Fabrication and evaluation of a printhead with integrated electrodes for electrohydrodynamic jet printing on insulating substrate

Yanqiao Pan1, 2, * and Liangcai Zeng1, 2
1 Key Laboratory of Metallurgical Equipment and Control Technology of Ministry of Education, Wuhan University of Science and Technology, Wuhan 430081, China
2 Hubei Key Laboratory of Mechanical Transmission and Manufacturing Engineering, Wuhan University of Science and Technology, Wuhan 430081, China
*Corresponding author’s e-mail: panyanqiao@wust.edu.cn

Abstract. Printhead with integrated electrodes plays an important role in electrohydrodynamic jet (E-jet) printing, which can remove the effect of substrate and may be potentially used in fabrication of microelectrodes for flexible electronics. In this paper, we propose a printhead with integrated electrodes to achieve E-jet printing on insulating substrate. Evaluation of the printhead is performed by E-jet printing tests of ethylene glycol in order to validate correctness of design and feasibility of printing on insulating substrate. The printing tests on silicon indicate that the design of proposed printhead with integrated electrodes is correct which can work normally. However, influence of repulsive force by remaining electric charges on substrate is the main cause for failure tests on PET substrate. This research can provide a lesson to implement E-jet printing on insulating substrate in future.

1. Introduction
Electrohydrodynamic jet (E-jet) printing has excellent abilities to fabricate functional micro/nano scale structures for flexible electronics [1]. It is well known that insulating substrates, e.g., PET, PI, are usually utilized in flexible electronic devices [2]. However, insulating substrate cannot be adopted as an electrode directly and can also bring some obstacles for stable performance of E-jet printing. Bu et al. found insulating substrate will affect jet’s direction and lead jet’s bending in space [3]. This indicates that insulating substrate can influence printing position accuracy and consistency. Therefore, it is attractive and challenging to achieve E-jet printing on insulating substrate in a controlled manner.

Actually, the principle of E-jet printing is not complex [4]. When a meniscus on nozzle orifice is applied with an appropriate electric field, shape of the meniscus will change gradually from a half sphere to a cone. Then, a micro jet will burst from cone, and finally deposit onto substrate to fabricate micro/nano scale pattern. E-jet printing has advantages of high resolution using large scale nozzle (inner diameter > 100 μm) and outstanding adaptability to solution with high viscosity (~10000 mPa·s) in drop-on-demand mode [5]. Some scholars have made some efforts on making it controllable and achieving high precision. Rogers et al. demonstrated high-speed printing ability at 1 kHz by setting a pulsed dc voltage and presented the capability to print 3–5 μm droplet size for an aqueous ink and 1–2 μm for a photocurable polymer ink [6]. Lin et al studied electrohydrodynamic deposition of polymeric droplets under low frequency pulsations. They successfully achieved circularly shaped polymeric droplets in an order with diameter of about 20 μm by a stainless capillary nozzle with 180 μm outer
diameter [7]. However, in these studies, the substrates are conductive. Some researchers changed type of applied voltage to print on insulating substrate. Wei et al. presented an ac-pulse modulated electrohydrodynamic jet printing technology and successfully fabricated electrical features and interconnects using silver nano-ink on highly insulating substrates [8]. However, this method needs very accurate control system which costs a lot but it can solve many information in real-time.

In this paper, we proposed a method by utilizing a printhead with integrated electrodes to achieve electrohydrodynamic jet printing on insulating substrate. The design and fabrication process are introduced in details. Evaluation of the printhead is performed by E-jet printing tests of ethylene glycol in order to validate the correctness of design and feasibility of printing on insulating substrate. The internal mechanisms of the printing process and corresponding results are analysed and discussed.

2. Methods and Materials

In this section, design of the proposed printhead with integrated electrodes will be introduced firstly. Then the experimental system for E-jet printing tests on two substrates is presented in details. Finally, the solution and observation equipment that are adopted in experiments are explained clearly.

The printhead with integrated electrodes consists of three parts, i.e., stainless nozzle, PMMA holder and copper electrode as shown in figure 1a. The stainless nozzle is ARQ-S-1020, which is one of the ARQUE serial products of TECDIA Company (Shanghai, China). The inner and outer diameter of the nozzle are 100 μm and 200 μm, respectively. Figure 1b shows the photo of the stainless nozzle. PMMA holder in figure 1a is an insulating clamping component which makes the nozzle and copper electrode to be a whole module. The distance between nozzle and copper electrode can be regulated from 0.4 mm to 1 mm. The copper electrode has a circular hole in the central part which diameter is 2 mm. The stainless nozzle and copper electrode are connected with the positive port and negative port of high voltage source, respectively. These three parts are in good centre accuracy with each other by accurate mechanical assembling method that can potentially fabricate functional micro/nano scale structures for flexible electronics.

Figure 1. (a) Schematic diagram of proposed printhead with integrated electrodes, (b) photo of the stainless nozzle.

Schematic diagram of electrohydrodynamic jet printing system is shown in figure 2, including a printhead with integrated electrodes, dc power supply, syringe pump, light source, high-speed camera and XY motion platform. The dc power supply (DW-P403, Dongwen Inc., China) provides high voltage between stainless nozzle and copper electrode. Solution is supplied to the nozzle by a syringe pump (11 Pico Plus, HARVARD Inc., USA). A high-speed camera (Basler A504k with microlens) and light source (MLS3, NAVITOR Inc.) captures the E-jet printing process. The X-Y motion platform with X-Y linear motor stage (THK Inc., Japan) which is controlled by the programmable controller (Parker Inc., USA) carries the aluminium plate and a substrate to gather the fallen jets.
Figure 2. Schematic diagram of electrohydrodynamic jet printing system.

