Printing of highly conductive solution by alternating current electrohydrodynamic direct-write

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Abstract. Electrohydrodynamic Direct-Write (EDW) is a novel technology for the printing of micro/nano structures. In this paper, Alternating Current (AC) electrical field was introduced to improve the ejection stability of jet with highly conductive solution. By alternating the electrical field, the polarity of free charges on the surface of jet was changed and the average density of charge, as well as the repulsive force, was reduced to stabilize the jet. When the frequency of AC electrical field increased, the EDW process became more stable and the shape of deposited droplets became more regular. The diameter of printed droplets decreased and the deposition frequency increased with the increase of voltage frequency. The phenomenon of corona discharge was overcome effectively as well. To further evaluate the performance of AC EDW for highly conductive solution, more NaCl was added to the solution and the conductivity was increased to 2810μs/cm. With such high conductivity, the problem of serious corona discharge could still be prevented by AC EDW, and the diameter of printed droplets decreased significantly. This work provides an effective way to accelerate industrial applications of EDW.

1. Introduction

As a typical manufacture technology of flexible electronics, electrohydrodynamic inkjet technology has been the most promising technique for the advantages of simple process, low cost and no pollution, which has attracted interests of researchers in many fields such as semiconductor transistors, organic light-emitting diodes, micro-/nano-sensing and bio-medical [1-3]. How to achieve the precision deposition of micro/nano structures has been the research hotspot in recent years [4-5].

Electrohydrodynamic Direct-Write (EDW) is a novel and simple inkjet technology in which a stable and straight jet is driven by electrical field to fabricate orderly micro/nano structure with a
shortened distance between nozzle and collector and a reduced voltage, which is helpful for decreasing the size of printed structures and increasing the material compatibility to promote its applications [6-8]. However, there are still some problems that hinder the industrial applications of EDW technique. A critical one is the ejection of highly conductive solution. In conventional EDW under DC electrical field, solution with high conductivity usually results in droplet breaking or corona discharge during the ejecting process. For highly conductive solution, the large density of free charges on the jet and the repulsive force will block the stable ejection of a single jet. The highly conductive jet discharges easily, thus it is difficult to direct-write orderly micro/nano structures [9]. The stable ejection of jet with highly conductive solution has been the key to promoting the applications of EDW.

The stable jet with highly conductive solution is the research hotspot for EDW technology, which has attracted wide interests and attentions. Yu [10] et al printed PANI with highly conductive solution on patterned architectures to control the deposition accuracy of nanofiber. Wang [11] et al fabricated conductive silver patterns on demand by pulsed electrohydrodynamic printing and prepared lots of micro/nano devices such as resistor, capacitor and inductor. Watanabe [12] et al achieved aligned nanofiber deposition with two parallel electrodes and discussed the relationship between solution conductivity and polymer structure. In recent years, AC electrical field was introduced to reduce the free charges on the jet and the repulsive force during EDW process effectively, which was beneficial to achieve stable motion of charged jet [13-15]. The rheology and deposition behaviors of jet under AC electrical field are significant to applications of EDW in the integration manufacturing of micro/nano system.

In this paper, an AC electrohydrodynamic direct-write method was proposed, where an AC electrical field, instead of a DC electrical field, was generated to print highly conductive solution. The influences of process parameters were studied.

2. Materials and Methods
The schematic diagram of EDW under AC electrical field was shown in figure 1, including an AC power source, a precise syringe pump, a stainless steel nozzle, a motion platform and a computer. The AC power source (HVP-402NP1, China) was used to generate a high electrical field between the nozzle and the collector. The stainless steel nozzle with an inner diameter of 0.26mm was used as the spinneret. The precise syringe pump (Harvard 11 Pico Plus, USA) was used to supply polymer solution to the spinneret. A silicon wafer, which was cleaned in acetone and deionized water before the experiment, was placed on the motion platform (Parker Linear Motor, USA) to act as the collector. The anode of the power source was connected to the nozzle, and the cathode was connected to the grounded collector. The trajectory and velocity of the motion platform was controlled by a computer. The ejection behavior of the charged jet was captured by a CCD camera (Sony SSC-DC80, Japan) and the morphology of the micro/nano structure was recorded by the optical microscope (Mitutoyo, Japan).

Figure 1. Schematic diagram of EDW under AC electrical field.
Polyethylene Oxide (PEO, $M_w=300\text{kg/mol}$) solution with a concentration of 3wt% was used as the material of the experiments. PEO powder was dissolved in the mixture of deionized water and ethanol ($v:v=2:1$) for the preparation of PEO solution. Then, NaCl was added to adjust the conductivity, which increased the amount of free charges on the jet.

