High-Viscosity Electronic Materials Printing Using Ultrasonic Inkjet System

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Abstract

The nozzle-less structure of the ultrasonic inkjet system is advantageous leading to printability of inks of a wide range of viscosities without clogging. In the ultrasonic inkjet head, dot placement accuracy of high-viscosity ink was as same as that of standard viscosity ink. The shape of meniscus changes as the viscosity increases. Using the printability of high-viscosity inks, the ultrasonic inkjet printing was applied for LED packaging by depositing silicone resin and fabrication of solution-processed organic thin-film transistors by printing high-viscosity organic conducting ink, PEDOT:PSS.

Introduction

In recent years, inkjet printing has been considered to be one of the most appropriate technologies for digital fabrication, because it is characterized by its non-contact process, which is favorable for fabricating multilayer devices. Moreover the inkjet system is on-demand printing technology with no mask plates, which makes the configuration simple while offering many advantages, for example, for large-area production. However, in the conventional inkjet printing methods such as the piezoelectric and the thermal inkjet printing methods, properties of printable ink are restricted and consequently the coverage of available materials and devices is also restricted [1]. On the other hand, the ultrasonic inkjet printing method is characterized by its high printability of inks with various properties (high viscosity, large particle inclusion, etc), because narrow nozzles and flow paths are not needed for the droplet formation [2, 3]. In the field of digital fabrication, the use of high-viscosity ink is desired in light of small ink spread and the forming of thick films on non-absorbing substrates. Accordingly, we examined the ultrasonic inkjet method that can eject high -viscosity inks. Using the inkjet method, silicone oils whose viscosities exceed 500 mPa·s were ejected at room temperature. We discuss the relationship between viscosities of inks and the state of ejection and meniscus. In addition, the method was applied for LED packaging and all-solution processed organic thin-film transistor.

Ultrasonic inkjet printing method

Figure 1 shows the fundamental principle of the ultrasonic inkjet head, where ultrasonic beam generated by transducers is focused on the free liquid surface by the acoustic lens and a droplet is ejected. The ultrasonic inkjet printing has the following advantages: 1. nozzle-less structure, which leads to less clogging and uniform dot size with no tail, 2. little restriction on ink properties, such as high viscosity and large-particle inclusion, 3. droplet size depends on the ultrasonic wavelength (the droplet diameter is approximately equal to the wavelength), that is, the size is controllable by transducer frequency, 4. simple head structure; no need of a narrow ink path and chamber. Therefore, the ultrasonic inkjet printing method enables ejection of a wide variety of inks that cannot be ejected by other inkjet methods. This characteristic is useful in pattering of electronic materials having various properties, which can be printed for device fabrication without dilution or addition for easy ejection.

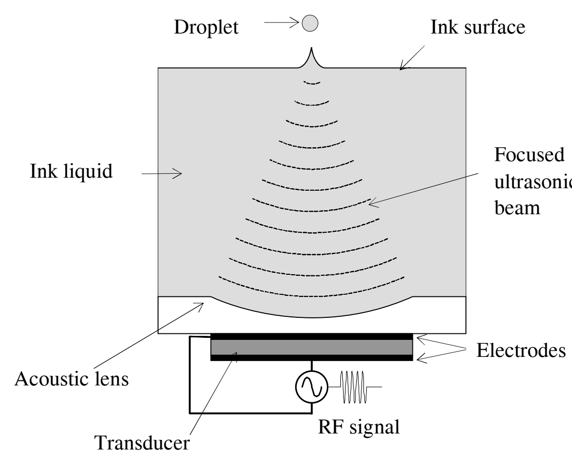


Figure 1. *Schematic of ultrasonic inkjet head*

Experimental

The ultrasonic inkjet printing device is composed of a single ejector operated at a transducer frequency of 12.5 MHz. The piezoelectric of the transducer is composed of lead titanate with 180 μm thickness. The acoustic lens with f-number 1.5 and an aperture of 2 mm was made of glass.

The standard viscosity liquids used were silicone oil with viscosities of 5, 50 and 500 mPa·s (manufactured by Brookfield Engineering).

Results and Discussion

Construction of apparatus for observation of dots

In this work, ejectability of high-viscosity inks was judged from dot placement accuracy. Figure 2 shows the apparatus for observation of dot placement. A CCD camera A was set above the ultrasonic inkjet head to observe deposited dots, and a transparent substrate, namely a glass substrate, was placed between the camera and the inkjet head; dots were observed from the back side. The substrate moved parallel to the head face at a constant speed. To synchronize the shooting by the CCD camera A with ink ejection, the ejection frequency was set at 15 Hz. This observation system saved image sequence of dots as still frames and the dot placement accuracy was analyzed from the images. Since the dot positions shifted easily on a non-absorbing substrate, the dot images were collected immediately after the deposition.

