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Rib-free Ink-Jet Printing Fabrication on Heterogeneous Surfaces

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Rib-free ink-jet printing device fabrication on hetrogeneous surfaces opens a new possibility for greener processes. The integrity of ink droplets was maintained with less than 3° contact angle differences on the heterogeneous surfaces after surface treatment. The breaking of color lines and color mixing were avoided by accelerating solvent evaporation. The inks were greatly conserved without the need for additional lithography process for rib structure. © 2010 The Japan Society of Applied Physics

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Ink-jet printing (IJP) technology has been widely used to fabricate thin-film transistors, color filters, polymer light-emitting diodes, light-emitting quantum dots, conductive tracks, and combinatorial materials discovery among others.[1–8] As the emerging calls for conserving energy and materials grow intensely around the world, the precision drop-on-demand (DoD) ink-jet printing technology has great potentials to answer such calls. Previous research focused on atomistic understanding of ink fluid physical properties.[9,10] The important inks parameters, namely viscosity, density, and surface tension, were identified to be critical for optimizing jetting results. The possibility of device fabrication on heterogeneous surfaces, such as metal on wafer or metal on glass, however, has not been fully examined.[11] In particular, the issues of ink droplet deformation on heterogeneous surfaces and the droplet coalescence effect encountered during the jetting process have not been clarified. Droplet deformation on heterogeneous surfaces can be predicted by Young's force (d_{FY}) as

$$d_{FY} = [(\gamma_{sv} - \gamma_{sl})_A - (\gamma_{sv}\gamma_{sl})_B]\,dx, \quad (1)$$

where γ_{sv} and γ_{sl} are the surface tension of the solid–vapor and liquid–vapor interfaces, respectively; A and B represent two edges of a droplet's cross section on two different surfaces; x is the thickness of the droplet.[12] Equation (1) can be rewritten as eq. (2), where θ_A and θ_B represent the local contact angles at points A and B, respectively, and γ_{lv} is the surface tension of the liquid–solid interface:

$$d_{FY} = \gamma_{lv}(\cos\theta_A - \cos\theta_B)\,dx, \quad (2)$$

When the contact angle at point A is smaller than that of point B on a low-hysteresis surface, the droplet would shift to the direction of higher γ_{sv} owing to the net free energy. The large difference in contact angle may result in a deformed droplet, and may even shift the droplet's position. Consequently, the device fabrication may fail owing to the droplet distortion or void space between the interfaces of heterogeneous surfaces. Droplet coalescence may occur when continuous ink deposits form a line or a two-dimensional pattern. The continuous ink deposits may break up to form large sessile droplets from coalescing inks. This is because the lowest surface energy and constant contact angle are maintained by individual ink droplets. A single sessile water droplet was reported to form as rapidly as 20 µs, and such single sessile water droplets are deposited continuously on Plexigals.[13] One of the solutions is to define the print area using ribs, such as a color filter prepared by using the ink-jet printing process.[14–16] The results are limited by the rib's design and fabrication, which requires an additional lithography process. In this study, we explored rib-free ink-jet printing fabrication on chromium–glass heterogeneous surfaces. The issues of droplet deformation, line width variation, and droplet coalescence were addressed by controlling surface properties and solvent evaporation. Color resists were applied as the model for the rib-free ink-jet printing demonstration. Resists of primary red, green, blue colors were deposited sequentially to form color lines, covering 40 µm in subpixel width without ribs on a chromium-patterned glass substrate.

A Litrex 70L equipped with a Spectra SE-128 print head (FUJIFILM Dimatix) was used as the ink-jet printer. The volume of a deposited individual ink droplet was 20 ± 5 pL. A UV-curable color resist consisting of a pigment, acrylate-binding agents (monomers and oligmers), an initiator, and a surfactant was mixed with propylene glycol monomethyl ether acetate (PGMEA) as the primary solvent. The viscosity and surface tension of red (R), green (G), and blue (B) resists were adjusted to 7–13 Cp and 28 mN/m (Everlight Chemical Industrial), respectively. The particle size of the pigments was smaller than 100 nm. The solid content of these inks was kept between 20 to 23 wt %. The coffee ring effect of a color resist droplet was minimized by using co-solvent mixtures composed of the primary solvent and a high-boiling-point solvent, such as acetophenone.[17,18] The heterogeneous surface of a 4-in. glass substrate was made of patterned chromium (Cr) within 200 nm thickness on bare glass. The dimensions of each subpixel within the 4-in. glass substrate were approximately 40 × 160 µm, and each subpixel was separated by chromium 30 µm in width as a light blocking matrix. The color line was mildly baked at 90 °C for 15 to 30 min before the next jetting process. The final post baking was set at 230 °C for 45 min. Additional fixation was carried out by UV exposure at 250 mJ/cm^2 for 5 min. Contact angle was measured by using a KRÜSS universal surface tester GH-100. The surface profile was measured by using a KLA-Tencor Alpha-step IQ. Chromaticity, color difference, and optical density were determined by using a Photal LCF-6000.

