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Negatively Charged Ultrafine Black Particles of P(MMA-*co*-EGDMA) by Dispersion Polymerization for Electrophoretic Displays

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ABSTRACT: Polymeric particles ranging in diameter from 1 to 3.5 μm containing a black dye, Sudan black B, were prepared by dispersion polymerization in a methanol and water mixture. To control their electrophoretic mobility, a varying amount of charge control additives was added after first labeling them with a fluorescent moiety to trace their distribution by confocal laser microscopy. The particle size was found to be quite sensitive to both the change in polarity of the polymerization medium and the amount of polymeric stabilizer employed. At the same time, increasing the amount of charge control additives resulted in an increase in the particle size. On the other hand, the electrophoretic mobility exhibited a maximum or optimum point at ∼0.3 wt % of charge control additive. This is consistent with the fluorescence intensity profile obtained from the confocal laser measurement, which shows a decrease in the intensity of the particles with increasing concentration of charge control additives. The simple peak patterns in the cross-sectional profile obtained from the confocal laser measurement suggests that the charge control additives are mainly distributed inside of the particles.

Introduction

Recently, electronic papers have attracted a great deal of interest for use in information displays requiring low cost, low weight, good flexibility, and low power consumption. The fabrication of displays satisfying these requirements can be achieved in several different ways, such as by using cholesteric liquid crystal, twisting balls, mobile fine particles with liquid crystals, in-plane electrophoretic particles, or microencapsulated electrophoretic particles.[1−4] Among these different materials, in-plane electrophoretic particle displays and microencapsulated electrophoretic particle displays (EPDs) offer the advantage of lower manufacturing cost, since they can be fabricated by a roll-to-roll method. The electrophoretic display operates on the principle that certain particles, when suspended in a dielectric medium, can be electrically charged and thereby can be driven to migrate through the medium toward an electrode of opposite charge. The same principle can also be applied to colored dielectric particles suspended in a fluid medium that is either clear or of an optically contrasting color to compose a color display. In both cases, the chargeability of the particles is a key factor in determining the speed or efficiency of the electrophoretic displays. Thus, to control the chargeability of the particles, charge control additives providing a positive or negative charge are added to the particles. The most common charge control additives used to provide the particles with a positive charge are quaternary ammonium salts compounds, while salicylic acid-based metal compounds are usually employed for negative charge control. Generally, carbon black is the material of choice to produce the black color in most applications. However, carbon blacks are not readily adaptable to electrophoretic displays because they are conductive and because the density of carbon black ($\rho = 0.8$ g/cm^3) is not matched to that of the usual suspending medium ($\rho = 1.71$ g/cm^3). Monodispersed particles with diameters ranging from 0.01 to 1 μm can be prepared by miniemulsion, emulsion, and emulsifier-free emulsion polymerization.[5−11] Among these different techniques, seeded emulsion, polymerization, and dispersion polymerization are suitable for preparing monodispersed microspheres with diameters of several micrometers.[12−15] Seeded emulsion polymerization is a very attractive way of preparing microspheres starting from small seed particles, and this technique has been used to prepare particles having a well-defined morphology. However, it involves several complex steps and is relatively time-consuming as well as requiring well-controlled polymerization procedures. One the other hand, dispersion polymerization represents a possible alternative for obtaining micron size particles, since it is a much simpler process and can be performed in a single step. Dispersion polymerization typically involves the polymerization of monomers which are partially soluble in the liquid dispersion medium in the presence of a polymeric steric stabilizer, such as poly(ethylene oxide), poly(vinylpyrrolidone), poly(vinyl alcohol), poly(acrylic acid), and hydroxylpropylcellulose. The objective of this study is to report the synthetic procedures used for the in-situ dispersion polymerization of methyl methacrylate/ethylene glycol dimethacrylate containing black dye for electrophoretic displays and to examine the basic electrophoretic response of the black colored particles so prepared. The effects of the fluorescent-labeled charge control additives in relation to the electrophoretic mobility and image analysis are also discussed.

Experimental Section

Materials. Methyl methacrylate (MMA) was washed with 10 wt % aqueous sodium hydroxide solution and passed through an activated aluminum oxide column to remove the

