All-printed Organic TFT Backplanes for Flexible Electronic Paper

K. Suzuki, K. Yutani, M. Nakashima, A. Onodera, S. Mizukami, M. Kato, T. Tano,
H. Tomono, M. Yanagisawa, and K. Kameyama
Ricoh Co., Ltd.
16-1 Shinei-cho, Tsuzuki-ku Yokohama, 224-0035, Japan

Abstract

80 ppi, 160 ppi and 200 ppi all-printed organic thin film transistor (OTFT) backplanes were fabricated by a surface energy controlled ink-jet printing and several printing methods on plastic substrate. We developed the surface energy controlled ink-jet printing with ultraviolet (UV) irradiation on a novel polyimide film for high-resolution electrode patterning. Minimum line width was 15 μm and minimum space between two electrodes was 2 μm respectively. A 200 ppi all-printed OTFT backplane with a channel length of 5 μm showed high mobility over 0.1 cm^2/Vs with small-molecule organic semiconductor (OSC). To show the scaling capability, we have also fabricated a 300 ppi all-printed OTFT array on plastic substrate. Flexible electrophoretic displays (EPDs) driven by 80 ppi, 160 ppi and 200 ppi all-printed OTFT backplanes were also successfully demonstrated.

1. Introduction

In recent years, printing method has attracted much attention as a new fabrication of electronic devices because it has potential of low cost, low environmental impact and large area fabrication. Low cost and low environmental impact are because of small number of process steps, small amount of materials and high through-put. Several printing methods such as screen printing [1] and ink-jet printing [2, 3] have been developed to fabricate organic thin film transistors (OTFTs). However, there are some issue to overcome such as resolution, electric property, process integration, and reliability to fabricate electronic devices by printing. Recently, there were some reports of OTFT backplane to improve resolution and mobility for flexible electronic paper, 76 ppi all-printed OTFTs using an off-set-based high-resolution printing[4], 150 ppi OTFTs using photo-assisted surface modification technique with screen-printing [5], 200 ppi all-printed OTFTs using microcontact printing [6], and OTFTs with higher mobility of 0.4cm^2/Vs [7].

In previous work we developed a surface energy controlled ink-jet printing with ultraviolet (UV) irradiation on a novel polyimide film for high-resolution electrode patterning [8]. In this paper it was shown that minimum line width of electrode was 15 μm and minimum space between two electrodes was 2 μm. We successfully fabricated several resolution of all-printed OTFT backplane such as 80 ppi, 160 ppi [9] and 200 ppi [10] on plastic substrate applying this novel ink-jet technique. We used polymer organic semiconductor (OSC) for 80 ppi and 160 ppi OTFT backplanes, which has high solubility and high air stability [11] and small-molecule OSC for 200 ppi one, which exhibited high mobility of over 0.1 cm^2/Vs. Flexible electrophoretic displays (EPDs) driven by these all-printed OTFT backplanes were also successfully demonstrated. Japanese characters with 6 point were clearly displayed. To show the scaling capability, we have also fabricated a 300 ppi all-printed OTFT array without pixel electrodes on plastic substrate.

2. All-printed Organic TFT Backplane

2-1. TFT structure

Fig. 1 shows a schematic cross-section of the all-printed OTFT backplane with bottom-gate bottom-contact structure on plastic substrate. The gate electrode and the storage capacitor electrode were fabricated using Ag nanoparticles ink by the surface energy controlled ink-jet printing. The gate insulator was a novel polyimide film fabricated by spin coating. The source/drain (S/D) electrode consist of Ag were also fabricated by the surface energy controlled ink-jet printing. Small-molecule OSC for 200 ppi OTFTs and polymer OSC for 80 ppi, 160 ppi OTFTs were fabricated by ink-jet printing under ambient conditions respectively. After the formation of OSC layer, the insulator and pixel electrodes were fabricated by screen printing. All of these layers were fabricated by several printing processes under ambient conditions. Maximum process temperature was 180˚C.

