Sugar Fatty Acid Ester Surfactants: Structure and Ultimate Aerobic Biodegradability

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ABSTRACT: Ultimate aerobic biodegradabilities of an array of sugar ester surfactants were determined by International Standards Organisation method 7827, "Water Quality—Evaluation in an Aqueous Medium of the Aerobic Biodegradability of Organic Compounds, Method by Dissolved Organic Carbon" (1984). The surfactants were nonionic sugar esters with different-sized sugar head groups (formed from glucose, sucrose, or raffinose) and different lengths and numbers of alkyl chains [formed from lauric (C_{12}) or palmitic (C_{16}) acid]. Analogous anionic sugar ester surfactants, formed by attaching an α-sulfonyl group adjacent to the ester bond, and sugar esters with α-alkyl substituents were also studied. It was found that variations in sugar head group size or in alkyl chain length and number do not significantly affect biodegradability. In contrast, the biodegradation rate of sugar esters with α-sulfonyl or α-alkyl groups, although sufficient for them to be classified as readily biodegradable, was dramatically reduced compared to that of the unsubstituted sugar esters. An understanding of the relationship between structure and biodegradability provided by the results of this study will aid the targeted design of readily biodegradable sugar ester surfactants for use in consumer products.

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KEY WORDS: Alkylbenzene sulfonate, biodegradation, fatty acid ester, glucose, raffinose, sucrose, sugar, sulfonyl, surfactant.

Surfactant biodegradability is a crucial factor in determining whether their concentrations in the environment remain below detrimental levels. Surfactants derived from sugar fatty acid esters are attractive because of their ready biodegradability, low toxicity, low irritation to eyes and skin, and the renewable nature of the sugar and fatty acid starting materials. They are widely used in food, cosmetic, and pharmaceutical formulations (1–3). Physicochemical properties of these surfactants can be tailored to suit potential applications by varying the sugar head group size and the length and number of alkyl chains. As well as nonionic surfactants, analogous anionic sugar ester surfactants can be produced by incorporation of a sulfonate group. These anionic sugar esters are more water soluble than their nonionic counterparts and may more easily replace conventional anionic surfactants in product formulations. Many effects of structural variations on the physicochemical properties of sugar ester surfactants have been reported (4–6).

Sucrose fatty acid esters are rapidly biodegradable (7–13). However, the relationship between biodegradability and chemical structure of sugar ester surfactants has not been comprehensively studied. The aim of the current research was to investigate the ultimate aerobic biodegradation of surfactants derived from sugar fatty acid esters so as to develop an understanding of the relationships between surfactant structure and biodegradability. The biodegradabilities of an array of sugar ester surfactants in which the structure was systematically varied were determined. Structures of the surfactants studied are indicated in Scheme 1. Sugar head group size was varied from a monosaccharide (glucose) to a trisaccharide (raffinose). Alkyl chain length was varied between 12 (laurate) and 16 carbon units (palmitate). Structures with two alkyl chains attached were studied and structures where a sulfonate or alkyl side group had been attached in a position α to the ester bond were also included.

EXPERIMENTAL PROCEDURES

Materials. Analytical-grade reagents obtained from commercial suppliers were used in the preparation of mineral media for biodegradation cultures. Aerobically growing activated sludge was obtained from the CSIRO pilot-scale sewage treatment plant located at Lower Plenty, Melbourne, Australia. The standard soft (soft type D1238) and hard (hard type D0990) anionic surfactants were linear and branched-chain sodium salts of dodecylbenzene sulfonic acid, respectively, obtained from Tokyo Kasei (Tokyo, Japan).

Sugar surfactant synthesis. Sugar ester surfactants were synthesized using procedures described below. Column chromatography was conducted with silica gel 60 (230–400

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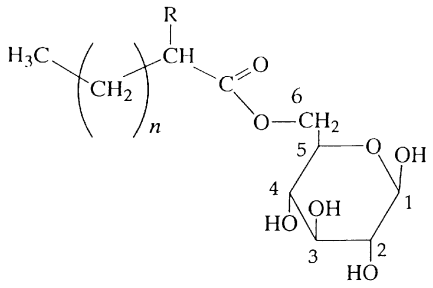
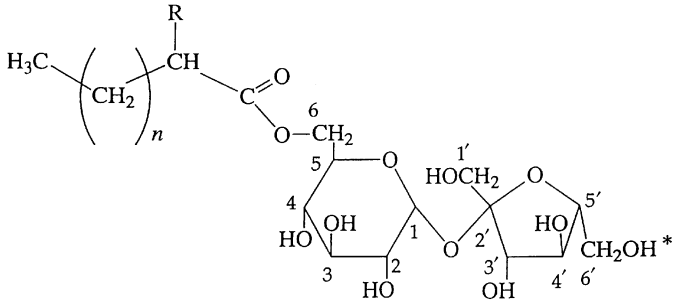
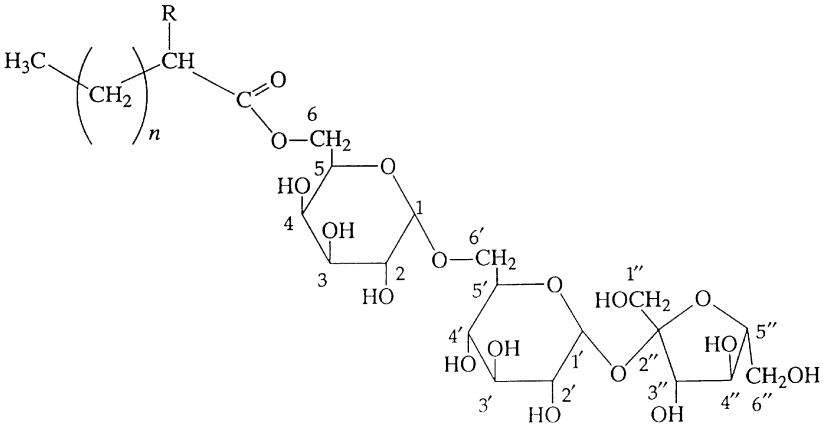
Glucose Esters



Sucrose Esters



Raffinose Esters



SCHEME 1. General structures of sugar fatty acid ester surfactants. R = H, $SO_3^-Na^+$, CH_3, or CH_2CH_3. n = 9 (laurates) or 13 (palmitates). The asterisk indicates the position where the second alkyl chain was attached to form di(α-sulfonyl alkyl) sucrose esters.

mesh) from Merck (Darmstadt, Germany). Proton (^{1}H) and carbon 13 (^{13}C) nuclear magnetic resonance (NMR) spectra were recorded on a Bruker AC200 spectrometer (Karlsruhe, Germany) in deuterated dimethyl sulfoxide (DMSO). Chemical shifts, δ (ppm), of the ^{1}H NMR spectral peaks are reported for each compound. Splitting patterns are represented as *s* (singlet), *bs* (broad singlet), *d* (doublet), *dd* (doublet of doublets), *t* (triplet), *m* (multiplet), or *c* (complex). Where appropriate, the coupling constant, *J* (Hz), between peaks is shown. The number of protons represented by each peak, determined by integration, and the proposed proton assignment are reported (see Scheme 1 for sugar group carbon numbering).

