Technical

New Amphoteric Surfactants Containing a 2-Hydroxyalkyl Group: IV. Performance of Amphoteric Surfactant/Soap Blends

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ABSTRACT

The performance of new amphoteric surfactants, *N*-(2-hydroxyethyl)-*N*-(2-hydroxyalkyl)-β-alanines (HAA) and their oxyethylated derivatives (HAA-nEO) was studied in blends with a fatty acid soap. The solubility, foaming power and fabric detergency were measured for blends of (Na-HAA or Na-HAA-nEO)/soap. Furthermore, lime soap dispersing power, toxicity to fish and biodegradability for HAA or HAA-nEO homologs were examined and evaluated. The blend of tallow soap and not less than 10% of $C_{12\text{-}14}$-Na-HAA exhibited better solubility behavior than tallow soap or tallow/coconut oil soap alone. Fabric detergency was improved by blending Na-HAA with soap. Soil deposition in Na-HAA/soap or Na-HAA/soap/Na_2CO_3 solution was measured. On the basis of the results for: (a) the measurements of redeposition; (b) interfacial tension between oil and water and (c) contact angle between oil and fabric in water, it was presumed that since Na-HAA adsorbed more or less in preference to soap on the surface of fabric in water, soil redeposition would be prevented in the Na-HAA/soap detergent solution.

INTRODUCTION

Extensive studies of soap-based detergents have been reported by Linfield and coworkers (1-7). They showed that amphoteric surfactants containing a large hydrophilic moiety such as sulfobetaines and sulfated quaternary ammonium derivatives were highly efficient as lime soap dispersants (8).

We have already presented the preparation and surface active properties of new amphoteric surfactants, *N*-(2-hydroxyethyl)-*N*-(2-hydroxyalkyl)-β-alanines (HAA) and their oxyethylated derivatives (HAA-nEO) (9,10). Furthermore, performance reports for the combination of HAA or HAA-nEO with anionic surfactants such as sodium linear alkylbenzene sulfonate (LAS), sodium lauryl sulfate (SLS) and sodium lauryl ether sulfate (LES) were also published (11). Various soap-based detergents with which these surfactants are blended at a certain ratio are now used as a silk and raw wool scouring agent and an industrial detergent.

In this paper, we wish to examine whether these amphoteric surfactants are useful as lime soap dispersants. Practical performance such as foaming power, fabric detergency and wet soiling for polyester/cotton cloth are evaluated for the (Na-HAA or Na-HAA-nEO)/soap and the (Na-HAA or Na-HAA-nEO)/soap/Na_2CO_3 systems. Furthermore, toxicity to fish and biodegradability are evaluated.

EXPERIMENTAL PROCEDURES

Materials

The amphoteric surfactants of Na-HAA and Na-HAA-nEO used in this paper are the same as previously reported (11). The fatty acid soap used was prepared by neutralization of distilled tallow fatty acid: neutralization value 201.6, saponification value 204.7, iodine value 45.0, m.p. 43.6 C and distilled coconut fatty acid: neutralization value 279.9, saponification value 281.0, iodine value 0.05, m.p. 33.6 C. The soap was dehydrated by an azeotropic distillation using benzene and was then dried in vacuo. Commercial grades of sodium carbonate and calcium carbonate were used (Kanto Chem. Co.).