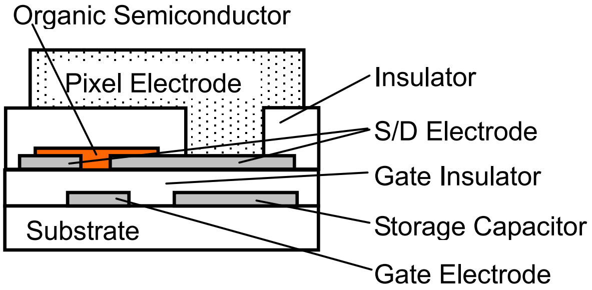


Fig.1 Schematic cross-section of the all-printed OTFT backplane

To achieve all-printed OTFTs we choose the several printing methods. The surface energy controlled IJ printing was used for achieving the small surface roughness, the fine electrode patterning, and the precise controlled channel length. The conventional IJ printing was used for the contamination-free printing because of non-contact printing without printing plate. The screen printing was used for the thick film formation.

2-2. Surface Energy Controlled Ink-jet Printing

To obtain the fine electrode patterns, we developed the surface energy controlled ink-jet printing technique (see Fig. 2). The novel polyimide film was fabricated on the plastic substrate by spin coating, whose surface had low surface energy after post-baking in N_2 condition at 180°C. After UV irradiation from super-high pressure mercury lamp through a photo mask from the front side of the substrate (Fig. 2a), the high surface energy area corresponding to electrode patterns and the low surface energy area were formed on the novel polyimide film surface (Fig. 2b). Hydrophilic Ag nanoparticles ink was ink-jetted onto the high surface energy area and spread over the edge of the area (Fig. 2c). After post-baking under ambient condition at 180°C, the fine electrode patterns were obtained. Using this novel polyimide as an insulating layer, additional fabrication process of wiring is only exposure process without such wet process as development and cleaning. Thus, our surface energy controlled ink-jet printing takes advantage of practical number of process steps.

Fig. 3 shows an optical micrograph of electrode patterns overlaid with the gate insulator (a) without and (b) with the surface energy controlled ink-jet printing. Electrodes without the surface energy controlled ink-jet printing (Fig. 3a) show droplet-like shape and rough surface with interference fringes, which depends on the difference of the film thickness due to surface roughness of electrodes. On the other hand, electrodes with the surface energy controlled ink-jet printing (Fig. 3b) show photo mask-like shape and very smooth surface because of no interference fringes.

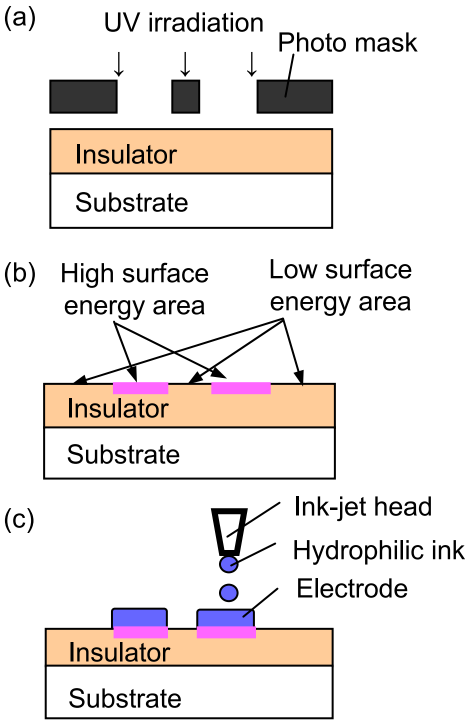


Fig.2 Schematic of the surface energy controlled ink-jet printing process (a) UV irradiation, (b) Formation of areas with different surface energy, (c) Fabrication of electrodes by ink-jet printing

