

# Functional Polymers for Colloidal Application. IX. The Dispersing Ability of Anionic and Zwitterionic Aniline–Formaldehyde Condensates to Dyes

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## SYNOPSIS

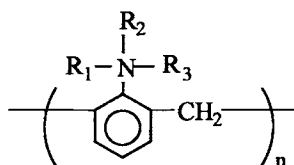
Anionic and zwitterionic aniline–formaldehyde condensates, *N*-(3-sodium propylsulfonate) aniline–formaldehyde condensates (PSAF), and *N,N*-dimethyl-*N*-(3-propylsulfonate) anilinium–formaldehyde condensates (DPSAF) are used as dispersants to disperse dyes (C. I. Navy Blue 79 and C. I. Brown 1), compared with the conventional dye dispersants—naphthalenesulfonate formaldehyde condensates (NSF). The dispersing properties of the condensates are assessed by the viscosity method, scanning electron microscopy (SEM), the final volume fraction after sedimentation, and adsorption behaviors. Compared to DPSAF, the PSAF and NSF with a lower molecular weight (PSAF-L, NSF-L) show significantly lower apparent viscosities of the paste of Navy Blue 79. Comparing the results of the viscosity method with those of SEM and the final volume fraction after sedimentation, it can be concluded that a better-dispersed paste displayed by SEM shows a lower viscosity and a smaller final volume fraction. The amount of NSF-L adsorbed on Navy Blue 79 is significantly higher than that of PSAF-L and DPSAF-H. However, the sulfonate on PSAF-L can be extended more deeply toward the bulk phase compared to that on NSF-L so that PSAF-L can result in a high effective thickness. These adsorption phenomena are used to interpret the dispersing ability. For Brown 1, the anionic dispersants can reach a similar minimum viscosity independent of their molecular weight, but the PSAF with high molecular weight shows a remarkably low optimum concentration (ca. 3% compared to PSAF-L 9%, NSFs 5%). © 1993 John Wiley & Sons, Inc.

## INTRODUCTION

Because of the diversified application in both conventional industries<sup>1–3</sup> (such as coatings, pigments, and agriculture) and high-tech industries<sup>4</sup> (such as magnetic slurry and photo applications), dispersing technology has been widely studied both for academic and industrial purposes. A system of particles dispersed in solution is thermodynamically unstable, thus dispersants are normally used to stabilize the system. The main function of the dispersant is to provide the particles with an electrostatic barrier<sup>5</sup> and/or with steric hindrance<sup>6</sup> to prevent the co-

agulation of particles. In addition to surfactants and polymers, naphthalenesulfonate formaldehyde condensates (NSF) are used as industrial dispersants. Because of the strong interaction of NSF with polar and nonpolar surfaces, NSF has been widely used for dispersing dyes, coal, and cement in aqueous systems.<sup>7–11</sup> Many kinds of dyes are constituted by an aromatic ring and an azo group.<sup>12</sup> For dispersing the particles of the dye, the adsorption of NSF on the dye arises mainly from the interaction between the naphthalene ring and the surface of dye. Aniline has an aromatic ring and amino group, so it can be expected to interact efficiently with the dye particles. In addition to the azo group, a dye always contains polar groups such as a nitro, ester, and/or hydroxyl group. If the amino group of the aniline is converted into an ionic group, the interaction between aniline and dye will be influenced.

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**PSAF** :  $R_1 = \text{none}$ ,  $R_2 = \text{H}$ ,  $R_3 = -(\text{CH}_2)_3\text{SO}_3\text{Na}$

**DPSAF** :  $R_1, R_2 = -\text{CH}_3$ ,  $R_3 = -(\text{CH}_2)_3\text{SO}_3^-$

**Scheme 1** The structure of *N*-(3-sodium propyl sulfonate) aniline-formaldehyde condensate (PSAF) and *N,N*-dimethyl-*N*-(3-propyl sulfonate) anilinium-formaldehyde condensate (DPSAF).

In this study, anilines react with formaldehyde to form condensates first, and, then, the amino group reacts with propane sultone to carry an anionic or a zwitterionic hydrophilic group (as shown in Scheme 1). The anionic and zwitterionic aniline-formaldehyde condensates are used as dispersants to disperse dye in the aqueous solution and the results were compared to NSF. The dispersing ability is assessed by viscosity methods and scanning electron microscopy (SEM) and correlated with the adsorption phenomena. These results are used to illustrate the influence of the charge and the structure of dispersants on their dispersing ability.

