# **Sucrose Esters and Sucrose Ester/Glyceride Blends as Emulsifiers**

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## **ABSTRACT**

The sucrose mono- and diesters of long-chain fatty acids form a class of food-grade emulsifiers with a wide range of hydrophilelipophile balance (HLB) values. Blends of sucrose mono- and ditallowate show a linear relationship between HLB and composition over the HLB range 10-16. Using blends of mixed glycerides and sucrose tallowate, the HLB range from 4 to 16 can be covered. The relationship between HLB water-solubility and composition in the sucrose esters is dependent on three factors: (a) the degree of substitution; (b) the alkyl chain length in the ester group; and (c) the presence of dienoic or trienoic acyl groups. Water-solubility of the esters studied ranged from soluble to insoluble; sucrose esters with C12 or unsaturated acyl groups act as solubilizers. The relationship between fatty acid profile and solubility was studied using sucrose esters prepared from tallow, palm kernel oil, soybean oil and coconut oil. The properties required of a range of emulsifiers designed for use in foods, cosmetics and pharmaceuticals are: (a) that they cover the range of HLB from 2 to 16+; (b) that at high HLB values (16+), they act as solubilizers; (c) low toxicity and irritancy. The sucrose esters, and sucrose ester/mixed glyceride blends, have the required combination of properties. The HLB range to 16+ can be covered without forming ethoxylated or propoxylated derivatives.

# **INTRODUCTION**

The task of selecting a suitable emulsifier for a particular system can often involve a great deal of experimentation. The hydrophile-lipophile balance (HLB) system proposed by Griffin (1) and Griffin and coworkers (2, 3) offers a means of selecting a suitable emulsifier based on the hydrophilic-lipophilic balance. The determination of the HLB values for new emulsifiers is normally made by comparison of their emulsifying properties with those of surfactants of known HLB values. The HLB value allows a prediction of the action that may be expected from an emulsifier, e.g., a low value (3-6) will be a water-in-oil emulsifier, an intermediate value (8-13) will be an oil-in-water emulsifier, and a high value (15-18) will act as a solubilizer.

Various methods of estimating the HLB from contributions of the active groups in the surfactant molecule have been reported (4, 5). However, these methods are not applicable to sucrose esters and HLB values have been determined experimentally by the emulsification (1) and by the titration method (6). Kawakami (7) presented an equation for the evaluation of HLB values from the composition of the surfactant molecule. Davies (8) calculated the HLB from the chemical formulae of substances using group numbers.

A rapid method by measurement of the heat of hydration  $(\Delta H)$  has been reported by Racz and Orban (9). An investigation by Wachs and Hayano (10) revealed a correlation between HLB value and the logarithm of critical micelle concentration. In systems using nonionic surfactants as emulsifiers, a linear relationship was found to exist between the HLB value and the temperature of phase inversion (11).

A titration method has been reported by Greenwald et al. (12). The nonionic compounds are solubilized in a dioxane/benzene solution and titrated to a cloud point with water, but the method suffers from a lack of reproducibility. The method is also not applicable to ester adducts. This method was further developed (6) to overcome the difficulties experienced with the dioxane solvent by using dimethyl-formamide.

The application of the HLB concept to guide the choice of nonionic surfactant for an emulsification was developed by Atlas Chemicals using the Honeywill-Atlas (now Atlas Chemical Industries Inc., Wilmington, DE) 'Span' and 'Tween' ranges of sorbitan esters and ethoxylated sorbitan esters and is well known (13). Here a series of products with a range of HLB values from 2 to 16 is available.

In this paper, it is shown that a similar range of HLB values can be covered using sucrose esters and sucroglycerides without resorting to ethoxylation or propoxylation. Sucrose esters of widely different degrees of hydrophilic behavior can be prepared by varying: (a) the alkyl chain length of the ester groups; (b) the degree of unsaturation of the ester groups; (c) the number of ester groups per molecule.

As an example, a sucrose diester containing tallowate ester groups, prepared by the method described below, is almost insoluble in water. A sucrose ester containing cocoate ester groups is water-soluble at room temperature.

Contrary to statements by Research Group IV of Daiichi Kogyo Seiyaku Company (14), the degree of substitution of the sucrose is not the single'major factor affecting watersolubility of the sucrose ester. Alkyl chain length and the presence of dienoic or trienoic acyl groups are equally important.

The solubilization effect of the sucrose monoesters containing a C12 alkyl group is considered below: The presence of dienoic or trienoic acyl groups has a similar effect, the monoester solubilizing the diester. The fatty acid profiles of the triglycerides used, and of the sucrose mono- and diesters obtained from them are given in Tables I and II, respectively.

