/L)	HLB
SDBS	$C_{18}H_{29}SO_3Na$	348.5	522	11.7
SDS	$C_{12}H_{25}OSO_3Na$	288	1455	40
Tween20	$C_{12}H_{23}O_2C_6H_{11}O_4(CH_2CH_2O)_{20}$	1226	60	16.7
Tween40	$C_{16}H_{31}O_2C_6H_{11}O_4(CH_2CH_2O)_{20}$	1282	29	15.6
Naphthalene	$C_{10}H_8$	128.19	–	–

Fig. 1. Experimental apparatus for the volatilization of naphthalene (1, constant temperature controller; 2, Plexiglas cover; 3, temperature–humidity meter; 4, volatilization container).

in centrifuge tubes of about 8 mL. The amount of naphthalene added was in excess of its apparent solubility. Tubes were sealed and then horizontally shaken at 150 rpm at $25 \pm 0.5\,^{\circ}$C for 48 h to reach equilibrium conditions. The tubes were subsequently centrifuged at 4000 rpm for 20 min to separate the undissolved naphthalene at 25 °C. Naphthalene particles adhering to the glass walls were carefully removed. An appropriate amount of aliquot was carefully withdrawn from each tube and diluted with proper ratio for the measurement of naphthalene.

Parallel to the solubility study, the CMC of each surfactant solution were determined by measuring the surface tension of surfactant solution over a wide concentration range and noting the inflection in the plot of surface tension versus the nonionic surfactant concentration. Surface tensions of surfactant solutions were determined with a Model 20 surface tensionmeter (ZJHY-80) at 25 °C water temperature. Detailed procedures can be found elsewhere [20]. The CMC values of single surfactant were obtained through the conventional plot of the surface tension versus surfactant concentration. The CMC values of the mixed surfactant were determined by the plot of surface tension as a function of the nonionic surfactant concentrations while the anionic surfactant concentration was kept constant (550 mg/L for SDS and 600 mg/L for SDBS).

2.4. Batch study on the volatilization of naphthalene

A stock solution of naphthalene solution was prepared by dissolving a predetermined amount of naphthalene (0.025 g based on its solubility) in 1 L deionized water in 1 L ground-glass stoppered flask. Ultrasonication was used to aid the complete dissolution of naphthalene. Then 75 mL of this initial solution were added into a series of 100 mL ground-glass stoppered flasks. Surfactant solutions with different concentrations and proportions of mixed surfactants were added to the flasks and sealed with parafilm. Each prepared mixture was then taken into a volatilization container (Fig. 1) after the liquids reached complete mixing (usually within 20 min) at room temperature ($25 \pm 0.5\,^{\circ}$C). The volatilization loss of naphthalene was determined by analyzing the concentration of naphthalene remained in aqueous solutions. Samples were collected at a 30 min interval until 8 h at the con-

clusion of the volatilization study. The 8 h concentration data were used to estimate the total volatilization loss during each experiment. For each surfactant or surfactant mixture, at least triplicate test chambers were employed.

2.5. Naphthalene measurement and error analysis

Samples were quickly collected for naphthalene measurement by a UV spectrophotometer (UV-2401PC, Shimadzu). To maintain the total volume of the solution, the measured sample was poured back into the volatilization container immediately after measurement. Naphthalene concentrations were determined at 276 nm and pH 6.6–7.3, with the detection limit of 8 μg/L. The background UV absorption of each test surfactant (Tween40–SDS, Tween40–SDBS, Tween20–SDS and Tween20–SDBS) was corrected by calibrating with naphthalene-free solutions containing the same surfactants at the test concentration.

Potential errors were quantified during the volatilization experiments. With surfactant-free solutions, water volatilization loss ranged from 1.87% to 1.88%. The error introduced by volatilization container due to adsorption was estimated to be less than 0.01%. In the presence of mixed surfactants at different concentrations, water volatilization loss was less than 0.10% (container error included), indicating that only minimal errors were introduced.

