Microcup® displays: Electronic paper by roll-to-roll manufacturing processes

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Abstract — Rolls of flexible displays or electronic paper have recently been prepared by a high-speed roll-to-roll manufacturing process based on SiPix's novel Microcup® and top-sealing technologies.[1-5] Both Microcup® electrophoretic displays (EPDs) and LCDs have been demonstrated. The display rolls are format flexible and may be cut into desirable size and shape for a variety of applications. High-performance flexible passive-matrix Microcup® EPDs having a wide range of threshold voltages have also been demonstrated.

Keywords — *Electronic paper, Microcup® electrophoretic display, EPD, EPID, passive matrix threshold, active matrix, dispersed LCD, PDLC, top sealing, reflective display, bistability, roll to roll, plastic display, format flexibility.*

1 Introduction

A flexible and durable reflective display with low power consumption, high contrast ratio, fast response time, thin and lightweight, and low manufacturing cost has been pursued for many years for various applications such as cellular phones, PDAs, e-signs, and e-books. Among various reflective display technologies, electrophoretic display (EPD or EPID) has been considered as one of the most promising and actively investigated in the past few years. In addition to the traditional EPDs disclosed since 1969,[6–10] several interesting approaches including microencapsulated EPDs,[11–13] in-plane EPDs,[14–16] and reverse emulsion EPDs,[17] have been disclosed for improvements particularly in substrate flexibility and potential cost reduction. However, so far, none of the prior art EPD technologies has achieved business success partly because of (1) the high cost associated with the inefficient manufacturing processes and (2) the technical challenges in the making of passive-matrix electrophoretic displays (PMEPDs).[6,10,18–20] Most electrophoretic fluids do not have the required threshold characteristics to suppress or eliminate the undesirable cross-talk or cross-bias among adjacent pixels during matrix driving. An additional conductor layer[6] or grid electrode[10,18] has been employed to suppress the cross effects. PMEPDs have been demonstrated, but the manufacturing cost for such multilayer electrode structures is very high. Alternatively, magnetic particles and a magnetic electrode have been proposed to provide the required threshold effect,[19] also at the expense of manufacturing cost. An electrophoretic fluid having inherent threshold characteristics has been reported[20] with tradeoffs in, for example, response time, operation voltage, image uniformity, and display longevity.

SiPix Imaging, Inc., has recently disclosed a roll-to-roll EPD manufacturing process based on novel Microcup® and top-sealing technologies.[1–4] Ultra thin, ultra light, durable, and flexible EPDs with excellent format flexibility and many desirable display features have been manufactured on a continuous plastic web at very high speed and low cost. Low-voltage driving active-matrix and segment EPDs and passive-matrix Microcup® EPDs having threshold voltages ranging from 5 to 50 V have also been demonstrated for a variety of applications. The inherent threshold characteristics of the SiPix Microcup® PMEPDs has been achieved by optimizing the particle-particle, particle-sealing, and particle-Microcup® interactions without the need of complex electrode structure or magnetic particles/electrodes. No tradeoff in colloidal and shelf life stability was observed.

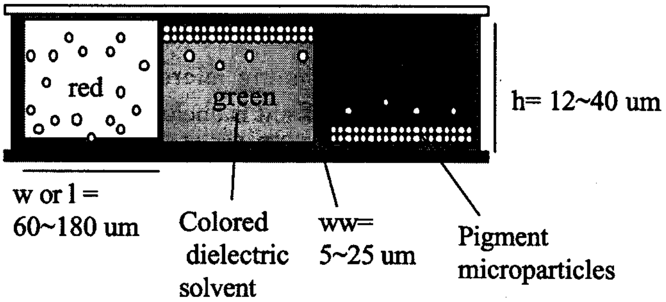

FIGURE 1 — Schematic cross-section of a color Microcup® EPD. Each Microcup® is isolated and seamlessly top-sealed.

Format flexible Microcup® LCDs[5] have also been prepared by the same roll-to-roll top-sealing technologies with much smaller Microcups.® This new type of LCD is in a way a mono-dispersed PDLC[21] (polymer dispersed liquid crystal) with superior cost structure, physico-mechanical properties, and switching characteristics including fast-response rate, low orientation field, and negligible hysteresis.

2 Microcups® and roll-to-roll manufacturing processes

As can be seen in the schematic drawing of a typical Microcup® EPD (Fig. 1), an electrophoretic fluid comprising

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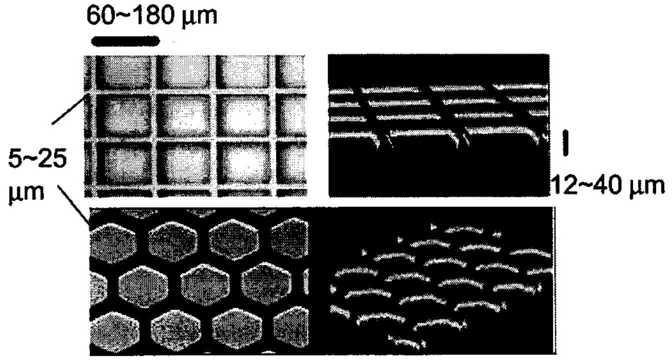

FIGURE 2 — Surface profiles of some typical Microcup® arrays as measured by a Wyko NT1000 surface profiler.

charged pigment (TiO_2)-containing microparticles dispersed in a colored dielectric solvent is enclosed and seamlessly sealed in the Microcups.® Depending on the application, the dimension of Microcups® may vary in the range of 60–180 µm (*w* or *l*) × 12–40 µm (h) × 5–25 µm (ww) for EPDs (Fig. 2). Much smaller Microcups® are employed for LCD applications [Figs. 3(a) and 3(b)].