The adopted solution is the commercial ethylene glycol (Wuhan Shenshi Chemical Engineering Co., Ltd, China). It is a common solution to study the E-jet printing and the physical properties are summarized in Table 1. To evaluate the feasibility and performance of proposed printhead, two substrates (silicon and PET substrate) are adopted in E-jet printing tests. In the first test on silicon substrate, the detail of pulsed rectangular voltage are duty cycle 1%, bias voltage 1 kV, amplitude 0.1 kV, frequency 3 Hz. The distance between nozzle and copper electrode is 0.4 mm. In the second test on PET substrate, the detail of pulsed rectangular voltage are duty cycle 1%, bias voltage 1.8 kV, amplitude 0.1 kV, frequency 3 Hz. Now the distance between nozzle and copper electrode is 1 mm. In both tests, the supply rate, printhead-to-substrate distance and substrate speed are kept the same and they are 300 nl/min, 1 mm and 1 mm/s, respectively. To decrease the effect of solution evaporation as much as possible and capture 2D/3D images of patterns, printed samples were subsequently observed under a confocal laser scanning microscopy (VK-X200, Keyence Inc., Japan). The other experimental conditions are shown as follows: the room temperature is 25°C and the relative humidity is 30%.

Table 1. Physical properties of the ethylene glycol.

| Name          | Density     | Viscosity | Surface tension | Electrical conductivity |
|---------------|-------------|-----------|-----------------|------------------------|
| Ethylene glycol | 1109 kg/m³  | 20 mPa s  | 48 mN/m         | 7.6e-5 S/m             |

3. Results and Discussion

Figure 3a shows the E-jet printing image when the substrate is silicon. The jet is vertical to the substrate and a droplet is achieved on the substrate due to the multiple jets. This indicates that the design of proposed printhead with integrated electrodes is correct and can work normally. Figure 3b shows the E-jet printing image when PET substrate is adopted. The jet is always not vertical to the substrate and the droplets on substrate are also disordered and coarse.

It is meaningful and necessary to explain the results, which determines whether fabrication of flexible devices on insulating substrate can be controlled or not. The whole printing process on PET substrate is replayed one frame by one frame. We can know that the jet is not always bending in a same angle. When the printing starts, the jet is more likely vertical to the PET substrate. After a very short while, the jet starts to “dance” and “bend” gradually. This is different from traditional way that utilizing the bending instability in electrospinning when working distance is high. This is mainly due to effect of repulsive forces by remaining electric charges in printed droplets on PET substrate. When jetting starts for a short while, the charges on substrate cannot affect and change the jet’s direction. However, repulsive forces by electric charges may push the jet and make it bending gradually. This will bring more risk of the instability of jet’s path simultaneously. Therefore, only by the proposed printhead with integrated electrodes still cannot overcome the obstacles of insulating substrate. Some
controlling methods should be combined together to implement E-jet printing on insulating substrate in future.

Figure 3. (a) the E-jet printing image when the substrate is silicon, (b) the E-jet printing image when the substrate is PET.

4. Conclusion
A printhead with integrated electrodes is proposed and successfully fabricated in order to remove the effect of insulating substrate. The printing test on silicon indicates that design of proposed printhead with integrated electrodes is correct and can work normally. However, the bending jet and coarse droplets in printing test on PET substrate show that insulating substrate can still bring difficulties to the printhead. The reasons are mainly due to influence of repulsive forces by remaining electric charges on substrate. This can provide a lesson to researchers, and some controlling methods should be combined together to implement E-jet printing on insulating substrate in future.

Acknowledgements
This research was financially supported by National Natural Science Foundation of China (51705380), Hubei Provincial Natural Science Foundation of China (2017CFB129), Open Fund of Hubei Key Laboratory of Mechanical Transmission and Manufacturing Engineering at Wuhan University of Science and Technology (2017A10) and Cultivating Project for Young Scholar at Wuhan University of Science and Technology (2017xz006).

References
[1] Onses M S, Sutanto E, Ferreira P M, Alleyne A G and Rogers J A 2015 Small 11 4237-4266
[2] Kaltenbrunner M, Sekitani T, Reeder J, Yokota T, Kuribara K, Tokuhara T, Drack M, Schwödiauer R, Graz I and Bauer-Gogonea S 2013 Nature 499 458-463
[3] Bu N, Huang Y and Yin Z 2013 Advanced Materials Research, 684,352-356.
[4] Pan Y Q, Chen X Y, Zeng L C, Huang Y A and Yin Z P 2017 J. Micromech. Microeng 27 125004
[5] Jang Y, Hartarto Tambunan I, Tak H, Dat Nguyen V, Kang T and Byun D 2013 Appl. Phys. Lett. 102 123901
[6] Mishra S, Barton K L, Alleyne A G, Ferreira P M and Rogers J A 2010 J. Micromech. Microeng 20 095026
[7] Xu L, Wang X, Lei T, Sun D and Lin L 2011 Langmuir. 27 6541-6548
[8] Wei C, Qin H, Ramirez-Iglesias N A, Chiu C-P, Lee Y-s and Dong J 2014 J. Micromech. Microeng 24 045010