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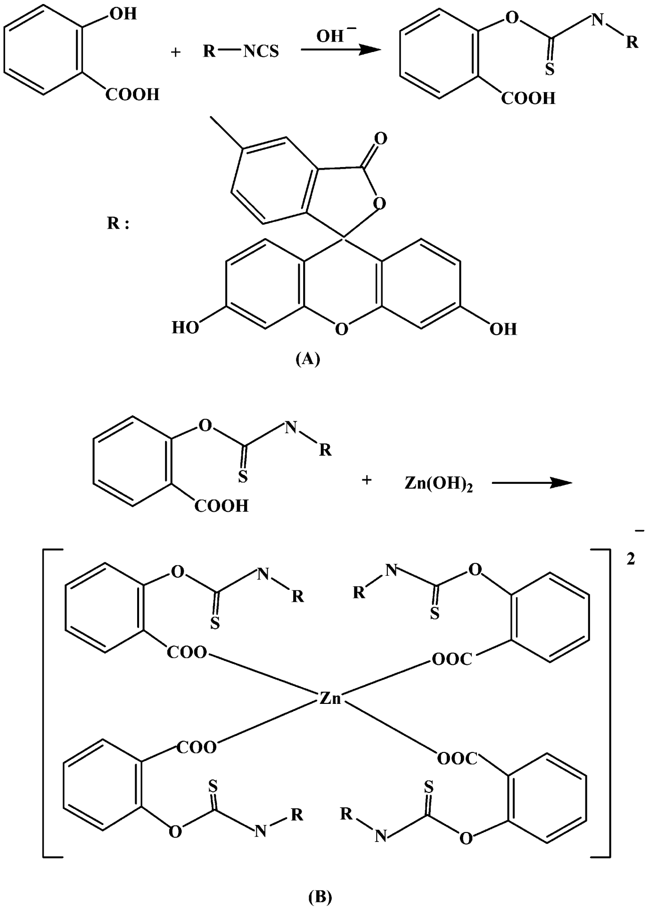

Figure 1. Reaction schemes for the labeling procedure of the charge control additives with fluorescence dyes.

inhibitor. Ethylene glycol dimethacrylate (EGDMA), poly(vinylpyrrolidone) (PVP, $M_w = 40\,000$), salicylic acid, $Zn(OH)_2$, fluorescein isothiocyanate (FITC), ethanol, tetrahydrofuran (THF), and Sudan black B were purchased from Aldrich Chemical Co. The 2,2′-azobis(2-methylpropionitrile) initiator (AIBN) was purchased from Junsei and recrystallized from methanol. The dielectric fluid, poly(chlorotrifluoroethylene) (Dp ≈ 4−10), was purchased from Halocarbon Product Corp. All other chemicals used in this study were of reagent grade.

Preparation of the Fluorescent-Labeled Charge Control Additives. As shown in Figure 1A, 100 mL of a 2.5 wt % aqueous salicylic acid solution was adjusted to pH 8.5 by adding 0.1 N sodium hydroxide solution. The fluorescence dye (FITC) was dissolved in THF at a concentration of 1 mg/mL. Subsequently, 100 mL of the dye solution was added to the salicylic acid solution and stirred for 4 h at 40 °C, and the free FITC was removed by ethanol extraction. The negative charge control additives were prepared by employing a protocol based on the method published by Yamanaka et al. (Figure 1B).[16−18]

Dispersion Polymerization of MMA and EGDMA in the Presence of Sudan Black B and Fluorescent-Labeled Charge Control Additives. The standard reaction conditions used in this study are given in Table 1. In a typical example, poly(vinylpyrrolidone) (1.5 g) and Sudan black B (0.3 g) were dissolved in a mixture of 240 mL of methanol and 60 mL of water under nitrogen at 65 °C in a 500 mL four-neck round-bottomed flask equipped with mechanical stirrer. A mixture of 28.5 g of MMA, 1.5 g of EGDMA, 0.3 g of AIBN, and 0.03 g of fluorescent-labeled charge control additives was added, and the polymerization was allowed to proceed for 24 h. Dispersion polymerization is very sensitive to various experimental conditions, such as the composition of the polymerization medium, the amount and composition of the monomers, the temperature, the amount and type of initiator, and the concentration of stabilizer. In this work, the MMA/EGDMA weight ratio in the monomer mixture was fixed at 95:5. The concentration of initiator and the polymerization temperature were also kept constant throughout the entire study, as shown in Table 1, while the composition of the reaction medium (the ratio of methanol to water) and the amount of Sudan black B were varied. After the completion of the polymerization process, the reaction mixture was cooled, diluted with methanol, decanted, washed several times with methanol and water, and dried in a freeze-drier.

Table 1. Standard Reaction Conditions for Dispersion Polymerization of P(MMA-*co*-EGDMA)

	components	amounts
monomer	MMA	28.5 g
	EGDMA	1.5 g
dispersion medium[a]	methanol	240−270 mL
	water	60−30 mL
dye[b]	Sudan black B	0.3−1.5 g
fluorescent-labeled CCA[c]	FITC-SA	0.03−0.3 g
stabilizer[d]	PVP	0.6−3 g
initiator	AIBN	0.3 g

[a] Total dispersion medium is 300 mL. [b] Dye concentration is varied from 1 to 5 wt % of total monomer base. [c] Fluorescent-labeled charge control additives. [d] Stabilizer concentration is varied from 2 to 10 wt % of total monomer base.