3. Results and discussion

3.1. Synergetic solubilization of naphthalene by mixed surfactants

Micellar solubilization of naphthalene in mixed anionic–nonionic surfactants and corresponding single nonionic surfactant were determined. Fig. 2 shows the apparent water solubility (S_w^*) of naphthalene as a function of the concentrations of the nonionic surfactant (X). The concentrations of anionic SDS and SDBS were kept constant (550 and 600 mg/L, respectively). As can be seen from Fig. 2, significant solubility enhancement was noted in all cases when an anionic surfactant co-existed. The
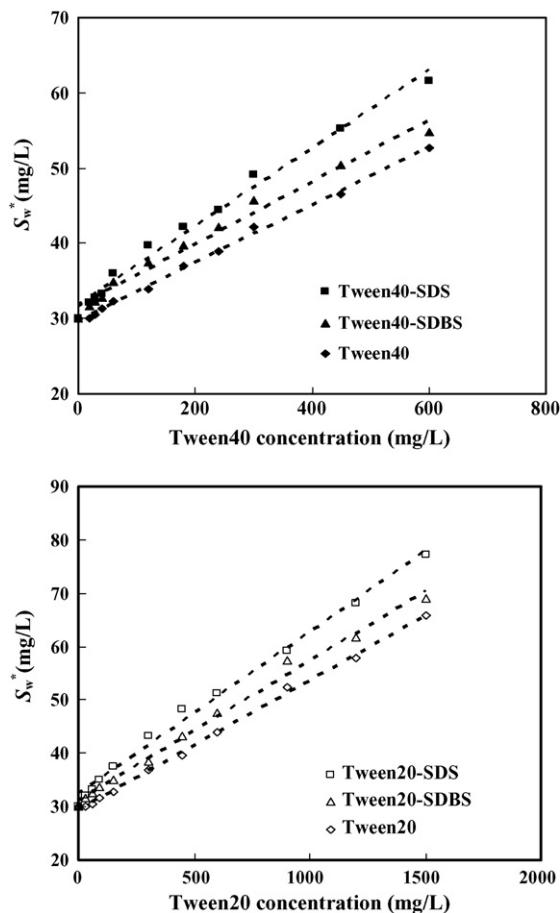
Fig. 2. Water solubility enhancement of naphthalene by mixed surfactants.

Table 2
Enhanced aqueous solubility of naphthalene by single anionic and mixed anionic–nonionic surfactants and values of K_{mc} and CMC

Surfactant	Regression equation $(X > CMC)$	R	K_{mc} (mL/g)	CMC (mg/L)
Tween40	$S_w^* = 0.039X + 29.70$	0.999	1217	29
Tween40–SDBS	$S_w^* = 0.041X + 31.49$	0.991	1279	4
Tween40–SDS	$S_w^* = 0.048X + 31.79$	0.992	1498	0.5
Tween20	$S_w^* = 0.024X + 29.60$	0.999	749	60
Tween20–SDBS	$S_w^* = 0.027X + 31.21$	0.998	842	8
Tween20–SDS	$S_w^* = 0.030X + 32.57$	0.995	936	6

general linear relationship between apparent solubility (S_w^*) and nonionic surfactant concentration (X) can be expressed as [5]:

$$\frac{S_w^*}{S_w} = 1 + X_{mn}K_{mn} + X_{mc}K_{mc} \tag{1}$$

where S_w^* is the apparent water solubility of naphthalene at the total surfactant concentration of X ($X = X_{mn} + X_{mc}$), S_w the intrinsic solubility of naphthalene in "pure water", X_{mn} the concentration of the surfactant as monomers, and X_{mc} the concentration of the surfactant in micellar form. The K_{mn} term is the partition constant of the solute between monomers and water while the K_{mc} term is the corresponding partition constant of the solute between micelles and water.