A schematic process flow of a typical SiPix roll-to-roll display-manufacturing process is shown in Fig. 4. For example, rolls of Microcup® EPDs have been prepared by (1) coating a radiation curable resin composition on a conductor film, (2) embossing and hardening the resin composition, (3) filling the Microcups® with an electrophoretic fluid, (4) seamlessly top-sealing the filled Microcups® by the SiPix one-pass or two-pass top-sealing process[1–5] that will be discussed later, and finally (5) laminating the top-sealed Microcups® with a release liner or a second conductor film. An optical micrograph of the cross-section of a top-sealed square Microcup® array with a 3–6 µm sealing/adhesive layer is shown in Fig. 5. Color EPDs may be realized by overlaying a color filter onto the monochrome display or by selectively filling or printing R,G,B color fluids in predetermined Microcups® with registration, followed by one of the SiPix top-sealing processes.

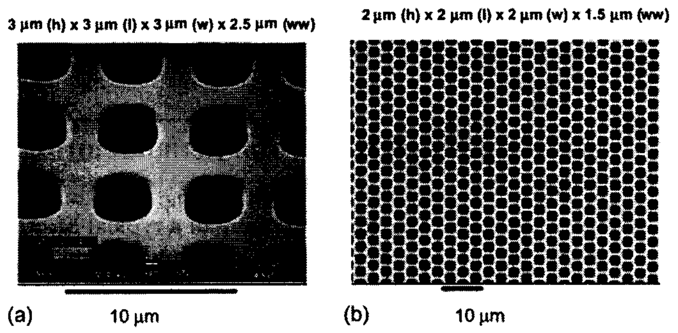

FIGURE 3 — SEM micrographs of two typical Microcup® arrays for LCD applications.

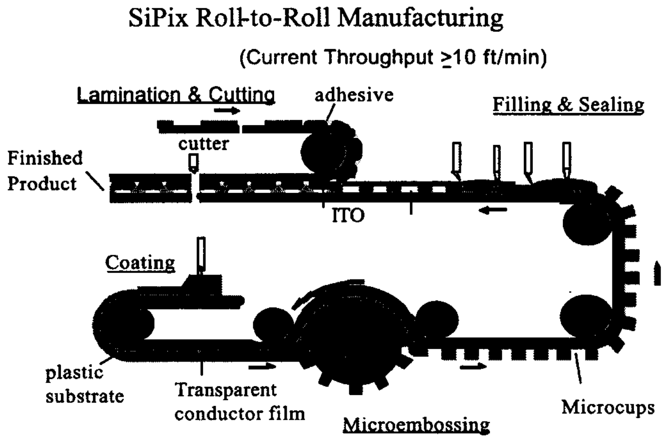

FIGURE 4 — Schematic process flow of the SiPix roll-to-roll manufacturing process

3 Pigment-containing microparticles

SiPix proprietary charged pigment particles are narrowly dispersed, submicron TiO_2-containing microparticles in, for example, a high-density halogenated dielectric solvent.[22–25] The microparticles are density matched very closely to the dielectric solvent by coating or encapsulating TiO_2 primary particles (diameter $\approx$0.3 µm) with a highly crosslinked polymer matrix.[25–26] Excellent colloidal stability has been observed even after centrifuged at 1000g for more than 30 min.

4 SiPix's filling and top-sealing processes[1–5,22–24]

The filling and top-sealing processes are the critical steps of the roll-to-roll manufacturing process. The traditional edge-sealing and vacuum-filling processes for assembling a LCD cell are not useful at all for the Microcup® structure. Directly laminating a conductor film onto the filled Microcups® typically results in a poor sealing with unacceptable defects such as de-wetting, de-lamination, trapped-in air pocket or void, and non-uniform image quality. To enable the roll-to-roll manufacturing of Microcup® displays, two high-speed continuous filling and top-sealing processes have been developed (Figs. 6 and 7). High-quality seamless

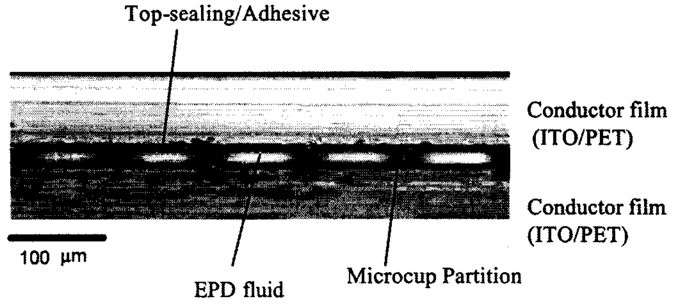

FIGURE 5 — Cross-section optical micrograph of a typical SiPix top-sealed Microcup® EPD array prepared by a roll-to-roll process.

