

# Relationship of Chemical Classification and Hydrophile-Lipophile Balance of Surfactants to Enhancement of Foliar Uptake of Iron

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Seventy-three surfactants were selected for investigation, to assess the relation of surfactant hydrophile-lipophile balance (HLB) and surfactant chemical class upon accumulation of Fe from the chelate, FeNaHDTPA, by chrysanthemum foliage. An HLB in the range of 15 to 16 is the first criterion for selecting an effective nonionic surfactant. Chemical class should follow. The HLB relationship does not exist in the case of ionic surfactants; therefore an

investigation by chemical class is the first tool. An investigation of representative surfactants in six ionic class categories indicated that the sulfate, amide, and quaternary ammonium chloride categories were relatively inactive. A few active surfactants were found among the sulfonated and amine categories. The most active category was phosphate surfactants. Ten surfactants increased Fe accumulation 2.5-fold or more and were not phytotoxic.

All essential plant nutrients may be, at least partially, absorbed through the foliage of plants. Wittwer (1964) listed the nutrients according to foliar absorption rate as follows: urea  $N > K \approx Ca > Mg > Mn \approx Zn > Cl > P \approx S > Fe \approx Mo$ .

Foliar uptake of nutrients can be stimulated by temperature, oxygen level, and light energy where the process is an active one (Yamada *et al.*, 1964). Uptake may be stimulated by the presence of other chemicals in the spray. Urea has been used in combination with  $FeSO_4$  and Fe chelates to enhance correction of Fe chlorosis (Wallace 1962) and P uptake (Yamada, 1962). Yamada (1962) suggests that the enhancement by urea is due to an increase in the permeability of the cuticular membrane.

Certain surfactants are also credited with increased nutrient uptake. Hager and Seeley (1965) found that a 0.03% concentration of Triton B 1956 increased foliar uptake of Fe from several inorganic and organic Fe salt sprays applied to rose plants by observing the reappearance of green color. Comparing individually a nonionic, a cationic, and an anionic surfactant at 0.1% in solution with  $NH_4H_2PO_4$ ,  $NaH_2PO_4$ , or  $KH_2PO_4$ , Boroughs and Labarca (1962) found that in all cases the surfactant increased foliar uptake and translocation of P by coffee plants. Fisher and Walker (1955) found a sevenfold increase in P absorption by apple leaves using Triton X-100. Wallace (1962) increased Fe uptake in citrus foliage by using Vatsol OT at a concentration of 0.05%.

Not all surfactants are effective. Even those which are effective in one situation may not be effective in other situations. Tween 80 was reported by Swanson and Whitney (1953) to reduce P uptake in bean plants. Barrier and Loomis (1957) reported no increase in P uptake in soybeans from the use of 0.1% Triton B-1956.

Out of the foregoing irregularities and the great potential surfactants have as an adjunct in nutrient sprays, a need arose for developing a method of screening the more than 3000 surfactants.

Surfactants have often been classified according to their hydrophile-lipophile balance (HLB). Theoretically this value can range from slightly above 0 for a molecule which is mostly oil soluble to slightly less than 20 for a molecule which is almost completely water soluble (Atlas Chemical

Industries, 1963). Researchers, including Gakenheimer (1966) and Behrens (1964), have correlated surfactant HLB with the ability to improve herbicidal activity. Since surfactants enhance foliar uptake of nutrient ions, and it has been possible to relate surfactant HLB to effectiveness of other biological sprays, surfactant HLB may be related to nutrient ion uptake. The objective of this study was to assess the role of surfactant HLB and chemical classification of surfactant upon the accumulation of Fe by chrysanthemum foliage.

## MATERIALS AND METHODS

A series of four studies was carried out. In the first, HLB values of 4.3, 8.1, 12.3, and 15.0 were obtained with Span 80 (HLB 4.3) alone or in combination with Tween 80 (HLB 15.0) at a surfactant concentration of 1%. These surfactants, with all others used in the investigation, are further described in Table I. To these spray solutions was added Fe in the form of the chelate, ferric sodium hydrogen diethylenetriamine pentaacetate (FeNaHDTPA), at a concentration of  $6.63 \times 10^{-3} M$  FeNaHDTPA (370 mg. of Fe per liter). The spray solution was labeled with  $^{59}Fe$  to yield a specific activity of  $1.91 \times 10^{-3} \mu c.$  per  $\mu g.$  of Fe. Plants were treated with these sprays to investigate the possible relationship between nonionic surfactant HLB and Fe accumulation.