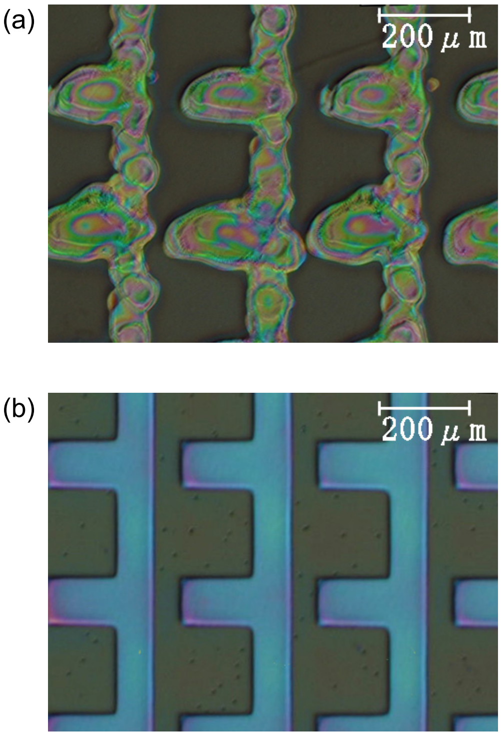


Fig. 3 Optical micrograph of electrode patterns (a) without and (b) with the surface energy controlled ink-jet printing

To investigate the minimum space between two electrodes, we prepared a photo mask with various line and space patterns. After UV irradiation on the novel polyimide film through the photo mask, hydrophilic Ag nanoparticles ink was ink-jetted. Controlling with ink-jet printing conditions such as drop size and ink volume per unit line, two kinds of electrodes with different thicknesses were fabricated. Fig. 4 shows the dependence of the yield of electrodes separation on designed space against electrode thickness. Yield means the ratio of separation between the two electrodes and was determined by using an optical microscope with 100 points in one sheet. Fig. 4 shows that minimum space up to 2 μm (designed) could be fabricated using surface energy controlled ink-jet printing with UV irradiation on the polyimide film. It is superior to conventional ink-jet method.

We also examined alignment margins of electrode fabrication by this ink-jet technique, increasing the distance between the center of high surface energy line pattern and that of ink droplet impact position every 10 μm. Fig. 5 shows that the lines in 80 μm width can be successfully formed same as photo mask patterns apart from the center of the ideal impact position by a distance of 50 μm or less. This is because the hydrophilic ink droplets, which land onto the low surface energy area, could be drawn into the high surface energy area.

Thus, the surface energy controlled ink-jet printing technique possesses such unique features as good surface roughness, high-resolution patterning and high alignment margin because of using a photo mask.

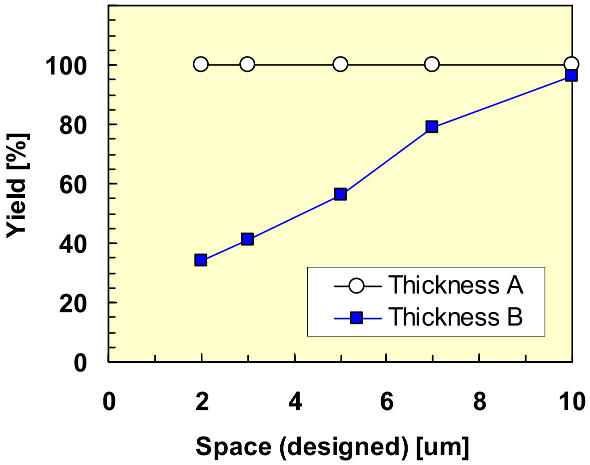


Fig. 4 Yield of electrodes separation on designed space against electrode thicknesses