The contact angles of the three color inks on both untreated and treated chromium and glass surfaces are listed in Table I. The overflow of ink was found on the untreated chromium patterned glass surface, as shown in Fig. 1(a). Surface modification was carried out by using HFC-128 and EGC-1700 solutions (from 3M). The different surface-

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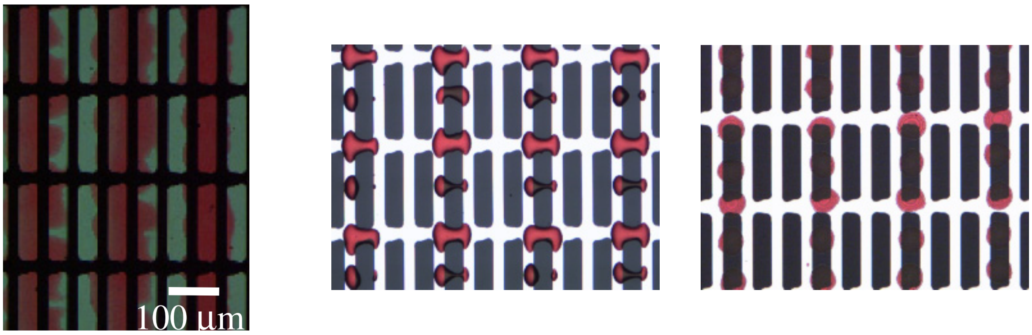

(a) (b) (c)

Fig. 1. (Color online) (a) Smearing of ink on untreated surfaces; (b) deformed droplets on HFC-128-treated surfaces; (c) droplets on EGC-1700-treated surfaces. Photos were taken in the transmission mode for (a), and reflection mode for (b) and (c).

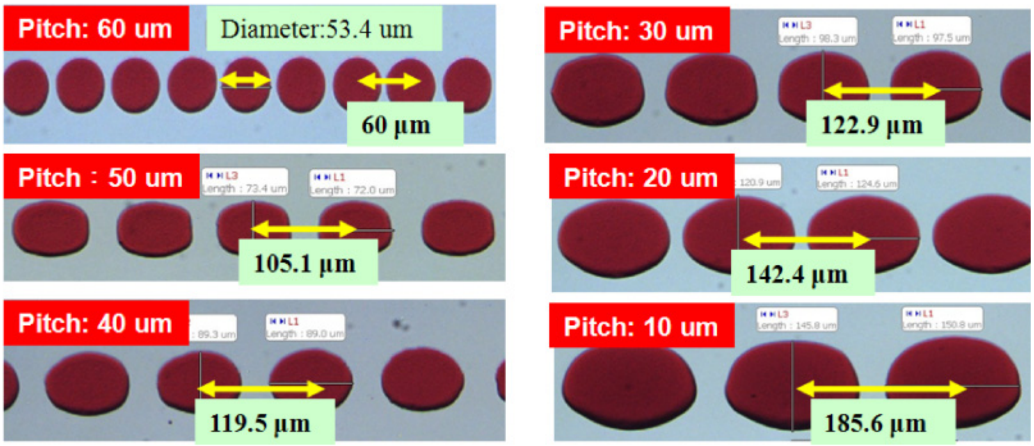

Fig. 2. (Color online) Sessile red ink droplets on the EGC-1700-treated glass with various dot pitches. The printing direction was from left to right.

Table I. Contact angles (deg) of treated and untreated chromium and glass surfaces (measured at 25 °C).

	Untreated surface	HFC-128	EGC-1700
	Chromium		
R	13	37	33
G	11	42	35
B	12	40	38
	Glass		
R	22	47	32
G	19	51	34
B	19	55	35

bonded conditions for surface treatment reagents yielded various contact angles on chromium and glass surfaces. The size of an ink droplet was observed to be 60 µm when the contact angle was controlled within the range of 30 to 50°. The deformation of an ink droplet was observed when the contact angle differed by more than 5° on heterogeneous surfaces, as shown in Fig. 1(b).[19] The integrity of individual droplets was maintained, when the contact angle differed by less than 3°, as shown in Fig. 1(c). Thus, a 60 µm ink droplet can be prepared at a 30° contact angle with a 3° difference on heterogeneous surfaces.

