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Fabrication of self-recoverable flexible and stretchable electronic devices

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ABSTRACT

In this paper, we developed an EHD printing process for the fabrication of high-resolution self-recoverable flexible and stretchable electronics using low-melting-point metal inks. Three different metal inks were tested for their printability on four different substrates separately to demonstrate the capability of EHD printing technology. EHD printing enables low-cost direct fabrication of metallic conductors with sub-50 µm resolution, which represents a promising way to create electronic features with metallic conductivity and excellent flexibility and stretchability. When properly designed, the EHD printed electronics provided a stable resistance under hundreds of bending cycles and many stretching and releasing cycles with high tensile strain, which demonstrates their good flexibility and stretchability in electronics applications. The printed electronics was capable of self-healing under low temperature treatment to recover from failures without sacrificing its electrical properties. Moreover, a high-resolution capacitive sensor array was designed and fabricated. A Finite Element Analysis (FEA) model was developed to study the performance of the designed touch sensor. The results from FEA model agreed well with experimental results, which demonstrated the high-resolution capability of the EHD printing for the direction fabrication of flexible and stretchable devices.

1. Introduction

Printing is a powerful tool to fabricate electronics. Current printing technologies that were used in fabricating electronics include inkjet printing [1,2], 3D printing [3–6], direct writing [7–9], dry printing [10], spray coating [11,12], spin coating [13], screen printing [14], dual-trans printing [15] and soft lithography [16]. A variety of the materials have been used in those printing technologies, such as conductive polymers [10,17], metal nanoparticles/nanowire [18–20], carbon tube [21,22], and liquid metal or low melting point metal [4,8,23,24]. Those materials offer good performance for their specific electronics application. However, many limitations still existed, for example, the electronics printed with polymers doesn't have very good conductivity; electronics printed with metal nanoparticle/nanowire need to perform post-process to improve its conductivity; liquid metal can offer good conductivity and self-healing property [25,26], but it is very expensive comparing with materials mentioned above, and the resolution of the printed features is generally at millimetre scale and packaging is needed to protect the devices. With those limitations, most of those printing technologies cannot offer both high-resolution and high conductivity in electronics. Only a few printing technologies, for example, soft lithography based contact printing can provide high resolution, but the fabrication process is very complex and molds/masks are generally needed, which increase the time and cost of the overall manufacturing process.

Electrohydrodynamic (EHD) is a cost-effective, high-resolution printing method [27–29], which can produce fine droplet or jet that have much smaller size than the nozzle dimension by inducing electric field to the printing system. Many materials have already be applied to EHD printing, such as polymeric materials [30,31], biomaterials [32], metal nanoparticles [33], and some studies [34–36] have demonstrated the capability of fabricating electronics with EHD printing. However, to the best of our knowledge, no study has been reported on direct EHD printing of molten metal inks for the fabrication of electronic devices.

In this work, we studied the fabrication of flexible and stretchable electronics by direct EHD printing of molten metal inks. Micro-scale metallic electronics were successfully fabricated with sub-50 µm resolution and without performing any post-processing. The printed electronic features were tested under cyclic bending test and stretching tests, and the results demonstrated excellent flexibility after hundreds of bending cycles with a bending radius of 10 mm, and during stretching/releasing cycles with a large tensile strain of 70%. Moreover, the printed electronic devices were capable of recovering from failure by heating the sample above the eutectic temperature of the metal ink and applying a small pressure. The EHD printed molten metal features provides a promising method for mask-less fabrication of electronics with metallic conductivity, excellent flexibility and stretchability, and capability of self-recovery.

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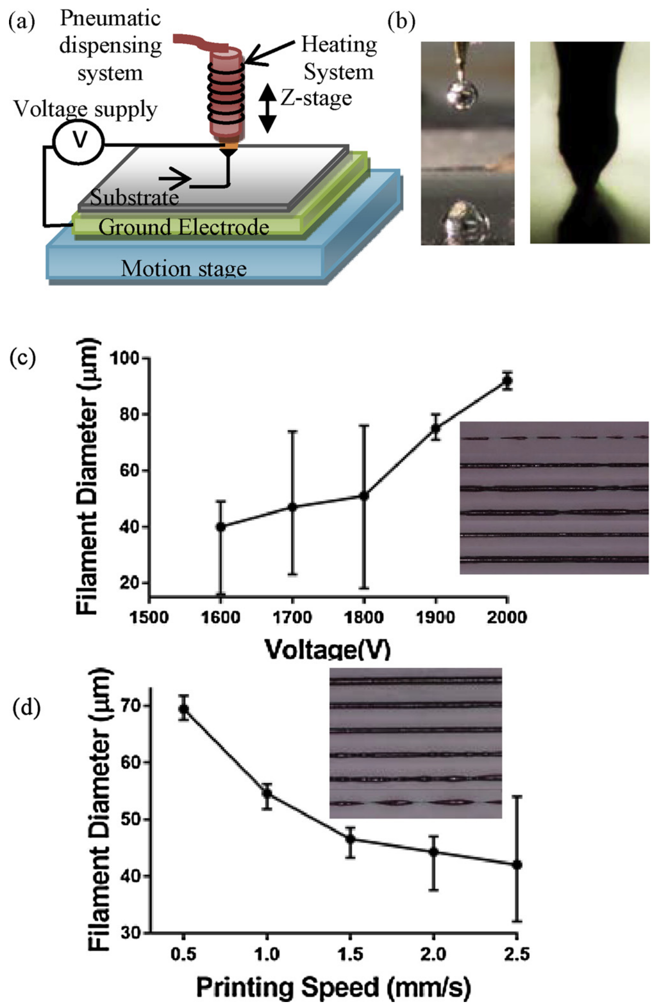

Fig. 1. (a) Schematic of EHD printing system. (b) Droplet printed by extrusion method (left) and cone shape (right) of EHD metal printing. (c) The diameter of printed metallic filaments on glass slide at different voltages of 500, 1600, 1700, 1800, 1900, and 2000 V with a constant speed of 0.5 mm/s. (d) Printed metallic filaments on glass slide at printing speeds of 0.5, 1, 1.5, 2, 3, 4 mm/s with a constant printing voltage of 1900 V.