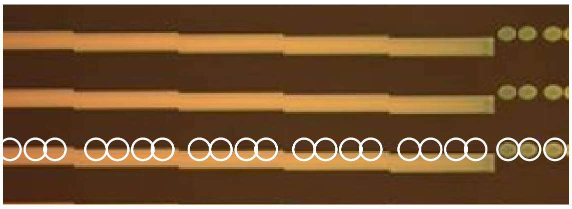


Fig. 5 Alignment margin of ink droplets ejected from IJ head onto the surface energy controlled polyimide film. White circles show impact positions of ink droplets

2-3. Organic TFT Backplane

We fabricated a 80 ppi and a 160 ppi OTFT backplane with polymer OSC, which consisted of stilbene polymer with triarylamine unit and long-chain alkyl group. Fig. 6 shows an optical micrograph of a 160 ppi (pixel size 159 μm×159 μm) all-printed OTFT array on flexible substrates after polymer OSC fabrication. Using the appropriate solvent of polymer OSC and optimizing ink-jet conditions, OSC profile was like a coffee stain and separated with each others at 159 μm pitches without any bank structure. After OSC printing, insulator and pixel electrode were fabricated by the screen printing.

To shrink the TFT size to 200 ppi (pixel size 127 μm × 127 μm) from 160 ppi (pixel size 159 μm × 159 μm), we have optimized TFT design such as the line and space of the gate electrode and the S/D electrode. However, it is difficult to achieve fine electrode patterning and low resistivity at same time. We optimized the electrode thickness and the curing time. The electrode thickness was controlled by ink-jet condition such as ink volume per unit line. Fig. 7 shows an optical micrograph of a 200 ppi all-printed OTFT array on plastic substrate after S/D electrode fabrication. It is shown the minimum line width and space of the gate line was 30 μm and 15 μm, respectively. Those of the source line were same. A minimum width of the source electrode was 15 μm, channel length was 5 μm. Resistances per unit length of the gate line and the source line were 1 kΩ/cm and 0.5 kΩ/cm at 30 μm line width respectively, which were enough for 3.2 inch EPDs.

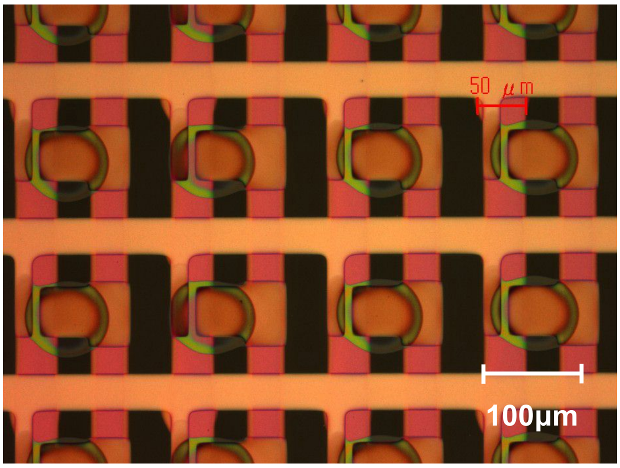


Fig. 6 Optical micrograph of a 160 ppi all-printed OTFT backplane on flexible substrate after OSC fabrication (Channel length L = 5 μm)

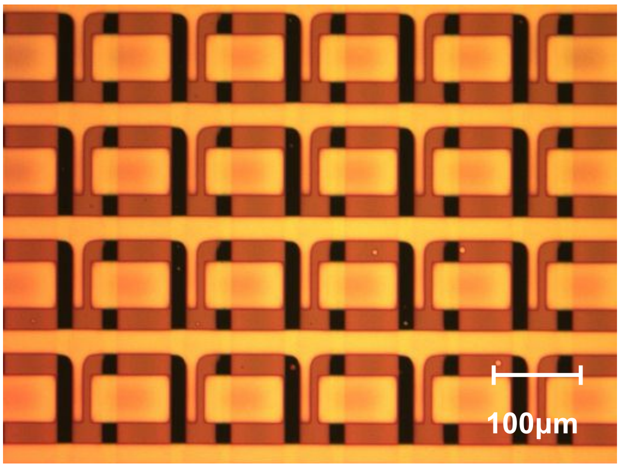


Fig. 7 Optical micrograph of a 200 ppi all-printed OTFT backplane on flexible substrate after S/D electrode fabrication (Channel length L = 5 μm)