The second challenge was encountered when the ink was continuously deposited on the surface. The color line broke up and formed large sessile droplets from coalescing ink, as shown in Fig. 2. Dot pitch was defined by the distance between two sequential deposits. The shorter the pitch was, the larger the droplets were formed. Intact ink droplets were obtained at a 60 µm dot pitch in our experiment. In order to minimize the droplet coalescence effect, droplet fluidity needs to be further suppressed. The surface boundary condition can be altered by either applying high-hysteresis surface treatment[12] or accelerating solvent evaporation. With limited options of surface treatment reagents, the latter method was applied to secure an ink droplet in a desired position. By heating the stage at 60 °C to accelerate solvent evaporation, the dot pitch can be reduced to 30–35 µm without the breaking of any line, as shown in Fig. 3. An additional benefit was found when the second color resist jetted parallel to the first color line, as shown in Fig. 4. The solvent evaporation helped to secure the position of ink droplets preventing their overflow to the first color line, which was very often caused by the large contact angle difference between the solid ink line and the surface-treated substrate. The result is critical for the manufacturing process to avoid undesirable color mixing, or material displacement. The remaining color resists were fabricated on EGC-1700 treated surface by the same processes at an elevated temperature. The transmission and reflection photos were shown in Fig. 5. The thicknesses of the red, green, and blue resists were about 1.2, 1.0, and 1.0 µm, respectively. The

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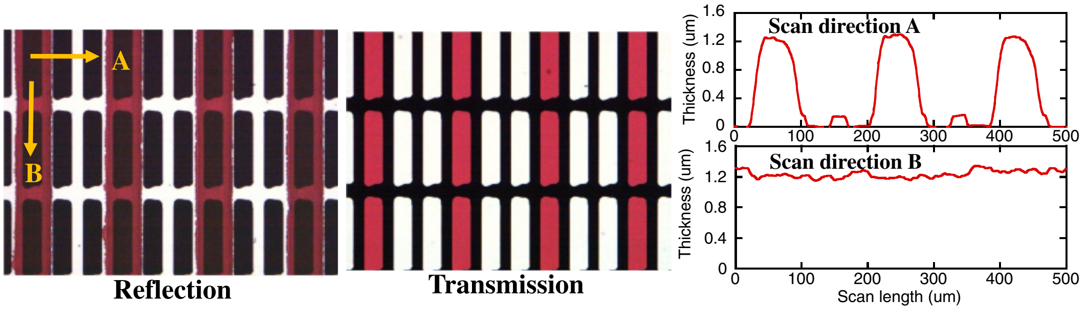
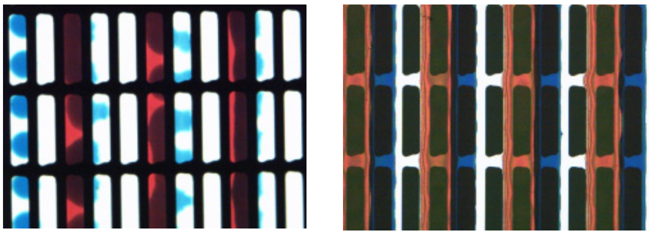
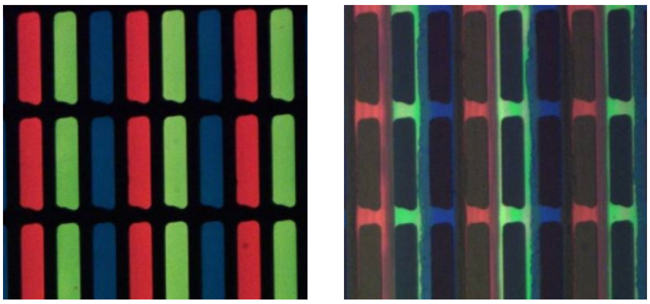


Fig. 3. (Color online) Color line prepared at elevated temperature and its surface profile.



(a) (b)

Fig. 4. (Color online) Reflection photos for second blue color printed at (a) room temperature and (b) 60 °C.



(a) (b)

Fig. 5. (Color online) (a) Transmission and (b) reflection photos of 4-in. panel.

panel's average chromaticities (x, y) were (0.6413, 0.3298), (0.1373, 0.1561), and (0.3356, 0.5612) for red, green, and blue, respectively. According to the Commission Internationale de l'Eclairage (CIE) chromaticity diagram, which was provided by the National Television Standards Committee (NTSC), our chromaticity was estimated to be $53.5 \pm 0.7\%$, which was higher than current 4-in. color filter's requirement.

In summary, our work demonstrates that the color lines can be independently prepared without the use of the ribs. A less than 3° contact angle difference on heterogeneous surfaces keeps the ink droplets intact. The width of a color line can be reduced to less than 60 μm by maintaining the ink contact angles between 30 to 50°. The undesirable breaking of color lines and color mixing can be avoided by accelerating solvent evaporation at an elevated temperature. In comparison with current ink-jet printing processes, the rib-free ink-jet printing process opens a new way to designing printing patterns on heterogeneous surfaces. For larger-pattern-area preparation, free standing lines can serve as their own ribs, which enable a state-of-the-art fabrication process without unnecessary wastes. Hence, the rib-free ink-jet printing process offers a promising technology contributing to a greener world.

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