(i) Sucrose 6-laurate. Prepared by treatment of sucrose with lauric acid, triphenylphosphine, and diisopropyl azodicarboxylate in dimethylformamide (14), purified using chloroform/methanol/acetic acid/water (78:11:8:2, by vol) as the chromatography eluent, and freeze-dried to give a white hygroscopic powder. ^{1}H NMR (DMSO): δ 0.85 (3H, *t*, *J* = 6.3 Hz, CH_3), 1.24 (16H, *s*, 8 × chain CH_2), 1.51 (2H, *bs*, β CH_2), 2.30 (2H, *t*, *J* = 6.6 Hz, α CH_2-CO), 3.04 (1H, *t*, *J* = 8.8 Hz, H4), 3.18 (1H, *dd*, *J* = 9.6, 3.5 Hz, H2), 3.37 (2H,

s, H1′), 3.4–3.6 (3H, *m*, H6′ + H5′), 3.72 (1H, *t*, *J* = 7.2 Hz, H4′), 3.8–3.9 (2H, *m*, H5 + H3′), 4.00 (1H, *dd*, *J* = 11.2, 6.1 Hz, H6b), 4.22 (1H, *d*, *J* = 11.2 Hz, 1H6a), 5.10 + 5.15 (1H, 2 × *d*, *J*=3.2 Hz, H1) ppm; ^{13}C NMR (DMSO) δ 172.6, 104.0, 91.4, 82.6, 77.0, 74.4, 72.6, 71.3, 70.2, 69.9, 63.4, 62.6, 62.2, 33.1, 31.0, 28.8, 28.7, 28.5, 28.3, 24.0, 22.0, 13.8. A small amount of the material was recrystallized from ethyl acetate to give sucrose 6-laurate as a white powder, mp 151–154°C, literature value: 150–152°C (15).

The other sugar fatty acid esters were prepared by potassium carbonate-catalyzed transesterification of the appropriate sugar and methyl fatty acid ester in dimethylformamide at 115°C and 70–100 mm Hg pressure (16).

(ii) Sucrose palmitate. Prepared from sucrose and ethyl palmitate. The product was purified using chloroform/methanol/water (82:18:1, vol/vol/vol) as the chromatography eluent, and freeze-dried to give a white powder. ^{1}H NMR (DMSO): δ 0.86 (3H, *t*, CH_3), 1.25 (24H, *s*, 12 × chain CH_2), 1.50 (2H, *bs*, β CH_2), 2.3 (2H, *m*, α CH_2-CO), 3.0–5.1 (~20H, H2–H6 + H1′–H6′), 5.15–5.2 (1H, *d*, H1) ppm.

(iii) Raffinose laurate. Prepared from raffinose (after azeotropic removal of water of hydration with benzene in dimethylformamide) and methyl laurate. The product was purified using chloroform/methanol/acetic acid/water (50:10:8:2, by vol) as the chromatography eluent, and freeze-dried to give a white powder. Elemental analysis gave C 52.3%, H 7.8%; $C_{30}H_{54}O_{17}$ requires C 52.2%, H 7.9%. ^{1}H NMR (DMSO): δ 0.86 (3H, *t*, *J* = 7.2 Hz, CH_3), 1.25 (16H, *s*, 8 × chain CH_2), 1.50 (2H, *bs*, β CH_2), 2.28 (2H, *m*, α CH_2-CO), 3.0–4.4 (H2–H6 + H2′–H6′ + H2″–H6″ + OH), 4.6–5.2 (H1 + H1′ + OH) ppm. Melting properties: softened at ~100°C, darkened at 150°C, melted at 192–196°C (dark brown).

(iv) Raffinose palmitate. Prepared from raffinose (after azeotropic removal of hydrated water with toluene in dimethylformamide) and methyl palmitate. The mixture was extracted several times with petroleum ether to remove unreacted methyl palmitate. The product was a white powder. ^{1}H NMR (DMSO): δ 0.86 (3H, *t*, CH_3), 1.25 (24H, *s*, 12 × chain CH_2), 1.50 (2H, *bs*, β CH_2), 2.24 (2H, *m*, α CH_2-CO), 3.0–5.0 (~23H, H2–H6 + H2′–H6′ + H2″–H6″ + OH), 5.06–5.2 (2H, *bs*, H1 + H1′) ppm.

(v) Sucrose α-methyl laurate and sucrose α-ethyl laurate. Prepared from the α-alkyl lauric acids obtained by treatment of lauric acid with the appropriate alkyl iodide, lithium diisopropyl amide, and hexamethylphosphoramide in dry tetrahydrofuran at 0°C under a stream of argon (17). The α-alkyl lauric acids were obtained as pale yellow oils. Sucrose α-alkyl laurates were then prepared by treatment of sucrose with the appropriate α-alkyl lauric acid, triphenylphosphine, and diisopropyl azodicarboxylate in dimethylformamide.

The esterification of sucrose and α-methyl laurate to obtain sucrose α-methyl laurate proceeded with only a small yield. The product was isolated using chloroform/methanol/water (60:39:1, vol/vol/vol) as the chromatography eluent. ^{1}H NMR (DMSO): δ 0.82 (3H, *m*, CH_3), 1.05 (3H, *d*, CH_3), 1.20 (~18H, *s*, 8 × chain CH_2), 1.25 (<1H, *c*, β CH_2), 2.26 (<1H, *m*, α CH-CO), 3.0–5.0 (>12H, *m*, H2–H6 + H1′–H6′ + impurities), 5.1–5.2 (1H, *m*, H1) ppm.