A concentration of 1% of surfactant was selected for study on the basis of an investigation of Tween 80 concentrations up to 5%. The rate of Fe accumulation became nearly linear at a surfactant concentration of 1% (Figure 1). Higher concentrations were avoided because of phytotoxicity of some surfactants. Results from tests with Aerosol AY, Gafac PE 510, and Pluronic L 121 confirm these data.

In the second study an investigation into the relationship of surfactant HLB to Fe accumulation was broadened to include other nonionic classes as well as several ionic classes of surfactants. Each experiment included a control treatment sprayed with  $1.66 \times 10^{-3} M$  FeNaHDTPA (92.5 mg. of Fe per liter) labeled with  $^{59}Fe$  at a specific activity of  $2.71 \times 10^{-3} \mu c.$  per  $\mu g.$  of Fe. The other treatments involved the same Fe solution in combination with a different surfactant for each treatment at a final concentration of 1%. Increases in Fe accumulation above the control level were related to ionic form and HLB.

In the third study representative surfactants from eight

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**Table I. Chemical Classification, Source, Phytotoxicity, and Accumulation Index of Surfactants**

Chemical Category and Class	Surfactant	Ionic Form <sup>a</sup>	Source <sup>b</sup>	Burn	AI	HLB
Nonethoxylated hydrocarbons						
1. Sorbitan tristearate	Armotan TS	N	3	No	0.7	2.1
2. Sorbitan monooleate	Span 80	N	4	No	0.8	4.3
3. Sorbitan monolaurate	Armotan ML	N	3	No	1.5	...
4. Diglycol laurate		N	3	No	1.3	...
5. Glyceromonooleate		N	3	No	1.3	3.8
6. Sucroglyceride	Sucroglyceride T-110	N	8	No	1.5	...
Ethoxylated hydrocarbons						
7. Ethoxylated monooleate	PEG 200 MO	N	3	No	1.2	7.9
8. Ethoxylated monooleate	PEG 600 MO	N	3	No	2.1	13.5
9. Ethoxylated monooleate	PEG 1000 MO	N	3	No	3.7	15.4
10. Ethoxylated monostearate	PEG 600 MS	N	3	No	2.8	13.6
11. Ethoxylated monostearate	PEG 1000 MS	N	3	No	2.4	15.6
12. Ethoxylated distearate	PEG 1540 DS	N	3	No	2.9	14.8
13. Ethoxylated distearate	PEG 6000 DS	N	3	No	1.7	18.4
14. Ethoxylated oleic acid	Ethofat 0-15	N	3	Yes	1.8	...
15. Ethoxylated alcohol	Alkanol OE	N	7	Yes	1.2	...
16. Ethoxylated alcohol	Alkanol OJ	N	7	Yes	1.5	...
17. Ethoxylated alcohol	Tergitol 15-S-9	N	14	Yes	1.4	...
18. Polyalkylene glycol ether	Tergitol XH	N	14	No	2.0	...
19. Ethoxylated polyoxypropylene	Pluronic L 31	N	17	No	1.2	...
20. Ethoxylated polyoxypropylene	Pluronic L 121	N	17	No	3.5	...
21. Ethoxylated polyoxypropylene	Pluronic F 38	N	17	No	1.9	...
22. Ethoxylated polyoxypropylene	Pluronic F 127	N	17	No	1.6	...
23. Ethoxylated sorbitan monolaurate	Tween 21	N	4	No	1.5	...