Measurements of Physical and Surface Active Properties

The rate of dissolution and the heat of dissolution were measured in the following way: the blended solid surfactant (500 mg) was pressed under 40 kg/cm^2 to make a pellet (13 mm diameter, 2.6 mm high). The pellet was placed in 100 ml of water at a fixed temperature with stirring at 200 rpm for 10 min. After sampling with a pipette through a filter, the solution was evaporated to dryness and weighed. Solubility (S) was expressed as g/100 ml-H_2O. The heat of dissolution (ΔH) was calculated from the equation, $\log S = -\Delta H/2.303\ RT$. The Vickers hardness number was measured with a microhardness tester (Type MVK, Akashi Manufacture Co.). Foaming power was measured according to the Ross-Miles method as described in previous papers (9,10). Degreasing power was measured as mentioned in our paper (11); wool cloth (muslin, Japanese Industrial Standard, JIS L-0803) was soiled with olive oil (4%) and a 10-g piece of the test cloth was washed in 1 l of the test solution using a Terg-O-Tometer. After washing, the test cloth was extracted with benzene/ethanol (1:1 v/v) for 3 hr to determine the olive oil remaining in the test cloth. Fabric detergency was carried out using a Terg-O-Tometer and artificially soiled test cloths containing 8.0 ± 0.5% of soil (mineral soil/organic soil/carbon black = 49.75:49.75:0.50) as reported by Kashiwa et al. (12). Cotton test cloth (A-2, Senshokukizai Co.) and polyester/cotton (65:35) test cloth (C-8, Senshokukizai) were used. Soil removal was determined from the reflectance measurement. A wet soiling test was carried out by laundering the soiled and unsoiled fabrics in the same washing test solution; the amount of soil deposits transferred from the soiled fabric to the unsoiled one upon laundering was measured by a photoreflectometer (Hitachi SPR-2). Soil additional density (SAD) was then calculated from the reflectance of the soiled and unsoiled fabrics by the equation (13). $SAD = \log R_0/R_s$, where R_0 is the reflectance of the unsoiled fabric and R_s that of the soiled one.

The interfacial tension between the test solution and liquid paraffin (specific gravity, 0.855; viscosity, 20 cs at 20 C; laboratory reagent, standard) was measured using a ring method at 20 C. The contact angle between oil and fabric in water was measured according to the Berch et al. method (14). A small piece of the fabric was placed horizontally at a proper depth in water containing a surfactant of a fixed concentration in a glass cell (50 x 20 x 20 mm).

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A drop of liquid paraffin (0.01 ml) was injected from the underside with a microsyringe so that the oil drop that was lighter than water could be deposited on the underside surface of the fabric. After standing for 10 sec, the contact angle between the oil drop and the fabric was measured by a projection method. Lime soap dispersing power was determined in 358 ppm $CaCO_3$ hard water at 45 ± 0.5 C according to the Kuchertz method (15). Sodium oleate was added to a 0.1% solution of a dispersing agent until the transmittance of the solution became 60%.

Tests for Toxicity to Fish and Biodegradability

Toxicity tests were carried out on fish in a neutral solution at 20-22 C according to the JIS K-0102-1971 method. The method is based on ASTM D 1345-59 standards for evaluating acute toxicity of water to freshwater fish. The TL_{50} (median tolerance limit) was expressed as the concentration of surfactant (ppm) at which 50% of the test fish survived for either 24 hr or 48 hr. The fish used for this study were adult (0.15-0.45 g) *hi-medaka*, var. *medaka, Orizias latipes* (Family Cyprinodontidae).

The biodegradability was measured according to the JIS K-3363-1976 method. This method describes a shake culture test: 30 ppm of surfactant, culture medium and microorganisms of activated sludge were shaken in a flask at 25 ± 3 C. Acclimatization was carried out for 72 hr two times.

RESULTS AND DISCUSSION

Behavior Relating to Solubility of Na-HAA/Soap Binary Systems

The rate of dissolution for C_{12-14}-Na-HAA/soap and C_{12-14}-Na-HAA/soap/Na_2CO_3 is listed in Table I. C_{12}-, C_{14}-, C_{16}- and C_{18}- prefixed to the abbreviations used in this paper denote, respectively, the carbon number, 12, 14, 16 and 18, in the hydroxyalkyl group. For comparison, the rate of dissolution of tallow soap and the blended soap of tallow and coconut oil was also examined. A blend of not less than 10% of C_{12-14}-Na-HAA and tallow soap exhibited a higher solubility than tallow soap or the blended soap of tallow/coconut oil (7/3). The heat of dissolution calculated from the temperature dependence of solubility for Na-HAA/tallow soap blend showed a higher value than that of tallow soap alone. As amounts of C_{12-14} -Na-HAA increased, the solubility in water was improved. With respect to the hardness of a solid pellet, the hardness of the blended pellet became softer with increasing amounts of C_{12-14}-Na-HAA, whereas that of Na-HAA/tallow soap (1/9) was softer than that of tallow/coconut oil soap (7/3) or tallow soap alone. In experiments with the three-component system of C_{12-14}-Na-HAA/tallow soap/Na_2CO_3, the pellet became progressively harder with increasing amounts of Na_2CO_3. The system also had improved solubility.