Sucrose α-ethyl laurate was obtained in good yield. It was isolated using chloroform/methanol/water (81:18:1, vol/vol/vol) as the chromatography eluent. ^{1}H NMR (DMSO): δ 0.82 (6H, *m*, 2 × CH_3), 1.20 (16H, *s*, 8 × chain CH_2), 1.25 (4H, *c*, 2 × β CH_2), 2.26 (1H, *m*, α CH-CO), 3.0–4.32 (12H, *m*, H2–H6 + H1′–H6′), 4.32–5.06 (5H, *m*, OH), 5.06–5.20 (3H, *m*, H1 + OH) ppm.

Methyl monosodium α-sulfonyl fatty acid esters were prepared from monosodium α-sulfonyl fatty acids. These were prepared by treating the fatty acid with chlorosulfuric acid in tetrachloromethane (18,19). Methyl monosodium α-sulfonyl fatty acid esters were prepared by treating the monosodium α-sulfonyl fatty acid with methanol and sulfuric acid under reflux for 24 h. The refluxing solution was passed through a Soxhlet cup containing a 3-Å molecular sieve to maintain dry conditions. The mixture was cooled overnight and neutralized with Na_2CO_3; the solids were filtered and the solvents removed *in vacuo* to give solid product.

(vi) Methyl monosodium α-sulfonyl laurate. ^{1}H NMR (DMSO): δ 0.86 (3H, *t*, CH_3), 1.22 (16H, *s*, 8 × chain CH_2), 1.80 (2H, *bs*, β CH_2), 3.33 (1H, *dd*, *J* = 10.6, 4.4, α CH-SO_3Na), 3.56 (3H, *s*, O-CH_3) ppm.

(vii) Methyl monosodium α-sulfonyl palmitate. ^{1}H NMR (DMSO): δ 0.86 (3H, *t*, CH_3), 1.22 (24H, *s*, 12 × chain CH_2), 1.80 (2H, *bs*, β CH_2), 3.33 (1H, *dd*, *J* = 10.6, 4.4, α CH-SO_3Na), 3.56 (3H, *s*, O-CH_3) ppm.

(viii) Glucose monosodium α-sulfonyl laurate. Prepared from monosodium α-sulfonyl lauroyl chloride (20). Monosodium α-sulfonyl lauroyl chloride was prepared by treating monosodium α-sulfonyl lauric acid with thionyl chloride in benzene and a few drops of dimethylformamide. Glucose was treated with monosodium α-sulfonyl lauroyl chloride in pyridine at 80°C. Glucose monosodium α-sulfonyl laurate was isolated using chloroform/methanol/water (60:40:4, vol/vol/vol) as the chromatography eluent. ^{1}H NMR (DMSO): δ 0.86 (3H, *t*, CH_3), 1.22 (16H, *s*, 8 × chain CH_2), 1.80 (2H, *bs*, β CH_2), 3.0–5.4 (~7H, *m*, H1–H6 + α CH-SO_3Na) ppm.

The other sugar monosodium α-sulfonyl fatty acid esters were prepared by a method similar to that used to prepare the unsulfonated sugar esters described above. The appropriate sugar was transesterified with the appropriate methyl monosodium α-sulfonyl fatty acid ester, using sodium methoxide as the transesterification catalyst in dry dimethylformamide at 100°C and 40 mm Hg (21).

(ix) Sucrose monosodium α-sulfonyl laurate. Isolated using chloroform/methanol/water (60:40:10, vol/vol/vol) as the chromatography eluent. ^{1}H NMR (DMSO): δ 0.86 (3H, *t*, CH_3), 1.22 (16H, *s*, 8 × chain CH_2), 1.80 (2H, *bs*, β CH_2), 3.0–5.4 (~19H, H1–H6 + H1′–H6′ + α CH-SO_3Na + OH) ppm.

(x) Raffinose monosodium α-sulfonyl laurate. Isolated using chloroform/methanol/water (60:50:10, vol/vol/vol) as the chromatography eluent. ^{1}H-NMR (DMSO): δ 0.86 (3H, *t*, CH_3), 1.20 (16H, *s*, 8 × chain CH_2), 1.80 (2H, *bs*, β CH_2), 3.0–5.0 (~22H, H2–H6 + H2′-H6′ + H1″–H6″ + α CH-SO_3Na + OH), 5.0–5.25 (~3H, *m*, H1 + H1′) ppm.

(xi) Sucrose monosodium α-sulfonyl palmitate. Isolated using chloroform/methanol/water (60:50:10, vol/vol/vol) as the chromatography eluent. ^{1}H NMR (DMSO): δ 0.86 (3H, *t*, CH_3), 1.22 (25H, *s*, 12 × chain CH_2), 1.80 (2H, *bs*, β CH_2), 3.0–5.4 (~19H, H1–H6 + H1′–H6′ + α CH-SO_3Na + OH) ppm.

Sugar di(α-sulfonyl fatty acid) esters were prepared by the same method as the monoesters except that the sugar was reacted with two equivalents of methyl monosodium α-sulfonyl fatty acid ester.

(xii) Sucrose disodium di(α-sulfonyl laurate). Isolated using chloroform/methanol/water (60:50:10, vol/vol/vol) as the chromatography eluent. ^{1}H NMR (DMSO): δ 0.86 (6H, *t*, 2 × CH_3), 1.22 (32H, *s*, 16 × chain CH_2), 1.80 (4H, *bs*, 2 × β CH_2), 3.0–5.4 (~19H, H1–H6 + H1′–H6′ + 2 × α CH-SO_3Na + OH) ppm.

(xiii) Sucrose disodium di(α-sulfonyl palmitate). Isolated using chloroform/methanol/water (60:50:10, vol/vol/vol) as the chromatography eluent. ^{1}H NMR (DMSO): δ 0.86 (6H, *t*, 2 × CH_3), 1.22 (48H, *s*, 24 × chain CH_2), 1.80 (4H, *bs*, 2 × β CH_2), 3.0–5.4 (~16H, H1–H6 + H1′–H6′ + 2 × α CH-SO_3Na + OH) ppm.