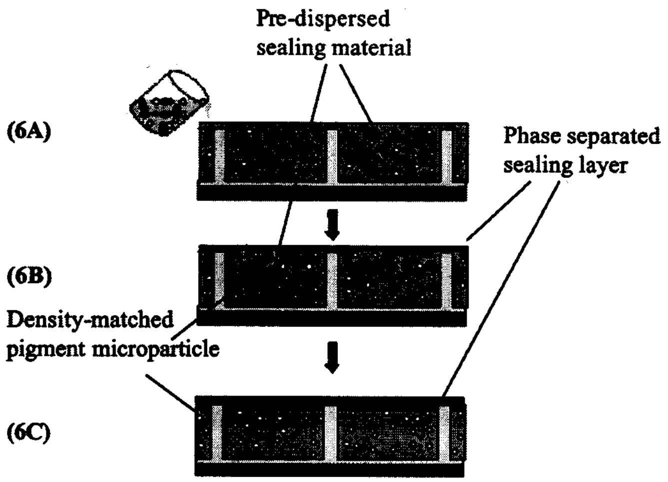

FIGURE 6 — Schematic SiPix one-pass top-sealing process: (a) Filling the Microcup® with an electrophoretic fluid comprising a pre-emulsified sealing composition; (b) Phase separation and floating of the lighter sealing composition to the top of the fluid; (c) Hardening the top-sealing layer.

sealing of the filled Microcups® has been demonstrated at a speed of more than 30 ft./min by both processes.

In the SiPix one-pass filling and top-sealing process (Fig. 6), a sealing composition is pre-emulsified in the electrophoretic fluid containing density-matched pigment microparticles and coated onto the Microcups® [Fig. 6(a)]. The sealing composition is optimized to allow fast phase-separation and creaming of the sealing layer to form a contiguous film with acceptable integrity on the top of the filled Microcups® [Fig. 6(b)]. The phase-separated sealing layer may then be hardened by methods such as solvent evaporation, moisture, thermal, and radiation curing to enclose the fluid in the Microcups® [Fig. 6(c)].

Alternatively, the Microcups may be filled and top-sealed by the SiPix two-pass top-sealing process (Fig. 7). The Microcups® are partially filled with an electrophoretic fluid [Fig. 7(a)] and subsequently overcoated with a top-sealing layer [Fig. 7(b)]. The top-sealing layer is then dried and hardened [Fig. 7(c)] by, for example, interfacial polymerization/crosslinking, moisture, radiation, and thermal curing, or their combinations.

To achieve an acceptable seamless top-sealing by either process, at least six critical parameters must be optimized: (1) the density difference between the top-sealing composition and the electrophoretic fluid; (2) the degree of incompatibility among the pigment microparticles, the dielectric solvent, and the top-sealing composition; (3) the surface tension and interfacial tension of all interfaces or surfaces involved; (4) the interaction between the top-sealing composition and the partition walls; (5) the rheology of both the top-sealing composition and the electrophoretic fluid; and (6) the integrity and physico-mechanical properties of the top-sealing layer during and after sealing. The one-pass sealing process is very attractive because of its simplicity. However, the two-pass process is very useful particularly when the sealing composition is at least partially compatible with the electrophoretic fluid. Microcup® LCDs have also been successfully prepared roll-to-roll by similar filling and top-sealing processes.[5]

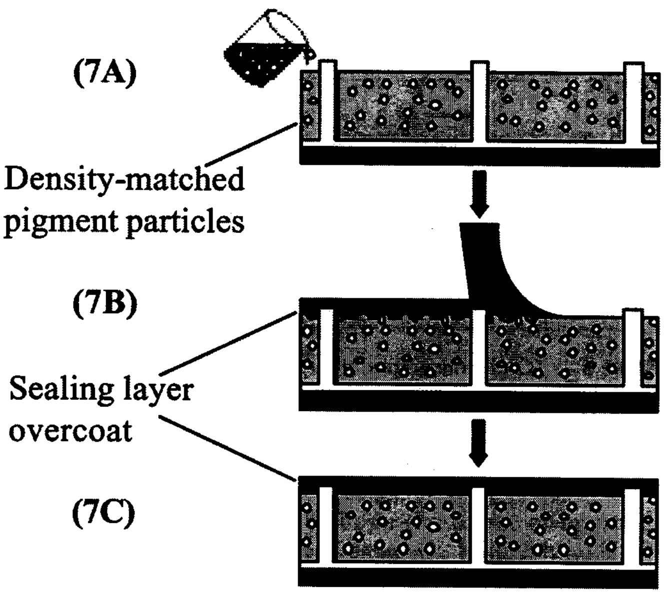

FIGURE 7 — Schematic SiPix two-pass top-sealing process: (a) Filling the Microcup® with an electrophoretic fluid; (b) Overcoating a light, incompatible sealing layer onto the partially filled Microcup®; (c) Phase separation and hardening of the top-sealing layer.