2. Characterization of EHD printing process

The EHD printing system consists four subsystems: pneumatic dispensing system, thermal control system, high voltage supply system, and precision three-axis motion system. Fig. 1(a) shows the schematic of the EHD printing system setup. The pneumatic dispensing system is controlled by a pressure regulator that provides a small pressure (0.1 psi) to assist the ink flow to the nozzle tip. The thermal control system includes heating rope, thermocouple, and a PID controller that can provide a maximum temperature of 400 °C to the system. The printing nozzle and the ground electrode are connected to a high-voltage supply, which can provide a maximum voltage of 10 K volts. The precision motion stage has three linear stages in XYZ direction with a repeatability and accuracy of 100 nm. A high-resolution camera is used to monitor the printing process. The printing nozzle with an inner diameter of 160 µm is used for EHD printing. The low-melting-point metal inks used in this study are Field's Metal (32.5% Bismuth, 51% Indium, 16.5% Tin), Wood’s Metal (50% Bismuth, 13.3% Tin, 26.7% Lead, and 10% Cadmium), and solder (32% Bismuth, 48% Tin, 20% Lead). Four different types of substrates, glass slide, PDMS, PET, and photo paper are used in this study to show the capability of EHD printing onto different substrates.

In EHD printing of molten metal, a voltage is applied to the system to provide the electrostatic force to eject ink out from the nozzle. When the applied voltage is larger enough, only a very small pressure is needed to keep the ink flow rate. In this study, a pressure of 0.1 psi is applied to the system. Fig. 1(b) compares the printing behaviors between direct extrusion based printing and EHD printing. It clearly shows that the diameter of the ejected droplet in direct extrusion is much larger than the nozzle size. However, for EHD printing, the formed cone shape and ejected filament have a smaller size than the nozzle diameter, which can print high resolution features.

The printing voltage and printing speed are the two critical printing parameters for EHD direct metal printing because both of them affect the printing resolution and the quality of the printed patterns. To find the feasible printing conditions for EHD direct metal printing, characterization experiments for Field’s metal were conducted. Fig. 1(c) shows the printing results under different voltages (500 V, 1600 V, 1700 V, 1800 V, 1900 V, and 2000 V respectively from top to bottom) on the glass slides with a constant printing speed of 0.5 mm/s. During the EHD printing, the electrostatic force overcomes the viscose force and surface tension force to eject ink out from the nozzle. Insufficient voltage (less than 1600 V) brings relatively low ink flow rate that results in discontinuous filaments as in the first line from the top in Fig. 1(c). At 1600 V, continous filaments are printed but with limited uniformity, indicating that the ink flow rate is still low and a larger voltage is needed to improve the printing quality. When the voltage is increased to 1900 V, uniform and the continuous filament is printed on the substrate. Increasing the voltage increases the ink flow rate. With a constant printing speed, filament printed under higher voltage has a larger line width as shown in the Fig. 1(c). Besides the applied voltage, printing speed also affects the printing behavior. When selecting a constant voltage, the printing system has a constant ink flow rate. Printing speed that is much less than the flow rate results in a large amount of ink collecting around the nozzle and a larger line width. On the other hand, too faster printing speed results in discontinuous or nonuniformity line. In a proper printing speed range, higher printing speed produces a continuous filament with smaller linewidth, which can be easily explained as the tensile strength of the printed filament can hold the filament from being torn apart that allows fewer materials printed in a unit length under higher printing speed condition. Fig. 1(d) shows the printing result under different printing speed (0.5, 1, 1.5, 2, 3, 4 mm/s) using a constant voltage of 1900 V. When the printing speed is 1.5 mm/s, good uniformity filaments with linewidth of about 45 µm are successfully printed.

Besides printing Field’s metal on glass, EHD printing is capable of printing a variant of metal inks onto different substrates. To demonstrate the capability of EHD printing of different materials onto different substrates, three different low-melting-point metal inks (Field’s metal, Wood’s metal and solder) and four different substrates (PDMS, PET, and photo paper) are selected to perform the experiment. Field’s metal, Wood’s metal, and solder are all eutectic alloys with the melting point of 60 °C, 70 °C, and 160 °C respectively. Due to the different melting points of printing materials, the printing temperatures are selected to 175 °C for Field’s metal and Wood’s metal, and 300 °C for the solder to ensure the good ink flowability. For printing onto different substrates, the printing voltage and printing speed have to be adjusted accordingly to achieve reliable printing behaviors, because different substrates have different permittivity, which will affect electric field strength around the nozzle tip. However, the overall effect of the printing voltage and printing speed on the printing behavior is similar to that present in Fig. 1. In our study, all three inks have successfully printed on the four different substrates that are widely used in flexible and stretchable electronics. The wettability of the molten metal inks on the substrate affects the adhesion between the printed ink and the substrate thus affect the printing performance. The wettability of three metal inks on four substrates was studied by measuring the contact angle between the interface of ink and substrate surface. The contact angle is measured by evaluating the drop image on the substrate, and