By Eq. (1), a plot of S_w^* against X will be bilinear, i.e. a straight line with a slope of S_wK_{mn} from $X = 0$ to $X = CMC$, followed by another straight line with a slope of S_wK_{mc} when surfactant concentrations exceed CMC ($X \geq CMC$). The latter is the case for this study, implying that the slope (K, dimensionless) of the solubility curve (Fig. 2) can be used to estimate the micellar partition coefficient (K_{mc} in mL/g) by the following equation:

$$K_{mc} = \frac{K \times 10^6}{S_w} \tag{2}$$

where the intrinsic solubility (S_w) of naphthalene is 32.05 mg/L and 10^6 is a unit conversion factor. A summary of linear regres-

sion equations from Fig. 2 along with the estimated K_{mc} values for various surfactant systems are listed in Table 2. As with Fig. 2, the synergistic effects on solubilization of naphthalene by a mixed surfactants solution are also apparent from the calculated K_{mc} values in Table 2. Results from Table 2 also indicate that naphthalene has a higher micellar partition coefficient in Tween40 than Tween20. For the same nonionic surfactant, SDS had a greater synergistic effect on solubility enhancement than SDBS.

In addition to the variations of K_{mc} values, it is noted that the CMC values of the mixed surfactant were significantly reduced. For instance, a CMC value of 29 mg/L for Tween40 was reduced to 4.0 and 0.5 mg/L when SDBS and SDS, respectively was present at the test concentrations. The reduced CMC values imply that anionic surfactants enhanced the micellization of nonionic surfactants.

The synergetic solubilization of naphthalene by a mixed surfactant solution at a given surfactant concentration can be further quantified by [21]:

$$\Delta S (\%) = \frac{S_w^* - S_{w1}^* - S_{w2}^*}{S_{w1}^* + S_{w2}^*} \times 100 \tag{3}$$

where ΔS is the synergistic solubilization ratio, a dimensionless parameter (%) measuring the synergistic effect on naphthalene solubility by a mixed surfactant solution; S_w^* the apparent aqueous solubility of the solute at a given surfactant concentration in the mixed surfactant solution; S_{w1}^* and S_{w2}^* are the apparent aqueous solubility of the solute at the same concentration of the anionic surfactant (SDS or SDBS) and nonionic solutions (Tween40 or Tween20), respectively. Solubility tests indicated that the S_{w1}^* of naphthalene in SDS (550 mg/L) and SDBS (600 mg/L) solutions were 1.19 and 0.66 mg/L, respectively. The ΔS of naphthalene in different mixed surfactant solutions will be discussed in the next section.

As observed in this study (Table 2), the synergistic solubilization was attributed to the increased micellar partition coefficients (K_{mc}) and the decreased CMC determined from our earlier solubilization studies using mixed anionic and nonionic surfactants [20,21]. It was suggested that the reduced CMC value is probably related to the free-energy reduction upon mixing two types of different surfactants in the aqueous solutions. Marszall [24] noted that the incorporation of ionic surfactant into the nonionic

Table 3
Synergistic effects of mixed surfactants on naphthalene partitioning

	Tween40 (mg/L)							
	20	30	60	120	180	300	450	600
$\Delta S\%$ (Tween40–SDS)	204.3	141.3	113.5	84.4	58.7	49.7	41.4	27.3
$\Delta S\%$ (Tween40–SDBS)	37.8	49.2	30.1	22.6	19.7	17.4	14.4	4.8
	Tween20 (mg/L)							
	30	60	90	150	300	450	600	900	1200
$\Delta S\%$ (Tween20–SDS)	204.3	193.2	146.1	125.0	98.8	70.6	45.0	31.9	23.9
$\Delta S\%$ (Tween20–SDBS)	77.1	55.3	43.2	32.4	27.6	21.0	15.2	11.7	9.6

micelles introduces electrostatic repulsion between micelles, thus hindering the coacervate phase formation and raising the cloud point. It is also likely that mixing caused the weakening of electrostatic self-repulsion for the anionic and the steric self-repulsion for the nonionic surfactant [20].