The HLB values of sucrose esters containing saturated alkyl chains in the ester moieties have been reported (14, 15) and a range of 6-15 HLB obtained with mixtures of sucrose mono- and diesters varying in composition from 30% mono/70% (di- and tri-) to 70% mono/ (30% di- and triesters). HLB values of sucrose esters containing unsaturated alkyl chains have not been reported previously.

However, preparation of pure mono- or diester of sucrose to food-grade standards (14) is a difficult matter, and we have found that an easier way in which to obtain a wide range of HLB values using sucrose esters is to blend these with mixed glycerides (the blend corresponding to the European Economic Community (EEC) definition of a

#### **TABLE I**

Fatty Acid Profiles **of Triglycerides a** (%)



aDetermined by formation of the methyl esters and GLC analysis.

# 'sucroglyceride') (16).

Using sucrose mono- and ditallowate and mixed glycerides prepared from tallow (iodine value [IV] 45), it is shown that the HLB range 4-16 can be covered, and that using mixtures containing varying proportions of sucrose mono- and ditallowate, the HLB range 8-16 can be covered. A similar HLB range (8-16+) is available using sucrose cocoate and mixed glycerides derived from coconut oil, and the same range of HLB can be covered using soybean esters and mixed glycerides from soybean oil.

Sucrose esters can be used in foods as emulsifiers, because they are nontoxic, tasteless, odorless and are digested as a blend of sucrose and fatty acids in the stomach. Sucrose esters can also be used in the pharmaceutical industry, in cosmetics, in food additives and various other areas where a nonionic, nontoxic, biodegradable emulsifier is required.

The ADI of sucrose esters has been established at 0-10 mg/kg by the FAO Food Additives Committee (17).

Sucrose esters are on the BIBRA (18) list of permitted indirect food additives, and are permitted food emulsifiers in the UK (SI 1833/80). They are listed in the EEC Directive as permitted food emulsifiers, and a specification to which they must conform is given (E473) (EEC Directive 78/663/EEC) (16).

# **EXPERIMENTAL**

## **Materials**

*Sucrose esters.* These were prepared from four triglycerides: (a) tallow, IV 45  $\pm$  2 (grade II) with edible certification; (b) coconut oil, IV,  $9 \pm 1$ ; (c) palm kernel oil, IV,  $19 \pm 5$ ; and (d) soybean oil, IV,  $130 \pm 10$ . The fatty acid profiles of the triglycerides are given in Table I. *Pine oil.* American 70% distilled material supplied by Meade and King Limited, Liverpool, England. *Cottonseed oil.* (Commercial) supplied by Edible Oils Limited, Liverpool. *Dimetbyl formamide (DMF).* Technical grade. Dried over molecular sieve for 5-7 days before use. *Benzene.* Reagent grade. *Toluene,* 'Analar' grade. *Etber.* Reagent grade, *n-butanone.* Reagent grade.

#### **Purification of Esters and Mixed Glycerides**

From sucroglycerides prepared as described (19, 20), mixed glycerides, mono- and diesters of sucrose were separated by large-scale column chromatography.

The column chromatography was run using columns 10 cm in diameter and 40 cm deep, filled with Merck silica gel, grade 7734. The columns were filled dry, using a glass tamping rod and a vibrator to achieve uniform filling and the silica gel deactivated by eluting with dry ether. The impure sample of sucroglycerides was dissolved in 'Analar'

#### **TABLE** II

Fatty Acid Profiles of Sucrose Mono- and Diesters<sup>a</sup> (%) (Purified by Large-Scale Column Chromatography)



aDetermined by formation of the methyl esters and GLC analysis.

### **TABLE 1II**

Analysis **of Sucrose Mono- and Diesters of Tallow, Coconut, Palm Kernel and Soybean** Oils



grade toluene (250 g in 250 mL toluene) and the solution poured onto the column. Three solvents were used for elution of components off the column. The mixed glycerides were eluted first from the columns using diethyl ether. The diesters were then eluted with dry n-butanone and the monoesters with *n*-butanone saturated with water. Successive 200 mL fractions were collected and portions of these evaporated on a Buchi evaporator. The progress of the separation was checked by thin layer chromatography. Each column of silica gel could be used twice. The large portions of eluate obtained were evaporated under reduced pressure and removal of residual solvent was effected in a vacuum oven. Analysis of the samples produced is given in Table Ill.

### **ANALYTICAL TECHNIQUES**

#### **Thin Layer Chromatography (TLC)**

This provides a method for rapid identification of the components present as well as visual estimation of relative concentrations. Glass plates (20  $\times$  20 cm or 5  $\times$  20 cm) coated with silica gel '60F254' of 0.25 mm layer thickness were used (Merck type 5737), without binder.