## EXPERIMENTAL

### Materials

Aniline (Wako, EP grade), *N,N*-dimethylaniline (Wako, EP grade), formaldehyde solution (Merck,

37 wt %, EP grade), and propane sultone (Tokyo Kasei, GR grade) were used without further purification. High and low molecular weight NSFs (Suparex YM, K powder) were also used as dispersants for comparison. C. I. Navy Blue 79 (E-Hwa Co., with BET surface area  $4.85 \pm 0.03 \text{ m}^2/\text{g}$ ) and C. I. Brown 1 (E-Hwa Co., with BET surface area  $1.53 \pm 0.04 \text{ m}^2/\text{g}$ ) were used as supplied. The water used in these experiments is ion-exchanged and distilled, with the pH value between 6.8 and 7.2 and the conductivity of  $4.4 \times 10^{-6} \Omega^{-1} \text{ m}^{-1}$ .

### Methods

#### *Synthesis and Characterization of Condensates*<sup>13</sup> (Table I)

(a) **Synthesis of *N*-(3-sodium propylsulfonate) Aniline-Formaldehyde Condensates (PSAF) (as Scheme 1)**. In a 500 mL reaction kettle, formaldehyde (37% 48.7 g) was added to the mixture of aniline (93 g) and HCl (28% 39.4 g) within 50 min. Condensation was carried at 95–100°C for 1 and 2 h. The temperature was decreased to 60°C and the mixture was neutralized with NaOH (38.8%, 32.2 g) and washed in warm water three times followed by the separation of the oil phase from the aqueous layer. The unreacted aniline was separated and water was removed at 170°C under the reduced pressure of 15 mmHg. Then, the residue was dissolved in the mixed solvent of 90% phenol and 10% acetone (150 g). To this solution, propane sultone (130 g) was added dropwisely, and the temperature was increased to 70–80°C to react for 5 h. After reaction, the mixture was poured into acetone. The precipitation was dissolved in water and the pH value of the solution was adjusted to 9–10. The water was evaporated and the residue was reprecipitated in acetone again to obtain the product of about 154 g.

**Table I** Reaction Conditions for Synthesizing the Dispersants<sup>a</sup>

Dispersants	HCHO/Monomer/HCl (Molar Ratio)	Reaction Time (h)	
		Condensation <sup>b</sup>	Opening <sup>c</sup>
PSAF-L	0.4/1.0/0.3	1.0	5.0
PSAF-H	0.6/1.0/0.3	2.0	5.0
DPSAF-L	3.0/1.0/1.0	5.0	8.0
DPSAF-H	3.0/1.0/1.0	10.0	8.0

<sup>a</sup> The data are from Ref. 13.

<sup>b</sup> The reaction temperature for condensation was maintained at 95–100°C.

<sup>c</sup> The reaction temperature for the ring-opening of propane sultone was maintained at 70–80°C.

**(b) Synthesis of *N,N*-Dimethyl-*N*-(3-propylsulfonate) Anilinium-Formaldehyde Condensates (DPSAF) (as Scheme 1).** *N,N*-Dimethylaniline (61.0 g) and HCl (30% 51.0 g) was mixed, and formaldehyde (37% 121.6 g) was added to the mixture during 50 min and the mixture was heated up to 95–100°C to reflux 8 or 10 h. Then, the temperature was decreased to about 60–70°C, and an aqueous solution of NaOH was added to neutralize HCl. The mixture was washed in water, and the water phase was pouring out and dried in a vacuum oven for 5 h, then the residue was dissolved in the mixed solvent of 90% phenol and 10% acetone (150 g). To this solution, propane sulfone (61.0 g) was added dropwisely, and the mixture was heated up to 70–80°C to react for 5 h. After the reaction, part of the solvent of the mixture was evaporated in a rotavapor under reduced pressure. Then, the concentrated mixture was poured into acetone for reprecipitation. The precipitate was filtered and dried, and the product of 95.0 g was obtained.

**(c) Characterization of Condensates.** PSAF and DPSAF dissolved in D<sub>2</sub>O were characterized by <sup>1</sup>H-NMR (Bruker WP 100). For PSAF, the protons on the benzene ring appeared at  $\delta = 6.5\text{--}8.0$  ppm and the methylene group between two benzene rings was found at  $\delta = 3.5\text{--}4.0$  ppm, and the other methylene groups—N(CH<sub>2</sub>)<sub>3</sub>—were distributed at 1.5–3.5 ppm.<sup>14</sup> Molecular weight distributions of PSAF (sodium salts) and DPSAF obtained under different reaction conditions were compared by the GPC method.<sup>15,16</sup> Two GPC columns (Waters Ultrahydrogel-120, 250, with length 30 cm, inner diameter 7.8 mm, and fully porous highly cross-linked hydroxylated polymer as packing material) were combined in series for the measurements. The water (GPC grade) was used as the mobile phase.

