Contrast Ratio of Colorant Film: Theoretical Consideration and Effect of Polymeric Binder

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ABSTRACT: We made theoretical approaches concerning the optical property of a pigment-containing system to understand the conditions under which an enhanced contrast can be achieved. Maintaining a uniform distribution of pigment particles in thin film well, as in millbase dispersion, is considered critical for a higher contrast. As a specific case, we estimated the effect of a polymeric binder on the contrast ratio. Chemical composition of the dispersion binder has a huge effect, and carboxylate in its long side chain plays a favorable role. Using this binder, we were able to obtain a smooth surface and uniformity, and an enhancement of contrast ratio in the colorant film. © 2010 Wiley Periodicals, Inc. J Appl Polym Sci 117: 428–433, 2010

Key words: contrast; dispersing binder; millbase; pigment; dispersions

INTRODUCTION

Color filters are an essential component for flat panel displays like liquid crystal displays. They generally consist of several primary colors like red, green, and blue. For their fabrication, until now, pigment dispersion has been widely adopted because of its durable, repeatable, and reproducible characteristics.[1,2] Generally, the dispersion process can be divided into three successive stages: pigment wetting, pigment disintegration, and pigment stabilization.[3–5] Pigments are disintegrated by breaking up pigment agglomerates with an external energy and then stabilized by creating repulsive forces between the particles. The repulsive force between the particles needs to be greater than the van der Waals attraction, otherwise flocculation occurs. All stages involve the aid of a dispersant. A dispersant contains anchoring groups, which interact with pigments like a bridge between particle surfaces. Stability of pigment dispersion is also influenced by a dispersion binder. A dispersion binder generally contains acidic groups which interact with amino end groups in the dispersant. The mixture of pigment particles, dispersant, dispersion binder, and solvent is often called millbase, and chemical balance plays an important role in maintaining its dispersion stability.[6,7]

Color filters are required to exhibit the properties of wide color gamut, high color brightness, and improved contrast ratio.[8–10] Among them, contrast ratio (CR) is important in determining the quality of color filters and the display device containing them. CR is a measure of a display system, representing the ratio of the luminance of the brightest color (white) to that of the darkest color (black) that the system is producing, and is typically defined as $T_{0°}/T_{90°}$, where $T_{0°}$ and $T_{90°}$ are transmittances of the system when optical axes of polarizer and analyzer correspond to $0°$ and $90°$, respectively (Fig. 1).[11,12] To achieve a high CR, light transmittance should be maximized under parallel polarizers and minimized under crossed polarizers. There have been several attempts to increase contrast by designing cell structures with a high aperture ratio and by optimizing the polarizers' retardation.[13,14] It has also been reported that light leakage in the black state caused by scattering media of displays is responsible for poor contrast.[15–17] Pigment particles are among the prime sources for this, and therefore, pigment dispersion is claimed to be closely related to the final CR.

However, notwithstanding that pigment dispersion profoundly affects the characteristics of the color filter, systematic literature on this subject is quite rare. It is because of the difficulties in obtaining and evaluating stable distribution of fine pigment particles in the final film as well as in the liquid millbase, which arises from its inherently
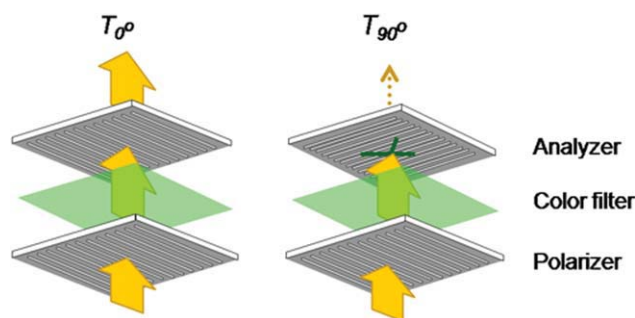
Figure 1 Schematic diagram for contrast ratio measurement. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

complex chemical and physical properties. In this article, we present theoretical approaches concerning CR as a measure to better understand the optical conditions affecting the CR of pigment-containing thin film. Then, we establish one specific experimental condition, with attention to chemical composition of dispersion binders. The findings explained here are applicable to other thin films containing particle distribution, and systematic experiments about other ingredients will follow based on our current approach.