5 Barrier properties of the top-sealing layer

For active-matrix EPDs (AMEPDs) and direct-drive EPDs, Microcups® may be formed on a non-patterned electrode layer, top-sealed and laminated onto a release liner. The resultant sandwiched EPD film may be converted by customers to a module by peeling off the release liner and subsequently laminating the top-sealed Microcups® onto a patterned electrode layer for direct-drive EPDs or a TFT backplane for AMEPDs. To preserve the dielectric solvent in the top-sealed Microcup® during shipping and storage, a sealing layer with satisfactory barrier properties is required. Figure 8 shows the TGA thermographs of a low-boiling-point dielectric solvent in open and as-sealed Microcup® arrays without any protecting layer above the top-sealing layer. As shown in Fig. 8, the onset temperature (T_{onset}) of solvent evaporation in the open Microcup® array is about 33°C (curve a). The same solvent was filled in the Microcups® and top-sealed with, for example, a highly crosslinked elastomer-acrylic interpenetrating network (IPN).[22–24,27] As it can clearly be seen that the T_{onset} was significantly increased to 179°C, 186°C, and 197°C when the thickness of the sealing layer is 1–2, 3–4, and 4–5 μm, respectively, as determined by cross-sectional SEM of the top-sealed Microcups®. A seamless sealing layer with good film integrity is evident from the high T_{onset} of the top-sealed samples.

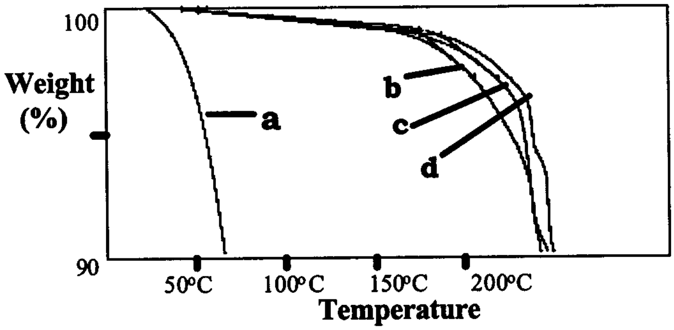

FIGURE 8 — TGA thermographs of a typical dielectric solvent in open Microcups® (curve a, Tonset = 33°C) and in a top-sealed Microcup® array; thickness of the sealing layer: 1–2 μm (curve b: T_{onset} = 179°C) 3–4 μm (curve c, T_{onset} = 187°C), and 4–5 μm (curve d, T_{onset} = 192°C). All the TGA were measured without a top substrate laminated onto the top-sealed Microcup®.

The activation energy of solvent permeation through the top-sealing layer was estimated by the TGA heating rate method[28] to be about 26 kcal/mol. The high T_{onset} together with the high barrier activation energy assure a long process green time of the as-sealed Microcups® since even a low-boiling-point dielectric solvent may be well preserved before the subsequent converting and lamination steps.

6 Format flexibility and edge sharpness

Other important features of Microcup® EPDs are its excellent format flexibility and physico-mechanical properties. The Microcup® partition walls are in fact the mechanical support of the display. Since each Microcup® is isolated and seamlessly top-sealed, the Microcup® may be cut into different sizes and shapes without the risk of losing fluid in the active area. Figure 9(a) shows an optical photograph of a direct-drive Microcup® EPD operating at ±15 V and a bending angle of about 120°. A near 180° viewing angle is evident. Figure 9B shows that the same direct drive Microcup® EPD maintained its switching performance even after the display was cut. The excellent format flexibility allows displays of different size and shape to be made from the same jumbo roll of Microcup® EPD prepared by the continuous roll-to-roll manufacturing process. Figure 10 shows the optical micrograph of a typical Microcup® EPD driven with a sharp-edge electrode pattern. It is evident that the electrophoretic composition in Microcups® can be partially addressed to exhibit an edge-sharpness significantly finer than the dimension of the Microcups®.

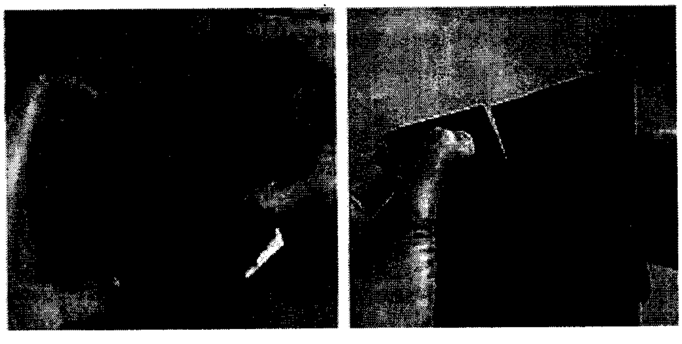

FIGURE 9 — Photographs of a direct-drive Microcup® EPD switched at ±15 V and (a) 120° bending angle; (b) after the display was cut.

