

## Towards Multi-color Microencapsulated Electrophoretic Display

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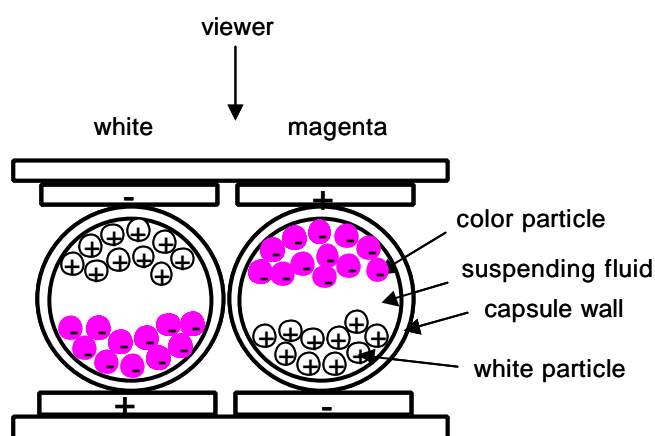
### Abstract

*In this paper, we present techniques to manufacture color electronic ink for multi-color electrophoretic display implementation. The charged color pigments have been prepared to have superior affinity for dielectric fluid. White TiO<sub>2</sub> nanoparticles were modified with poly(methyl methacrylate) copolymer for a microencapsulated electrophoretic display system, in order to reduce the density mismatch between nanoparticles and dielectric medium. These color balls and white pigment particle suspensions were microencapsulated through the typical microencapsulation technique. We fabricate the microcapsules to the single layer on flexible ITO substrate to test the multi-color electrophoretic display application.*

### 1. Introduction

The Flexible and rewritable reflective paper-like display with low power consumption and high contrast ratio, can be fabricated to, a low cost, therefore, it has stimulated the development of various applications such as low-power portable displays, PDAs, and e-books. There are several different technologies to realize the electronic paper such as a microencapsulated electrophoretic display (MC-EPD) [1,2], a twisting ball display [3], an electrochromic display [4], and an electrowetting display [5]. Many research groups have devoted to develop the electronic ink materials and other related technologies, because of the various advantages of the electrophoretic display (EPD). The first MC-EPD was reported by Comisky, et al. [1], and several displays have been developed up to the present. A schematic cross-sectional diagram of a part of the MC-EPD is represented in Figure 1. The electrophoretic medium comprises two different types of white and colored pigment particles, bearing charges of opposite polarity, in a colorless dielectric fluid. Optical contrast is achieved by moving charged particles

separately to the opposite electrode. From the front common electrode the viewer can observe white or color particles held electrostatically to the common electrode depending on the electric field and particle charge.



**Figure 1. Schematic illustration of microcapsule-type electrophoretic display.**

In this paper, we report the preparation of microcapsules which are containing cyan/white, magenta/white, and yellow/white pigment pairs dispersed in the clear dielectric fluid to realize the color MC-EPDs. The fabrication of the full-color MC-EPD is possible by this method through imposing the color microcapsules vertically or horizontally.

### 2. Experimental

We used the TiO<sub>2</sub> pigment particle with the average particle size of 0.42 μm made by Dupont (Ti-Pure, R-900) without further purification. Other commercially available reagent grade chemicals were also used without further purification.

The TiO<sub>2</sub>/Polymer hybrid particles were prepared through a two-stage dispersion polymerization technique [6]. Firstly, poly(vinyl pyrrolidone) was

dissolved in methanol with a mechanical stirrer under nitrogen condition. The 4 g of  $\text{TiO}_2$  particles were sonicated in the PVP methanol solution.

Mixture of methylmethacrylate and ethylene glycol dimethylate (EGDMA, Aldrich Co.) was poured into  $\text{TiO}_2$  dispersion and emulsified. Finally, a solution of initiator was added into the reactor. The polymerization was carried out at 65 °C for 6 hr.

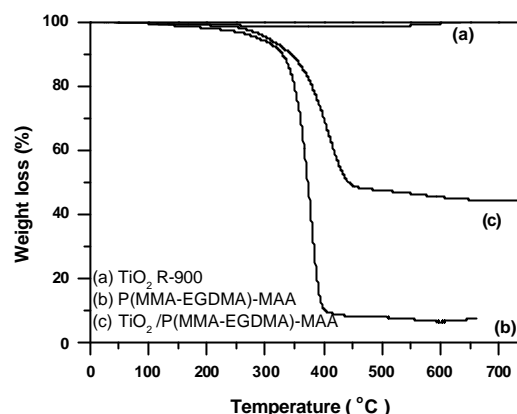
Secondly, 1.4 g of methacrylic acid (MAA, Aldrich Co.) to introduce the charge moiety on the  $\text{TiO}_2$ /Polymer composite surface, was added slowly into the reactor to incorporate the composite particle surface. After adding the MAA, the reactions were carried out for 12 hr at the same temperature. After the termination of the reaction, deionized water was added to the synthesized  $\text{TiO}_2$ /Polymer hybrid composite particles mixture, and shaken thoroughly.  $\text{TiO}_2$ /Polymer composite particles were washed with deionized water. The charged  $\text{TiO}_2$ /Polymer composite particles were dried and became the white electrophoretic ink particle successfully.