Another CCD camera B for observing a meniscus of ejecting droplets was set at the side of the inkjet head.

Though the ultrasonic inkjet head has no nozzles for ejection, for keeping ink surface and prevention of dust, the inkjet head has a top plate with a hole, 700 µm in diameter, for ejection.

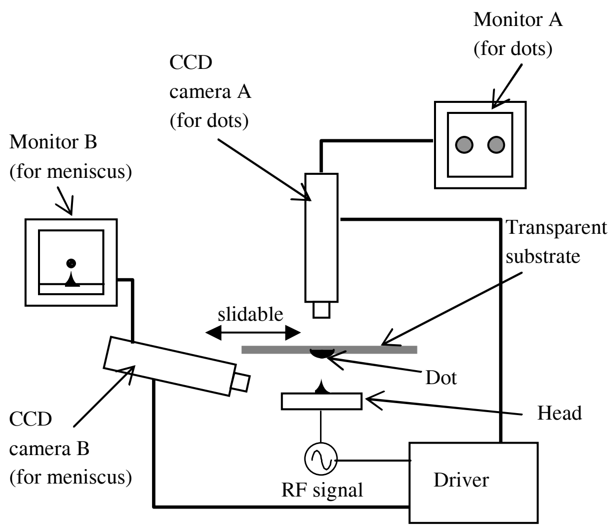
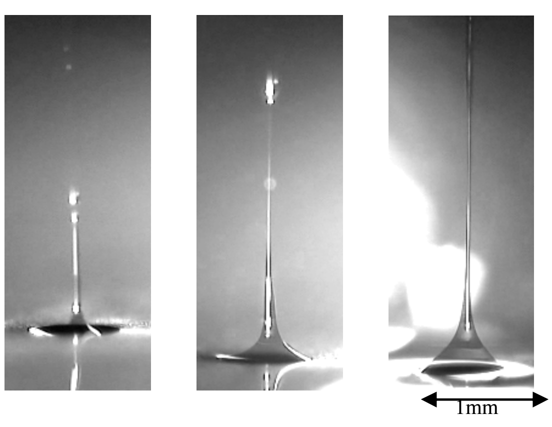


***Figure 2**. Schematic of an apparatus for observation of ejection*

Observation of ejections

Figure 3 shows the meniscus shapes at droplet formation. The lengths of menisci depend on the viscosity of the inks. The meniscus becomes longer in case of using ink with higher viscosity. The meniscus of 500 mPa·s ink is more than 2 mm.



(a) 5mPa·s ink (b) 50mPa·s ink (c) 500mPa·s ink

***Figure 3.** Stroboscopic images of menisci and droplets*

In our ultrasonic inkjet system, the diameters of droplets depend on the frequency of the head transducer. They are nearly equal to the ultrasonic wavelength in ink liquid: in the case of the inkjet head with frequency of 12.5 MHz, about 135-µm wavelength, the diameters of droplets and dots are about 135 µm and 270 µm, respectively. Figure 4 shows the diameters of droplets, measured form the images of menisci and droplets (Fig 3). Diameters of droplets of 5 mPa·s ink are near 135 µm, but those of droplets of higher-viscosity inks are about 200 µm. This is partially because the viscosity affects the diameter of droplet: menisci of higher-viscosity inks are longer than that of standard viscosity ink.

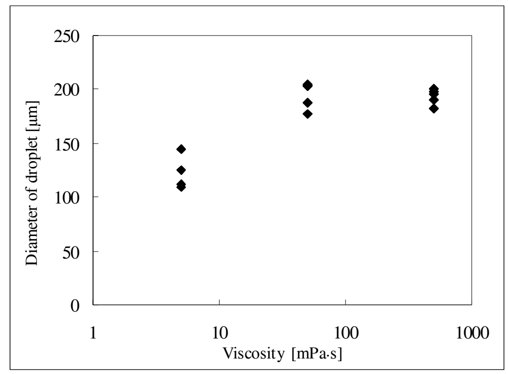


***Figure 4**. Logarithmic relationship between viscosities and dot diameters*

Dots observation

Figure 5 shows dots images of ink-viscosity at 5, 50 and 500 mPa·s. In the dot observation system, the substrate needs to be transparent because the CCD camera shoots from the back side of the dots. Furthermore, since standard viscosity liquids are also achromatic, the edges of dot images need to be detected clearly.

In order to suppress the spread of the deposited dots on the glass substrate, the substrates was coated with fluoropolymer to make the substrate surface oil-repellent. Therefore, the diameters of deposited dots are not discussed in this paper.

The gap between the substrate and the inkjet head should be set longer than the height of the meniscus: the gaps are 1.5, 2 and 3 mm for the standard viscosity liquids of 5, 50 and 500 mPa·s, respectively.