3. Materials and Methods

The ejection behavior of charged jet with highly conductive solution under DC electrical field was first investigated. For solution of high conductivity, the amount of free charges on the jet increased and the radius of jet decreased. The stretching length of the Tayler Cone became larger and the distance between the tip of the Taylor Cone and the collector was shorter, which brought more difficulties for the inducing of single jet with highly conductive solution under DC electrical field, as shown in figure 2. Under DC electrical field, because of the large repulsive force during the ejection process, the jet would have the problems such as whipping, shrinking and breaking.

![Figure 2. Ejection behavior of charged jet with highly conductive solution under DC electrical field.](image)

However, by utilizing the AC electrical field, the polarity of free charges on the surface of the jet was changed by the alternation of electrical field, and the average density of charge, as well as the repulsive force, was reduced, which contributed to improving the stability of charged jet with highly conductive solution. In our work, a series of EDW experiments with solution of different conductivities were carried out, and the influence of process parameters on the printed patterns was investigated.

![Figure 3. Printed patterns under various frequencies with a solution conductivity of 146.5μs/cm.](image)
Figure 4. Printed patterns under various frequencies with a solution conductivity of 770μs/cm.

Because of high conductivity, electrostatic discharge occurred when the distance between the nozzle and the collector was short. So the distance was fixed to be 3mm in our experiments. The positive voltage was 2600V, while the negative voltage was -1000V. The duty ratio was 50%, the solution supply rate was 50μl/hr, and the velocity of collector was 10mm/s. Figure 3 and figure 4 showed the printed patterns under various voltage frequencies when the conductivity of solution were 146.5μs/cm and 770μs/cm, respectively. When the voltage frequency of AC electrical field increased, the EDW process became more stable and the shape of deposited droplets became more regular. Experimental results showed that the diameter of printed droplets decreased with the increase of voltage frequency.

The effect of voltage frequency on the deposition frequency of droplets was further discussed, as shown in figure 5(a). When the voltage frequency increased, the deposition frequency had a trend of increasing. When the conductivity was 146.5μs/cm, the deposition frequency increased by leaps and bounds with the increase of voltage frequency. When the conductivity was 770μs/cm, however, the deposition frequency increased at a lower speed. The diameter of printed droplets was investigated as well, as shown in figure 5(b). The deposition diameter decreased with the increase of voltage frequency. With the increase of solution conductivity, the deposition of droplets became more unstable. Therefore, the diameter of printed droplets for the conductivity of 770μs/cm was larger than the diameter for the conductivity of 146.5μs/cm.

Figure 5. Effect of the voltage frequency on the diameter of printed patterns and the deposition frequency.
To further examine the performance of AC EDW with solution of high conductivity, more NaCl was added to the solution and the conductivity was increased to 2810μs/cm. With such high conductivity, there would be serious corona discharge in DC EDW. This problem could be well overcome under the AC electrical field. The printed patterns under various voltage frequencies were shown in figure 6. When the conductivity was 2810μs/cm, the diameter of printed droplets decreased and the deposition frequency increased significantly with the increase of voltage frequency, as shown in figure 7. Results showed that when the conductivity of solution was beyond a certain degree, the rheology and deposition behavior of jet was different. Because of the high density of charges, the droplets were broken up during the deposition process, thereby the deposition frequency increased and diameter of printed patterns decreased significantly compared with the results of solution with lower conductivity.

![Figure 6. Printed patterns under various frequencies with a solution conductivity of 2810μs/cm.](image)

![Figure 7. Effect of the voltage frequency on the diameter of printed patterns and the deposition frequency when the conductivity of solution was 2810μs/cm.](image)

4. Conclusions
AC electrical field was introduced into EDW to improve the stability of jet ejection. Under DC electrical field, due to the large repulsive force, it is difficult to induce jet ejection with highly conductive solution. However, with the introduction of AC electrical field, the free charges on the jet and the repulsive force were reduced effectively, which promoted the improvement of the stability of charged jet. When the frequency of AC electrical field increased, the printed patterns became stable and the shape of deposited droplets became regular. The diameter of printed droplets decreased and the deposition frequency increased with the increase of voltage frequency. With a solution conductivity of 2810μs/cm, the deposition frequency increased and the diameter of printed patterns decreased significantly. This work provides an effective way to improve the stability of jet ejection especially with highly conductive solution, which is available to push the applications of electrohydrodynamic inkjet technology in the fields of micro/nano system integration.
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