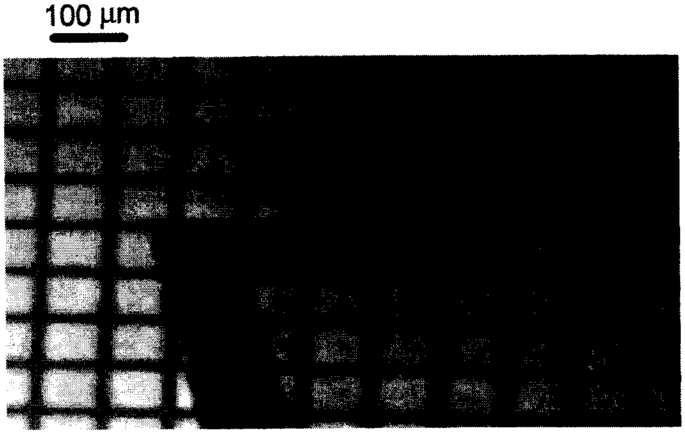

FIGURE 10 — Optical micrograph of a Microcup® EPD showing partially addressed Microcups® using a sharp-edged electrode pattern.

7 Converting the microcup® EPD rolls

Two types of Microcup® EPD films have been manufactured by the SiPix roll-to-roll process: (1) pre-laminated PMEPD rolls sandwiched between row and column electrode films [Fig. 11(a)], and (2) ready-to-laminate EPD rolls sandwiched between a release liner and a common electrode film for active-matrix (AMEPDs) or direct-drive EPDs [Fig. 11(b)]. The PMEPD films are as thin as 0.15 mm, as light as 0.4 g/in², and may be cut to desired size and formats, followed by asymmetrical cutting and stripping processes to expose the row and column electrodes for con-

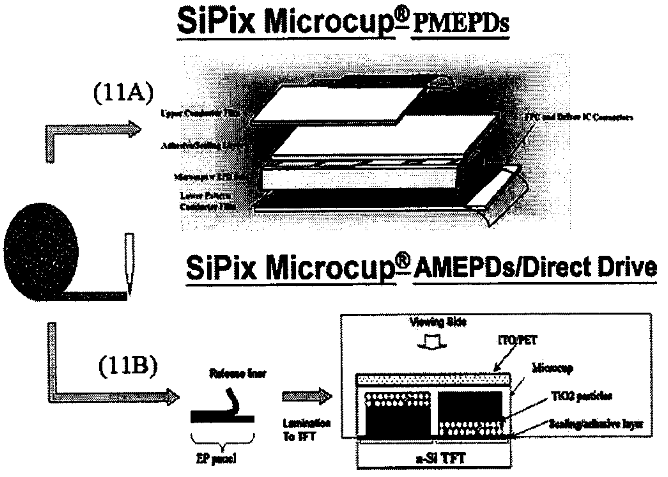

FIGURE 11 — Schematic drawing of two types of EPD rolls: (a) Prelaminated PMEPD between row and column ITO/PET films (b) Release liners/sealed Microcups® on common electrode for AMEPDs and direct-drive EPDs.

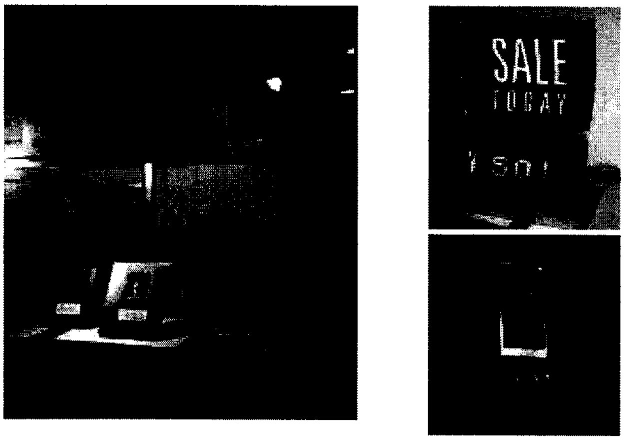

FIGURE 12 — SiPix EPDs in recent exhibitions and conferences. LCD/PDP International, Yokahama (10/12); IDW '02, Hiroshima (12/02); IDMC '03, Taipei (02/03).

nection to outside circuitry.[2] To prepare AMEPDs or direct-drive EPDs, the top-sealed Microcup® on common electrode film may be cut and laminated onto a patterned electrode film or a TFT backplane after the release liner is removed. Figure 12 shows some Microcup® PMEPD and AMEPD prototypes exhibited in recent international display conferences and exhibitions.

8 Operation temperature latitude

The switching rate of an EPD is inversely proportional to the viscosity of the electrophoretic fluid. As temperature decreases, the switching rate typically decreases significantly since the viscosity of a colloidal dispersion generally increases with decreasing temperature. The temperature effect is particularly significant if a phase transition or a critical flocculation phenomenon involved. SiPix's unique filling and top-sealing processes allow the electrophoretic composition be optimized independently from the surrounding partition walls and sealing layer. The excellent barrier properties of the top-sealing layer further allow the use of a low boiling-point solvent of low viscosity, which is relatively insensitive to the change of temperature in the range of interest. Figure 13 shows the effect of operation temperature on the optical response of a typical Microcup® EPD. The arrows represent the sequence of the heating and cooling cycle. It is evident that the optical response of the Microcup® EPD remains almost the same in the entire temperature range investigated. The temperature latitude may be further extended to –20°C optionally with a thermal compensation mechanism.