EXPERIMENTAL

Materials

C.I. pigment green 36 (G36) and C.I. pigment yellow 150 (Y150) were supplied by Dainichiseika Chemicals (Fig. 2). Disperbyk-2001 (Modified acrylate block copolymer, amine value 29, acid value 19, BYK-Chemie) was used as a dispersant without further purification. Monomers, including benzyl methacrylate (BzMA), methacrylic acid (MAA), and acrylic acid 2-(2-carboxy-ethoxycarbonyl)-ethyl ester (HOA-MS), 2,2′-azobisisobutyronitrile (AIBN) and propylene glycol monomethyl ether acetate (PGMEA) were purchased from Aldrich. For preparation of the color filter film, dipentaerythritol hexaacrylate (DPHA) was purchased from Kyoeisha Chemicals, and initiator (Irgacure369) and leveling agent (EFKA3288) were obtained from Ciba Specialty Chemicals.

Dispersion binder preparation

Acrylic ester copolymers for dispersion binders were prepared by free radical copolymerization following previous reports.[18–20] In a three-necked flask, PGMEA (150 mL) was preheated to 80°C. For PB-1, BzMA (42.5 g, 241 mmol), MAA (7.5 g, 87 mmol), and AIBN (4.9 g, 30 mmol) were premixed in PGMEA (50 mL), and then injected dropwise into the preheated PGMEA (150 mL) for 2 h. The mixture was stirred for another 4 h at 80°C, and then cooled

to room temperature to complete the reaction. For PB-2, BzMA (40 g, 227 mmol), MAA (5 g, 58 mmol), HOA-MS (5 g, 22 mmol), and AIBN (4.9 g, 30 mmol) were added, following the previous reaction. And, for PB-3, the same condition was applied, except that a larger amount of AIBN was used (7.6 g, 46 mmol) and that reaction time was reduced to 3 h. The molecular weights of the dispersion binders were measured by gel permeation chromatography (GPC) in THF using polystyrene as standard. Their acid values were determined by KOH titration and solid contents were measured after removing the solvent in hot-oven (200°C) for 1 h. These results are listed in Table I.

Millbase preparation

An oscillatory shaker (Asada Co) was used for preparation of millbases. A dispersing mixture containing G36 (6.67 g, 4.8 mmol), Y150 (4.09 g, 12.0 mmol), Disperbyk-2001 (2 g), and dispersion binder (5.38 g) was prepared and mixed with PGMEA (35 mL) in a 100 mL closed vessel. The mixture was vigorously stirred for prewetting, and then zirconia beads (diameter 0.2 mm, 10 g) were put into the dispersing mixture before loading to the shaker. After loading into the shaker, milling was continued for 20 h, then diluted with PGMEA (10 mL), followed by dispersion for another 2 h. The mean size of the millbase was determined by the dynamic light scattering method (HORIBA HA-500).

Colorant film preparation

Millbase (33.53 g) was mixed with DPHA (1.96 g), Irgacure369 (2.42 g), EFKA3288 (1.00 g), and PGMEA (4.23 g), and then thoroughly stirred for 3 h at room temperature. For preparation of the color filter film, the mixture was first applied on a glass surface by spin-coating. Its thickness was adjusted from 1.0 to 2.5 μm depending on the RPM of the spin-coater
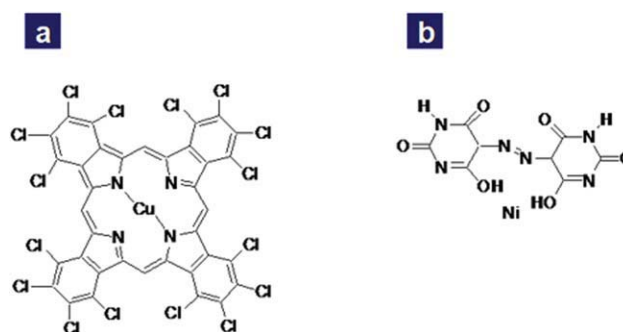

Figure 2 Chemical structure of organic pigments used in this study: (a) C.I. pigment green 36, (b) C.I. pigment yellow 150.

TABLE I
Chemical Composition for Dispersion Binders Used in this Study

Binder	Composition (wt %)			Mw (PDI)	Acid value	Solid content
	A	B	C			
PB-1	85	15	–	23,100 (1.73)	96	21.0 %
PB-2	80	10	10	22,500 (1.79)	88	20.7 %
PB-3	80	10	10	14,000 (1.81)	87	21.8 %

(500–900 rpm for 30 s). The film went through pre-baking (110°C, 30 min) and UV exposure (I-line, 365 nm, 100 mJ/cm^2, contact mask aligner). For complete crosslinking, thermal baking was carried out at 230°C for 30 min.