24. Ethoxylated sorbitan monooleate	Tween 80	N	4	No	1.8	15.0
25. Ethoxylated polysorbate monostearate	Armotan PMS-20	N	3	No	2.0	...
26. Ethoxylated sorbitol hexaoleate	G 1096	N	4	No	2.1	...
27. Ethoxylated trimethyl nonyl ether	Tergitol TMN	N	14	Yes	2.6	...
28. Ethoxylated isoocetyl phenoxy polyethoxy ethanol	Triton X-100	N	13	No	1.3	...
29. Ethoxylated nonyl phenyl ether	Tergitol NP-14	N	14	No	1.8	...
30. Ethoxylated nonyl phenyl ether	Tergitol NP-35	N	14	No	2.4	...
31. Ethoxylated nonyl phenyl ether	Tergitol NP-44	N	14	No	2.2	...
32. Ethoxylated nonyl phenyl ether	Tergitol TP-9	N	14	No	2.5	...
33. Ethoxylated hydroabietyl alcohol	Surfactant AD 400	N	11	No	2.8	...
34. Ethoxylated pale wood rosin	Surfactant AR 150	N	11	No	2.6	...
Sulfates						
35. Na 2-ethylhexyl sulfate	Tergitol Anionic 08	A	14	No	2.0	...
36. Na heptadecyl sulfate	Tergitol Anionic 7	A	14	No	1.5	...
37. Na lauryl sulfate	Duponol WAQ	A	7	No	1.0	...
38. Na lauryl sulfate	Duponol ME	A	7	No	1.4	...
39. Ethoxylated Na sulfated nonyl phenol	Alipal CO-433	A	9	No	1.5	...
40. Ethoxylated NH <sub>4</sub> alkyl sulfate		A	12	Yes	1.8	...
Sulfonates						
41. Polymerized organic salt of sulfonic acid	Darvan 1	A	15	No	0.5	...
42. Polymerized organic salt of sulfonic acid	Darvan 2	A	15	No	0.5	...
43. Na polymerized alkyl naphthalene sulfonic acid	Daxad 15	A	10	No	0.6	...
44. Na polymerized alkyl naphthalene sulfonic acid	Daxad 30	A	10	No	1.0	...
45. Dodecyl Na benzene sulfonate	Ultrawet K	A	2	No	2.7	...
46. Na diamyl sulfosuccinate	Aerosol AY	A	1	Yes	2.9	11.6
47. Na bistridecyl sulfosuccinate	Aerosol TR	A	1	No	1.6	7.0
48. Na distearyl sulfosuccinate		A	1	No	2.2	5.6
49. Na didodecyl sulfosuccinate		A	1	No	1.6	7.4
50. Na didecyl sulfosuccinate		A	1	No	1.4	8.2
51. Na diisopropyl sulfosuccinate		A	1	No	1.4	13.9
52. Oleic ester of Na isethionate	Igepon AP-78	A	9	No	1.8	...
53. Na <i>N</i> -methyl- <i>N</i> -oleoyltaurate	Igepon T-33	A	9	No	1.8	...
Amines						
54. <i>n</i> -Octadecylamine	Armeen 18D	C	3	No	3.3	...
55. Ethoxylated alkylamine	Peregal TW	C	9	No	2.3	...
56. Diethoxylated alkylamine	Ethomeen T/12	C	3	Yes	1.4	2.5
57. Diethoxylated alkylamine	Ethomeen T/15	C	3	Yes	2.8	9.0
58. Diethoxylated alkylamine	Ethomeen T/25	C	3	No	1.5	13.0+
59. Disoyamine	Armeen 2S	C	3	Yes	2.5	...
60. 1-Hydroxyethyl-2-heptadecenyl glyoxalidine	Amine 220	C	14	No	1.1	...
61. Product of cocoamine and crotonic acid	Armeen SZ	Z	3	No	1.1	...
62. Amine oxide	Aromox C/12	C	3	Yes	2.7	...
Amides						
63. C <sub>12</sub> to C <sub>14</sub> ammonia amide	Amide 20	C	12	Yes	2.2	...
64. C <sub>12</sub> diethanolamide	Amide 22	C	12	Yes	1.5	...

(Continued)