Foaming Power of Na-HAA/Soap Binary Systems

Figure 1 shows the foaming power against the component ratio (total concentration 0.2%) of (Na-HAA or Na-HAA-nEO)/tallow soap in 50 ppm $CaCO_3$ hard water at pH 10.5.

The foaming power of the blended systems of (C_{12-14}-Na-HAA or Na-HAA-nEO)/tallow soap did not show the synergistic effect observed for the binary systems of HAA/(LAS, LES or SLS) as stated in a previous paper (11). On the contrary, the foaming power for the binary system of C_{16-18}-Na-HAA-nEO/tallow soap exhibited a lower value than the individual surfactants, showing the minimum value at the component ratio of 0.75/0.25 to 0.5/0.5. The foam height increased in the order of Na-HAA < Na-HAA-2EO < Na-HAA-5EO.

Lime Soap Dispersing Power

Linfield and coworkers published the preparation and lime soap dispersant requirements (LSDR) for various types of surfactants (1-7). A soap-based detergent commonly has poor foaming power and poor detergency in hard water. Therefore, an investigation was undertaken to improve the properties of soap by various additives. The lime soap dispersing power for Na-HAA homologs and their oxyethylated derivatives is given in Table II.

The dispersing power of the ethylene oxide adducts of diethanolamides of fatty acids markedly increased up to ca. 10 mol with increasing mole numbers of ethylene oxide as described by Weil et al. (16). In experiments with the oxyethylated derivatives of Na-HAA, however, 2-mol adducts of C_{12-14}- and C_{16-18}-homologs exhibited the best dispersing power. Even if the mole number of ethylene oxide increased further, the LSDR remained unaffected.

Detergency of Na-HAA/Soap Binary Systems

HAA homologs and their oxyethylated derivatives have excellent surface active properties over a wide pH range. Their surface activities, however, varied with pH value as described in a previous paper (10). The dependence of detergency on pH value is illustrated in Figures 2 and 3.

Figure 2 shows the degreasing power of HAA for wool cloth (muslin) soiled with olive oil. C_{12-14}-HAA always gave a high degreasing power in the range of pH 7-11. The degreasing power of C_{12-14}-HAA-2EO increased as pH value increased up to 10-11. However, both HAA and HAA-2EO exhibited a remarkable drop in degreasing power

TABLE I

Rate of Dissolution for C_{12-14}-Na-HAA/Soap and/Na_2CO_3 System

Composition		Solubility after 10 min (g/100 ml)			Heat of dissolution	Vickers hardness number
		30 C	15 C	5 C	ΔH (Kcal/mol)	H_v (Kg/mm^2)
T^a	(–)	0.081	0.054	0.049	3.5	0.31
C^b/T	(3/7)	0.147	0.058	0.042	8.4	0.54
HAA^c/T	(1/9)	0.156	0.111	0.061	6.1	0.25
HAA/T	(3/7)	0.202	0.163	0.091	5.2	0.14
HAA/T/Na_2CO_3	(1.5/3.5/5)	0.109	0.077	0.072	2.7	1.04
HAA/T/Na_2CO_3	(0.5/4.5/5)	0.089	0.066	0.040	5.3	0.88

^{a}T = Tallow soap.
^{b}C = Coconut oil soap.
^{c}HAA = C_{12-14}-Na-HAA.