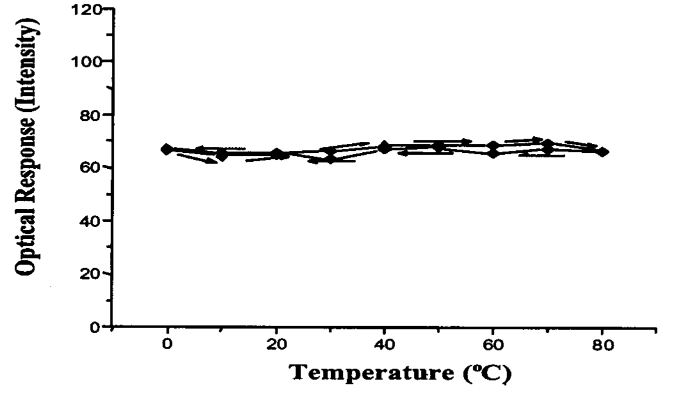

FIGURE 13 — Effect of heating and cooling cycle on the electro-optical response of a typical Microcup® EPD.

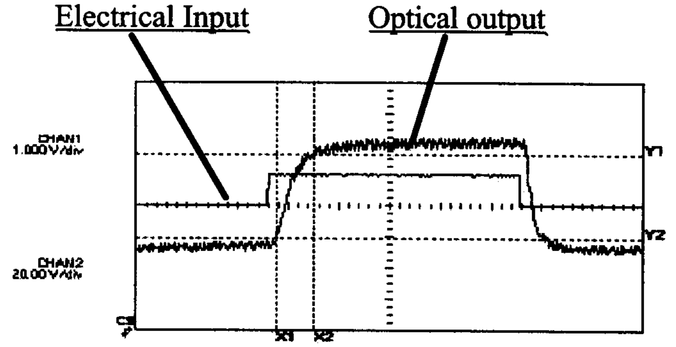

FIGURE 14 — Electro-optical response curves of the first-generation a-Si TFT Microcup® AMEPD driven at ±10 V, $t_{on} \approx 400$ msec.

9 Microcup® AMEPDs

In-plane-switching AMEPDs[14–16] and Microcapsule AMEPDs[29–35] have been reported recently. In comparison, the SiPix Microcup® structure and the roll-to-roll manufacturing processes provide an alternative solution with significant improvements in cost, process latitude, and performance. For example, a jumbo-roll release liner/top-sealed Microcup®/ITO/PET sandwiched film was cut to desirable format. The release liner was peeled off, and the top-sealed Microcup® film was laminated onto a commercially available 3.5-in. color QVGA reflective a-Si TFT backplane. Figure 14 shows the electro-optical response of the resultant a-Si TFT Microcup® AMEPD prototype. The optical response was recorded at a 90° angle to the EPD surface with the incoming light illuminating at 45°.[2] The contrast ratio, frame rate, and image bistability driven at about ±10 V are ≥8, ≤0.4 sec, and ≥24 h, respectively.

10 Passive-matrix microcup® EPDs

A PMEPD driven by traditional column and row electrodes has been generally considered a major technical challenge. The threshold characteristics, which are needed to suppress the undesirable cross effects among non-addressed pixels during multiplexing driving, does not exist in most electrophoretic systems. An electrophoretic fluid having inherent threshold characteristics has been reported,[20] however, with undesirable tradeoffs in response time, operation voltage, brightness, image uniformity, and display longevity.

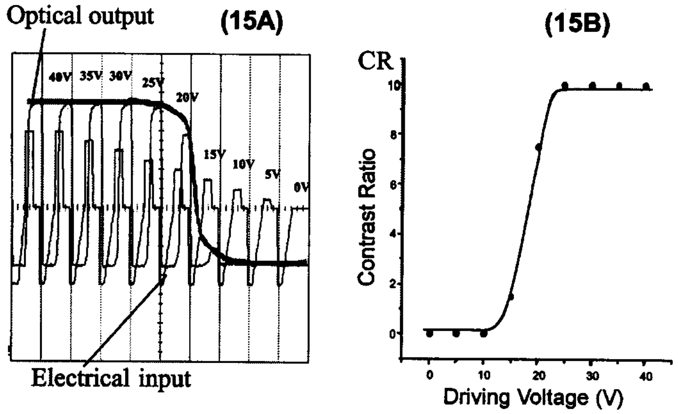

FIGURE 15 — (a) Electro-optical response and (b) contrast ratio–voltage curves of the first-generation Microcup® PMEPD.

In contrast, by optimizing the particle-particle, particle-sealing layer, and particle-Microcup® interactions, satisfactory threshold characteristics has been demonstrated without the tradeoffs.[1–4] High-performance Microcup® PMEPDs have been prepared at high throughput by the SiPix roll-to-roll process using column and row patterned ITO/PET films. Figure 15(a) showed the electro-optical response curve of a typical Microcup® PMEPD as a function of driving voltage from 0 to 40 V. Saturated optical signal could be reached at ≥30 V with a t_{on} of about 60 msec, but no signal was detected at all at ≤10 V. Figure 15(b) shows that the PMEPD exhibits a contrast ratio of about 10 at ≥25 V and a threshold voltage of about 10 V. Excellent image uniformity and shelf-life stability were also observed. By optimizing the composition of the electrophoretic fluid, the Microcups®, and the top-sealing/adhesive layers, high-performance passive-matrix Microcup® EPDs having threshold voltages ranging from 10 to 50 V have been demonstrated (Fig. 16).