Property characterization

Color coordinates (x and y) and brightness (Y) of the colorant film were measured using Photal OTS (UKA electronics), and its contrast ratio was measured by CT-1B (TSUBOSAKA). Film thickness was measured with a long scan surface profiler (KLA-Tencor P-11). To investigate pigment particle distribution in thin film, SEM images of the fractured cross-section in the color filter films were taken (Hitachi S-4200). The surface profile and topology were measured with atomic force microscopy (CSPM 4000).

RESULTS AND DISCUSSION

Theoretical consideration regarding contrast ratio

First, we consider the system which includes only polarizers, i.e., a polarizer and an analyzer. If a light incident on the polarizer has an intensity of I_0, the horizontal and vertical intensities are given by $I_h = I_0 \cdot t_{\parallel}/2$ and $I_v = I_0 \cdot t_{\perp}/2$, respectively, where $t_{\parallel}$ and $t_{\perp}$ are transmittances of the single polarizer for linearly polarized light when the plane of light polarization and polarizer's optical axis are parallel and perpendicular to each other, respectively. Then, intensity I after passing a polarizer is expressed by $I = I_h + I_v = I_0 \cdot (t_{\parallel} + t_{\perp})/2$. If we have an analyzer which is parallel to the polarizer, the intensities are given by $I_h = I_0 \cdot t_{\parallel} \cdot t_{\parallel}/2$ and $I_v = I_0 \cdot t_{\perp} \cdot t_{\perp}/2$, respectively. Then, we can obtain $I = I_0 \cdot (t_{\parallel}^2 + t_{\perp}^2)/2$. Rearrangement yields

$$\frac{I}{I_0} = \frac{1}{2} \cdot I_0 \cdot (t_{\parallel}^2 + t_{\perp}^2) = T_{0°} \qquad (1)$$

Similarly, when an analyzer is placed perpendicular to the polarizer, the intensities are given by $I_h = I_0 \cdot t_{\parallel} \cdot t_{\perp}/2$ and $I_v = I_0 \cdot t_{\perp} \cdot t_{\parallel}/2$, respectively. In this case, light intensity can be expressed as $I = I_0 \cdot t_{\parallel} \cdot t_{\perp}$. Then, we get the following

$$\frac{I}{I_0} = t_{\parallel} \cdot t_{\perp} = T_{90°} \qquad (2)$$

In all cases it is assumed that light does not change its polarization state while spreading through polarizers. The definition of CR gives

$$CR_{p+a} = \frac{T_{0°}}{T_{90°}} = \frac{t_{\parallel}^2 + t_{\perp}^2}{2 \cdot t_{\parallel} \cdot t_{\perp}} \qquad (3)$$

Now, we consider the case with a color filter film between a polarizer and an analyzer. We assume that horizontally polarized light at the input of the color filter has intensity I_h^{input}. After spreading through the film, the light will become partially depolarized. If the apparent component of polarization in the vertical plane has relative intensity Δ, we obtain $I_h^{\text{output}} = (1 - \Delta) \cdot I_h^{\text{input}}$ and $I_v^{\text{output}} = \Delta \cdot I_v^{\text{input}}$ at the exit of the color filter. For simplicity, we assume in the case of $\Delta \ll 1$ that the light intensities after a polarizer and the color filter, but before an analyzer, are given by $I_h \approx I_0 \cdot t_{\parallel}/2$ and $I_v = I_0 \cdot (t_{\perp} + \Delta \cdot t_{\parallel})/2$, respectively. When an analyzer is placed parallel to the polarizer, the intensities are given by $I_h \approx I_0 \cdot t_{\parallel} \cdot t_{\parallel}/2$ and $I_v = I_0 \cdot (t_{\perp} + \Delta \cdot t_{\parallel}) \cdot t_{\perp}/2$, respectively. From the relationship of $I \approx I_0 \cdot [t_{\parallel}^2 + (t_{\perp} + \Delta \cdot t_{\parallel})^2]/2$, we obtain the following

$$\frac{I}{I_0} = \frac{1}{2} \cdot \left[t_{\parallel}^2 + (t_{\perp} + \Delta \cdot t_{\parallel})^2 \right] = T_{0°} \qquad (4)$$