Biodegradation test method. Ultimate aerobic biodegradabilities were determined by the International Standards Organization (ISO) method 7827, "Water Quality—Evaluation in an Aqueous Medium of the Aerobic Biodegradability of Organic Compounds, Method by Dissolved Organic Carbon (DOC)" (22). In this study, 1- or 1.5-L biodegradation cultures were prepared in 2-L reagent bottles fitted with Drechsel bottle heads. The aqueous medium used to prepare the cultures had a pH of around 7 and contained the following minerals and micronutrients required to support bacterial activity: potassium dihydrogen phosphate (KH_2PO_4, 6.2×10^{-4} M), potassium hydrogen phosphate (K_2HPO_4, 1.25×10^{-3} M), sodium hydrogen phosphate (Na_2HPO_4, 1.27×10^{-3} M), ammonium chloride (NH_4Cl, 4.67×10^{-4} M), magnesium sulfate heptahydrate ($MgSO_4 \cdot 7H_2O$, 8.9×10^{-5} M), calcium chloride hexahydrate ($CaCl_2 \cdot 6H_2O$, 1.25×10^{-4} M), ferric chloride hexahydrate ($FeCl_3 \cdot 6H_2O$, 9.2×10^{-7} M), manganese sulfate tetrahydrate ($MnSO_4 \cdot 4H_2O$, 1.79×10^{-7} M), boric acid (H_3BO_3, 9.25×10^{-7} M), zinc sulfate heptahydrate ($ZnSO_4 \cdot 7H_2O$, 1.48×10^{-7} M), ammonium heptamolybdate tetrahydrate [$(NH_4)_6Mo_7O_{24} \cdot 4H_2O$, 2.98×10^{-8} M), iron chelate [$C_{10}H_{12}N_2O_8$-Fe(III)Na, 2.6×10^{-7} M), and yeast extract (0.15 mg/L). The test compounds were added to the culture media to give a concentration of 30 ppm dissolved organic carbon (DOC). Each medium was inoculated with 7 mL of fresh aerobic activated sludge to give a suspended solids content of about 30 mg/L. The cultures were placed on a shaker table in a temperature-controlled cabinet shielded from direct light and shaken continuously at about 140 to 150 rpm for the duration of the test. The motor driving the shaker table produced a considerable amount of heat with the result that it was impractical to maintain the cabinet temperature below a certain level. The temperature of the cabinet was maintained at the lowest possible level which was 27 ± 2°C. This is just outside the range specified in the standard method (20 to 25°C), but was consistent throughout all the tests. The higher temperature used should be taken into account when comparing results from this study with those reported from other studies, although only a small deviation is expected as a result of this slight temperature difference (23–25).

All biodegradation glassware was soaked in a mixture of concentrated nitric and sulfuric acids (1:1, vol/vol), then rinsed in copious amounts of distilled water and finally triply distilled water, and air-dried before use. Each batch of activated sludge was characterized by its suspended solids content and viable cell count. To determine the suspended solids content, 100 mL of the activated sludge was filtered through a preweighed filter paper. The filter paper and solids were then oven-dried, cooled, and reweighed. Viable cell counts were determined by 5-d, 25°C plate count agar tests (Microtech Laboratories, Melbourne, Victoria, Australia).

Each biodegradation run included duplicate cultures of the sugar surfactants, duplicate cultures of the soft and hard surfactant standards, and a blank culture containing only test medium and activated sludge inoculum. In some runs, an additional reference culture was included containing a standard material with known rapid biodegradability (sodium acetate) at a level of 30 ppm DOC in inoculated test medium. For each new surfactant being tested, both an inhibition test culture and an abiotic blank culture were also included. The inhibition test cultures contained the relevant new test material as well as sodium acetate, both at levels of 30 ppm DOC, in inoculated test medium. The abiotic blanks contained the new test material in an inoculated culture that was then sterilized by the addition of 4 mL/L of 4% hypochlorite solution. By means of these control cultures, processes other than biodegradation, which affect the rate of DOC loss from solution, could be detected.

Samples for DOC analysis (about 35 mL) were withdrawn from the cultures at the required intervals using graduated pipettes. The samples were centrifuged in glass tubes at 4,000 rpm (giving a centrifugal force of 2,275 × *g*) for 15 min. The supernatant was decanted from each sample into a clean test tube for DOC analysis. If analysis could not be performed immediately, samples were stored at 4°C.

DOC analyses were performed using an ASTRO Model 2001 computer-controlled Laboratory Organic Carbon Analyzer (League City, TX). This was calibrated using a standard solution of potassium hydrogen phthalate (0.02124 g is equivalent to 10.00 mg organic carbon) before each set of

samples was analyzed. The error in DOC measurement was found to be about ±3 ppm. The percentage of ultimate biodegradation of a surfactant which had occurred at any time was calculated as:

$$\%\text{degradation} = \left(\frac{(C_0 - C_t}{C_0}\right) \times 100 \quad [1]$$

where C_0 and C_t were the concentrations of DOC measured in the surfactant culture at time zero and time t, respectively. These concentrations were calculated from the level of DOC measured in the surfactant culture by subtracting the level measured in the corresponding blank culture. There is no pass mark specified in the ISO method, however, the OECD methods that are equivalent (26) specify that compounds giving a result of 70% degradation of DOC in 21 d should be regarded as readily biodegradable.

Treatment of data to obtain rate constants and half-lives. For a variety of different types of organic chemicals, including several surfactants, the rate of biodegradation can be described adequately by a simple pseudo first-order model, in which the rate of biodegradation of a material is directly proportional to its concentration (27–30). In this study, there was generally a linear region in plots of ln(DOC) vs. time, corresponding to the rapid biodegradation phase after acclimatization of the culture. The slope of the line of best fit through points in this linear region was used to obtain the pseudo first-order rate constant, k_1, for the biodegradation process using the first-order rate expression:

$$\ln[S]_t = \ln[S]_0 - k_1 t \quad [2]$$

where t is the reaction time, $[S]_0$ and $[S]_t$ are the concentrations of surfactant at time zero and time t respectively, and k_1 is the pseudo first-order rate constant. The half-life $t_{1/2}$ (the time by which the concentration is reduced to half the original concentration) was calculated from k_1 using the expression

$$t_{1/2} = \frac{(\ln 2)}{k_1} \quad [3]$$

RESULTS AND DISCUSSION

Validation of the biodegradation test method using standard soft and hard anionic surfactants. Biodegradation behavior of standard anionic surfactants, which had been well characterized previously, was determined to verify that the biodegradation test procedure was operating correctly and with a biodegradation potential comparable to that achieved in other similar studies. The standard surfactants used for this purpose were linear and branched-chain dodecylbenzene sulfonates. Linear alkylbenzene sulfonates are readily biodegradable (soft) whereas branched-chain alkylbenzene sulfonates are resistant to biodegradation (hard) (31).