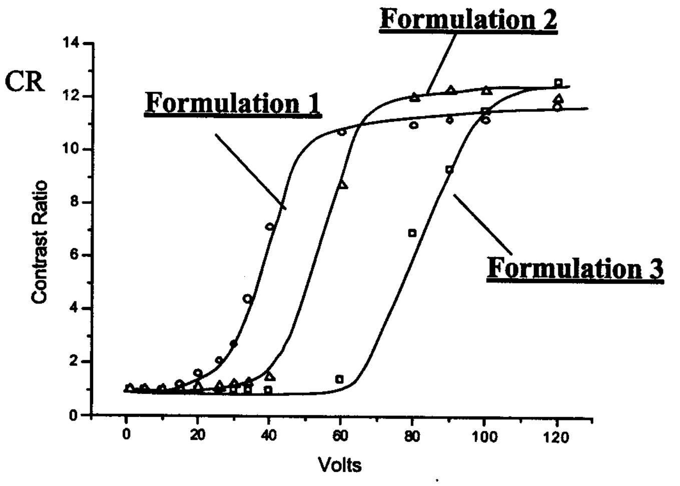

FIGURE 16 — Contrast-ratio (CR) *vs.* applied-voltage curves of three Microcup® PMEPDs showing high CR with threshold voltages ranging from 10 to 50 V.

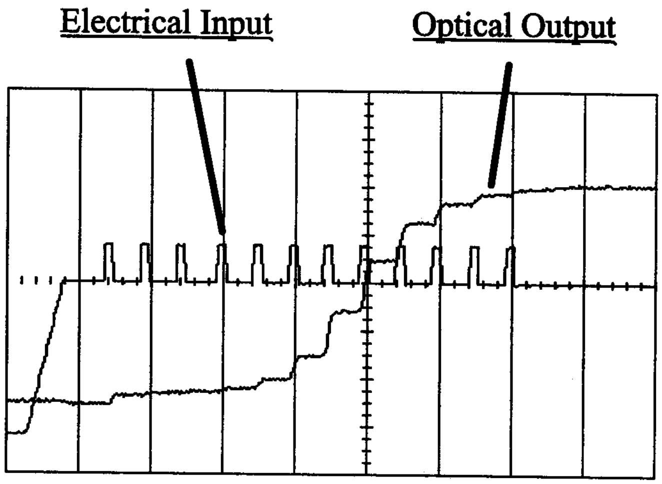

FIGURE 17 — Additive gray-scale rendition showing mid-tone image multistability of a Microcup® EPD.

It is evident that SiPix's electrophoretic composition, Microcup® structure and top-sealing process provide extremely wide formulation and process windows for the optimization of all the above-mentioned interactions and in turn enable high-performance PMEPDs using only row and column ITO electrodes on a plastic substrate. A 110-dpi 160 × 160-line Microcup® PMEPD prototype with a frame rate of 25–30 sec and a contrast ratio of about 10 has been demonstrated recently using conventional row and column STN drivers at 30 V. Faster frame rate may be achieved by using multiple drivers or at higher driving voltage.

11 Gray scale of microcup® EPDs

A microcapsule type of AMEPD capable of four-level gray scales has been demonstrated recently by an area modulation mechanism[35] at the expense of pixel resolution. A 4-bit gray-scale in-plane EPD with reasonable gray-scale bistability has also been demonstrated using pulse width and/or amplitude modulation.[16] Microcup® EPDs are capable of a significantly higher gray scale without the tradeoff in pixel resolution. As it can be seen from Fig. 17 that except for the initial few "wake-up" pulses, the optical response of each electrical pulse is stable and additive until a subsequent pulse is given. Because of the superior multistability of the mid-tone image of Microcup® EPDs, gray-scale rendition may be achieved by pulse amplitude (Fig. 15), pulse width [Fig. 18(a)], or pulse count [Fig. 18(b)] modulation mechanism, or their combinations. Six grayscale levels are shown in Fig. 18, although more than 10 gray levels have been demonstrated by either modulation mechanism. Since the resolution of a typical Microcup® is quite high, 50–180 μm or 120–500 cups/in. as compared to the pixel resolution of a typical LCD, high quality image may be achieved by further combining the area and/or amplitude-modulation mechanisms.

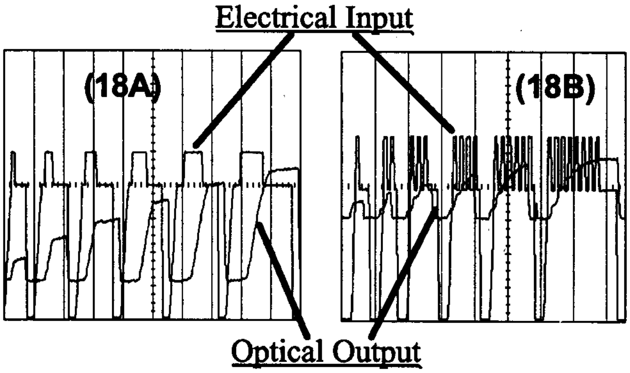

FIGURE 18 — Gray-scale rendition of a typical Microcup® EPD: (a) by pulse-width modulation with a variable pulse width from 20 to 120 msec; and (b) by pulse-count modulation at 20 V with a fixed pulse width of 15 msec.