Chloroform was used to make up solutions of standard mixtures of glycerides or sucrose esters. Samples were spotted on the plates using fine capillary tubes.

Development of the plates was effected by the usual upward elution technique in a tank containing a saturated vapor of the eluting solvent (n-butanone saturated with water). Spots were visualized by heating the plate at 120 C after spraying with 50% sulfuric acid in methanol or with a solution of urea (1 g) and phosphoric acid (4.5 mL) in nbutanone (48 mL) saturated with water.

# **Gas Liquid Chromatography (G LC)**

The GLC analysis of the sucrose esters run was under the following conditions. Column: glass 6 mm od  $\times$  2 mm id  $\times$ 50 cm long. Stationary phase: 3% Dexsil 300 on Chromosob WHP (80-100 mesh). Carrier gas: high purity nitrogen. Temperature program: 1 min at 70 C, 20.3 min programmed at 16 C/min, 3 min at 380 C (final temperature). GLC: Varian Model 3700.

The samples were silylated with a mixture of trimethyl chlorosilane (TMCS) and *N,O-bis-(trimethylsilyl)-acetamide*  (BSA). Tetracosane was used as an internal standard in

quantitative analysis. (Tetracosane is not derivatized by the silylating agents.) Standard solutions for calibration were prepared using mixed glycerides and sucrose esters obtained from small-scale column chromatography on silica gel.

The chromatograms for the various mono- and diesters studied are shown in Figures 1 and 2.

Resolution of the monoester peaks is higher than that obtained for the diester peaks by GLC. There is a possibility that 'mixed' diester molecules (i.e., with two different acyl groups) and positional isomers are present, giving rise to peak broadening.

The fatty acids profiles of the esters were obtained by transesterifying in methanol containing sodium methoxide, and GLC analysis of the methyl esters obtained.

The quantitative estimation of the fatty acids present is given in Table II. In general, there is a correlation between the fatty acid profiles of mono- and diesters.

## **H LB Determination**

The following methods were employed for the determination of HLB: (a) water number (6), and (b) emulsification (1).

### **Water Number**

The determination of HLB by water number was carried out as described elsewhere (6) using a DMF/benzene mixture as solvent. Samples of surfactants covering the HLB range 2-16 were obtained from Honeywill-Atlas Limited, and used to obtain a calibration graph. The water number for each sample was determined as follows.

One g of surfactant was dissolved in 25 mL of a DMF/ benzene mixture containing 5 parts of benzene in 100 parts of dry DMF. This was titrated with distilled water until a permanent turbidity was obtained. Since the temperature increase from the heat of solution was significant, it was necessary to cool the flask after each addition of water to maintain the sample at  $20 \pm 1$  C throughout. At a temperature of 20 C, all results were reproducible to within 0.5 units of HLB. At HLB values greater than 16 the titration end-point is uncertain.

From these results, a calibration graph was obtained by plotting water number against HLB (Fig. 3). From this, the HLB of a sucrose ester or glyceride could be determined. (When using a mixture of impure sucroglycerides, it was necessary to filter the sample before titration. This pro-





FIG. 1. GLC chromatograms of mono- and diesters of tallow **and soybean** oil.

cedure produced no significant effect on the HLB value obtained.)

## **Emulsification**

The determination of HLB by emulsification (1) was carried out by dispersing the unknown emulsifier (5%) into an oil phase (15%) of known 'required HLB'. The known 'required HLB' oil phase was prepared by blending the appropriate ratios of pine oil ('required HLB' =  $16$ ) and cottonseed oil ('required HLB' =  $6$ ), enabling the range

6-16 HLB to be covered. 80% water was then added to the blend and homogenized for 1 min using a Janke-Kunkel Model KG homogenizer (Janke and Kunkel AG, West Germany; supplied by Sartorius Instruments, Coulsdon, Surrey, UK) at minimum speed. Emulsion stability was compared on a series of samples at 12 hr and 24 hr after preparation. The blend which performed best was assumed to have an HLB value approximately equal to the 'required HLB' of the oil phase. A large number of experimental emulsions were made, from which an average HLB value for the unknown was finally calculated.



FIG. 2. GLC chromatograms of mono- **and diesters** from coconut and palm kernel oil.

# RESULTS

## **Analysis of Purified Sucrose Esters Prepared by Column Chromatography**

The GLC analyses of the sucrose esters eluted are given in Table III.

The ratio of sucrose monester to sucrose diester in the starting product varied from 2.5:1 to 4:1, and a change in this ratio will affect the HLB value obtained for the sucrose mono-/diester blend.

