



Fabrication of Flexible Devices by Inkjet Printing

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Abstract. Currently, flexible devices have a wide range of applications such as health managements, drug deliveries, and electronic skins. The inkjet printing is considered the most promising technology for manufacturing flexible devices due to its high resolution, large scale, and low cost. Here, we give a comprehensive review of the fabrication of flexible devices by inkjet printing. First, we introduce the different types of inkjet printing, including continuous inkjet printing and on-demand inkjet printing. In addition, we describe flexible substrates and nano-metallic inks for preparation of flexible devices. Lastly, the perspectives for future development of flexible devices are proposed.

Keywords: Inkjet printing · Flexible device · Flexible substrate

1 Introduction

In recent years, the application of flexible devices has been increasing, such as sensors [1], memristors [2], capacitors [3], diodes [4], circuit boards and solar cells [5]. Especially with the rapid development of flexible wearable devices, there is an urgent need for a feasible printing method that can print high-quality flexible devices. However, the traditional plate-printing and evaporation technologies have been unable to meet the high quality and efficiency required for the production of flexible devices due to issues of high cost, complex processes, and poor-quality control. Compared to traditional methods, inkjet printing requires no contact, no mask, and low cost [6]. The inkjet printing shows great potential in the manufacture of flexible devices: (a) No plate making, the digital process is simple and fast. (b) Non-contact deposition, substrate and pattern layout flexible. (c) High resolution, meets the accuracy of device. This paper summarizes the recent progress of inkjet printing in fabrication of flexible devices, including the design principle of different types of inkjet printing technology, the common flexible substrates, and nano-metallic inks. It provides a reference for the preparation of flexible devices.

2 Inkjet Printing

Inkjet printing is contactless, pressure-free and plateless. Because of the high precision of printing and easy control of ink raw materials, it is very suitable for manufacturing flexible devices. Different inkjet printing technologies have evolved, including continuous inkjet printing and on-demand inkjet printing (Fig. 1) [7].

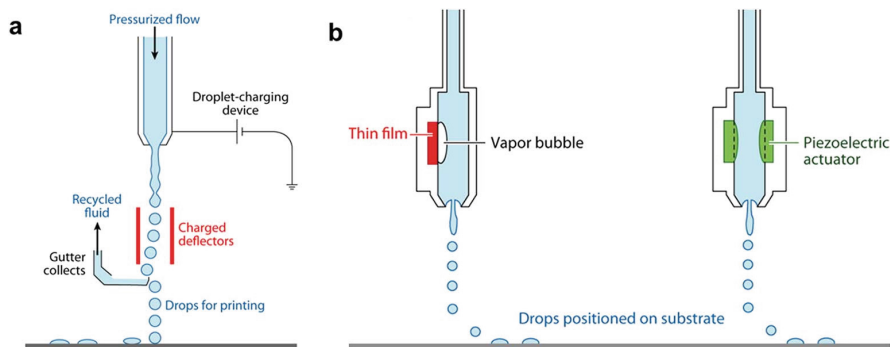


Fig. 1. Continuous inkjet printing (a) and on-demand inkjet printing (b) with hot bubble (left) and piezoelectric (right) printhead

2.1 Continuous Inkjet Printing

Continuous inkjet printing refers to the continuous injection of ink droplets through a pressure chamber equipped with ink. Under the action of ultrasonic driving signal generated by the piezoelectric crystal, the ink flow is decomposed into continuous drops of ink by high-frequency oscillation. A charging electric field controlled by a graphic output signal is provided at the nozzle. The ink droplet is selectively charged when passing through the nozzle [8].

In binary deflection inkjet printing. The droplets participating in recording are not affected by the deflecting magnetic field and keep straight. But the trajectory of charged ink droplet in multi-value deflection inkjet printing is transformed under the action of electric field to reach substrate. Uncharged ink droplets remain in a straight line until they are intercepted by an interceptor and enter the recovery system. Unlike binary deflection, the value of ink drop deflection in multi-value inkjet printing can be changed.

2.2 On-Demand Inkjet Printing

In on-demand inkjet printing, ink droplets are sprayed only for a specified period of time [9]. It has a small inkjet speed, around 30 kHz [10]. But the droplet diameter can reach 10–500 μm . On-demand inkjet printing can be divided into hot bubble inkjet printing and piezoelectric inkjet printing.

Hot Bubble Inkjet Printing. The ink chamber of the hot bubble inkjet printing system is equipped with a film heater and covered with a layer of ink. As the printing system works, the film heater causes the ink to form steam bubbles in an extremely short time. The expanding bubbles push out ink and form ink droplets [11]. Then the film heater cools, the ink fills the nozzle by capillary action. Injection speed is greatly limited because the ink chamber must be refilled with ink before the next drop can be sprayed out. The initial driving pressure is close to the saturated vapor pressure of the liquid at its superheating limit. So Clogged nozzles are a major problem [12].

Piezoelectric Inkjet Printing. Piezoelectric inkjet printing is similar to hot bubble inkjet printing, the difference is that the formation method of ink droplets is different.