After S/D electrode printing, small-molecule OSC was fabricated by ink-jet method. Patterning is important both for polymer OSC and for small-molecule OSC to obtain a high on/off current ratio of TFT. Small-molecule OSC is more difficult to print than polymer OSC because of the crystallization in the channel region of TFT. We optimized the OSC ink formulation such as the solvent and the concentration. We also controlled the surface energy of the gate insulator by the pre-treatment and the novel polyimide.

Fig. 8 shows the transfer characteristics of a 200 ppi all-printed OTFT with W/L = 57 μm/5 μm after OSC printing and after pixel electrode printing. Mobility of 0.25 cm^2/Vs, Vth of 1.3 V, an on/off current ratio of 10^6 at Vds = -20 V were obtained after small-molecule OSC printing. Although a slight reduction in the on-current and a small Vth shift were observed after pixel electrode printing, mobility of 0.12 cm^2/Vs, Vth of 3.9 V were still maintained, which is sufficient for driving high resolution EPDs.

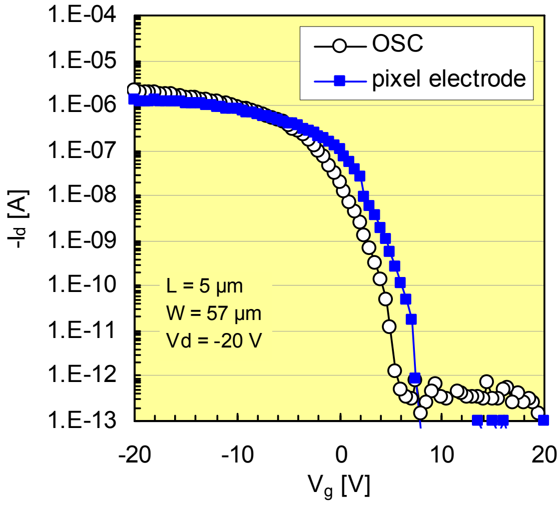


Fig. 8 Transfer characteristics of a 200 ppi all-printed OTFT after OSC printing and after pixel electrode printing (Channel length L = 5 μm)

We have also fabricated a 300 ppi (TFT pitch: 85 μm × 85 μm) all-printed OTFT array on plastic substrate without insulator and pixel electrode (see Fig. 9). We optimized the OTFT design such as the common structure of storage electrode, the line and space of electrodes, and optimized OSC ink-jet condition. The minimum channel length L was 3 μm, the minimum line width of source line was 20 μm.

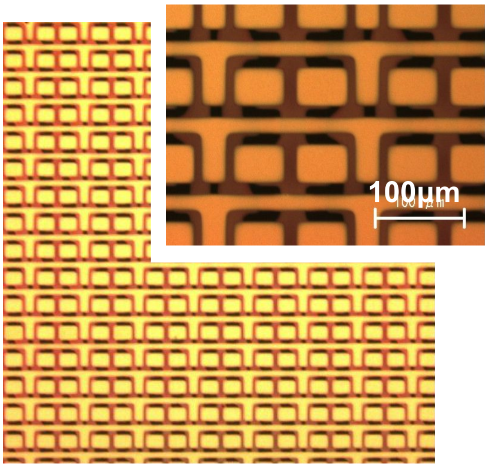


Fig. 9 Optical micrograph of a 300 ppi all-printed OTFT array on flexible substrate after S/D electrode fabrication (Channel length L = 5 μm)

3. Application to Flexible EPDs

For demonstration of flexible EPDs, electrophoretic sheets were laminated with all-printed OTFT backplanes on plastic substrate, which had several kinds of resolution (see Fig. 10, Table 1.). Large display size was 8 inch diagonal with pixel number of 500×400 and higher resolution was 200 ppi with pixel number of 540 × 360. As shown in Fig. 10, Japanese characters with 9 point were successfully driven by a 200 ppi all-printed

OTFT backplane. The applied voltages of select line and data line were 30 Vpp and 20 Vpp, respectively. Furthermore, 6-point Ming-style font characters were displayed with bended EPDs (see Fig. 12).