The reduced CMC values will directly impact solubilization of hydrophobic solute in the liquid through the formation of more micelles at a given mass of surfactant. In the meantime, incorporation of anionic surfactant into micelles of nonionic surfactant will likely induce morphological and/or structural changes that are favorable for micellar partitioning, hence the increased micelle partitioning coefficients observed in this study (Table 2). Experimental evidence has been obtained for a mixed cationic and anionic surfactant system where such mixture is capable of producing distinct wormlike micelles [25]. Additionally, it was interesting to note that synergism of mixed surfactant became less pronounced at higher concentrations of the nonionic surfactants (Table 3). The exact reason is not clear, but it is possible that as surfactant concentration increases, the kinetics of micelle aggression and growth may lag behind and the structure and geometry of the mixed surfactant micelles may not be in its optimal form for hydrophobic partitioning of naphthalene.

3.2. Effect of single surfactants on volatilization of naphthalene from static water

The 8 h volatilization loss of naphthalene in four single surfactant solutions at 25 °C and relative humidity of 60–75% are shown in Fig. 3. It is apparent from Fig. 3 that all four tested surfactants significantly reduced the volatilization loss of naphthalene compared to a surfactant-free control (i.e. ~65% at 0 × CMC). The reductions increased as the concentrations increased for all four surfactants. On the number of CMC basis, the percent reductions were in the order of SDS > SDBS > Tween20 > Tween40. The volatilization of naphthalene in two anionic surfactants was lower than in nonionic surfactants on the CMC basis, because SDS and SDBS have significantly higher CMC than that of nonionic Tween surfactants (Table 1). Results shown in Fig. 3 are in agreement with the data on solubility enhancement. This is expected because micellar solubilization will reduce both the apparent Henry's law constant of naphthalene and the mass transfer across the liquid film

[13–15], thereby decreasing the volatilization of naphthalene in surfactant solution.

3.3. Effect of mixed surfactants on volatilization of naphthalene from static water

The effects of mixed surfactants (Tween40–SDS, Tween40–SDBS, Tween20–SDS and Tween20–SDBS) on volatilization of naphthalene at the same experimental condition as described previously are shown in Fig. 4. In this study, the concentrations of the nonionic Tween40 and Tween20 varied at above the CMCs, but the concentrations of anionic SDS and SDBS were kept constant at below or near the CMCs (i.e. 550 mg/L for SDS and 600 mg/L for SDBS).

Fig. 4 clearly reveals that volatilization of naphthalene was decreased as the concentration of nonionic surfactant increased. The presence of SDS in Tween surfactant solutions inhibited more volatilization of naphthalene than when SDBS was present. To further quantify whether two mixed surfactants exhibited synergistic effect on the volatilization of naphthalene, i.e. whether volatilization in the mixed surfactant solutions was greater than the combined amount of volatilization in each single surfactant solution, the residual concentration of naphthalene was used as a measure of synergism. Analogous to ΔS, the synergetic inhibitory effect of mixed surfactant on the volatilization of
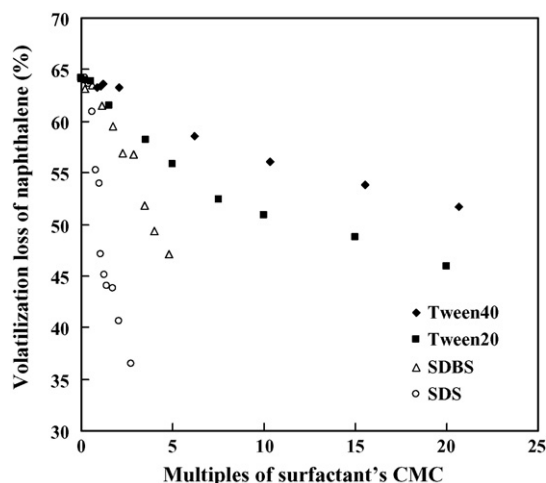