12 Microcup® LCD: A new type of dispersed LCD[5]

Conventional dispersed liquid-crystal systems typically have a broad distribution of size and shape of LC domains.[21] The resultant PDLC device often exhibits a transmittance–voltage characteristic curve of low gamma with a reorientation field of 1–10 V/μm and a significant hysteresis. The disadvantages of such a device include low level of multiplexing in passive-matrix addressing, slow frame rate, poor reproducibility in gray scale, and trade-off between contrast ratio and operating voltage. An improved system, PELC (polymer-encased liquid crystal)[36] in which liquid crystal is enclosed in hollow monodispersed latex particles have been disclosed to address the disadvantages associated with the broad size distribution. However, the multiple-step synthesis and purification processes involved in the preparation of the PELC system are not straightforward and often quite time-consuming particularly when a dye is used to improve the contrast ratio of displays of thin cell gaps.

Figure 19(a) shows the optical response curves of a conventional photo-induced PDLC film containing 30/70 (w/w) of Merck E7/Norland 65 with a cell gap of 4.5 μm. Figure 19(b) shows the curves of a Microcup® LCD comprising a four-layer sandwich of Microcup® array 3A (Fig. 3), each of four layers filled with 3 wt.% of a dichroic dye, BlueAB2 (from Funktionfluid) in Merck BL006 (cell gap ~16 μm), and subsequently top sealed. The cell gap of the photo-induced PDLC film was selected so that the LC loading was about the same as that of the four-layer filled and top-sealed Microcup® LCD. A significant hysteresis loop was detected in the former [Fig. 19(a)], but negligible hysteresis was found in the Microcup® LCD [Fig. 19(b)]. The t_{ON} and t_{OFF} of the four-layer Microcup® LCD at 40 V are about 0.5 and 11 msec, respectively.

Further improvements in switching performance may be achieved by optimizing the compositions and structure of the Microcup® and LC phases. A Microcup® LCD having a threshold voltage of about 3 V, a clearing voltage of 10–12 V, a reorientation field of less than 1 V/μm, a t_{ON} of 0.5 msec, and a t_{OFF} of 11 msec has been demonstrated.[5]

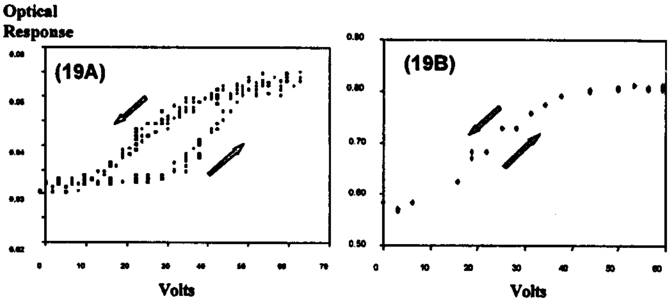

FIGURE 19 — Electro-optical response curves of (a) a traditional photo-induced PDLC and (b) a four-layer Microcup® LCD using the Microcup® array of Fig. 3A.

13 Conclusions

High-performance Microcup® electronic paper or displays have been prepared by the SiPix roll-to-roll manufacturing process at very low cost and high throughput. The unique Microcup® structure and top-sealing processes results in super physico-mechanical properties and format flexibility. They also allow the compositions of the electrophoretic fluid, the Microcups®, and the top-sealing and adhesive layers be optimized independently for optimum display performances.

Plastic PMEPDs having threshold voltages ranging from 10 to 50 V with a sharp gamma, fast response time, super image bistability, and wide temperature latitude have been demonstrated by using simple row/column electrode matrices. Satisfactory multistability of mid-tone images and high levels of gray scale have also been demonstrated without tradeoffs in pixel resolution and display longevity. Microcup® AMEPDs based on a-Si TFT have also been demonstrated. The top-sealing layer has shown great barrier properties for a satisfactory green time and process window for the converting and lamination processes. The first-generation prototype has shown a contrast ratio of ≥8 and a frame rate of ≤ 0.4 sec. at an operation voltage of ≤ ±10 V.

A new type of dispersed liquid-crystal system, Microcup® LCD, has also been developed using the SiPix roll-to-roll process. The display has shown negligible hysteresis, fast-switching rate, and a low reorientation field.

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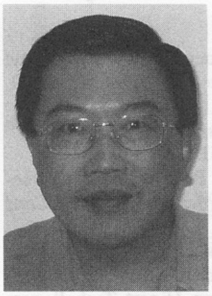

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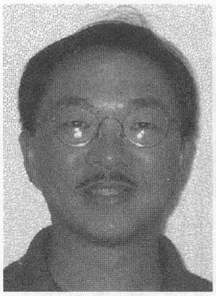

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