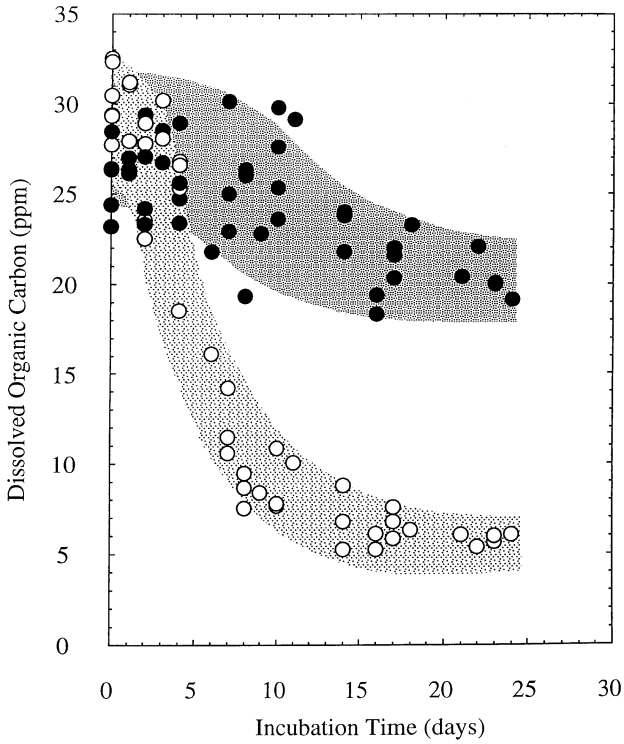


FIG. 1. Combined results for biodegradation of standard soft (○) and hard (●) anionic surfactants. The soft anionic was linear sodium dodecylbenzene sulfonate and the hard anionic was branched sodium dodecylbenzene sulfonate. The shaded regions represent the overall range of biodegradation behavior in each case.

The biodegradation behavior of these standard surfactants was determined in six separate runs. Figure 1 shows all the biodegradation profiles observed in these runs combined on the same graph. The final biodegradation percentages attained in each test are summarized in Table 1. The degree of scatter among points on the biodegradation curves shown in Figure 1 is greater than the error in the DOC measurements (±3 ppm). The additional scatter represents the variability in biodegradation behavior under the conditions of this test method.

The extent of biodegradation of the soft anionic surfactant after 25 d ranged from 72 to 81% with an average of 79%. These results are broadly consistent with those obtained previously. Results from similar measurements of linear alkylbenzene sulfonate biodegradation in inoculated media were between 40 and 100%, with most being between 55 and 85% (31). In this study, the extent of biodegradation attained for the hard anionic surfactant after 25 d ranged between 13 and 49% with an average of 27% (31). These results are also consistent with results obtained in previous studies, which were between 10 and 32% (31). The wide variation in results previously obtained for these standard surfactants is attributed to differences between experimental parameters, such as the aqueous medium composition, the type of inoculum, and the temperature employed, in each study. A clear difference exists,

TABLE 1
Summary of Ultimate Aerobic Biodegradability, Pseudo First-Order Rate Constants (k_1), and Half-Lives ($t_{1/2}$) of Biodegradation for Surfactants Determined by ISO Method 7827-1984 (E)[a]

Substrate	Ultimate biodegradability (% DOC)	k_1 (d-1), mean (range)	$t_{1/2}$ (d), mean (range)
Soft anionic	79	0.13	5.5
	(72–81)	(0.093–0.15)	(4.6–7.4)
Hard anionic	27	0.018	44
	(13–49)	(0.010–0.030)	(22–69)
Sodium acetate	98	>3.0	<0.23
	(94–100)		
Sucrose laurate	99	9.6	0.072
	(96–100)	(all >2.3)	(all <0.3)
Raffinose laurate	99	>1.5	<0.46
Sucrose palmitate	100	>1.7	<0.41
Raffinose palmitate	99	>2.5	<0.23
Sucrose methyl laurate	76	0.23	3.0
Sucrose ethyl laurate	99	0.97	0.71
Sucrose α-sulfonyl laurate	84	0.11	6.4
	(68–90)	(0.078–0.16)	(4.4–8.9)
Raffinose α-sulfonyl laurate	93	0.16	4.2
Glucose α-sulfonyl laurate	92	0.14	4.8
Sucrose α-sulfonyl palmitate	95	0.16	4.4
Sucrose di(α-sulfonyl laurate)	76	0.075	9.2
Sucrose di(α-sulfonyl palmitate)	84	0.096	7.2

[a]See Reference 22. DOC, dissolved organic carbon.

however, between the ranges of results for the soft surfactant and the hard surfactant. Because results of the current research are consistent with prior work, and a clear differentiation between readily degradable and resistant surfactants was shown, the biodegradation test method was performing in accordance with expectations.

Biodegradation behavior of the standard surfactants was also characterized by pseudo first-order rate constants, k_1, and half-lives $t_{1/2}$ of ultimate biodegradation, which provide a numerical basis on which the biodegradation of different compounds can be compared. Values of these parameters determined for the biodegradation of compounds in this study are summarized in Table 1. For the standard soft anionic surfactant the rate constants ranged between 0.093 and 0.15 d^{-1} (half-lives were 4.6 to 7.4 d), with an average of 0.13 d^{-1} (average half-life: 5.5 d). For the standard hard anionic surfactant the rate constants were between 0.010 and 0.031 d^{-1} (half-lives were 22 to 69 d), with an average of 0.018 d^{-1} (average half-life: 44 d). These results provide an indication of the degree of variation in these parameters that can be expected between readily and not readily biodegradable surfactants, and of what differences can be considered significant.