Characterization of the Black Particles. The DLS instrument was a laser light-scattering photometer from Brookhaven Instruments Inc., fit with a vertically polarized He−Ar laser at 632.8 nm and a digital correlator (BI-2000 AT). Samples for the measurement were prepared by dispersion microspheres in water. The prepared samples were filtered through a hydrophilic PTFE filter, unless otherwise mentioned. Filtered solutions were collected into dust-free glass tubes. The measurements were carried out at 25 °C at the following detection angles: 60°, 70°, 80°, 90°, 100°, 110°, and 120°. To avoid multiple scattering, the microsphere concentration (2×10^{-5} g/mL in water) was adjusted at each detection angle, to yield around 200 000 counts/s along measuring times ranging between 120 and 200 s. SEM (FE-SEM, Hitachi S-4300) specimens were prepared by redispersing the cleaned microspheres in methanol and placing a drop of the suspension on a piece of cover glass, which was mounted with double-adhesive tape on a carbon stud. The drop was dried in air at room temperature and then sputter-coated under vacuum with a thin layer of gold. A Biorad MRC 1024 (CLSM), equipped with an argon laser and a Zeiss axiovert 100 microscope, was used to investigate the distribution of the fluorescent-labeled charge control additives and the morphology of the polymeric particles. By redispersing the dried particles in suspending fluid, the samples for CLSM were prepared. A few drops of the solutions were dried on a slide glass, and then the particles were redispersed by adding a drop of suspending fluid on top of the dried particles and covering them with a cover glass. In this operation, the solutions should be dilute enough for single layers of particles to be acquired on the object glass. The percent dye incorporated into the electronic ink particles was determined with UV/vis spectrometry by sufficiently swelling the electronic ink particles in tetrahydrofuran (THF) for 5 days with stirring. The dried, black electronic ink particles were swelled in THF, and the concentration of THF was adjusted to 1 mg/mL. The dye content in the black electronic ink particles was determined by measuring the absorbance at the absorption maximum (λ_{max}) at 598 nm. The zeta-potential was measured by means of a Melvern Zetasizer 2000. The electrophoretic mobility (μ) was calculated by converting the zeta-potential, ζ, using the Smoluchowski relation $\zeta = \mu\eta/\epsilon$, where η (= 2.0 cP) and ϵ (= 2.6) are the viscosity and dielectric constant of the suspending fluid, respectively.

Results and Discussion

Effect of Water Concentration. Sudan black B was added to a reaction mixture containing methyl meth-

Table 2. Incorporation of Dye into Polymer Particles as a Function of Water Concentration

	monomer (g)					
sample ID	MMA	EGDMA	Sudan black B (g)	solvent ratio (M/W)[a]	absorbance (intensity)[b]	dye (%)[c]
A-1	28.5	1.5	0	95/5		
A-2	28.5	1.5	0.3	95/5	0.113	0.7
A-3	28.5	1.5	1.5	95/5	0.594	3.7
A-4	28.5	1.5	0	90/10		
A-5	28.5	1.5	0.3	90/10	0.118	0.73
A-6	28.5	1.5	1.5	90/10	0.632	3.93
A-7	28.5	1.5	0	85/15		
A-8	28.5	1.5	0.3	85/15	0.129	0.8
A-9	28.5	1.5	1.5	85/15	0.648	4.03
A-10	28.5	1.5	0	80/20		
A-11	28.5	1.5	0.3	80/20	0.150	0.93
A-12	28.5	1.5	1.5	80/20	0.776	4.83

[a] M/W (methanol/water). [b] Absorbance at λ_{max} 598 nm. [c] Calculated by UV calibration curve.

Table 3. Incorporation of Dye into Polymer Particles as a Function of Stabilizer Concentration

	monomer					
sample ID	MMA	EGDMA	Sudan black B (g)	stabilizer (g)	absorbance (intensity)[a]	dye (%)[b]
B-1	28.5	1.5	0.3	2	0.080	0.15
B-2	28.5	1.5	0.3	4	0.086	0.16
B-3	28.5	1.5	0.3	6	0.113	0.21
B-4	28.5	1.5	0.3	8	0.118	0.22
B-5	28.5	1.5	0.3	10	0.123	0.23
B-6	28.5	1.5	1.5	2	0.396	0.74
B-7	28.5	1.5	1.5	4	0.418	0.78
B-8	28.5	1.5	1.5	6	0.546	1.02
B-9	28.5	1.5	1.5	8	0.610	1.14
B-10	28.5	1.5	1.5	10	0.717	1.34

[a] Absorbance at λ_{max} 598 nm. [b] Calculated by UV calibration curve.