**Table I. Chemical Classification, Source, Phytotoxicity, and Accumulation Index of Surfactants (Continued)**

Chemical Category and Class	Surfactant	Ionic Form <sup>a</sup>	Source <sup>b</sup>	Burn	AI	HLB
Quaternary ammonium salts						
65. Dialkyl dimethyl ammonium chloride	Arquad 2C-75	C	3	No	1.7	13.0+
66. C <sub>12-18</sub> trimethyl ammonium chloride	Arquad 12-33	C	3	Yes	1.9	...
67. Cetyl betaine	Product BCO	Z	7	No	1.0	...
68. <i>p</i> -Diisobutyl phenoxyethoxyethyl dimethyl benzyl ammonium chloride	Hyamine 1622	C	13	No	0.4	13.0+
Phosphates						
69. Organic phosphate ester	Gafac RS-710	A	9	No	2.5	...
70. Organic phosphate ester	Gafac RM-710	A	9	No	1.9	...
71. Organic phosphate ester	Gafac PE-510	A	9	No	3.2	...
72. Lecithin	Centrophill IP	Z	5	No	3.0	7.0
73. Ethoxylated alkyl phosphate	Zonyl A	N	7	Yes	2.6	...
<sup>a</sup> A anionic, C cationic, N nonionic, Z zwitterionic.						
<sup>b</sup>						
1. American Cyanamid Co.	7. E. I. du Pont de Nemours & Co.	13. Rohm and Haas Co.				
2. Atlantic Refining Co.	8. Fats and Protein Research Foundation	14. Union Carbide Chemical Co.				
3. Armour & Co.	9. General Aniline Co.	15. R. T. Vanderbilt Co., Inc.				
4. Atlas Powder Co.	10. W. R. Grace Co.	16. Witco Chemical Co.				
5. Central Soya	11. Hercules Powder Co.	17. Wyandotte Chemicals Corp.				
6. Diamond Alkali Co.	12. Procter & Gamble Distributing Co.					

broad categories of surfactant classes were tested as in the second study, to determine relationships between chemical structure and Fe accumulation enhancement.

Throughout this investigation the plant material used was *Chrysanthemum morifolium*, between 3 and 5 weeks in age and in a vegetative state. In the first two studies the cultivar Indianapolis White was used, while in the third it was Indianapolis Yellow. Plants in the first three studies were not deficient in Fe.

The fourth study was established to verify the results of the earlier studies by using an Fe-deficient plant. Giant Betsy Ross was selected because of its tendency toward Fe deficiency. The average level of Fe in these plants at the start of treatment was 44 p.p.m. based on the dry weight of the leaf and stem tissue.

Six replications of one plant each were used for each treatment. Prior to treatment, the soil surface in 3-inch Styrofoam cups containing the plants was coated with paraffin to prevent root uptake of the spray solutions. The upper and lower surfaces of all leaves on each plant were sprayed with 50 ml. of solution—i.e., beyond the point of runoff. A separate Windex household sprayer was used for each treatment.

After spraying, the plants were set on a laboratory bench at an air temperature of 21° C. to dry. When dry, they were returned to the greenhouse (usually 1 hour).

The greenhouse temperature was maintained at 16° C. nights and 21° C. days. The plants were grown for 72 hours in shallow trays of water such that water could be absorbed through the base of the pot, then taken to the laboratory, cut off at the soil level, washed for 30 seconds in 0.2N HCl, and rinsed under tap water.

The washing procedure was developed from a preliminary experiment in which plants were sprayed with a solution containing  $6.63 \times 10^{-8}M$  FeNaHDTPA (<sup>59</sup>Fe-labeled) and 1% surfactant of the sulfosuccinate class, which is highly effective in enhancing foliar uptake of Fe. After the spray dried, the plants were washed with 0.2N HCl for 10 to 180 seconds. The wash solutions were counted and results (Figure 2) indicated that surface deposits of <sup>59</sup>Fe NaHDTPA were removed within 10 seconds. Therefore, a 30-second wash in 0.2N HCl was accepted as more than adequate to remove all Fe left on the surface of the leaves.

Washed plants were ashed at 500° C. for 12 hours, and

the ash was dissolved in 5 ml. of 20% HCl, evaporated to dryness, and finally taken up in 1 ml. of 1.5N HNO<sub>3</sub>. The solution was passed through fine filter paper and 0.1 ml. was transferred to 1.5 sq. inches of filter paper. When dry, this paper was placed in 20 ml. of scintillation solution (0.5% PPO plus 9.93% POPOP in toluene) and counted four times for 30 minutes.

After all counts for background were corrected, six replications were averaged within each treatment. Each surfactant treatment average was divided by the control treatment average to determine accumulation index (AI) values. When the activity of the control is equal to the surfactant treatment, the AI value is 1.0. AI values of 0.5 and 2.0 indicate one half and twice as much Fe uptake in the surfactant treatment as the control treatment.

## RESULTS

**Study 1.** Iron uptake increased in direct proportion to HLB values in the range of 4.3 to 15.0 (Figure 3). The optimum HLB for this nonionic sorbitan monooleate family, therefore, falls above 15.0.