In all biodegradation tests performed in this study, ultimate biodegradation of the additional reference material, sodium acetate, reached >94% within 3 d with an average rate constant >3.0 d^{-1} and half-life of <0.23 d. Since this compound is known to be readily biodegradable, these results provide further confirmation that the test is operating correctly. In all runs the abiotic blanks showed no significant decrease in DOC, for any of the surfactants, over the test period. This indicates that there is no measurable loss of material from the cultures by abiotic processes such as adsorption. None of the materials in this study was found to decrease DOC loss from cultures containing sodium acetate, indicating that they do not inhibit biodegradation of this reference material.

Suspended solids content of the activated sludge averaged 5.4 g/L, and varied between 1.6 and 7.5 g/L. Bacterial count of the activated sludge averaged 19 million colony-forming units (CFU)/mL, and varied between 11 million and 31 million CFU/mL. Although these parameters varied from one run to the next, this variation was not reflected in the biodegradation profiles of the standard soft and hard surfactants that were reproduced fairly consistently as shown in Figure 1.

Sugar ester biodegradation and the effects of structural variations. Turning from the results for the standard surfactants to those for the sugar ester surfactants, the biodegradation behavior for the various structural members of this surfactant class were compared. The results of different biodegradation runs were combined to illustrate the effects of structural changes on the biodegradation of sugar ester surfactants. Final biodegradation percentages for all compounds are summarized in Table 1.

First, consider the nonionic surfactant sucrose laurate, formed from esterification of sucrose and lauric acid, and the anionic analog sucrose sulfonyl laurate, in which a sulfonate group is attached α to the ester bond. In Figure 2, all biodegradation profiles for sucrose laurate and sucrose

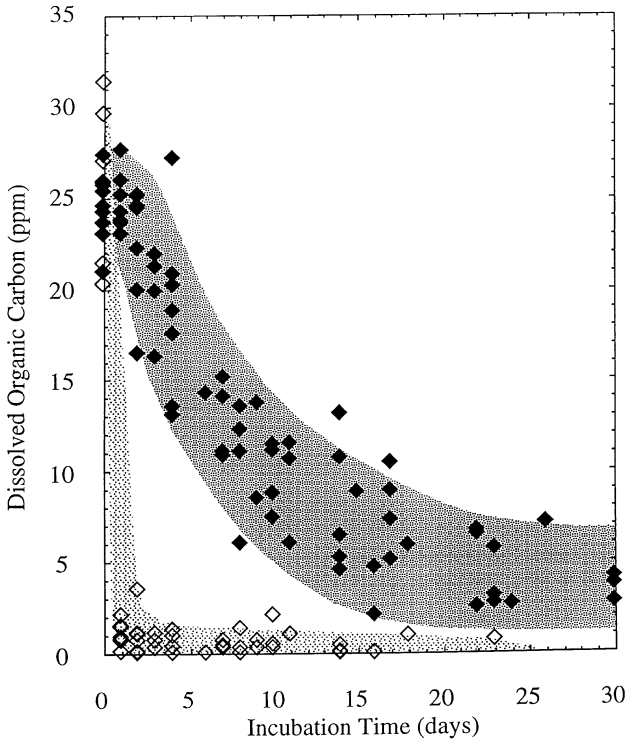


FIG. 2. Combined biodegradation results for sucrose laurate (◇) and sucrose α-sulfonyl laurate (◆) from all determinations. The shaded regions represent the overall range of biodegradation behavior in each case.

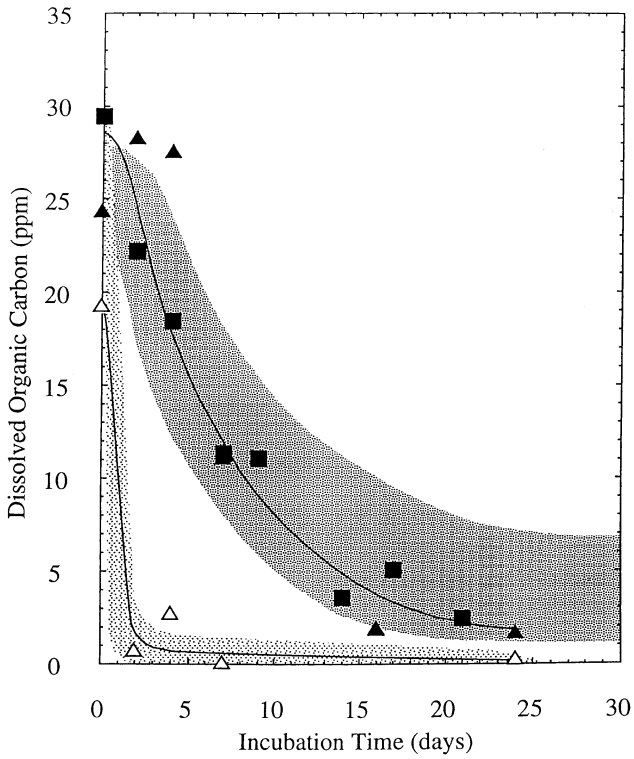


FIG. 3. Biodegradation of both sulfonated and unsulfonated sugar ester surfactants according to head group size: raffinose laurate (△), raffinose α-sulfonyl laurate (▲), and glucose α-sulfonyl laurate (■). For comparison, shaded regions representing the overall range of biodegradation behavior for sucrose laurate (lighter stippling) and sucrose α-sulfonyl laurate (darker stippling) are also shown.

sulfonyl laurate, obtained in eight separate biodegradation runs, are combined on one graph. As was the case for the standard surfactants, there is some scatter in the results, but it is clear that sucrose laurate is rapidly degraded within 1 d. Sucrose sulfonyl laurate degrades more slowly, with an average extent of biodegradation after 25 d of 84%.

Ready biodegradability of sucrose esters, as observed for sucrose laurate in this study, has previously been established (7–13). Biodegradation of α-sulfonated sugar esters has not previously been determined. However, biodegradabilities of other substances containing an α-sulfonyl group have been reported. Primary biodegradation of methyl α-sulfonyl fatty acid esters was reported to be complete within 3–10 d under aerobic conditions (10,32–35). Their ultimate biodegradation reaches about 80% within 20 d (35,36). These results are similar to those observed for α-sulfonated sugar esters in this study. It is evident that addition of the α-sulfonate group produces an ester that is biodegraded at a dramatically reduced rate compared to the unsulfonated analog. It is also known that chemical stability of the ester bond toward chemical hydrolysis is increased by the presence of a sulfonate group in the α position (32,37,38).