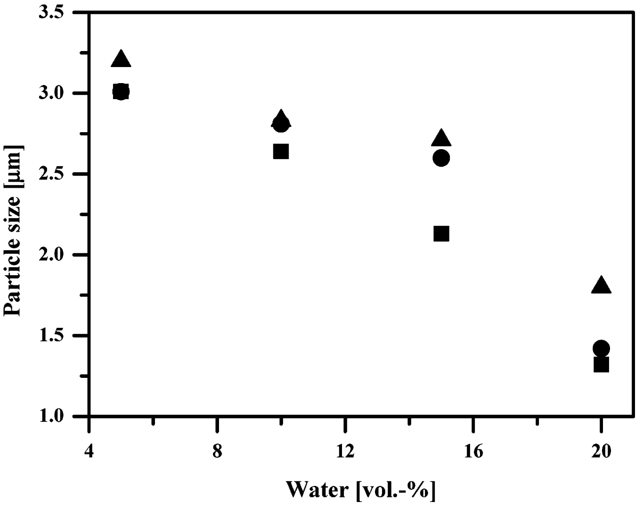

Figure 2. Effect of the P(MMA-*co*-EGDMA) particle size (fixed angle = 90°) on the concentration of water and Sudan black B concentrations in the polymerization mixture at a PVP concentration of 10 wt %: (■) no Sudan black B, (●) 1 wt % Sudan black B, and (▲) 5 wt % Sudan black B.

acrylate, elthylene glycol dimethacrylate, poly(vinylpyrrolidone), and azobis(isobutyronitrile) (1 wt %) and then polymerized in a mixture of water and methanol. The DLS average diameter was calculated through the quadratic cumulants method from the set of autocorrelations and the following contstants: $\kappa = 1.38 \times 10^{-23}$ m^2 kg/(s^2 K), $\lambda = 632.8$ nm, $n_m = 1.333$, $T = 298.15$ K, and $\eta = 0.01002$ g/(cm s). The intensity autocorrelation functions measured for microspheres at various scattering angles are shown in the Supporting Information (Figure S-1a). The correlator uses the average intensity and average squared intensity to calculate a baseline. The relative difference between the calculated and measured baselines can be used as an indicator of dust particles. Baseline differences should never exceed a fraction of a percent. Typical differences between the measured and calculated baselines were less than 0.5%. Measured average diameters (D_{DLS}) as a function of scattering angle are shown for the microspheres in the Supporting Information (Figure S-1b). For a dilute (2×10^{-5} g/mL in water), monodisperse suspension of scatters, the measured average diameter (D_{DLS}) should be independent of the scattering angle, which suggests a distribution of particle sizes. As shown in Figure 2, increasing the amount of water increases the polarity of the reaction medium, resulting in a decrease in the size of the Sudan black B containing poly(methyl methacrylate-*co*-ethylene glycol dimethacrylate) particles. While the particle size was sensitive to the composition of the water/methanol mixture reaction medium, the size distribution did not show much variation in the experimental range. The addition of Sudan black B does not seem to have not much effect on the particle size. As shown in Figure 2, the particles containing Sudan black B show a slightly larger size than those without Sudan black B. Even with the addition of 5 wt % of Sudan black B, the increase in the particle size is only marginal. The increase in the particle size with increasing amount of water in the reaction medium can be understood from the point of view of the solubility of the ingredients in the methanol–water mixture. Methanol and water are both good solvents for PVP, and consequently, its solubility is not significantly affected by the variation in the water content. However, water is certainly a somewhat poorer solvent for P(MMA-*co*-EGDMA) than methanol. With increasing water content, the critical chain length for the precipitation of the growing polymer chain would be expected to decrease, and the adsorption rate of the stabilizer to P(MMA-*co*-EGDMA) would likely increase. Therefore, both the rate of nuclei formation and the rate

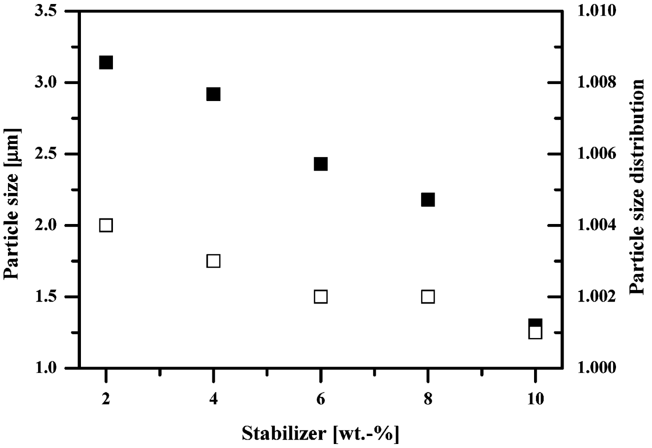

Figure 3. Effect of the P(MMA-*co*-EGDMA) particle size (fixed angle = 90°) on the concentration of stabilizer in the polymerization mixture at a water concentration of 20 vol % and Sudan black B concentration of 1 wt %: (□) particle size and (■) particle size distribution.