### Assessments of the Dispersing Abilities of the Condensates

The dispersed dye/water systems were prepared by using different condensates as dispersants. Table II shows the formulation used in these experiments. For example, the dispersant was dissolved in water (6 g) and mixed with the dye (C. I. Navy Blue 79, 50 g), then balls (ball no. 50, diameter 10 mm, 160 g) were added for milling. The mixtures were milled mechanically by centrifugal ball mills (Retsh) at 600 rpm for 60 min maintained at the room temperature. All the prepared pastes were assessed by the following methods:

**(a) Rheological Method.** Ten milliliters of the dispersed paste was put in the adapter with an SCS-

**Table II Formulation for Dispersing C.I. Navy Blue 79 and C.I. Brown 1. in Water<sup>a</sup>**

Material	Weight (g)		
	Navy Blue 79	Brown 1 <sup>b</sup>	
		A	B
Navy Blue 79	50	—	—
Brown 1	—	50	50
Water	6	6	20
Milled ball	160	160	160
Dispersant	0–28.6%	0–21.3%	

<sup>a</sup> The solid content of wet cake: C.I. Navy Blue 79: 35%; C.I. Brown 1: 47%.

<sup>b</sup> Formulation A is for NSF and PSAF, and formulation B is for DPSAD.

31 spindle, and its viscosity was measured using a rotating cylindrical viscometer (Brookfield DVII LVT). The apparent viscosities (under a fixed shear rate) were used as a parameter to assess the dispersing ability of the prepared dispersants. All the viscosities were measured at 25°C.

**(b) Scanning Electron Microscopy (SEM).** The dispersed paste (ca. 1 g) was coated on a thin copper plate (the wet film thickness 12.5  $\mu\text{m}$ ). After drying (in the room for 1 day), a small piece of the paste (ca. 4  $\times$  4 mm) was cut and put on an aluminum sheet (with diameter of 20 mm) and electrodeposited with a layer of gold. The surface of the coating film was observed by an SEM electron probe microanalyzer (Hitachi F-4010).

**(c) Sedimentation.** The dispersed pastes were put (ca. 40 mL) into 50 mL graduated cylinders, and the tops of the cylinders were sealed. The heights of the precipitates were recorded after each time interval; thus, the rate of precipitation and the final volume ratio were measured.

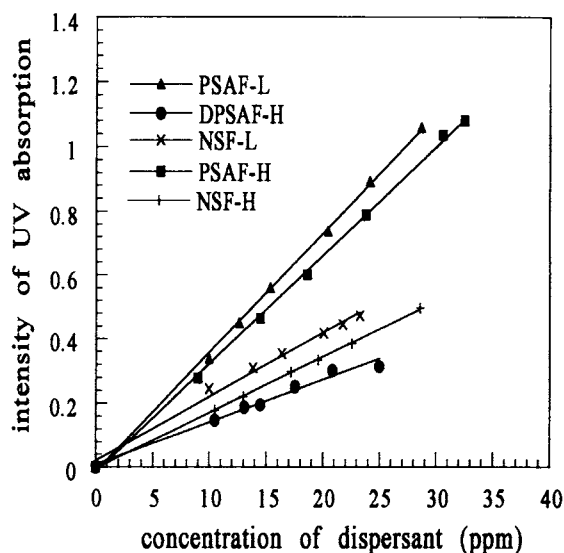
### Characterization of the Adsorption of the Condensate

The adsorption of the dispersants onto the surface of the dispersed dye surfaces was measured as follows: In a 50 mL bottle, 5 g of dye was put into 50 mL of the aqueous solution of dispersant with different concentrations. After shaking in a thermal bath (25°C, 350 rpm) for 48 h, the suspension was centrifuged by a DuPont Sorvall RC-5B at 15,000 rpm for 60 min at 25°C. Meanwhile, the intensity of UV absorption at 232 nm for various of dispersants (PSAF, NSF, and DPSAF) was measured as a function of the known concentration of dispersants