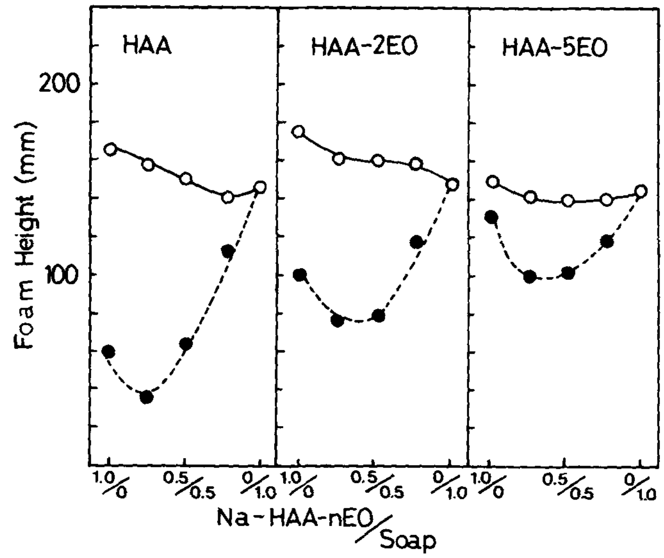

FIG. 1.

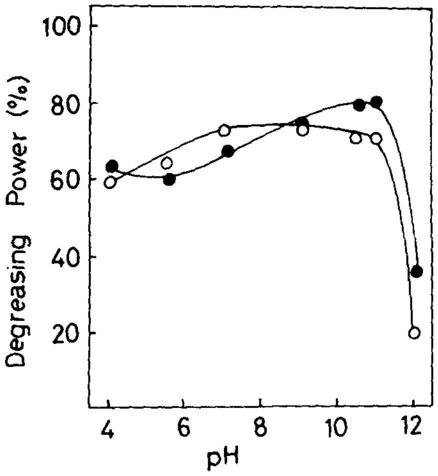

FIG. 2.

at pH 12.

Figure 3 shows the dependence of fabric detergency on pH for cotton and polyester/cotton cloths soiled with an artificial soil. The detergency of *N*-dodecyl-β-alanine (C_{12}-β-alanine) was also measured for comparison. The detergency of HAA and HAA-2EO was best in the range of pH 7-11. The detergency at pH 12 showed a lower value analogous to the result of degreasing power as already mentioned. C_{12}-β-alanine also gave lower detergency at pH 12. The detergency of HAA and HAA-2EO for both cotton and polyester/cotton cloths was better than C_{12}-β-alanine in any pH range.

The relationship between fabric detergency and component ratios of Na-HAA/soap and Na-HAA-nEO/soap binary systems is shown in Figure 4. Any combination of the binary systems showed a synergistic effect. Particularly, enhancement of detergency for polyester/cotton cloth was attained by blends of soap and either Na-HAA or Na-HAA-nEO.

Effect of Na-HAA/Soap Blends on Soil Deposition on Fabric

In order to prevent soil deposition in a detergent solution, a variety of additives such as sodium tripolyphosphate or carboxymethylcellulose are commonly used in commercial detergents. Furthermore, the surface of a synthetic fiber is altered by a surface modifier to change lipophilic properties in order to prevent soil deposition. The contact angle (θ) between oil and fabric in water is defined as:

$$\cos\theta = (\gamma_{WS} - \gamma_{LS})/\gamma_{LW}, \qquad [1]$$

where γ_{WS} is the interfacial tension between water and fabric, γ_{LS} that between oil and fabric and γ_{LW} that between oil and water. When the contact angle approaches 180°, it means the oily soil which was adhered to the surface of the fabric is removed spontaneously (17). The removal of oily soil from fabric is rendered easier as the contact angle between oil and fabric in water becames larger. In addition, if the oily soil collides with fabric in the washing solution, the adsorption on the fabric and some affinities between soil and fabric would be diminished (18). Hence, wet soiling is prevented. The contact angle between oil and fabric in water became larger in the tests with nylon fiber, to which an amphoteric surfactant containing a polyoxyethylene group was incorporated, and therefore wet soiling was prevented as previously reported (19). The contact angle between liquid paraffin and fabric in the solution of Na-HAA/soap blend and the interfacial tension between the solution of Na-HAA/soap blend and the interfacial tension between the solution of Na-HAA/blend

TABLE II

Lime Soap Dispersing Power for Na-HAA and Na-HAA-nEO

Compound	LSDR[a] (g)
C_{12-14}-Na-HAA	0.27
C_{12-14}-Na-HAA-2EO	0.25
C_{12-14}-Na-HAA-5EO	0.29
C_{12-14}-Na-HAA-11EO	0.32
C_{16-18}-Na-HAA	0.80
C_{16-18}-Na-HAA-2EO	0.21
C_{16-18}-Na-HAA-5EO	0.24
C_{16-18}-Na-HAA-11EO	0.27
FA-20[b]	0.25[c]

[a] LSDR = lime soap dispersant requirement: grams of agent required to maintain dispersion of lime soap curd from 1 gram of sodium oleate.