Table 1. Specifications of flexible EPDs driven by the all-printed OTFT backplane

Resolution	80 ppi	160 ppi	200 ppi
Display size (diagonal)	8 inch	3.2 inch	3.2 inch
Pixel number	500 × 400	432 × 288	540 × 360
Pixel size	318 × 318 μm	159 × 159 μm	127 × 127 μm

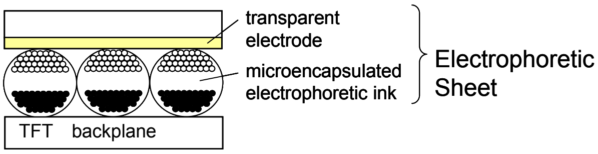
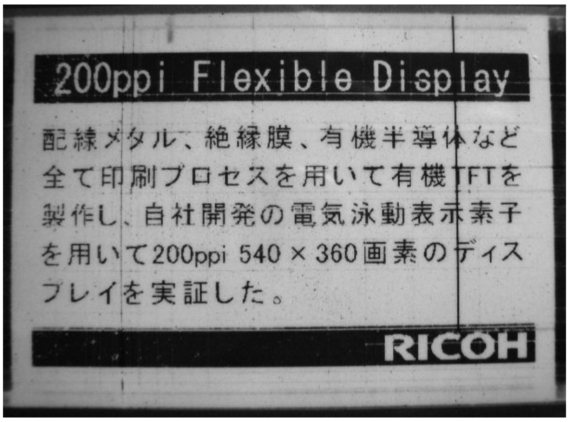


Fig. 10 Schematic structure of flexible EPDs



200ppi Flexible Display

配線メタル、絶縁膜、有機半導体など
全て印刷プロセスを用いて有機TFTを
製作し、自社開発の電気泳動表示素子
を用いて200ppi 540 × 360画素のディス
プレイを実証した。

RICOH

Fig. 11 Photograph of a 3.2 inch flexible EPD driven by a 200 ppi all-printed OTFT backplane (540 × 360 pixels)

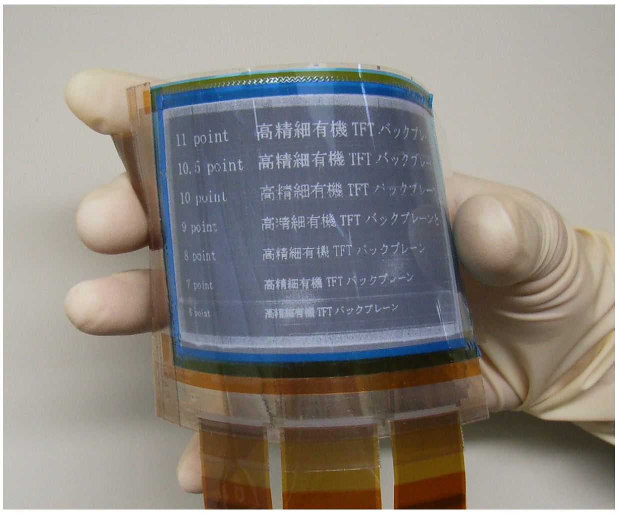


Fig. 12 Photograph of a 3.2 inch flexible EPD driven by a 200 ppi all-printed OTFT backplane

4. Conclusions

We have succeeded in fabricating 80, 160, 200 ppi all-printed OTFT backplanes and demonstrated flexible EPDs driven by these backplanes. The resolution of 200 ppi and mobility of over 0.1 cm^2/Vs are enough for a monochrome electronic paper. To achieve such a high-resolution and high-mobility backplane on plastic substrate, we have improved the surface energy controlled ink-jet printing with UV irradiation for Ag electrodes and the conventional ink-jet printing for small-molecule OSC. We have also fabricated a 300 ppi all-printed TFT array on plastic substrate. This printing technology is promising for high-resolution, low-cost and low-environmental impact manufacturing process.