Piezoelectric inkjet printing relies on the mechanical action of piezoelectric films to produce pulses [13]. When an electric current is passed through the piezoelectric actuator, it creates a tiny deformation. Therefore, the volume of the ink chamber is reduced, and the ink is extruded from the nozzle. By changing the driving voltage of the piezoelectric actuator, the speed and volume of the jet droplet can be controlled [14].

3 Flexible Substrates of Flexible Devices

Compared with conventional devices, flexible devices can be bent, folded and even transformed into any shape. The substrate of a flexible device is the basis for its flexibility. It can be made from various materials such as paper, conductive glass, and polymer.

3.1 Paper

The paper substrate enables the device to work normally under extreme deformation conditions such as folding, and is easy to recycle, without causing environmental pollution and waste of resources [15]. For its porosity, functional materials are coated on the surface to improve its printability. For example, paper coated with a layer of resin is suitable for inkjet printing. The resin layer helps the paper substrate capture ink particles and absorb excess liquid, resulting in a narrow resolution substrate surface [16]. But the hygroscopicity of paper substrate is worth considering. If the moisture is absorbed by the paper, the device is prone to failure. And during the sintering process, the paper made of cellulose will begin to decompose at a temperature of 100 °C.

3.2 Conductive Glass

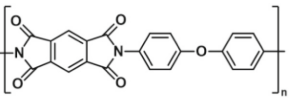
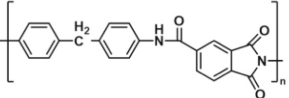
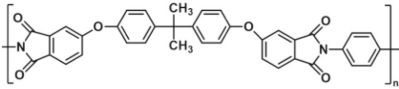
In contrast to a soft paper substrate, conductive glass is a rigid substrate. Conductive glass is divided into volume conductive glass and surface conductive layer glass. The composition of volume conductive glass usually contains basic oxide, silicon oxide and titanium oxide. Surface conductive layer glass refers to the glass surface covered with a layer of conductive film. For example, a film of indium tin oxide (ITO) is plated on a sodium-calcium or boron-silicate substrate glass [17]. It has good transparent conductivity, high transmittance and low resistivity in the visible spectrum, and is widely used in display devices, solar cells and other photoelectric devices. The glass substrate gives the device good electrical and optical properties. With the rapid development of flexible devices, it has a wide prospect.

3.3 Polymer

Polyimide material has comprehensive properties, and its main chain contains the imide ring and nitrogen five-membered heterocyclic ring (Table 1). Among them, PEI has excellent high temperature resistance, corrosion resistance and electrical properties, is widely used in the manufacture of flexible device substrate [18]. PI contains two acyl imide monomers bonded to nitrogen. It is known for its high thermal stability, which also

has excellent dielectric properties and an inherently low coefficient of thermal expansion [19]. In many industrial applications, PI is used to replace glass, metal and even steel. PAI is a polymer obtained by modifying PI. The heat-resistant aromatic heterimide groups and the flexible amide groups are arranged in regular alternations [20]. Polymers provide excellent mechanical properties, heat resistance and creep resistance for flexible devices.

Table 1. Properties of polyimide materials

| species | structure | Tg(°C) | Td(°C) | σ (MPa) | ε (kV·mm ⁻¹) |
|---------|---|--------------|--------|----------------|--------------------------------------|
| PI |  | >250 | >500 | >100 | 3 ~ 4 |
| PAI |  | 270 ~ 300 | >450 | >175 | 3.9 |
| PEI |  | >215 | >518 | >110 | 3.2 |

4 Nano-metallic Inks

At present, conductive inks such as graphene [21], carbon nanotubes [22], nano-metallic inks [23], and conductive polymers are developed [24]. Among them, Nano-metallic inks are most widely used in the preparation of flexible devices, because they have better stability and a high load of 4.0% wt. That means more metal can be deposited each time through inkjet printing. Au, Ag, Cu and other metal nanoparticles can be used to prepare conductive inks. They can be synthesized by physical milling, but the size is not uniform. Reduced metal salts are more reliable in controlling particle size. Compared with Au, Ag nanoparticles have lower resistivity and lower production cost, and have become a popular material for preparing conductive layers of flexible devices. Although the cost of copper is low and the conductivity is high, the high oxide formation rate of copper NPs requires the protection of the precious metal antioxidant shell. In order to improve the performance of ink injection and stability. Dispersant, surfactant, thickener and stabilizer are often added to nano-metallic inks according to the actual demand. The surface design and modification have endowed the nano metallic inks more special properties for greater applications in flexible devices.

5 Conclusions and Prospects

In recent years, inkjet printing has made breakthroughs in convenience, high resolution, mass production, and low cost. Inkjet printing is becoming a common tool for manufacturing flexible devices. However, it needs to overcome new problems to meet the new requirements of flexible devices: (a) Improve the inkjet printing equipment to realize the control of a finer volume of ink droplets. (b) Optimize the substrate materials to improve compatibility with ink. (c) Improve the ink formula to enhance printing quality. Embracing advancements in equipment, substrate materials, and ink formulation will unlock new possibilities, propelling the field forward and empowering researchers and manufacturers to create increasingly sophisticated and high-performance flexible devices.