[b] Oleyl alcohol adduct of 20 ethylene oxide moles.

[c] The literature value of LSDR for a commercial grade of the oxyethylated oleyl alcohol (20 mol) (Peregal O) is 0.298 (15).

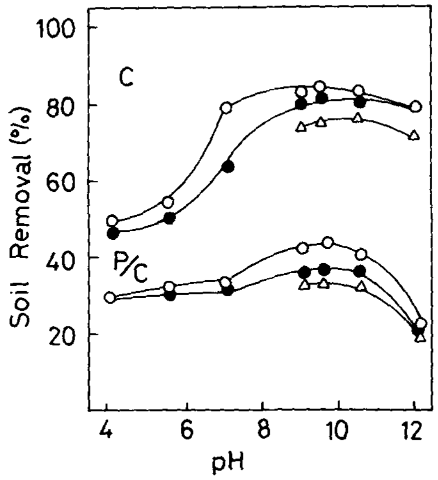

FIG. 3.

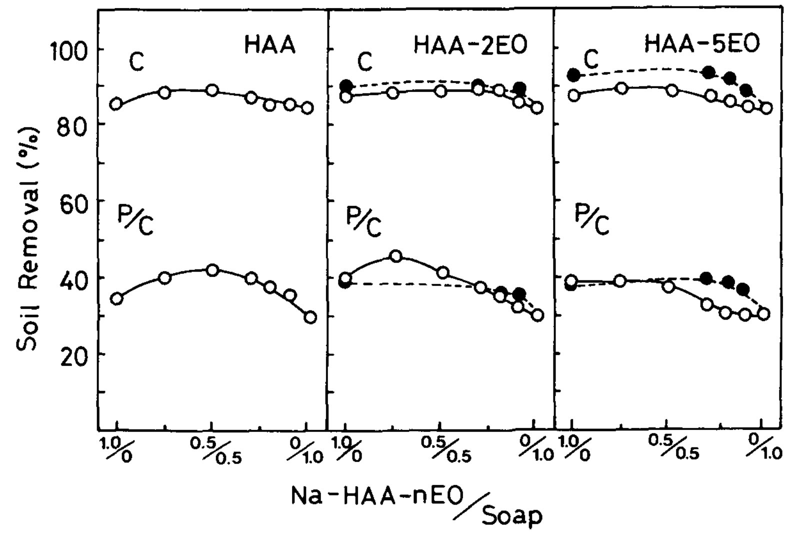

FIG. 4.

and liquid paraffin are listed in Table III.

The interfacial tension of the Na-HAA/soap blend was lower than that of the individual surfactants, showing a definite synergistic effect. The contact angles between oil and fabric in solutions of soap-free Na-HAA were the largest. The contact angle depended on the structure and component ratio of Na-HAA homologs. The contact angle for the binary system of Na-HAA/soap was higher than for soap alone and had a tendency to approach that of Na-HAA alone. Since the contact angle generally increased as the interfacial tension between oil and water (γ_{Lw}) decreased, it was presumed from the results in Table IV that the addition of a component which had a lower interfacial tension did not necessarily exhibit a higher contact angle. Nakamura reported C_{12}-β-alanine had stronger adsorption than sodium lauryl sulfate in the binary

TABLE III

Interfacial Tension and Contact Angle for Na-HAA Derivatives/Soap Mixed Systems at 20 C