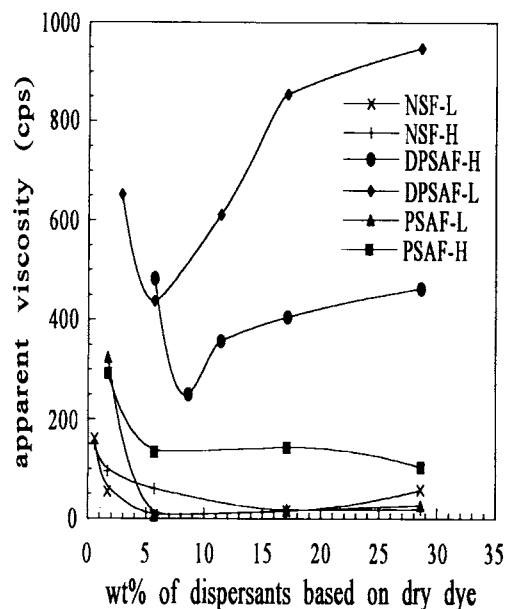
(ppm) (Fig. 1). The equilibrium concentrations of dispersants in the bulk phase (the supernatant after centrifugation) were determined by UV absorption (Jasco 7850) at 232 nm in terms of the adsorption calibration curve of Figure 1.

## RESULTS AND DISCUSSION

The aniline condensates were prepared at 95–100°C in the presence of HCl.<sup>13</sup> A low concentration of HCl (ca. 0.3 molar ratio of aniline) is used as the catalyst instead of the high concentration of H<sub>2</sub>SO<sub>4</sub> to avoid the side reaction of sulfonation on the aromatic ring. The sulfonyl group of the title compound is obtained by the ring-opening reaction of propane sultone at 70–80°C without an acid catalyst. The formation of condensates can be characterized by NMR as well as by GPC. Both PSAF and DPSAF have the methylene peak at  $\delta = 3.5\text{--}4.0$  ppm and have the peaks at  $\delta = 2.7\text{--}3.4$  and  $1.7\text{--}2.2$  ppm after the reaction of propane sultone with AF and DAF. Conventional NSF is a low molecular weight polymer and was qualitatively characterized by thin-layer chromatography<sup>9c</sup> or by GPC.<sup>16</sup> For neither case was molecular weight calculated; only the shape of molecular weight distribution in GPC or the number of spots on thin-layer plate was shown. If the molecular weight of NSF is calculated by GPC



**Figure 1** The calibration curve for the intensity of UV absorption at 232 nm as a function of the concentration of dispersant (ppm) for various of dispersants (PSAF, NSF, and DPSAF).



**Figure 2** The apparent viscosity (cps) of the dispersed C. I. Navy Blue 79 as a function of the concentration of dispersants added (wt % based on dry dye) for NSF, PSAF, and DPSAF.

based on the calibration standard of polyethylene glycol, the calculated molecular weight is tremendously high and does not make sense. The molecular weight of DPSAF-H and DPSAF-L was characterized by GPC in the previous study. The area of the peaks that appeared at a lower elution time is significantly higher for DPSAF-H (high MW) than that for DPSAF-L (low MW). The reaction time is longer for DPSAF-H than for DPSAF-L.

For a solution with a low solids content, the Einstein-Stokes equation<sup>17,18</sup> can be used to express the viscosity of the solution as a function of  $\eta_s$  (the viscosity of the solvent) and the volume fraction of the solid. For a dispersing system with high solids content, the shear rate ( $\gamma$ ) and the shear stress ( $\tau$ ) have a relationship of  $\gamma = k\tau^n$ .<sup>19</sup> On a plot of  $\tau$  vs.  $\gamma$  of a plastic flow, the intercept and the apparent viscosity ( $\tau/\gamma$ ) are useful parameters to estimate the extent of dispersion of a system. For a well-dispersed system, the aggregates of particles are smaller and less associated, and the viscometer spindle senses a smaller hindrance from aggregates at a given shear rate. Consequently, a smaller shear stress and a lower viscosity were detected.<sup>20</sup> In contrast, the agglomerates of a poorly dispersed system form a network that hinders the spindle and results in a higher shear stress and a higher viscosity.

In this study, a Brookfield DVII LVT (a low-shear-rate viscometer), with a spindle of SCS-31

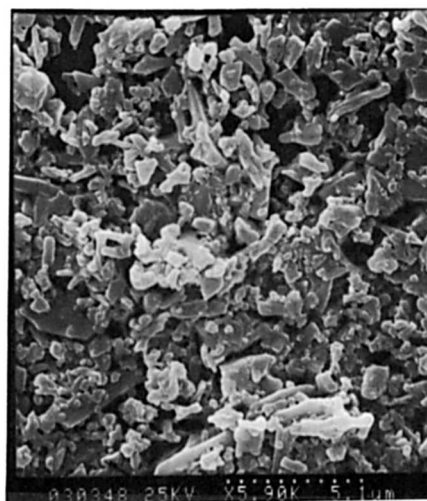
was used to measure the apparent viscosity values. Figure 2 shows the apparent viscosity as a function of dispersant concentration for C. I. Navy Blue 79 dispersed in water using various dispersants. At a low concentration of dispersant, the viscosity decreases as a function of increasing dispersant concentration for each plot. There exists a minimum in apparent viscosity (called minimum viscosity) for DPSAF-L and DPSAF-H; however, the viscosity

remains constant after the minimum viscosity for PSAF-H, NSF-H, NSF-L and PSAF-L is reached.