The separation of glycerides from sucrose esters was efficient with very little glyceride present in the sucrose esters.

# **H LB Values of Sucrose Esters/Mixed Glyceride Blends (from Tallow IV = 45)**

The HLB values are quoted in Table IV. Figures obtained by the water number and emulsification procedures are



FIG. 3. **Calibration graph: Water number and** HLB.

quoted, and in general the second method produced slightly higher values. The values ranged from an HLB of 16+ for the sucrose ester to 4.0-5.0 for the mixed glycerides.

# **HLB Values of Blends of Sucrose Monoesters and Sucrose Diesters (from Tallow, IV = 45)**

By the column chromatography process, described above, it proved possible to separate the sucrose mono- and diesters. Using these samples of mono- and diesters, a series of blends were made up and the HLB value determined. The results are shown in Figure 4. Again, a linear relationship was observed between composition of the blend and the HLB value. In this instance the HLB range covered was 8.0-16+.

# **HLB Values of Blends of Sucrose Esters and Mixed Glycerides (from Coconut Oil, IV = 9)**

In a similar manner, a plot of HLB against composition was obtained for blends of coconut esters and mixed glycerides. The relationship was nonlinear in this case. The results are given in Table IV, and cover the range of HLB values from 8 to 16+.

#### TABLE IV

**Relationship** Between Sucrose Ester/Glyceride **Content and HLB of Sucroglycerides ex Tallow IV 45 and Coconut** Oil



aFormed o/w and w/o emulsions at different HLB values.



**FIG. 4. Graph of HLB against composition for sucrose monotallowate/sucrose ditallowate blends.** 

# **H LB Values of Blends of Sucrose Esters and Mixed Glycerides (from Palm Kernel Oil, IV = 19)**

The results are given in Table V and indicated that, because of the high HLB value of the mixed glycerides, only the range 13.5-16+ could be covered.

# **HLB Values of Blends of Sucrose Esters and Mixed Glycerides (from Soybean Oil, IV = 130)**

Here a rather wider range from HLB value 7-16+ could be covered. The results are given in Table VI.

#### **Physical Properties of Sucrose Esters from Tallow (IV 45)**

Listed in Table VII are the physical properties of sucrose tallowate esters, with a mono- to diester ratio of ca. 2.8:1.

The sucrose tallowate esters are slightly hygroscopic in the powder form, and melt to give a viscous liquid. They are moderately stable to thermooxidative degradation, but darken in color if heated above 120 C. They can be bleached by ultraviolet (UV) light. In an aqueous medium, alkaline hydrolysis occurs if the pH is higher than 8.5-9.0 units. It is believed that the sample used for physical property measurements contained a small amount of higher (tri-, tetra-) sucrose esters.

#### TABLE V

**HLB of Mixed Glycerides, Sucrose Mono- and Diesters and Sucroglycerides from Palm Kernel** Oil



#### TABLE VI

**HLB of Mixed Glycerides, Sucrose Mono- and Diesters and** Sucroglycerides **from Soybean** Oil

Blend no.	% Ester	% Mixed glycerides	<b>HLB</b>
	$100$ mono-	n	$16+$
	100 di-	o	$16+$
	100 (mono- and di-)	0	$16+$
2	70	30	16.0
3	60	40	14.6
	50	50	14.0
5	40	60	12.0
6	30	70	11.0
7	20	80	9.5
8	10	90	8.5
9	0	100	7.0

# **Solubility and Foam Height**

In Table VIII are listed the water-solubilities of the mono-, di-, and mixed esters prepared from the four triglycerides studied. In addition, foam heights (Ross-Miles, 21) are quoted for the mixed esters. The ratio of mono-/diesters varied from ca.  $2.5:1$  to  $4:1$ .

# **DISCUSSION**

# **HLB Values of Mixtures of Sucrose Mono- and Diesters (Tallow, IV = 45)**

The results shown in Figure 4 indicate that a range of HLB values from 8 to 16+ can be covered by varying the ratio of mono- and diester. Since it is possible, by varying the conditions of the extraction procedure given (20), to produce sucrose tallowate of a range of compositions, it is also possible to produce a series of emulsifiers covering the HLB range 8-16 from sucrose tallowates, without the necessity

#### TARI E VIII

Solubility in **Water and Foam Height of Sucrose Esters** 

#### TABLE VII

#### Physical **Properties of Sucrose Tallowate** Esters



<sup>1</sup>Two thermal transitions observed.

bThermooxidative stability. Darkening of color occurs above 120 C. Sucrose esters are susceptible to degradation by alkaline hydrolysis at pH 9 and above.