Composition		Interfacial tension[a] (dyn/cm)	Contact angle (θ)[b]	
			(C)[c]	(P/C)[d]
C_{12-14}-Na-HAA/Soap[e]	100/0	8.2	158	143
	75/25	3.2	157	143
	50/50	2.4	155	143
	30/70	2.5	155	143
	20/80	3.2	153	142
	10/90	3.1	154	142
	0/100	6.2	148	138
C_{12-14}-Na-HAA-2EO/Soap	100/0	4.9	160	146
	75/25	4.4	153	147
	50/50	4.2	153	146
	30/70	4.1	153	143
	20/80	4.2	152	142
C_{12-14}-Na-HAA-5EO/Soap	100/0	6.5	159	146
	75/25	5.2	155	147
	50/50	4.3	155	147
	30/70	5.2	154	146
C_{16-18}-Na-HAA-2EO/Soap	100/0	15.1	158	150
	75/25	6.3	154	147
	50/50	5.8	153	144
	30/70	6.1	153	144
	20/80	6.0	152	142
	10/90	6.1	152	142
C_{16-18}-Na-HAA-5EO/Soap	100/0	7.5	159	151
	30/70	7.4	153	143
Blank[f]			138	121

[a] Aqueous solution of 0.2% surfactant in liquid paraffin.

[b] 0.2% Surfactant for liquid paraffin/fabric.

[c] (C) Cotton.

[d] (P/C) = Polyester/cotton.

[e] Tallow soap.

[f] In water without surfactant.

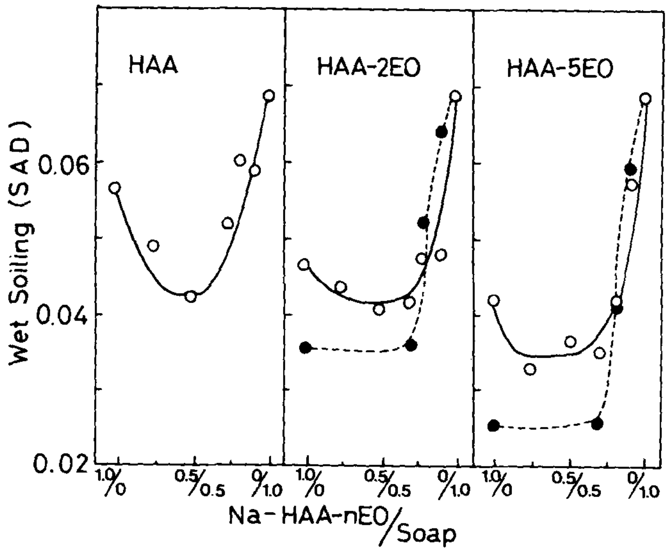

FIG. 5.

system of C_{12}-β-alanine and sodium lauryl sulfate at the nitrogen/solution interface (20). On the basis of his conclusion and our experimental results, it is inferred that Na-HAA or Na-HAA-nEO is preferentially adsorbed, rather than soap, on the surface of fabric in the solution of (Na-HAA or Na-HAA-nEO)/soap blend.

Figure 5 shows the result of wet soiling which was calculated by the difference of reflectance between the soiled and unsoiled cloths upon laundering. Wet soiling was prevented by blending Na-HAA or Na-HAA-nEO with soap. Na-HAA-nEO exhibited better prevention effects than Na-HAA. One reason soil removal was improved is the lime soap dispersing power of Na-HAA. The other reason is the prevention effects of wet soiling resulting from the preferential adsorption of Na-HAA on the fabric surface.

Table IV shows soil removal, wet soiling and interfacial tension of (Na-HAA or Na-HAA-2EO)/soap blends and their Na_2CO_3-containing blends. Particularly, soil removal for polyester/cotton was found to be higher at the composition of $C_{12\text{-}14}$-Na-HAA/soap/Na_2CO_3 (0.03/0.07/0.10), compared to that of a commercial soap. Furthermore, the Na-HAA/soap/Na_2CO_3 systems used in a proper component ratio gave the smallest wet soiling.

Evaluation of Toxicity to Fish and Biodegradability

The results of a toxicity test on fish are summarized in Table V. The TL_{50} value of $C_{12\text{-}14}$-HAA was higher than that of C_{12}-β-alanine and the TL_{50} value of $C_{16\text{-}18}$-HAA showed a remarkably lower value than $C_{12\text{-}14}$-HAA and C_{12}-β-alanine. The TL_{50} values of the oxyethylated derivatives of $C_{12\text{-}14}$-HAA homologs decreased with increasing mole numbers of ethylene oxide, whereas those of the oxyethylated derivatives of $C_{16\text{-}18}$-HAA homologs increased slightly with increasing mole numbers of ethylene oxide.