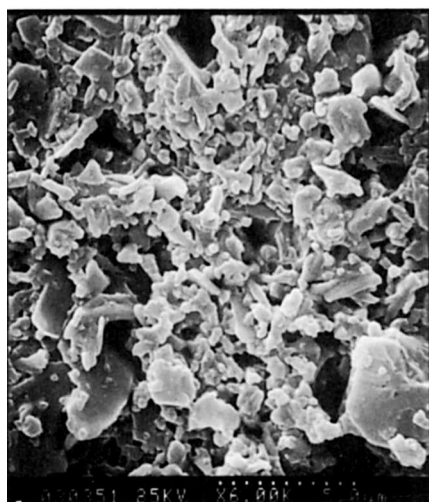
The minimum viscosity that PSAF-H, NSF-H, NSF-L, and PSAF-L can reach (ca. 140, 20, 10, and 6 cps, respectively) is remarkably lower than that for DPSAF-H or DPSAF-L (ca. 450 and 250 cps, respectively). For PSAF, the condensate with a lower molecular weight can reach a lower minimum viscosity, whereas for DPSAF, the higher the mo-



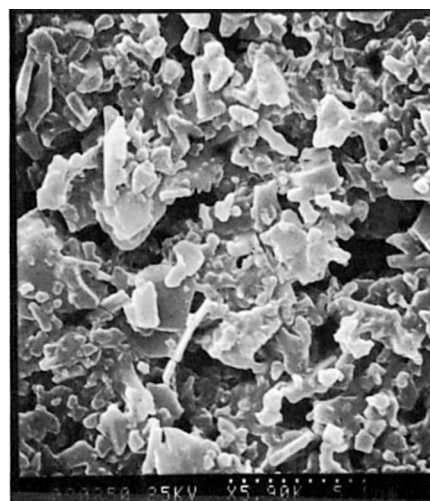
(a) 1.7%



(b) 5.7%



(c) 17.1%



(d) 28.6%

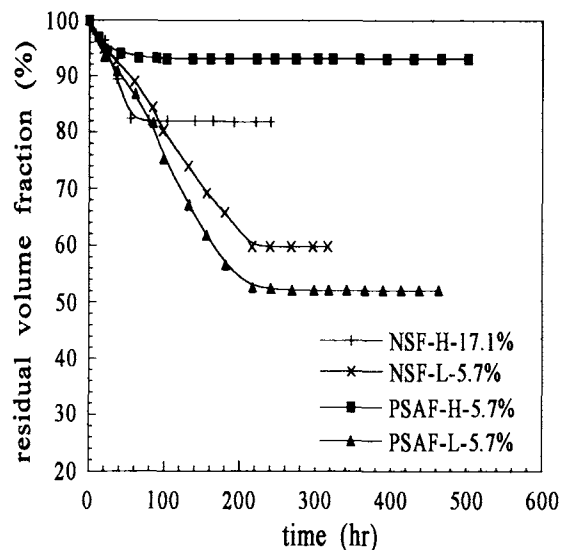
**Figure 3** The SEM pictures for C. I. Navy Blue 79 dispersed in water by different concentrations of PSAF-L: (a) 1.7%; (b) 5.7%; (c) 17.1%; (d) 28.6%.

molecular weight one can reach, the lower the minimum viscosity. The concentration for a dispersant to reach the minimum viscosity (called optimum concentration) is ca. 7–10 wt % for each dispersant. For industrial applications, a good dispersant is expected to reach a lower minimum viscosity accompanied by a lower optimum concentration.

Scanning electron microscopy (SEM) can be used to monitor the surface of the film of dispersed particles so that the state of the dispersed particles in solution can be assessed. Figure 3 shows the SEM (magnification of 6000 times) for C. I. Navy Blue 79 dispersed in a water system using PSAF-L as dispersants at different concentrations, which corresponds to the viscosity plots for PSAF-L in Figure 2. Obviously, the particles in Figure 3(b) are more homogeneously dispersed than are those in Figure 3(a), (c), and (d). Comparing this result with the plot of viscosity vs. the concentration of dispersant for PSAF-L in Figure 2, it can be concluded that, under a comparable condition, a suspension with a lower viscosity is more homogeneously dispersed.