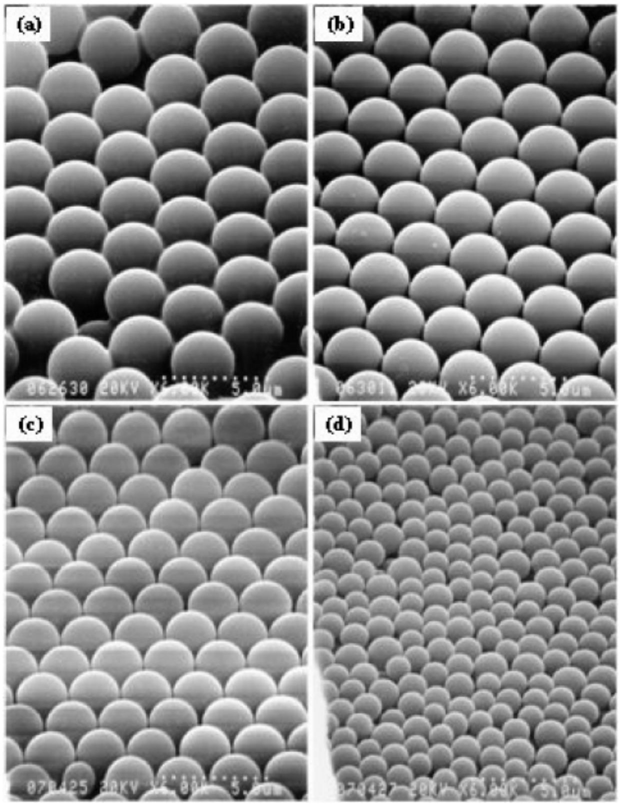

Figure 4. Scanning electron micrographs of P(MMA-*co*-EGDMA) particles prepared with different concentration of PVP stabilizer (water:methanol = 20:80, 1 wt % of Sudan black B): (a) 2 wt % PVP, (a) 4 wt % PVP, (c) 8 wt % PVP, and (d) 10 wt % PVP.

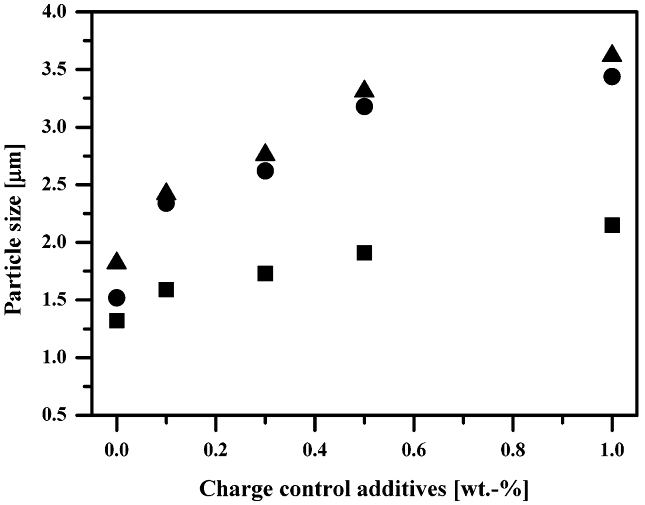

Figure 5. Effect of the P(MMA-*co*-EGDMA) particle size (fixed angle = 90°) on the concentration of the fluorescent labeled charge control additives in the polymerization mixture at a water concentration of 20 vol % and PVP concentration of 10 wt %: (■) no Sudan black B, (●) 1 wt % Sudan black B, and (▲) 5 wt % Sudan black B.