Fig. 3. Influence of single surfactants on the volatilization loss of naphthalene from static water in 8 h.
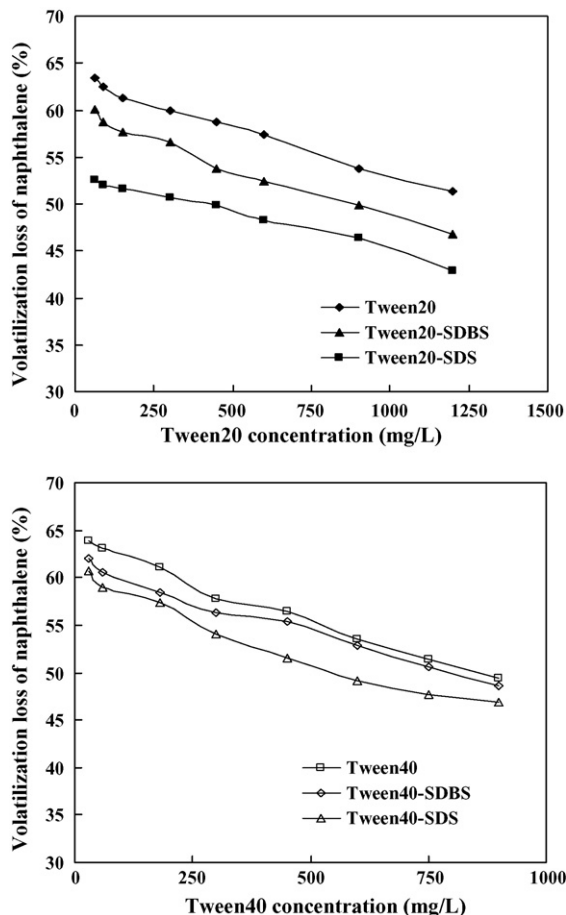
Fig. 4. Influence of mixed surfactants on the volatilization loss of naphthalene from static water in 8 h.

naphthalene is evaluated based on:

$$\Delta C\,(\%) = \frac{C_{\mathrm{w}} - C_{\mathrm{w1}} - C_{\mathrm{w2}}}{C_{\mathrm{w1}} + C_{\mathrm{w2}}} \times 100 \qquad (4)$$

where ΔC is termed the synergistic inhibitory capacity on naphthalene volatilization, a dimensionless parameter (%) related to the synergism. A larger ΔC denotes a higher level of synergism between two surfactants (i.e. these two surfactants synergistically inhibit the volatilization of naphthalene from static water). In a given mixed surfactant system, C_{w} is the increased concentration of the residual naphthalene in the mixed surfactant solution relative to surfactant-free solution; C_{w1} and C_{w2} are the increased concentration of the residual naphthalene in an anionic surfactant (SDS or SDBS) solution and a nonionic solution (Tween40 or Tween20), respectively. At the low concentrations of anionic surfactants used in this study, the value of C_{w1} in either SDS or SDBS solution was 0 mg/L. The ΔC values along with ΔS, calculated using Eqs. (4) and (3), respectively, are listed in Table 3 for both Tween40 and Tween20 solutions. It is evident that, for both nonionic surfactants, synergistic effects existed when either SDS or SDBS was present in the aqueous solution. Results in Table 3 also indicate that the synergism was more pronounced at lower concentrations than higher concentrations of the nonionic surfactants. The synergistic effects on enhanced

solubilization and reduced volatilization were generally higher for SDS than SDBS.