Figure 4 shows the residual volume fraction (the height ratio of the precipitates/the total volume) as a function of time for each dispersant at its optimum concentration (NSF-H at 17.1%, NSF-L at 5.7%, PSAF-H at 5.7%, and PSAF-L at 5.7%). Both the pastes dispersed by DPSAF-H (5.7%) and DPSAF-L (9.0%) were stable and did not precipitate after a standing of 2 weeks. Obviously, the dispersant that can reach the lower viscosity (PSAF-L-5.7% and NSF-L-5.7%) has the smaller final volume fraction. The dispersed pastes prepared by using the above-mentioned dispersants were coated on a metal plate; thus, the dispersed particles can be observed by SEM (Fig. 5). The results show that the pastes dispersed by the dispersants that reach the lower viscosity (PSAF-L-5.7% and NSF-L-5.7%) display the more homogeneously dispersed particles. From the above-mentioned results, it can be concluded that a better-dispersed paste shows a lower viscosity and a smaller final volume fraction.

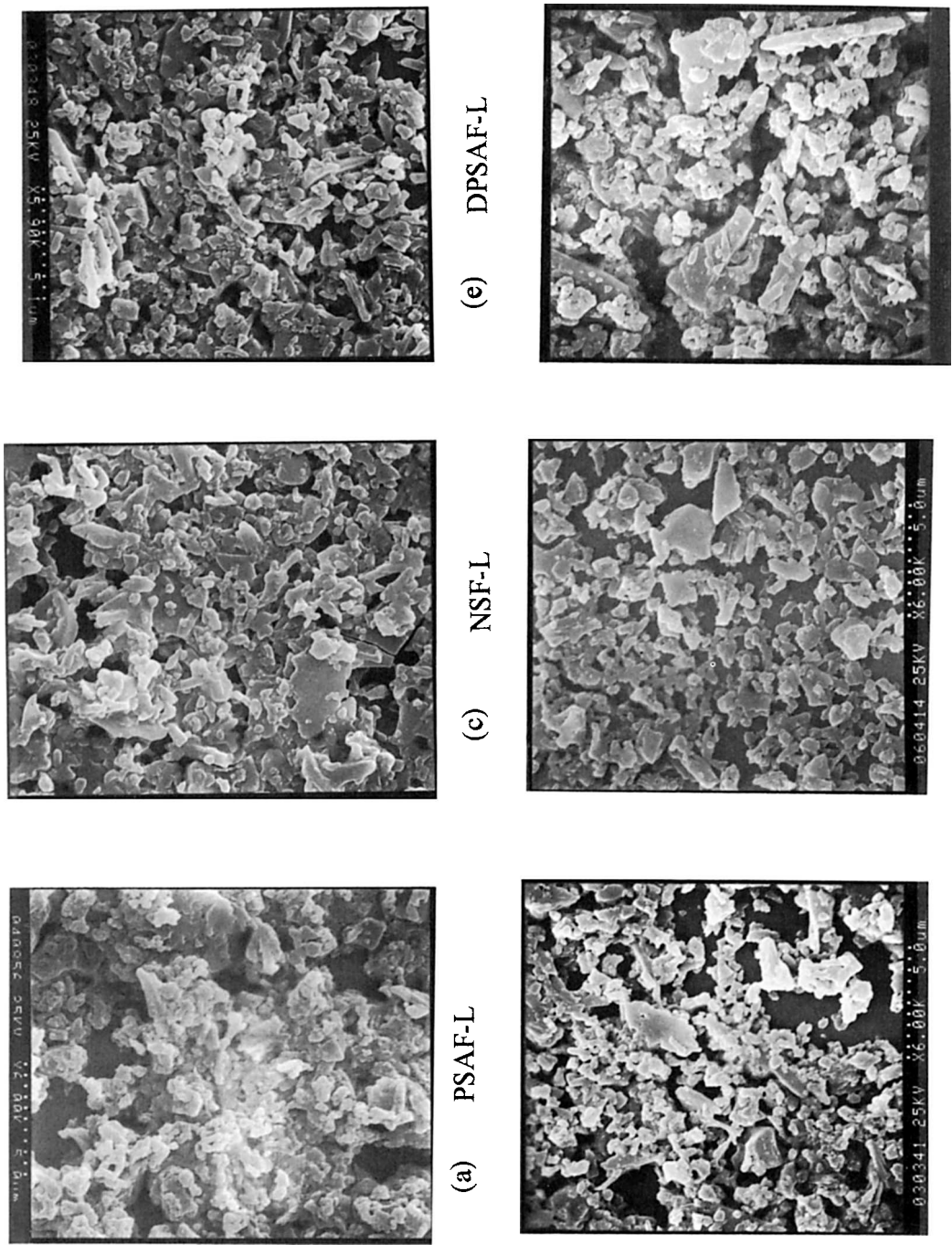
The stability of particles of adsorption is closely related to the effective thickness of the adsorption layer, which, in turn, is the function of the amount of dispersants adsorbed on the particles and of the orientation of the dispersant at the adsorption layer.<sup>21,22</sup> Figure 6 shows the adsorbed amount vs. the concentration of the dispersant in the bulk phase for C. I. Navy Blue 79 dispersed by NSF-L, PSAF-L, and DPSAF-H at different concentrations corresponding to the viscosity plot in Figure 2. Obviously, the amount of NSF-L adsorbed is significantly higher than that of PSAF-L and DPSAF-H.