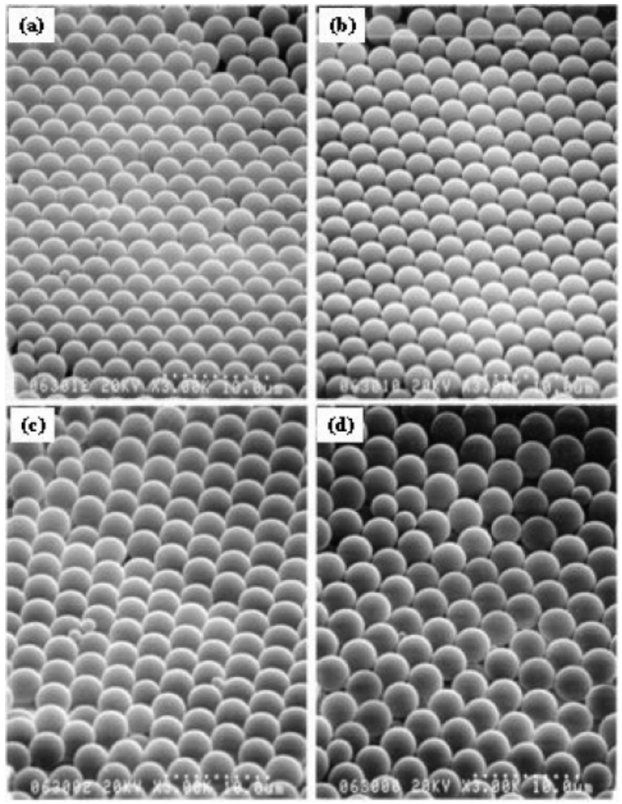

Figure 6. Scanning electron micrographs of P(MMA-*co*-EGDMA) particles with the different concentration of fluorescent-labeled charge control additives (water:methanol = 20:80, 5 wt % of Sudan black B, 10 wt % of PVP): (a) 0.1, (b) 0.3, (c) 0.5, and (d) 1.0 wt %.

of adsorption of the stabilizer would tend to increase, resulting in smaller particles. Table 2 shows the incorporation of the dye molecules into the polymer particles, which indicates how much of the dye is present in the final polymer particles. The sample A-2, which is prepared in a 95/5 methanol−water volume fraction, shows about 70% of incorporation, which means that 30% of the dye disappeared somewhere outside of polymer particles. However, the addition of water to the polymerization media has a significant effect on the dye holding or incorporation ability. As the water fraction is increased, the incorporation of dye is increased, attaining a level of as much as 96% of the theoretical value. The results clearly demonstrate that the addition of water is the key parameter involved in improving the incorporation of dye inside of the polymer particles.

Effect of Stabilizer Concentration. The stabilizer concentrations were varied from 2 to 10 wt % based on the total dispersion medium. Table 3 shows the effects of the stabilizer concentration on the incorporation of dyes into the final particles. The sample B-1, which was