6. Acknowledgements

The authors would like to thank all the members of the development team in Ricoh for their devoted supports and valuable discussions.

References

1) Z. Bao, Y. Feng, A. Dodabalapur, V. R. Raju, and A. J. Lovinger: High-Performance Plastic Transistors Fabricated by Printing Techniques, Chem. Mater., pp. 1299-1301, Vol. 9, (1997).

2) H. Sirringhause, T. Kawase, R. H. Friend, T. Shimoda, M. Inbasekaran, W. Wu, and E. P. Woo: High-Resolution Inkjet Printing of All-Polymer Transistor Circuits, Science, pp. 2123-2126, Vol. 290 (2000).

3) A. C. Arias, J. Daniel, B. Krusor, S. Ready, V. Sholin, and R. Street: All-additive ink-jet-printed display backplanes: Materials development and integration, Journal of the SID pp. 485-490, Vol. 15, No. 7 (2007).

4) T. Okubo, Y. Kokubo, K. Hatta, R. Matsubara, M. Ishizaki, Y. Ugajin, N. Sekine, N. Kawashima, T. Fukuda, A. Nomoto, T. Ohe, N. Kobayashi, K. Nomoto, and J. Kasahara: 10.5-in VGA All-printed Flexible Organic TFT Backplane for Electrophoretic Displays, Proc. IDW'07, pp. 463-464 (2007).

5) H. Maeda, H. Honda, M. Matsuoka, M. Nagae, T. Suzuki, K. Ogawa, and H. Kobayashi: 10inch Flexible Active-Matrix QR-LPD for Fast Image Refreshing with Printed OTFTs, Proc. IDW08, pp. 1469-1472 (2008).

6) K. Matsuoka, O. Kina, M. Koutake, K. Noda, H. Yonehara, K. Nakanishi, and K. Yase: High Resolution 200 ppi LCD Driven by Entirely Printed Organic TFT, Proc. IDW'09, pp. 717-720 (2009).

7) N. Kawashima, N. Kobayashi, N. Yoneya, H. Ono, T. Fukuda, T. Ohe, Y. Ishii, A. Nomoto, M. Sasaki, and K. Nomoto: A High Resolution Flexible Electrophoretic Display Driven by OTFTs with Inkjet-Printed Organic Semiconductor, Proc. SID'09, pp. 25-27 (2009).

8) T. Tano, H. Tomono, H. Kondoh, and K. Fujimura: Organic Thin-film Transistors with a Novel Polyimide Gate Insulator, Proc. AMLCD2004, pp. 37-40 (2004).

9) K. Suzuki, K. Yutani, A. Onodera, T. Tano, H. Tomono, A. Murakami, M. Yanagisawa, K. Kameyama, and I. Kawashima: A 160 ppi All-printed Organic TFT Backplane for Flexible Electrophoretic Displays, Proc. IDW'08, pp. 1477-1478 (2008).
10) K. Suzuki, K. Yutani, M. Nakashima, A. Onodera, S. Mizukami, M. Kato, T. Tano, H. Tomono, M. Yanagisawa, and K. Kameyama: A 200 ppi All-printed Organic TFT Backplane for Flexible Electrophoretic Displays, Proc. IDW'09, pp. 1581-1584 (2009).
11) T. Yamaga, T. Sagisaka, and Y. Akiyama: Development of Amorphous Polymer as High FET Mobility Materials, Abstracts of Papers, MRS 2007 Fall Meeting G1.10 (2007).