**Figure 4** The residual volume fraction (the height ratio of the precipitates/the total volume) as a function of standing time for different dispersants at their optimum concentrations.

This result can be used to interpret the low minimum viscosity obtained by NSF-L. The greater adsorption of NSF-L can provide more charges on the surface of dye, resulting in an increase in the electrostatic repulsion, thus preventing the coagulation of the particles.<sup>1,23</sup> The adsorption amount for PSAF-L is smaller compared to that of NSF; however, the sulfonate on PSAF-L can be extended more deeply toward the bulk phase compared to that on NSF-L, so that PSAF-L can result in a high effective thickness. (There exist four members of atoms between the aromatic ring and sulfonate in PSAF-L, but the sulfonate is directly anchored on the aromatic ring in NSF. Thus, it probably can be postulated from the structure that the sulfonate extends more deeply toward the bulk phase due to its strong interaction toward water.) This probably is the reason why PSAF-L with a smaller adsorption amount can reach a minimum viscosity as low as can NSF-L. The reason for the greater adsorption of NSF-L compared to PSAF-L can be explained by the stronger interactions between the naphthalene rings of NSF and the surface of Navy Blue 79 and Brown 1, which contains benzene rings.

Brown 1, a dye, was dispersed by those six dispersants following the same formulation. For NSF and PSAF, the paste can reach a very low viscosity (Fig. 7), whereas for DPSAF, the viscosity is too large; thus, the water amount was increased from 6 to 20 g so to decrease the viscosity to the measurable range (Fig. 8). For Brown 1, NSF-H and NSF-L