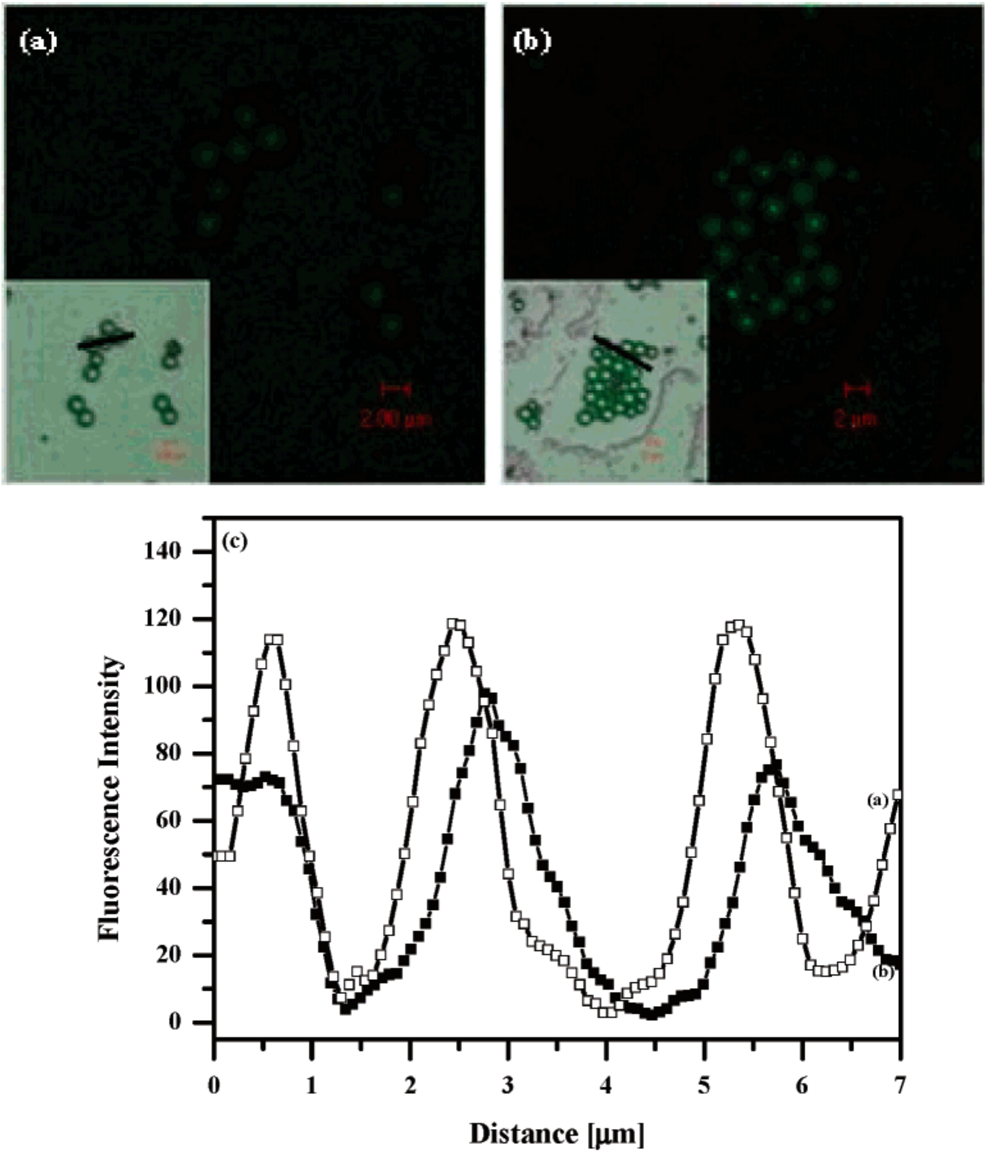

Figure 7. Confocal laser scanning microscopy images of polymeric particles for different amounts fluorescent-labeled charge control additives: (a) 0.3 wt % and (b) 0.5 wt %. (c) Cross-sectional view of fluorescence intensity: black dot = 0.5 wt %; white dot = 0.3 wt %.

prepared with 2 wt % of PVP, shows about 15% of incorporation, which means that 85% of the dye fed disappeared outside of polymer particles. However, the addition of 10 wt % of PVP to the dispersion medium had a dramatic effect on the dye incorporation ability. As the stabilizer concentration was increased, the incorporation of dye was enhanced, attaining a level of as much as 75% of the theoretical value. On the other hand, the increasing the dye feed levels in the range of 1−5 wt % did not have any significant effect on the amount of dye in the polymerized particles. As shown in Figure 3, the resulting particle sizes increased with decreasing stabilizer concentration. As the stabilizer content is increased, both the particle size and its distribution become smaller. The variation of the stabilizer content may affect both the extent of aggregation of the nuclei by changing the adsorption rate of the stabilizer and the viscosity of the dispersion medium. Figure 4 shows scanning electron micrographs of the P(MMA-*co*-EGDMA) black particles prepared in a mixture of methanol and water (ratio of 8:2) with different concentrations of stabilizer. The P(MMA-*co*-EGDMA) particles with fairly narrow size distributions were obtained, and their size ranged from 1 to 3 μm depending on the stabilizer content. The multiangle dynamic light scattering resulting average diameter is summarized (see Supporting Information).

Effect of Fluorescent-Labeled Charge Control Additives. In order for the polymer particles to exhibit an electrophoretic response, fluorescent-labeled charge control additives are necessary. Therefore, polymer particles containing fluorescent-labeled charge control additives were polymerized in an 80/20 (methanol/water) mixture, while varying the concentration of the fluorescent-labeled charge control additives, from 0.1 to 1 wt % at a constant PVP concentration of 10 wt %. Since the charge control additive is an ionic in nature, it may have a certain effect on either the particle size or size distribution. In this set of experiments, P(MMA-*co*-EGDMA) particles were prepared by adding a variable amount of charge control additives ranging from 0.1 to 1 wt %, while the concentration of the stabilizer, PVP, was fixed at 10 wt %. The particle size increased from approximately 1.5 to 3.5 μm as the amount of charge control additives was increased up to 1 wt %, as shown in Figure 5.

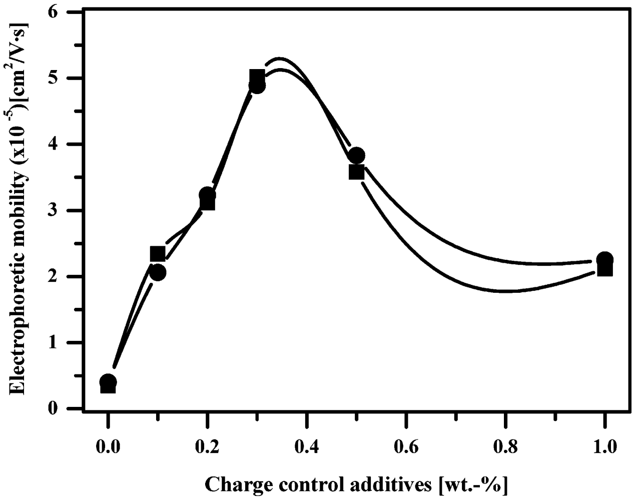

Figure 8. Effect of charge control additives in polymeric particles on electrophoretic mobility: (■) 3 wt % of Sudan black B and (●) 5 wt % Sudan black B.