**Study 2.** The AI values for each surfactant are listed in Table I. In Figure 4 the AI values of members of three similar nonionic surfactant classes (ethoxylated fatty acids) are plotted against HLB values. If the three classes are treated as one and a curve of best fit is drawn to illustrate the relationship of AI to HLB, an optimum HLB value of 15 to 16 is indicated. There are not sufficient points to establish optimum HLB values for each class; however, the existing points indicate that optimum values for individual classes could range from HLB 13 to 16.

In Figure 5 an over-all AI-HLB relationship is presented for the three classes illustrated in Figure 4, two additional nonionic classes, and the sorbitan monooleate data from study 1. Again, the curve of best fit shows an optimum HLB value in the range of 15 to 16.

Twelve ionic surfactants representative of several classes are presented in Figure 6. The HLB values of three of these were not available; however, their solubility properties indicated that they were 13 or above. These three are presented in a separate column on the right side of the figure. It is readily apparent that no single curve can fit all the points. Apparently the HLB of ionic surfactants is not related to AI in such a manner that several classes may

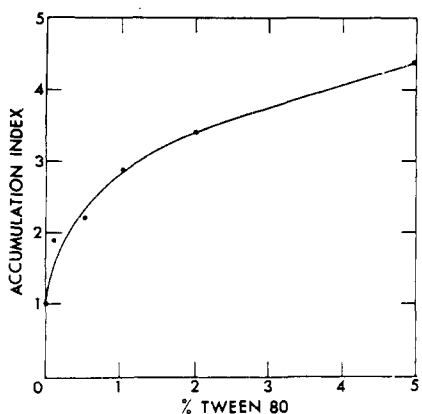


Figure 1. Influence of Tween 80 concentration in foliar sprays upon Fe accumulation

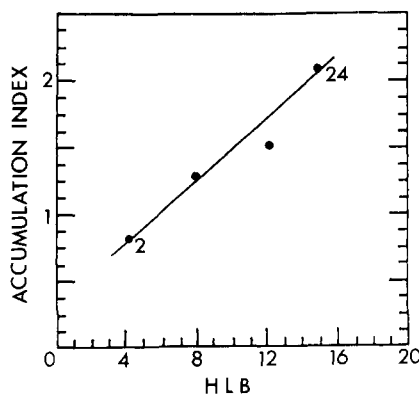


Figure 3. Relationship of HLB within sorbitan monooleate class and foliar Fe accumulation

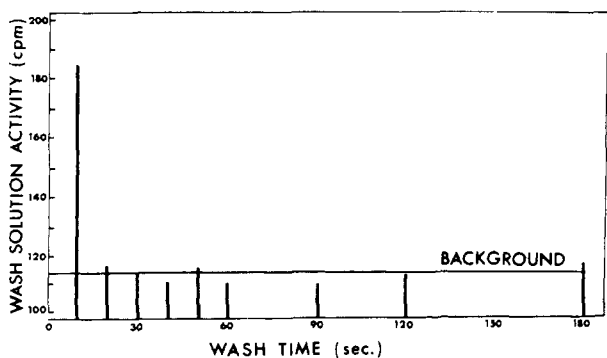


Figure 2. Levels of  $^{59}\text{Fe}$  activity in wash solutions after 10 to 180 seconds of wash time

be represented by a single optimum HLB value as is the case for nonionic surfactants.

Six surfactants plotted in Figure 6 are closely related in chemical class. All are dialkyl sulfosuccinates. The alkyl chain length is specific for each surfactant and ranges from three to 18 carbons in length. Although six classes were involved, it was felt that they were similar enough that an AI-HLB relationship indicative of a single class might be found. The results are plotted in Figure 7. Apparently the six classes do not fit a single AI-HLB relationship.

**Study 3.** The results of screening 73 surfactants representative of eight broad chemical categories are presented in Table I. In general the nonethoxylated hydrocarbons proved to be poor adjuncts for Fe chelate sprays. On the other hand, eight of the 28 ethoxylated hydrocarbons enhanced Fe uptake in excess of 2.4 times. These eight represent five groupings of chemical classes: ethoxylated fatty acids, ethoxylated polyoxypropylene, ethoxylated trimethyl nonyl ether, ethoxylated nonyl phenyl ether, and ethoxylated diterpenes.