(a) PSAF-L

(b) PSAF-H

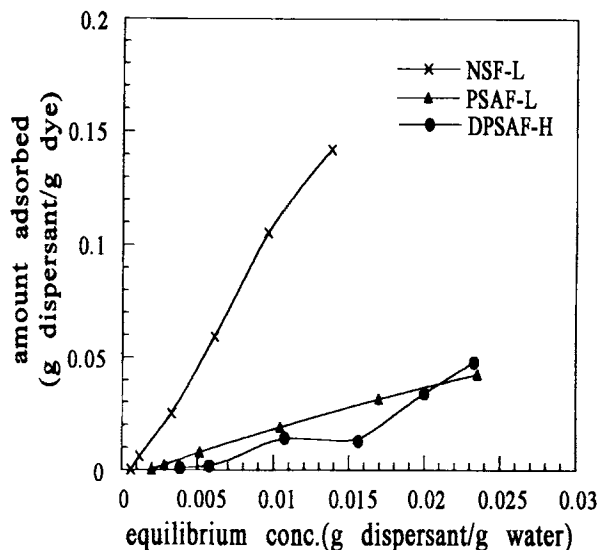
(c) NSF-L

(d) NSF-H

(e) DPSAF-L

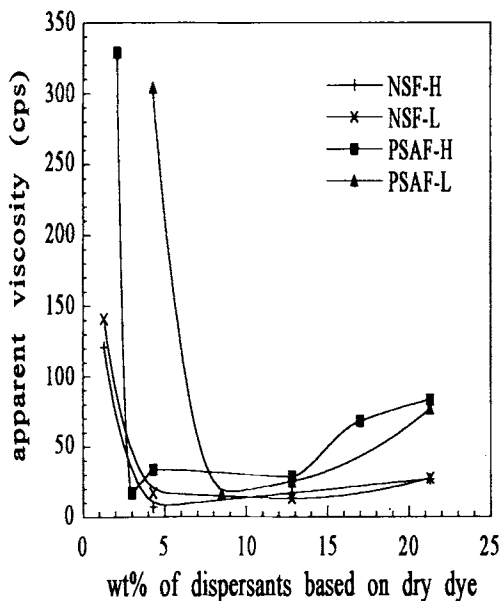
(f) DPSAF-H

**Figure 5** The SEM pictures for C. I. Navy Blue 79 dispersed in water by (a) PSAF-L, (b) PSAF-H, (c) NSF-L, (d) NSF-H, (e) DPSAF-L, and (f) DPSAF-H at the optimum concentration of each.

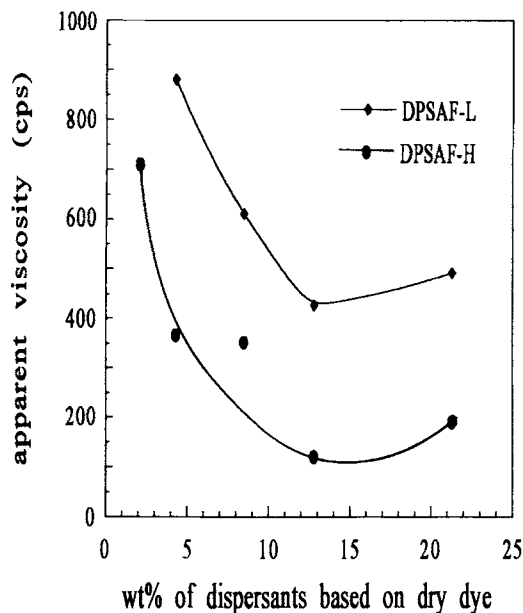


**Figure 6** The amount of dispersant adsorbed (g dispersant/g dye) as a function of the equilibrium concentration of dispersants (NSF-L, PSAF-L, and DPSAF-H).

can reach a similar minimum viscosity (ca. 10 cps) with a similar optimum concentration (ca. 5%) regardless of their molecular weights; PSAF-H and PSAF-L reach a similar minimum viscosity, whereas the optimum concentration is much lower for the PSAF with high molecular weight (PSAF-H, 3% compared to PSAF-L 9%). Even if the water content was increased for DPSAF, the apparent viscosity



**Figure 7** The apparent viscosity (cps) of the dispersed Brown 1 as a function of the concentration of dispersants added (wt % based on dry dye) for NSF and PSAF.



**Figure 8** The apparent viscosity (cps) of the dispersed Brown 1 as a function of the concentration of dispersants added (wt % based on dry dye) for DPSAF.

was still remarkably higher compared to that for NSF and PSAF. The DPSAF with higher molecular weight (DPSAF-H) can reach a lower viscosity compared to DPSAF-L, but the optimum concentrations for DPSAF-H and DPSAF-L are the same.

Both PSAF and DPSAF have a grafted sulfonate group, whereas the sulfonate group of NSF is on the backbone of condensates. The main difference between PSAF and DPSAF is that the former is anionic as is NSF, whereas the latter is zwitterionic. From the above-mentioned results, PSAF has an excellent dispersing ability both for Navy Blue 79 and Brown 1 as NSF; however, the dispersing ability of DPSAF is significantly worse compared to NSF.

### SUMMARY

Compared to DPSAF, the PSAF and NSF with a lower molecular weight (PSAF-L, NSF-L) show significantly lower apparent viscosities of the paste of Navy Blue 79. Comparing the results of SEM and the viscosity method, it can be concluded that a suspension with a lower viscosity is more homogeneously dispersed. Also, the dispersant that can reach the lower viscosity (PSAF-L-5.7% and NSF-L-5.7%) has the smaller final volume fraction.

The amount of NSF-L adsorbed on Navy Blue 79 is significantly higher than that of PSAF-L and DPSAF-H. The greater adsorption of NSF-L can



provide more charges on the surface of dye, resulting in an increase in the electrostatic repulsion, thus preventing the coagulation of the particles and resulting in the low minimum viscosity. The adsorption amount for PSAF-L is smaller compared to that of NSF; however, the sulfonate on PSAF-L can be extended more deeply toward the bulk phase compared to that on NSF-L so that PSAF-L can result in a high effective thickness.

For Brown 1, the anionic dispersants can reach a similar minimum viscosity independent of their molecular weight, but the PSAF with high molecular weight reaches a remarkably lower optimum concentration (ca. 3% compared to PSAF-L 9%, NSFs 5%).

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