Similarly, when an analyzer is placed perpendicular to the polarizer, the intensities are expressed by $I_h \approx I_0 \cdot t_{\parallel} \cdot t_{\parallel}/2$ and $I_v = I_0 \cdot (t_{\perp} + \Delta \cdot t_{\parallel}) \cdot t_{\parallel}/2$, respectively. Then, the relationship $I \approx I_0 \cdot t_{\parallel} \cdot (2 \cdot t_{\perp} + \Delta)/2$ yields

$$\frac{I}{I_0} = \frac{1}{2} \cdot t_{\parallel} \cdot (2 \cdot t_{\perp} + \Delta) = T_{90°} \qquad (5)$$

By combining eq. (4) and eq. (5), the CR will be presented as

$$CR_{p+CF+a} = \frac{T_{0°}}{T_{90°}} = \frac{t_{\parallel}^2 + (t_{\perp} + \Delta \cdot t_{\parallel})^2}{t_{\parallel} \cdot (2 \cdot t_{\perp} + \Delta)} \qquad (6)$$

TABLE II
$T_{0°}$, $T_{90°}$ and Contrast Ratio of Each PR Made of
Different Dispersion Binders at the Same Film
Thickness (1.5 μm)

Binder	Contact Ratio	$T_{0°}$	$T_{90°}$
no CF	5,020	500.8	0.0998
PB-1	2,343	270.4	0.1154
s/b PB-2	2,588	273.6	0.1057
s/b PB-3	2,799	272.1	0.0972

In the case of $t_{\parallel} \gg t_{\perp}$, eq. (3) becomes $CR_{p+a} = t_{\parallel}/2 \cdot t_{\perp}$. Also, in the case that $t_{\parallel} \gg t_{\perp}$ and $\Delta \ll 1$, eq. (6) becomes $CR_{p+CF+a} = t_{\parallel}/2\cdot(t_{\perp} + 0.5 \cdot \Delta)$. Therefore, by combining these relationships, we achieve the following,

$$\frac{CR_{p+CF+a}}{CF_{p+a}} = \frac{t_{\perp}}{t_{\perp} + 0.5 \cdot \Delta} = \left(1 + \frac{0.5 \cdot \Delta}{t_{\perp}}\right)^{-1} \quad (7)$$

When a color filter was placed between two polarizers, we observe a significant decrease in the CR (see in Table II). Since the CR decreases by half after insertion of a color filter, eq. (7) simplifies to

$$\Delta = 2 \cdot t_{\perp} \quad (8)$$

Therefore, it is critical to reduce Δ, i.e., partial depolarization, to circumvent the drop in the contrast ratio after inserting the color filter.

Light depolarization is closely related to the optical uniformity of the colorant film. In the presence of pigment particles it is assumed to be dimmed, having a haze or scattering while proceeding through the CF film.[21,22] The nature of this scattering is thought to be diffractive because the sizes of nonuniformity can be compared with or less than wavelength. Indeed, there should be diffraction on the optical nonuniformity with sizes equivalent to wavelength. Because of diffraction light polarization as well as light, the direction is changed randomly in the current case.

The uniformity is affected by the status of both film volume and its surface. Volume uniformity can be improved by achieving smaller $\|n_p - n_0\|$, where n_p and n_0 are the refractive indices of particles and base material, respectively. However, as n_p is given by the inherent nature of pigment, it is hard to change it unless we produce a new class of pigment structure. If we reduce film thickness and particle concentration, the CR would be improved because of a decreased diffractive loss. But, this is also restricted because of the requirement for the color gamut and color saturation at a given display system. Apparently, we should crush pigment particles into smaller ones. With the advent of advanced

grinding techniques, we obtain nanoscale particles. However, from an inherent nature of pigment particles, we have often encountered limitation to make finer particles than 50 μm. Also, smaller particles deteriorate the physical properties of colored film, especially thermal and chemical stabilities. Often the dispersion stability of the millbase breaks down after its thin film is fabricated. Thus, it is important to optimize the condition which will maintain the uniformity even after the film is formed. We have been trying to develop chemical compositions of the millbase. In the following section, as a specific case, we consider the effect of polymeric binder on CR.