Synergistic effects on volatilization can be directly attributed to the alteration of micellization process as described as early. Micellization minimizes the aqueous phase naphthalene concentrations that are in direct equilibrium with overlying vapor phase. The formation of micelles and the presence of micelles adjacent to the liquid film also affects the mass transfer coefficient particularly for naphthalene with a high H whose air/water exchange resistance will predominately lie in the water film across the interface [26,27]. In single surfactant system, we have experimentally demonstrated that suppression of VOC volatilization was correlated to the decreased ratio (α) of VOC concentration in liquid film at the interface to that in the bulk liquid [16,17]. The effects of surfactant on the mass transfer coefficient of VOCs were tested in an aeration device by Chern and Chou [15]. Under static conditions, however, micelles are capable of retaining more hydrophobic solute in the bulk liquid while micelles near the interface will likely impede the escape of volatile molecule from the liquid phase.

3.4. Relationship between synergic solubilization ratio and the synergetic inhibitory capacity

Data shown in Table 3 are further analyzed by plotting the two parameters that are related to the volatilization and micellar solubilization, i.e. the synergetic inhibitory capacity (ΔC) and the synergic solubilization ratio (ΔS), both of which are caused by the same mixed surfactant solution above the nominal CMC. The resulting plot (Fig. 5) reveal a significant positive linear relationship between ΔC and ΔS ($p < 0.05$). Fig. 5 also indicates that the synergism was more pronounced in Tween20 than Tween40, and SDS than SDBS.

The vapor–liquid partitioning of VOCs in mixed surfactant micelle systems is a complex but important process. This is partially because mixed surfactants are commonplace in the natural environment and mixed surfactants have recently shown to hold some more desirable properties than single surfactants [19,20].
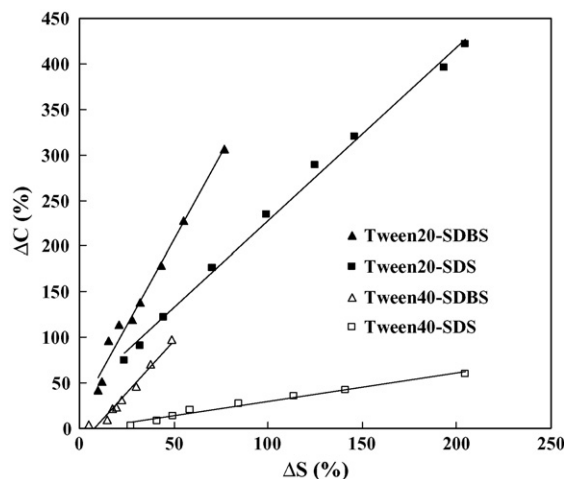

Fig. 5. Relationship between two parameters, ΔS (the synergistic solubilization ratio) and ΔC (the synergistic inhibitory capacity on naphthalene volatilization).

Results from the present study illustrate the synergistic interactions between anionic and nonionic surfactants in the solubility enhancement and reduced loss of naphthalene volatilization. This implies that mixed anionic–nonionic surfactants can be employed in environmental remediation to formulate the needed solubility and volatility of volatile and semivolatile compounds in aqueous solutions. Although more mechanistic studies are needed, our study provided the first quantitative examination into such synergistic effects due to mixed surfactants.

4. Conclusions

(1) Volatilization of naphthalene from static air/water interface was restricted in solutions of surfactants at concentrations greater than the CMC. Compared with single surfactant systems, the mixed surfactant solutions, Tween40–SDS, Tween40–SDBS, Tween20–SDS and Tween20–SDBS led to the synergetic inhibitory effects on naphthalene volatilization.

(2) Over the range of experimental concentrations, mixed surfactant solutions containing SDS had less pronounced loss of naphthalene volatilization than aqueous solutions containing SDBS.

(3) The synergism noted for the mixed surfactants is attributed to the decrease of CMC and the increase of micellar partition coefficients (K_{mc}) of the solutes between micelle and aqueous phase.

(4) The synergic solubilization ratio (ΔS) has a positive linear correlation with the synergetic inhibitory capacity (ΔC) on naphthalene volatilization in the presence of mixed surfactants ($R^2 = 0.971$–0.996, $p < 0.05$).

Acknowledgements

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