To improve their affinity for the dielectric fluid, the color pigment particles were coated with the functionalized natural wax (Montan Wax®). Microcapsules were prepared by in situ polymerization [7].

### 3. Results and Discussion

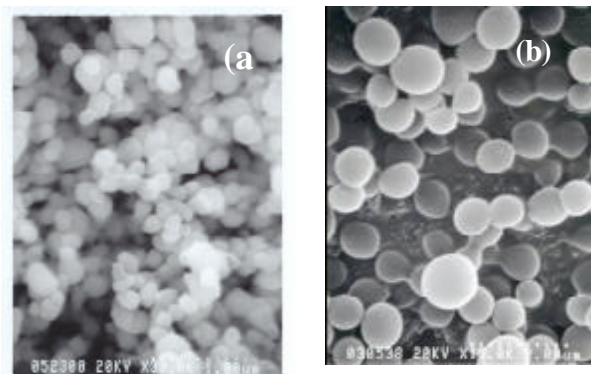
The white and color pigment particles were modified because of the density difference between the pigment particles and dielectric fluid and their poor affinity for the electrophoretic fluid.

Thermogravimetric analysis (TGA) measurement was carried out to determine the core pigment particle content. Figure 2 shows TGA thermograms of the raw  $\text{TiO}_2$ , the reference copolymer, and the nanohybrid particles at a heating rate of 5 °C/min with nitrogen purging. Until it was heated up to 500 °C,  $\text{TiO}_2$  particles remained in its initial state. The reference copolymer starts to degrade at around 300 °C and decomposes at 430 °C. Obviously, the onset temperature of thermal decomposition of the hybrid particle is shifted toward to the higher temperature than that of the copolymer, indicating that the enhancement of the thermal stability of the hybrid sample. The thermogram of nanohybrid shows that around 43 % of residue occurred at above 400 °C, due to the degradation of the copolymer coated on  $\text{TiO}_2$  particles.



**Figure 2.** TGA thermograms of bare  $\text{TiO}_2$  particle (a); PMMA copolymer (b); and  $\text{TiO}_2$ /P(MMA-co-EGDMA-co-MAA) hybrid composite particle (c).

Figure 3 presents the SEM images of (a) the raw  $\text{TiO}_2$  particles which have irregular shapes and (b) the  $\text{TiO}_2$ /Polymer hybrid submicron-sized microspheres. The surface morphology of the nanohybrid particles turned into spherical shape after coating process. We found that the surface morphology of the nanohybrid particles became spherical after coating process. The size of bare  $\text{TiO}_2$  particles was determined to be 425 nm from a particle size analyzer, whereas the size of the nanohybrid particles was around 625 nm.

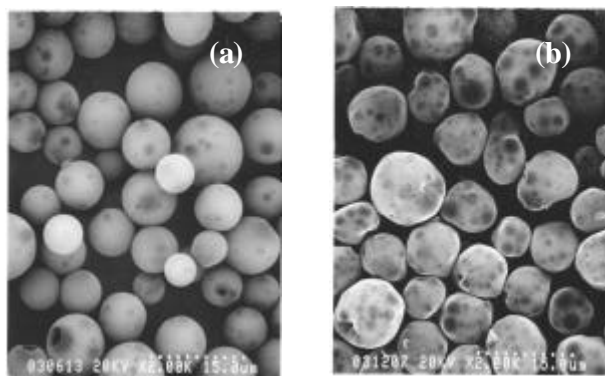


**Figure 3.** SEM photograph of bare  $\text{TiO}_2$  particle (a);  $\text{TiO}_2$ /P(MMA-co-EGDMA-co-MAA) hybrid composite particle (b).

As shown in Figure 4, the morphology of yellow ball was changed after polymer coating. There are holes and collapses on the surface of the raw yellow microsphere particles. The wax-coated particles exhibit that the wax film covers the holes and

collapses of the raw particles. The mean particle size of each color particles is determined to 6.2  $\mu\text{m}$  for magenta particle, 7.9  $\mu\text{m}$  for yellow particle, and 3.4  $\mu\text{m}$  for cyan particle, respectively.