The sulfate surfactants were relatively poor, while members of two sulfonated classes, the dialkyl sulfosuccinates and alkylbenzene sulfonate, were very effective.

Within one category of N-containing surfactants, the amines, Fe accumulation was stimulated in excess of 2.5 times, while in two other categories, the amides and quaternary ammonium salts, it was not.

All phosphate surfactants tested yielded AI values of 2.5 or greater except Gafac RM 710, which had a value of 1.9. This is the only chemical category where all candidates tested greatly enhanced Fe uptake.

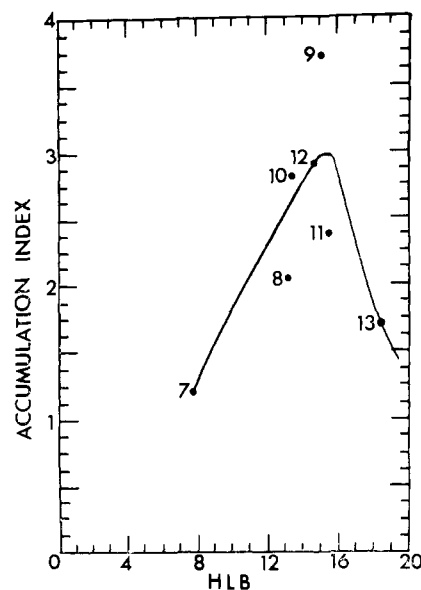


Figure 4. Relationship of HLB within three closely related ethoxylated fatty acid classes and foliar Fe accumulation

**Study 4.** The results from this study are presented in Table II. Values for nine surfactants are contrasted between trials on normal Indianapolis White plants and on Fe-deficient Giant Betsy Ross plants to determine if the earlier relationships existed for the latter. In general, AI values are smaller for the Fe-deficient cultivar. However, independent of Pluronic L 121 and Armeen 18D, the relative ranking of the surfactant AI values remains the same. This indicates that conclusions drawn from the normal plant trials apply to deficient plants as well.

Pluronic L 121 and Armeen 18D did not go into true solution nor form a true dispersion. This accounts for the low degree of reproducibility. The lower AI values witnessed for Fe-deficient plants could be due either to a greater resistance to Fe uptake by deficient plants or to a greater ease of Fe accumulation in deficient plants, which in turn suppresses the role of the surfactant. More work is needed on this point.

Based on the conditions of these studies the following 10 surfactants in decreasing order are found to be the best adjuncts to Fe chelate sprays: PEG 1000 MO, Gafac PE 510, Centrophill IP, PEG 1540 DS, Surfactant AD 400, PEG 600 MS, Ultrawet K, Surfactant AR 150, Tergitol TP-9, and Gafac RS 710. PEG 1540 DS and PEG 600 MS were first dissolved in sufficient 2-propanol to yield a final concentration of 10% and then added to water.

#### DISCUSSION

Thirteen surfactants were involved in two separate, but identical, experiments. The standard deviation of each pair of observations was determined and expressed as a decimal fraction of the mean. These values range from 0.025 to 0.283 and the average of all 13 is 0.179 or 17.9%.

Variation of this magnitude is typical in this type of surfactant response. It indicates that the AI values in Table I, on the basis of one standard deviation, may be taken to be within  $\pm 17.9\%$  of the true value. The assumption here is that all surfactants behave similarly.

To bring some of the surfactants (1, 5, 10, 12, 13, 48, 56,

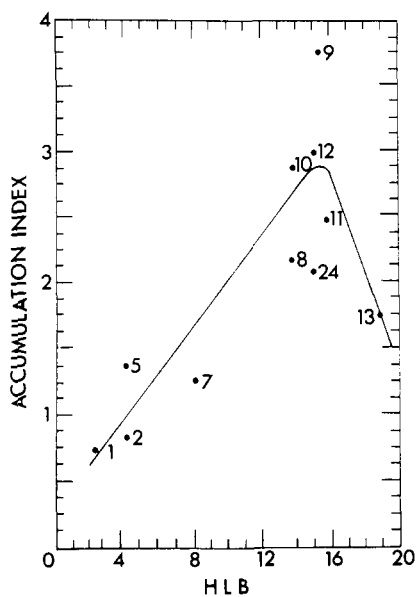


Figure 5. Relationship of HLB of 11 non-ionic surfactants, representative of six classes, and foliar Fe accumulation

and 57) into aqueous solution it was first necessary to dissolve them in sufficient 2-propanol to give a final concentration of 10%. To determine the influence of 2-propanol on Fe uptake, a 10% solution of 2-propanol was tested and found to have an AI of 1.06. It was concluded that 2-propanol alone has no influence upon Fe accumulation.