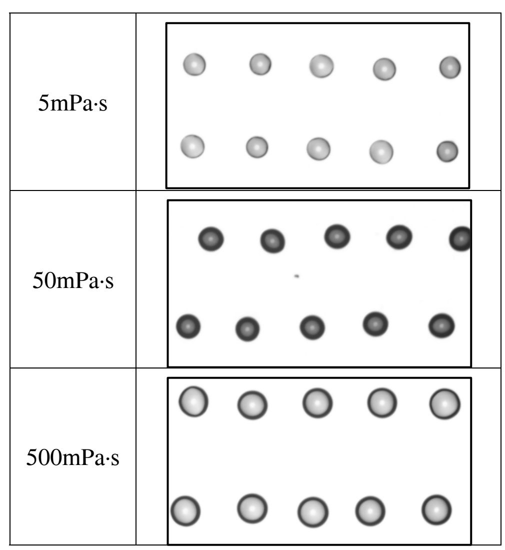
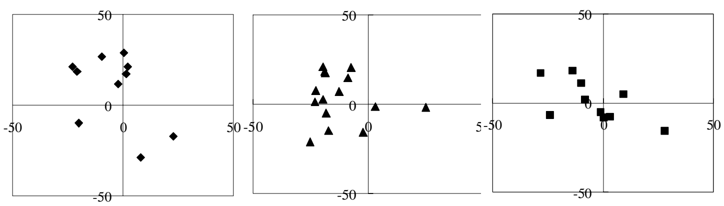


***Figure 5.** Dot images of ink viscosity at 5, 50 and 500 mPa·s*

Figure 6 shows placement errors of dots for each viscosity. The placement of the dots is defined by the center of gravity calculated from the images, which are measured from the images shot by the apparatus for dot observation shown in Fig.2.

For all viscosities, dot placement accuracy was within ±30 μm. This shows that in the ultrasonic inkjet system dot placement accuracy of high-viscosity ink is almost the same as that of standard viscosity ink.



(a) 5mPa·s gap:1.5mm (b) 50mPa·s gap:2mm (c) 500mPa·s gap:3mm

Figure 6. *Placement errors of ink viscosity of 5, 50 and 500 mPa·s*

High-viscosity ink ejection

RF signals are applied to the transducer of an ultrasonic inkjet head. The power of the RF signals is defined by burst time and applied voltage. Figure 7 shows the relationship between the burst time and the minimum applied voltage for droplet ejection. The minimum voltage for the ejection is determined to be the voltage where the placement errors of the depositions are within a quarter-dot-diameter. This shows that the ejections of high-viscosity ink need very high-power voltage.

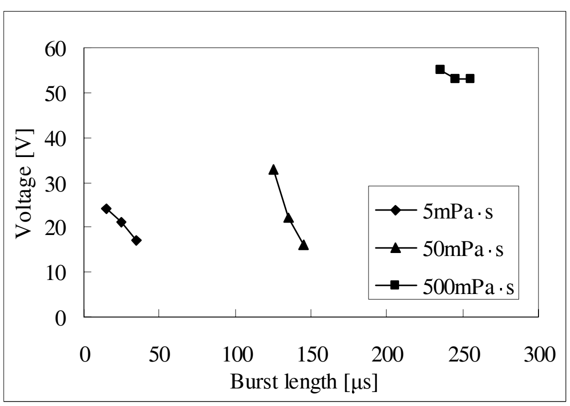


Figure 7. *Relationship between burst time and applied voltage for ink ejection*

To compare the power of ejection among viscosities, we introduce "standardizable energy density". Standardized energy density (ρ) is determined by standardizing the energy value for ejecting droplets per unit area in the focused ultrasonic beam width (W), using a standard value. The focused ultrasonic beam width (W) is represented by equation (1): a simple equation of ultrasonic field. Fl, λ and D are a focal length of an acoustic lens, an ultrasonic wavelength in ink liquid and an aperture size, respectively. The standardizable energy density (ρ) is determined by equation (2). Vth and BST are the minimum applied voltage for droplet ejection and burst time, respectively.

$$W = 2.44 \times Fl \times \lambda / D \quad (1)$$

$$\rho \propto Vth^2 \times BST / w^2 \quad (2)$$

Figure 8 shows the logarithmic relationship between viscosity and standardizable energy density. This shows that the ejections of high-viscosity ink need such high power because high-viscosity ink has large attenuation.