Cyan, magenta, and yellow microcapsules containing the polymer-coated  $\text{TiO}_2$  (negative polarity) and each color particles (positive polarity) suspension as a core material, were manufactured. The obtained microcapsules have the diameter of 50 ~ 200  $\mu\text{m}$ . The microcapsules show a good mechanical strength and thermal property. The apparent optical images of the color microcapsules are observed with an optical microscope. As shown in Figure 5, white pigment and color particles are suspended in suspending fluid in a capsule wall.

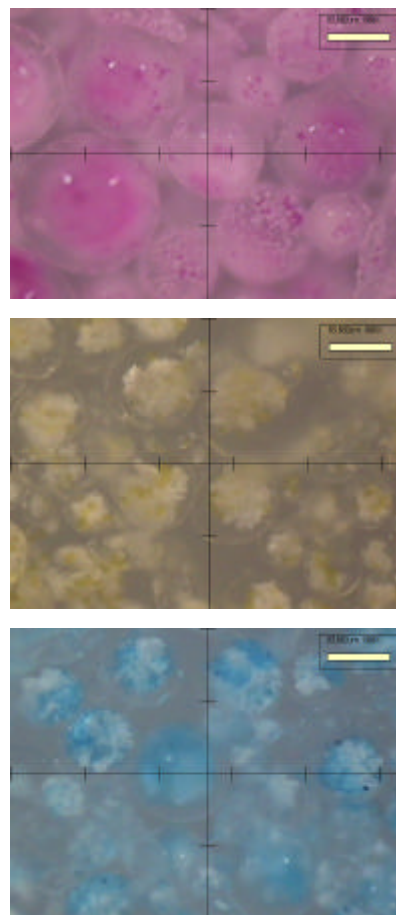


**Figure 4. SEM photograph of bare yellow ball (a); the SEM photograph of the functionalized wax coated yellow ball (b).**

To investigate the movement of the particles in the microcapsule, we have made a simple electrophoretic display cell. It is fabricated between two ITO coated transparent electrodes with 200  $\mu\text{m}$  gap space.

Figure 6 is the microphotograph of microcapsules suspending both magenta and white particles under electric field. Positive DC electric field ( $E = 250$  V/mm) was applied to the upper electrode, then the white particles moved fast in the upward (Figure 6a). When the field was revised, the magenta particles came to the upper electrode and white particles were pulled back (Figure 6b). Based on the electrophoretic mobility by zeta potential measurement, the response time is about 0.3 sec. But there is a large difference between the two values of the response time. The other response time, an actually measured time of white pigment particles in the capsules was measured to about 5 s under a DC 50 V power. Ota et al. [10]

observed the difference between the two types of electrophoretic mobility; one was estimated by using the Stokes' law and the other was calculated from the charge-to-mass ratio. Even though they obtained the two different electrophoretic mobility values, the reason was not explained clearly.

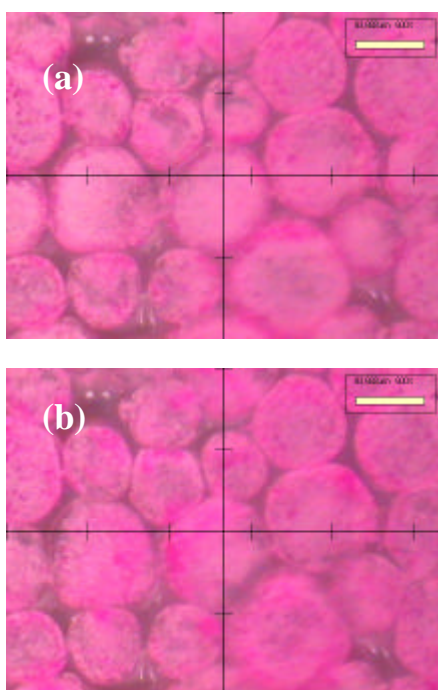


**Figure. 5 Optical microscope photograph of the color microcapsules containing white and magenta (a); white and yellow (b); and white and blue particles (a).**

From the investigation, we found that the particle separation influences the bright contrast. Some particles move together (e.g., white and magenta particles). Colloidal particles owe their stability against aggregation solely to the fact that their surfaces are charged and hence repel each other..

#### 4. Summary

The color encapsulated electronic ink was prepared by in situ polymerization utilizing urea/melamine and formaldehyde as wall materials. The white particles modified with PMMA copolymer and the color (magenta, yellow and cyan) particles modified with wax material were found to have superior affinity for suspending medium, and they were provided the different polarities. The particles moved in the microcapsules each electrodes by applying a DC electric field ( $E = 250 \text{ V/mm}$ ). When the field was reversed, the particles were pulled back reversibly.



**Figure 6. Microcapsule layer in the electric field ( $E = 250 \text{ V/mm}$ ) ((a) negative field is applied to the upper electrode, (b) positive field is applied to the upper electrode).**

#### 5. References

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