Upon adding the dye, the particle size increased, as compared with the case where no dye was added, and an almost 2-fold increase was observed at a high concentration of charge control additive. Since the maximum amount of dye added was 5 wt %, such an increase cannot be explained by a simple increase in the solid content. Multiple factors including the change in interfacial tension and the change in the electrolyte concentration and viscosity of the medium caused by the addition of either the dye or the charge control additive seem to play a role in determining the particle size. The electron micrographs show quite narrow size distributions regardless of the amount of charge control additive added, as shown by the micrographs in Figure 6.

Confocal Laser Scanning Microscopy (CLSM). Confocal laser scanning microscopy was used to verify the DLS and SEM results. In addition, the concentrations of the fluorescent-labeled charge control additives distributions were characterized, and the fluorescence intensity across the polymeric particles was determined quantitatively. As illustrated in Figure 7, a homogeneous distribution of the fluorescence intensity was observed throughout the particles in each case. The particle size of the polymeric particles was found to be about 2.5−3 μm. These particle sizes are in good agreement of with the DLS and SEM results. At a high concentration of fluorescent-labeled charge control additives (0.5 wt %), the fluorescence intensity of the interior of the polymeric particles is rather weak. The cross-section profile of the fluorescence intensity (Figure 7c) along the diameter gives information about the distribution of the fluorescent-labeled charge control additives. There are three peaks demonstrating that the fluorescent-labeled charge control additives are confined to the interior of the polymeric particles. The reduction in the fluorescence intensity at a high concentration of fluorescent-labeled charge control additives (0.5 wt %) is probably due to the isolated disintegration of charge control additive molecules. At a high concentration of fluorescent-labeled charge control additives (0.5 wt %), the fluorescence intensity is lower than that observed in the case of a lower concentration of charge control additives, indicating that not all of the charge control additives are not incorporated into the particles and that some of them are lost from the particles probably due to polymerization or their diffusion into the dispersing medium.

Electrophoretic Mobility. Figure 8 shows the effect of the charge control additive on the electrophoretic mobility of the polymer particles. The electrophoretic mobility was measured using a polymer particles dispersion, which was obtained by redispersing the synthesized particles after cleaning and drying them. The polymer particles in this study were spherical with a uniformly smooth surface (Figure 6). The electrophoretic mobility value increased as the charge control additive concentration increased. However, it showed a maximum and then decreased as the charge control additive concentration was further increased. The maximum value of the electrophoretic mobility was -5.202×10^{-5} cm^2/(V s) at a charge control additive content of 0.3 wt %. Increasing the concentration of the charge control additives increases the number of effective charging sites of the polymer particles, resulting in them having higher electrophjoretic mobility. However, the charge control additive used in this study is a Zn complex which is quite soluble in water. Therefore, some of the charge control additive migrates from the particle to the water phase during the washing process, even though the particles are completely cleaned and dried before use. This migration of the charge control additive has the combined effect with the increase in the charge density of causing the electrophoretic mobility to show a maximum as shown in Figure 8.

Conclusions

The dispersion polymerization of methyl methacrylate and ethylene glycol dimethacrylate in the presence of nonpolymerizable dyes in a methanol and water mixture was studied to obtain monodisperse black particles ranging in size from 1 to 3.5 μm. The particle size was found to decrease with increasing concentration of the stabilizer. The most important role of the stabilizer is its effect during the particle formation step, where it determines the particle stability and the continuous viscosity. The polarity of the dispersion medium and the polymerization rate have a significant effect on the particle size, and consequently, the polarity of the reaction medium and ultimately the particle size can be controlled by using mixed solvents. The effects of the charge control additives were significant. The appropriate additions of charge control additives results in an increase in electrophoretic mobility. The concentration dependency of the fluorescent-labeled charge control additives on its deposition behavior in the polymeric particles was successfully examined and thereafter quantified by image analysis. In further applications, monodispersed black particles are expected to be very useful in electrophoretic displays (see Supporting Information).

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Supporting Information Available: Multiangle dynamic light scattering were applied to determine the average particle diameter: schematic illustrations for in-plane electrophoretic display cells (a) and schematic operation mechanisms (b); optical microscopy of (a) initial state, (b) applied voltage, (c) no applied voltage of state (b), and (d) reverse applied voltage; photograph of an in-plane electrophoretic display cell, with (a) no bias voltage applied and (b) −50 V applied. This material is available free of charge via the Internet at http://pubs.acs.org.

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