Consequences of other structural changes considered in this study are illustrated by similar graphs that combine results from several runs. Figure 3 shows biodegradation profiles measured for sugar ester surfactants, analogous to those discussed above, formed from glucose or raffinose in place of sucrose. Shaded regions representing the range of biodegradation profiles for sucrose laurate and sucrose α-sulfonyl laurate are included on the same graph to aid comparison. Glucose is a monosaccharide, sucrose a disaccharide, and raffinose a trisaccharide. By interchanging these sugars the surfactant head group size surfactant head group was varied. The effect of changing the sugar head group size was determined for both unsulfonated (nonionic) and sulfonated (anionic) surfactants. Biodegradation profiles were very similar to those for sucrose-based surfactants discussed in the preceding paragraphs. Thus, the effect of these changes in sugar head group size on the biodegradability of both the unsulfonated and the sulfonated surfactants was not significant under these conditions.

Sugar ester surfactants with different alkyl chain lengths were formed from fatty acids containing different numbers of carbon atoms. Figure 4 shows the biodegradation profiles determined where the alkyl chain length was increased from C_{12} (laurate) to C_{16} (palmitate). Again both unsulfonated and sulfonated materials are represented. Biodegradation profiles of the palmitates are very similar to those determined for the equivalent laurate-based surfactants. Thus, variation in chain length was not found to have a significant effect on biodegradability under the con-

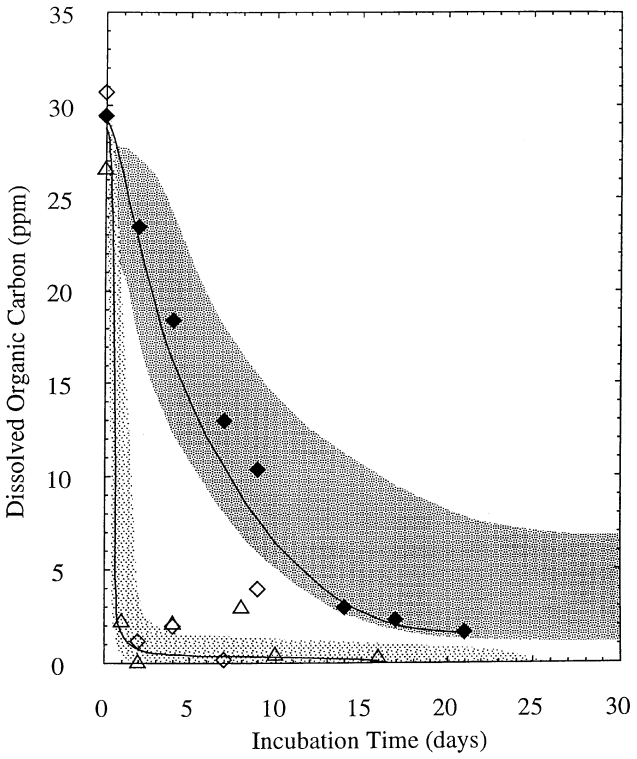


FIG. 4. Biodegradation of sulfonated and unsulfonated sugar ester surfactants with a different alkyl chain length: sucrose palmitate (◇), sucrose α-sulfonyl palmitate (◆), and raffinose palmitate (△). For comparison, shaded regions representing the overall range of biodegradation behavior for sucrose laurate (lighter stippling) and sucrose α-sulfonyl laurate (darker stippling) are also shown.

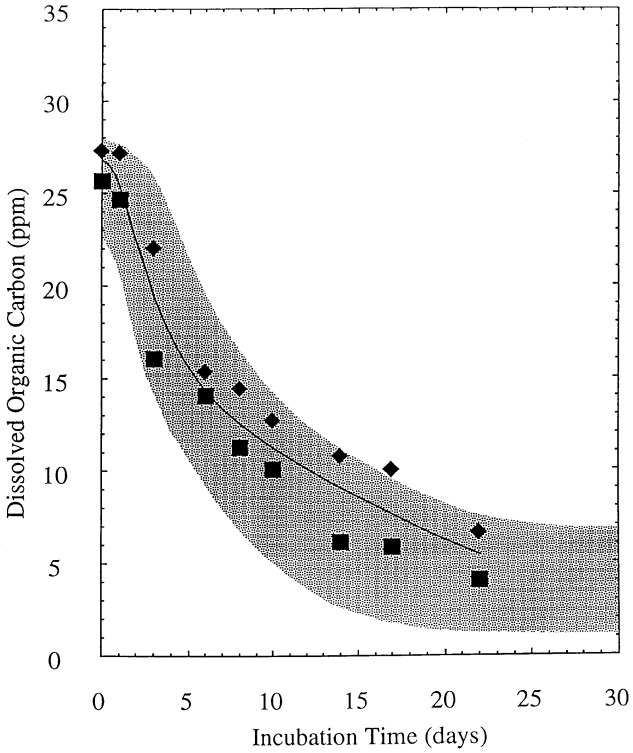


FIG. 5. Biodegradation results for sulfonated sugar ester surfactants which contain two alkyl chains: sucrose di(α-sulfonyl laurate) (◆) and sucrose di(α-sulfonyl palmitate) (■). For comparison, a stippled region representing the overall range of biodegradation behavior for sucrose α-sulfonyl laurate is also shown.

ditions of this test. Previous studies reported slight differences between primary and ultimate biodegradabilities of sucrose laurate, palmitate, and stearate (11,12). The first of these studies (12) used the Sturm method and followed ultimate biodegradation by measuring CO_2 evolution. In this method, CO_2-free air is bubbled through cultures, prepared in an aqueous mineral medium, which are incubated at 22–24°C. Decreased rates of biodegradation were observed with increased chain lengths. The second study (11) used a river water die-away test method and followed primary biodegradation by surface tension measurements. Slightly increased rates of biodegradation were observed with increased chain length. These apparently contradictory findings may have arisen because each study employed different analytical methods to indicate the extent of biodegradation. Thus, the stage of biodegradation observed was different in each case. For example, primary biodegradation, indicated by analysis of surface tension reduction, may occur slightly more rapidly in the case of long-chain sugar esters, while ultimate biodegradation, indicated by CO_2 evolution, may be completed more rapidly in the case of short-chain sugar esters. In addition, the previous investigations used test methods that resulted in lower overall biodegradation potentials than that employed in the present study. The effect of this is that in the two earlier studies, biodegradation of the materials occurred over a longer time frame, such that small differences in biodegradability were observed with greater resolution. Thus, it is possible that there may be slight differences in biodegradation rates of sugar ester surfactants with different chain lengths, which did not shown up on the time scale of the test used in our study. Any such effects are unlikely to be of significance in real environmental situations, such as in sewage treatment plants, where the biodegradation potential is higher than that of these screening tests.