Effect of polymeric binder on particle distribution

There are many components affecting the pigment distribution in a color filter, including pigment size, dispersant, and dispersion binder. Several works have reported about pigment dispersion depending on dispersants.[23,24] However, few reports have been made on dispersion binder, though we can hardly overlook the fact that the nature of the binder has a large influence. More importantly, we notice that chemical composition of the binder exerts a critical effect on pigment distribution in the film state.

Specifically, we consider typical green color filter films for liquid crystal displays. It is generally accepted that both green and yellow pigments are necessary for expansion of the color gamut as required. We milled together pigment G36 and Y150 (75:25, wt %) to satisfy color coordinates of (x, y) = (0.281, 0.590) based on the 1931 CIE chromaticity diagram. This is a required specification for a green color filter of typical liquid crystal displays. Comilling proves effective in optimizing the dispersion in the solvent media, even though they have different particle sizes and physical properties.

Three dispersion binders were prepared depending on the ratio and type of monomers used. PB-1 is a common acrylic polymer having co-monomers of BzMA and MAA. For PB-2 and PB-3, an additional monomer, HOA-MS, was added to form copolymers with BzMA and MAA. PB-3 was polymerized to give a smaller molecular weight than PB-2. Their molecular weights, acid values, and solid contents were measured and listed in Table I. We observed no traces of their derivatives of low molecular weight in GPC measurement, thus confirming the synthesis of polymeric binders.

Colorant films were prepared and their spectral properties were compared in terms of their color property, brightness, and contrast ratio (Fig. 3). As shown in Figure 3(a), the three films exhibit almost identical spectral ranges, as they include the same amount of pigment in their film preparation. We observe almost identical brightness (Y) for all
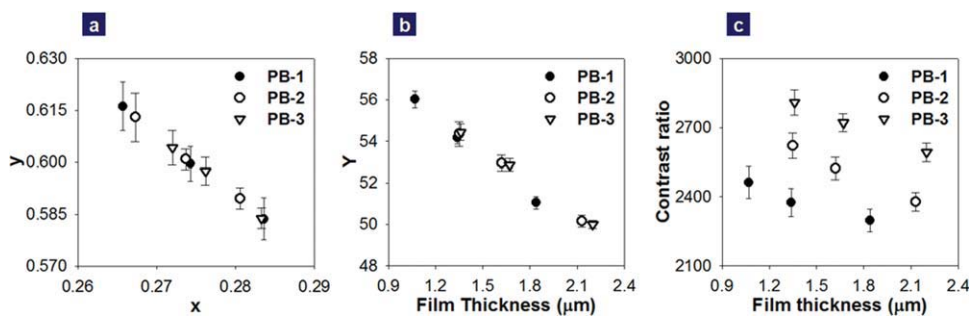
Figure 3 (a) Color coordinates (x, y), (b) brightness (Y), and (c) contrast ratio of green-colored color filter films fabricated using different dispersion binders (PB-1, PB-2, PB-3).

colorant films. We also observe that brightness linearly decreases with film thickness [Fig. 3(b)]. As for the CR, however, we noticed a significant difference depending on dispersion binders. Dispersion binders, PB-2 and PB-3, containing HOA-MS are found to exhibit a significant increase in CR by up to 19.5%, compared with PB-1. PB-3 with a smaller molecular weight shows the highest CR.

For a more systematic study, we separately measured $T_{0°}$ and $T_{90°}$ of the films with thicknesses of 1.5 μm (Table II). The three films have identical $T_{0°}$ values, but there exist noticeable differences in $T_{90°}$ depending on the dispersion binders used. This, as a consequence, leads to different CRs. We ascribe this result to the effect of dispersion binder having a long side-chain. It has previously been reported that carboxylate functional groups strongly interact with the pigment surface, producing a high adsorption density. When carboxylate is attached at the long side-chains, it binds more effectively to the surface of pigment particles.[25–27] In the same way, HOA-MS-containing binders enable an enhanced interaction between dispersant and binder or between pigments and binder, from the formation of more effective bonding. It results in a decrease in $T_{90°}$, leading to a higher CR. It is surprising that a small change in chemical composition of the dispersion binder severely affects the contrast of the colorant film. However, despite a slight improvement, long side-chain binders cannot prevent the CR from dropping by colorant films.