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References

1. Han, S.-T., et al.: An overview of the development of flexible sensors. *Adv. Mater.* **29**(33), 1700375 (2017)
2. Zhang, H., et al.: Research progress of biomimetic memristor flexible synapse. *Coatings* **12**(1), 21 (2021)
3. Zhao, W., Jiang, M., Wang, W., Liu, S., Huang, W., Zhao, Q.: Flexible transparent supercapacitors: materials and devices. *Adv. Func. Mater.* **31**(11), 2009136 (2020)
4. Lian, C., et al.: Flexible organic light-emitting diodes for antimicrobial photodynamic therapy. *npj Flex. Electron.* **3**(1), 18 (2019)
5. Cheng, Y.-B., Pascoe, A., Huang, F., Peng, Y.: Print flexible solar cells. *Nature* **539**(7630), 488–489 (2016)
6. Zhang, F., et al.: Reactive material jetting of polyimide insulators for complex circuit board design. *Addit. Manuf.* **25**, 477–484 (2019)
7. Huang, T.-T., Wu, W.: Scalable nanomanufacturing of inkjet-printed wearable energy storage devices. *J. Mater. Chem. A* **7**(41), 23280–23300 (2019)
8. Shah, M.A., Lee, D.-G., Lee, B.-Y., Hur, S.: Classifications and applications of inkjet printing technology: a review. *IEEE Access* **9**, 140079–140102 (2021)
9. Basirico, L., Cosseddu, P., Fraboni, B., Bonfiglio, A.: Inkjet printing of transparent, flexible, organic transistors. *Thin Solid Films* **520**(4), 1291–1294 (2011)
10. Peng, X., et al.: Simulation of a hemispherical chamber for thermal inkjet printing. *Micromachines* **13**(11), 1843 (2022)
11. Peng, X., et al.: Design of h-shape chamber in thermal bubble printer. *Micromachines* **13**(2), 194 (2022)
12. Kim, T., et al.: Inkjet-printed stretchable single-walled carbon nanotube electrodes with excellent mechanical properties. *Appl. Phys. Lett.* **104**(11), 113103 (2014)
13. Ko, S.H., Chung, J., Pan, H., Grigoropoulos, C.P., Poulidakos, D.: Fabrication of multilayer passive and active electric components on polymer using inkjet printing and low temperature laser processing. *Sens. Actuators A* **134**(1), 161–168 (2007)

14. Beedasy, V., Smith, P.J.: Printed electronics as prepared by inkjet printing. *Materials* **13**(3), 704 (2020)
15. Rida, A., Yang, L., Vyas, R., Tentzeris, M.M.: Conductive inkjet-printed antennas on flexible low-cost paper-based substrates for RFID and WSN applications. *IEEE Antennas Propag. Mag. Propag. Mag.* **51**(3), 13–23 (2009)
16. Ozcan, A., Tozluoglu, A., Kandirmaz, E.A., Tutus, A., Fidan, H.: Printability of variative nanocellulose derived papers. *Cellulose* **28**(8), 5019–5031 (2021)
17. Rudzik, T.J., Gerhardt, R.A.: Effect of spark plasma sintering current and voltage on the microstructure and electrical properties of borosilicate glass-indium tin oxide composites. *Adv. Eng. Mater.* **22**(5), 1901431 (2020)
18. Lei, W., et al.: Fabrication of electrospun polyetherimide/polyaniline self-supporting microfiber membranes as electrodes for flexible supercapacitors via in-situ polymerization. *Colloids Surf. A* **651**, 129796 (2022)
19. Butnaru, I., Serbezeanu, D., Bruma, M., Sava, I., Gaan, S., Fortunato, G.: Physical and thermal properties of poly(ethylene terephthalate) fabric coated with electrospun polyimide fibers. *High Perform. Polym. Polym.* **27**(5), 616–624 (2015)
20. Mallakpour, S., Zadehnazari, A.: Synthesis and characterization of novel heat stable and processable optically active poly(amide–imide) nanostructures bearing hydroxyl pendant group in an ionic green medium. *J. Polym. Environ. Polym. Environ.* **21**, 132–140 (2012)
21. Deng, D., Feng, S., Shi, M., Huang, C.: In situ preparation of silver nanoparticles decorated graphene conductive ink for inkjet printing. *J. Mater. Sci. Mater. Electron.* **28**(20), 15411–15417 (2017)
22. Tortorich, R., Choi, J.-W.: Inkjet printing of carbon nanotubes. *Nanomaterials* **3**(3), 453–468 (2013)
23. Raut, N.C., Al-Shamery, K.: Inkjet printing metals on flexible materials for plastic and paper electronics. *J. Mater. Chem. C* **6**(7), 1618–1641 (2018)
24. Kraft, U., Molina-Lopez, F., Son, D., Bao, Z., Murmann, B.: Ink Development and Printing of Conducting Polymers for Intrinsically Stretchable Interconnects and Circuits. *Adv. Electron. Mater.* **6**(1), 1900681 (2019)