Figure 5 shows biodegradation profiles of surfactants where two α-sulfonated alkyl chains are attached to one sugar. It was not possible to test the corresponding unsulfonated surfactants with two alkyl chains attached by this test method, since these are not sufficiently soluble in the test medium. Again, biodegradation profiles are similar to those of single-chain surfactants. There is no significant difference in the biodegradability of di(α-sulfonyl laurate) and di(α-sulfonyl palmitate). In the case of sulfonated surfactants, the presence of two alkyl chains, rather than one, did not affect biodegradability significantly.

The only structural effect evident at this stage was reduced biodegradability imparted to these sugar esters by the presence of an α-sulfonate group. To gain some indica-

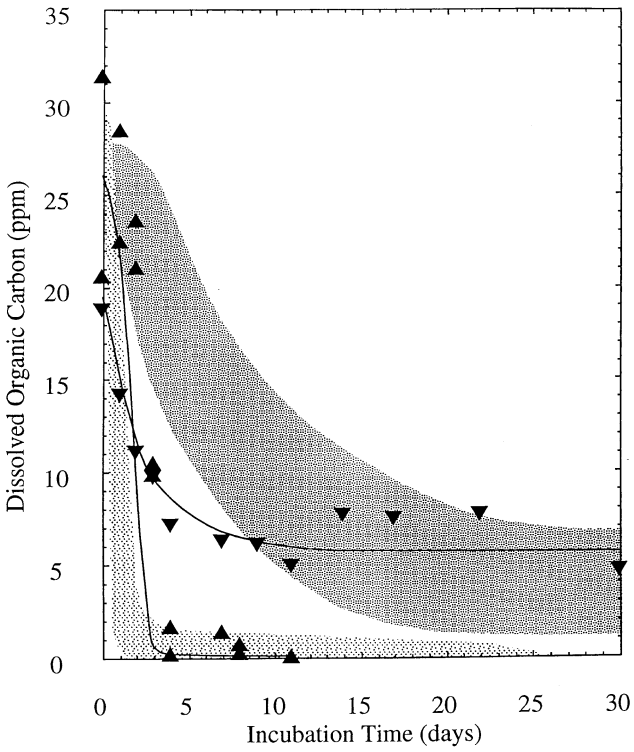


FIG. 6. Biodegradation results for sugar ester surfactants with an alkyl side group attached adjacent to the ester bond: sucrose α-ethyl laurate (▲) and sucrose α-methyl laurate (▼). For comparison, shaded regions representing the overall range of biodegradation behavior for sucrose laurate (lighter stippling) and sucrose α-sulfonyl laurate (darker stippling) are also shown.

tion of whether the inhibitory effect of the α-sulfonate group is due to its charge or to steric hindrance, biodegradabilities of sugar esters with uncharged groups present in the α position were determined. Figure 6 shows biodegradation profiles for sucrose α-methyl laurate and sucrose α-ethyl laurate. Both show biodegradation rates between those of the sucrose laurate and sucrose α-sulfonyl laurate. Thus, the presence of these uncharged groups has also reduced the biodegradability of these esters relative to the underivatized sucrose ester, but not as dramatically as the presence of a sulfonate group in the same position. This indicates that the inhibitory effect of the α-sulfonate group is unlikely to be solely due to its charge but rather also is a consequence of steric effects. The different electronic properties of these subtsituents may also affect biodegradability. Reasons for biodegradability differences were the subject of considerable further investigation described in separate publications (39,40).

Observations made in the preceding paragraphs are also reflected in the pseudo first-order rate constants and corresponding biodegradation half-lives determined for the surfactants in Table 1. In most runs sampling was not done frequently enough to determine rate constants for the unsulfonated sugar ester surfactants. These were all greater than 1.5 d^{-1}, giving corresponding half-lives of less than 0.3 d. On one occasion, the biodegradation of sucrose laurate was followed on a short enough time scale to determine a rate constant for its ultimate degradation. This was found to be 9.6 d^{-1}, resulting in a half-life of 0.072 d. In contrast, the rate constants determined for sulfonated sugar esters were considerably lower. The range of rate constants for sucrose α-sulfonyl laurate was from 0.078 to 0.16 d^{-1} (half-lives: 4.4 to 8.9 d), with an average of 0.11 d^{-1} (average half-life: 6.4 d). Rate constants and half-lives for all other sulfonated sugar esters were in the above ranges determined for sucrose α-sulfonyl laurate. These were very similar to the ranges of rate constants and half-lives of the soft anionic surfactant standard. Rate constants and half-lives for the biodegradation of sucrose α-methyl laurate and sucrose α-ethyl laurate were between those of the unsulfonated and sulfonated sugar ester surfactants.

In conclusion, changes to the sugar head group size, the alkyl chain length, and the number of alkyl chains attached to one sugar head group had no significant effect on the biodegradability of this class of surfactants. The only structural variation that did have a significant effect was the presence of side groups on the alkyl chain adjacent to the ester bond. These groups decreased the rate and extent of biodegradation of the sugar esters. This effect was greatest in the presence of an α-sulfonyl group. When α-ethyl or α-methyl groups were attached, the inhibitory effect was weaker, but nonetheless significant. Further studies, described in separate publications (39,40), indicated that these structural changes affect biodegradability of this class of surfactants through their effects on the pathways followed during biodegradation. The biodegradation rate of sulfonated sugar esters was similar to that of the soft anionic standard, linear alkylbenzene sulfonate. Biodegradation of these surfactants was sufficiently rapid and complete to classify them as readily biodegradable, but they do not have the advantage of very rapid biodegradability of unsulfonated sugar esters.

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