Fernly reported amphoteric surfactants such as ammoniocarboxylates and sulfobetaines had excellent biodegradability (21). Eldib also concluded C_{12}-β-alanine and *N*-stearyl-β-amino dipropionic acid showed good biodegradability of 95-100% after acclimatization for 10 days by an activated sludge test (22). The biodegradation of $C_{12\text{-}14}$-Na-HAA was 98.5% as a mean value of 7-8 days and that of $C_{12\text{-}14}$-Na-HAA-2EO was 72.2% after 2 days and 95.5% as a mean value of 7-8 days. These new amphoteric surfactants

TABLE IV

Soil Removal in Na-HAA or Na-HAA-2EO, Soap and Na_2CO_3 System

Composition	g/100 ml water	Soil removal (%) (C)[a]	Soil removal (%) (P/C)[b]	Wet soiling[c] (SAD)[d]	Interfacial tension (dyn/cm)
$C_{12\text{-}14}$-Na-HAA-Soap	= 0.060/0.140	87	40	0.0462	2.3
	= 0.039/0.091	82	33	0.0484	5.1
	= 0.030/0.070	80	31	0.0628	8.1
	= 0.021/0.049	65	29	0.0742	11.7
$C_{12\text{-}14}$-Na-HAA/Soap/Na_2CO_3	= 0.039/0.091/0.07	89	48	0.0409	2.5
	= 0.030/0.070/0.054	85	36	0.0517	—
	= 0.030/0.070/0.10	86	41	0.0572	2.8
	= 0.021/0.049/0.13	70	30	0.0860	4.1
$C_{12\text{-}14}$-Na-HAA-2EO/Soap/Na_2CO_3	= 0.020/0.080/0.054	85	37	—	—
	= 0.020/0.080/0.10	86	42	—	—
	= 0.010/0.090/0.054	86	40	—	—
	= 0.010/0.090/0.10	86	42	—	—
Soap/Na_2CO_3	= 0.10/0.10	81	31	—	—
$C_{16\text{-}18}$-Na-HAA-2EO/Soap	= 0.06/0.140	90	38	0.0355	—
	= 0.039/0.091	86	31	0.0517	—
	= 0.030/0.070	82	26	0.0600	—
	= 0.021/0.049	72	24	0.0685	—
$C_{16\text{-}18}$-Na-HAA-2EO/Soap/Na_2CO_3	= 0.039/0.091/0.07	88	44	0.0355	—
	= 0.030/0.070/0.10	82	40	0.0256	—
	= 0.021/0.049/0.07	78	36	0.0742	—
Control detergent[f]	= 0.2	86	35	0.0696	—

[a] C = cotton.
[b] P/C = polyester/cotton.
[c] Redeposition for P/C.
[d] SAD = soil additional density.
[e] Soap = tallow soap.
[f] Commercial soap-based detergent.

TABLE V

Toxicity Test to Fish[a]

Compound	TL_{50} (ppm)[b]	
	24 hr	48 hr
$C_{12\text{-}14}$-HAA	48	47
$C_{12\text{-}14}$-HAA-2EO	23	23
$C_{12\text{-}14}$-HAA-5EO	25	24
$C_{12\text{-}14}$-HAA-11EO	25	20
$C_{16\text{-}18}$-HAA	3	3
$C_{16\text{-}18}$-HAA-2EO	4	3
$C_{16\text{-}18}$-HAA-5EO	4	4
$C_{16\text{-}18}$-HAA-11EO	14	13
C_{12}-β-Alanine[c]	36	34
NP-9[d]	10	--

[a] The fish used were adults of *hi-medaka*, var. *medaka*, *Oryzidas latipes* (family Cyprinodontidae).

[b] TL_{50} = median tolerance limit.

[c] Commercial grade of *N*-lauryl-β-alanine.

[d] Commercial grade of nonyl phenol adduct of 9 mol ethylene oxide.

of β-alanine types containing hydroxy groups exhibited good biodegradability.

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