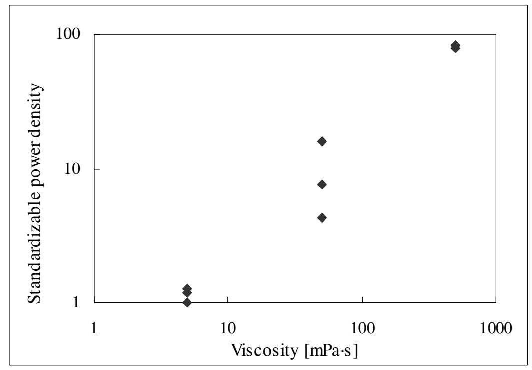


Figure 8*. Logarithmic relationship between viscosity and standardizable energy density*

Application

LED packaging and microlens fabrication by inkjet printing

With our ultrasonic printing device, we have confirmed the printability of silicone resin (340 mPa·s).

Packaged LED structures with encapsulant domes were fabricated using the ultrasonic inkjet printing method. Uncured resin was printed on the lead frame on which a blue LED chip is mounted (Fig. 9). The inkjet ejection is in the upper direction. The dome structure was shaped using surface tension of the resin and the edge of the substrate of a lead frame.

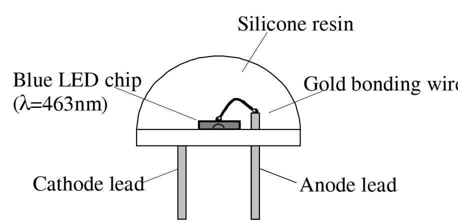
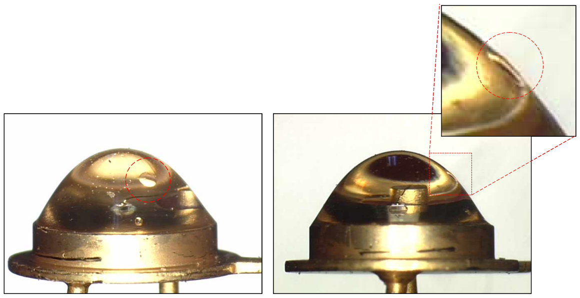


Figure 9*. Schematic cross-section image of LED package structure*

Furthermore, a microlens composed of the same resin was deposited on the dome surface (Fig. 10). The formed microlens diameter is 0.4 mm and the height is 0.04 mm (the aspect ratio: 0.1). The illumination intensity is increased (50%) at the microlens deposited area (θ= 15 °). The results confirm the possibility of light control of an LED device by a microlens fabricated using inkjet printing. This method will lead to fabricating optimized lens structures and on-demand modification techniques for the LED devices.



a) Front view (b) Side view

Figure 10. *Microlens structure deposited on the encapsulant dome.*

White LEDs were also fabricated using the ultrasonic inkjet printing method for forming phosphor dispersed encapsulant dome. [4]

All-solution-processed organic thin-film-transistor

PEDOT: PSS dispersion is a widely used organic conductor liquid dispersion. PEDOT: PSS dispersion is suitable for gate electrode material of top-gate OTFT owing to its low cure temperature, which avoids degradation of organic material components. Printing of PEDOT:PSS water dispersion was performed with the ultrasonic inkjet printing system.

Printing the top gate using PEODT:PSS using the ultrasonic inkjet printing method, a top-gate organic thin-film transistor (Fig. 11) was fabricated and transistor characteristic (p-type) was obtained (Fig. 12). The field-effect mobility from the saturated regime was 2.5×10^{-3} cm^2/Vs, on/off drain current ratio was 1.8×10^2, and threshold voltage was 3V.

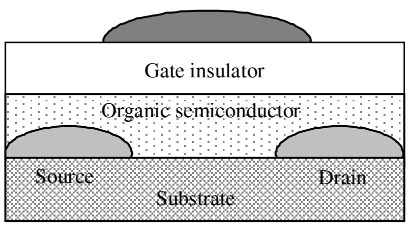


Figure 11. *Schematic of cross-section of the organic thin-film transistor*

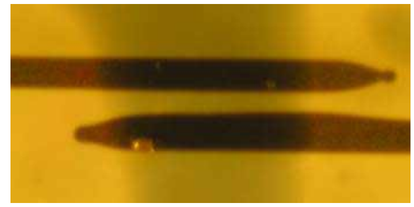


Figure 12. *Top view of the fabricated all-solution-processed organic thin-film transistor.*

Conclusion

The nozzle-less ultrasonic inkjet system keeps the same dot placement accuracy for high-viscosity ink as for standard viscosity ink. The shape of meniscus changes as the viscosity increases. High power is necessary to eject high-viscosity ink. Also, it a contraption is necessary for high ejection frequency.

If these issues concerning the ultrasonic inkjet system can be resolved, it may be widely applied in the field of digital fabrication.

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Biography

Yuko Nomura graduated from Tsuda College, Tokyo, Japan, in 1992. Since 1992 she has been with the Corporate Research & Development Center, Toshiba Corporation, Kawasaki, Japan. Her research interests include inkjet printing and electrophotography. Her work has focused on the development of methods and applications of ultrasonic inkjet printing.