COleate esters are more soluble than palmitate/stearate esters.

of separating out the pure mono- and ditallowate esters. A similar process is possible with the sucrose esters of the other three oils studied, but the HLB range available is in the region above 16.

Where the use of sucrose esters, without the addition of mixed glycerides, as emulsifiers is desired, then the linear relationship between composition and HLB value of the sucrose tallowate esters enables the ratio of mono-/diester to be determined for the required HLB values.

#### HLB **Values of Sucrose Ester/Mixed Glyceride Blends**

If a wider HLB range is desired (4-16) than that obtained from the sucrose ester blends, then from the data given in Tables IV, V and VI an emulsifier of an HLB value required within this range can be prepared by blending the appropriate sucrose ester with the corresponding mixed glycerides. Again the relationship between composition and HLB value is linear for sucrose tallowate, enabling the composition of the blend required for a given HLB value to be determined. Narrower ranges of HLB values can be covered with the other three glycerides studied. Depending on the HLB value, either water-in-oil or oil-in-water emulsions can be obtained (22).

## **Relationship between Water Solubility and H LB of Sucrose Esters from Coconut Oil, Palm Kernel Oil and Soybean Oil**

The HLB values and water-solubilities obtained for the



SS = Slightly soluble; S = soluble; I = insoluble; E = emulsion; and D = dispersible.

mono- and diesters obtained by column chromatography are given in Tables IV-VIII. These results demonstrate very clearly the enhanced water solubility obtained for the monoesters by reducing the alkyl chain length from C16-C18 to C12, or by incorporating unsaturated alkyl chains. Thus the monoesters obtained from these three glycerides are water-soluble at 22 C, whereas the tallow monoester is insoluble. The diesters show the effect in a more pronounced manner; the presence of linoleic esters (with soybean oil) is insufficient to render the diesters water-soluble, even though their HLB value is above 16.

In the mixture of sucrose mono- and diesters from these triglycerides, a small portion of monoesters is sufficient to solubilize the diester, and the monoester/diester mixtures obtained are water-soluble at 22 C. The presence of up to 50% of the diester can be accommodated without the mixed ester product losing its water-solubility. Monoesters containing either C12 or unsaturated alkyl groups are effective solubilizers (23). The esters containing a high proportion of C12 alkyl groups are low softening point waxes and rather more difficult to handle than the sucrose tallowates, as are those containing a high proportion of unsaturated alkyl groups.

The monoesters obtained from these three glycerides act as solubilizers and are suitable for use in cosmetic and handcare products.

Using blends of mixed glycerides and sucrose esters of varying glyceride/ester ratios, it is possible to produce a series of emulsifiers covering the HLB ranges: from tallow (IV 45), HLB 4.0-16.0; from coconut (IV 9), HLB 8.0-  $16+$ ; from palm kernel (IV 19), HLB 13.5-16+; and from soybean (IV 130), HLB 7.0-16+.

With the blends made from mixed glycerides plus sucrose esters of tallow (IV 45), a linear relationship exists between composition and HLB value, Hence the compositon required for an emulsification can be determined by interpolation, if the HLB value required is known.

For sucrose esters prepared from tallow (IV 45), a linear relationship exists between composition (mono- to diester ratio) and HLB value. The HLB range covered is from 8 to 16+. Since the HLB values of the diesters of coconut, palm kernel and soybean are 16+, this relationship cannot be determined for mono-/diester blends.

A good agreement was found, in general, between HLB values determined by the water titration and emulsification techniques. The reproducibility of the HLB values determined by the water titration technique was ca. 0.5 units.

The water-solubility of the sucrose esters containing C12, C18 (2) and C18 (3) alkyl groups and their high HLB values (16) indicated that they could find application as solubilizers.

If it were desired to extend the HLB range in values below 8, using sucrose esters, then diesters of C18-C20 fatty acids would probably give HLB values of this order.

The ability to produce sucrose esters with high HLB values (16+) and either water-solubility or oil-solubility indicated that as emulsifiers for water-in-oil emulsions they should possess unique properties.

From the water-solubility, HLB value and fatty acid profiles, it can be seen that solubilization effects occur either when: (a) sucrose monoesters containing C12 alkyl chains in the ester group are present; (b) sucrose monoesters containing dienoic or trienoic acyl groups are present.

Thus the sucrose esters prepared from coconut, palm kernel, or soybean oil are water-soluble at 22 C, even though they contain sucrose palmitate and stearates which are insoluble in water at 22 C. It is clear that the sucrose monoesters confer these properties, solubilizing the diesters and insoluble C16-C18 monoesters.

#### ACKNOWLEDGMENT

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