A surfactant HLB to Fe accumulation relationship cannot be demonstrated for ionic surfactants, possibly because HLB is not taking into account all of the pertinent characteristics of the ionic surfactants. It is difficult, if possible, to assign a workable HLB value to ionic surfactants.

Enhancement of Fe accumulation was still increasing at Tween 80 concentrations up to 5%. This value is well beyond the critical micellar concentration (CMC), indicating that the action of surfactants is not solely that of altering surface phenomena at the leaf surface—i.e., lowering surface tension. It is more likely that surfactants are most active in either the cuticle, where a dual solubility in the wax platelets and the aqueous cutin matrix is important, or at the point of the outer epidermal cell membrane, where a dual solubility again is important, this time in the lipoidal and proteinaceous layers of the membrane.

The importance of the characteristic of polarity is pointed out in the third study, where surfactant effectiveness is frequently increased by the presence of an ethylene oxide

Table II. Fe Accumulation Indexes for Normal and Fe-Deficient Cultivars of *Chrysanthemum morifolium*

Surfactant	AI	
	Normal	Fe-deficient
PEG 1000 MO	3.7	2.5
Pluronic L 121	3.5	1.4
Armeen 18 D	3.3	0.7
Gafac PE 510	3.2	1.9
Surfactant AD 400	2.8	1.9
Surfactant AR 150	2.6	1.5
G 1096	2.1	1.5
Product BCO	1.0	1.4
Darvan 1	0.5	0.7

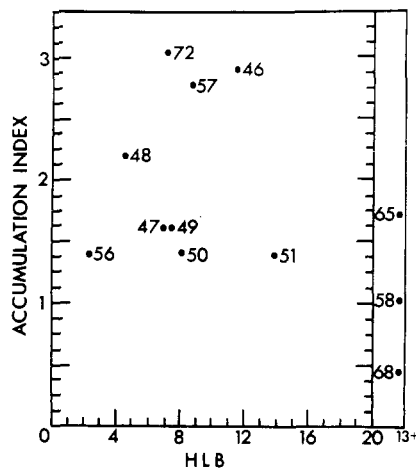


Figure 6. Relationship of HLB of 12 ionic surfactants, representative of 10 classes, and foliar Fe accumulation

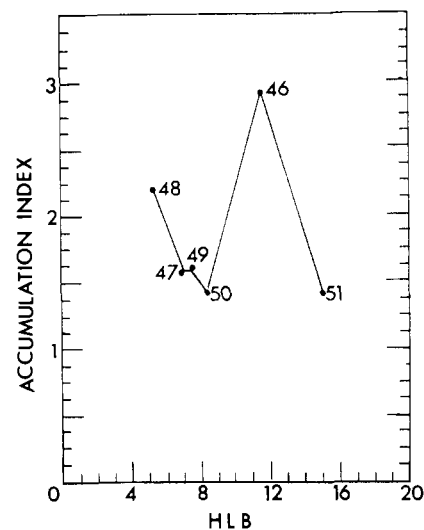


Figure 7. Relationship of HLB of six closely related anionic, dialkyl sulfosuccinate classes, and foliar Fe accumulation

chain which in turn increases HLB, or water solubility. Less polar surfactants above HLB 16 result in reduced effectiveness.

The one category of surfactants showing greatest activity are those containing phosphate. It is conceivable that activity of these compounds is in the lipoidal fraction of the plasmalemma which abounds in phospholipids. It is even more feasible that a surfactant such as Centrophill IP (AI 3.0) should be active in this region, since lecithin—i.e., Centrophill IP—is a constituent of the phospholipid group.

The role of surfactants in altering surface tension at the leaf-solution interface should not be completely disregarded. Out of 73 surfactants tested only nine were not identified with some level of enhancement of Fe uptake. Since these surfactants were representative of all categories tested, it is likely that activity is not entirely due to specific solubility or carrier properties of the surfactant but is due in a minor degree to the facility to alter surface phenomena.

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