To examine volume uniformity of pigment particles, we have taken scanning electron microscope (SEM) images of the fractured cross-section of the color filter films (Fig. 4). Grain size in the cross-section varies from 70 to 200 nm, but in the SEM images we do not see a big difference, though PB-1 exhibits slightly higher fractured roughness. However, in the atomic force microscope (AFM) measurement, root mean square roughnesses on the film surface were measured to be highest for PB-1 and lowest for PB-3, exhibiting 103.1 Å, 75.6 Å, and 51.5 Å for PB-1, PB-2, and PB-3, respectively (Fig. 5). It is clear that PB-3 produces the smoothest surface in colorant film. This implies that dispersion binders affect the uniformity of pigment particles and binders containing HOA-MS are more effective for uniform distribution of the particles. Often pigment dispersion is evaluated by particle size analysis in the liquid state.[28,29] When particle size was measured by the dynamic light scattering method, unlike what was expected, the three formulations show identical particle size distribution in liquids, with mean values of 80 nm. Therefore, we concur that dispersion binder affects the aggregational behavior effectively when the colorant film solidifies.

CONCLUSIONS

In conclusion, by analyzing the factors affecting the optical property of a pigment-containing system, we are able to elucidate the conditions under which
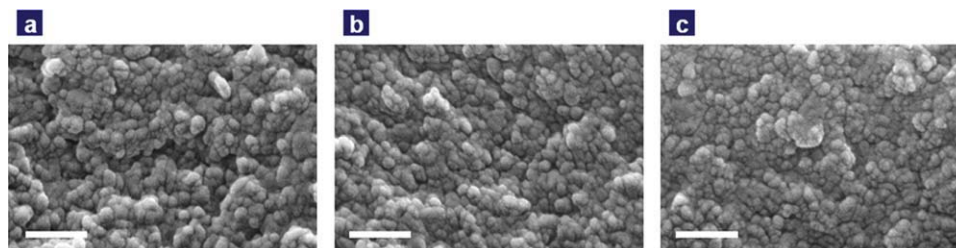

Figure 4 Scanning electron microscope (SEM) images of fractured cross-section of thin films which were made using dispersion binders of (a) PB-1, (b) PB-2, and (c) PB-3. Scale bar shows 500 nm.
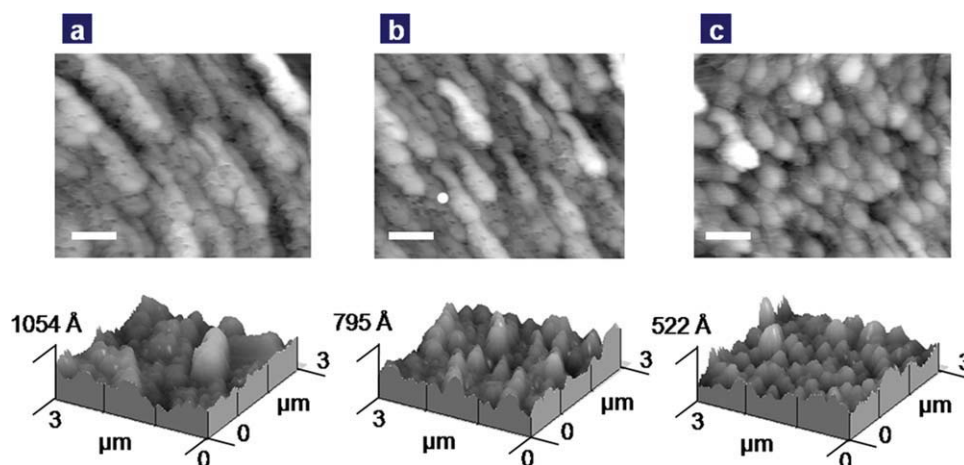
Figure 5 Atomic force microscope (AFM) images on the surface of the three thin films which were made using dispersion binders of (a) PB-1, (b) PB-2, and (c) PB-3. Scale bar shows 500 nm.

enhanced contrast can be achieved. Maintaining uniform distribution of pigment particles in a solid state of thin film well, as in millbase dispersion, is regarded as important. Chemical composition of dispersion binder has a huge effect, and carboxylate in a long side-chain is found to be favorable for high contrast. We believe that our contribution will prove beneficial in future approaches for pigment dispersion and provide momentum to successive research efforts for developing chemical composition and structure of the millbase. We are now conducting further experiments about dispersant binders having new chemical structures which will be reported in a separate article.

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