Conference on Nano/Micro Engineered and Molecular Systems
January 20-23, Xiamen, China

Bead-on-String Structure Formed by Electrohydrodynamic Printing

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***Abstract*— A Bead–on-String (B-S) structure which consists of droplet and filament is generated based on electrohydrodynamic printing system. The formation process of the B-S structure is demonstrated and discussed. Subsequently, the influence of the substrate moving speed on the B-S structure is investigated. The size of the droplet in the B-S structure decreases with the substrate moving speed. When the substrate moving speed is higher than the jetting speed, satellite droplet will appear and its number increases with the substrate moving speed. The effect of the solution concentration on the deposited patterns is also studied. A continuous line is printed with 3 wt% PEO solution. The B-S structure is generated when the concentration of the PEO solution is within 5-15 wt%. Moreover, the size of the droplet decreases with the increasing of the solution concentration. At the concentration of 18 wt%, nanofiber is produced and a pattern similar to the B-S structure is deposited on the substrate.**

***Index Terms*—electrohydrodynamic printing, Bead-on-String structure, deposited pattern**

I. Introduction

As one of the direct-write technologies, Electrohydro-dynamic(EHD) printing has recently attracted attention in patterning micro/nano-sized structures because of the following advantages: (i) EHD printing can directly deposit patterns on the substrate without the need for lithography processes; (ii) versatile materials, including metal nanoparticle [1, 2, 3], organics [4], ceramic [5] and even fragile biological materials [6], can be used as "ink" . (iii) The printing system is simple and easy to implement.

Numerous studies have been attempted to make fine patterns on the substrate. Jang-ung Park and John A. Rogers *et al.* [2] had yields dot-matrix complex patterns with diverse solutions. Patterns of continuous lines and other shapes were also achieved by printing at the speed that allowed the dots to merge. A straight-line pattern was printed on the agarose-coated glass plate with collagen solution by H. –S. Kim *et al.*[7]. D.-H. Youn *et al.* [1] also got a continuous line on the silicon substrate with the silver ink. When D. Y. Lee *et al.* [3] printed the nanopartile suspension onto a polyimide film without a guide ring, the dotted line patterns were yielded, and some of which were shaped like a "hook". However, there are few reports on the Bead-on-String (B-S) structure formed by EHD printing [8].

In this paper, we firstly demonstrate the formation process of the B-S structure, and subsequently investigate the effect of the substrate speed on the B-S structure. In addition, the patterns which are deposited with the solution of different concentration are discussed.

II. Experiments and Discussions

A. Experimental Setup

Figure 1 shows the schematic diagram of the experimental system. A nozzle-perpendicular-to-substrate configuration is used with a stainless steel capillary as the nozzle and a silicon wafer as the substrate. The nozzle diameter is 0.26mm (inner) and 0.51mm (outer), respectively. The distance between the nozzle and the substrate is fixed to 3.55mm. Polyethylene oxide (PEO: M_w=300,000g/mol, Dadi Fine Chemical Engineering, China) is dissolved in de-ionized water to form the solution. The solution with the concentration of 3-18 wt% is prepared. The positive electrode of high-voltage DC power supply (DW-P403-1AC, Tianjin Dongwen, China) is connected to the nozzle, and the negative electrode is connected to the substrate which is grounded. The substrate is put on a computer-controlled X-Y motion stage. The solution is supplied to the nozzle by a syringe pump (HARVARD-11plus, U.S.A) and formed cone-jet mode by the electrical force. A C CD video camera (SSC-DC80, Sony, Japan) is used to observe the printing process in real-time.

Electron micrographs of the nanofiber were obtained by using XL30 Field-Emission Environmental Scanning Electron Microscope (ESEM-FEG).

The subject is supported by National Hi-Tech Research and Development Program of China (2007AA04Z308) and the Key Project of Chinese Ministry of Education (708055).

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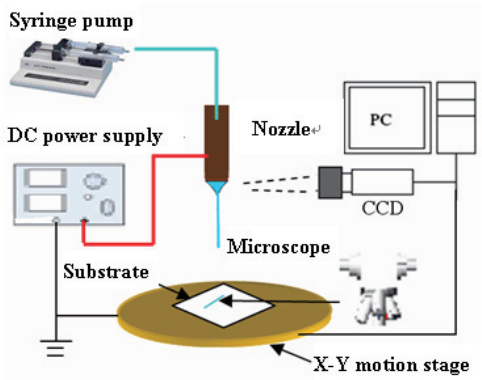

Fig.1. Schematic diagram of the experimental setup

B. Results and Discussion

1. Formation Process

The formation process of the B-S structure is shown in Fig. 2. When the electrical force locally overcomes the surface tension of the liquid, a cone will be formed at the end of the nozzle, from which a thin jet emerges. Before the jet breaks up it strikes to the substrate to form an intact jet, as shown in Fig. 2(a). When the substrate moves in the right direction, the intact jet will be connected with 1# droplet because of the surface tension of PEO solution, as shown in Fig. 2(b). As the moving of the substrate, the intact jet is elongated and its viscoelastic force will increase. With the pulling effect of the viscoelastic force the intact jet will "jump" from 1# droplet to 2# droplet, as shown in Fig. 2(c). During the "jump" process, the further elongated jet becomes filament and then deposits on the substrate. Figure 2(d) shows the B-S structure deposited on the silicon wafer, when the flow rate, the applied voltage, the substrate speed and the concentration of the solution is 50μl/h, 3.2kV, 5mm/s, and 15 wt% respectively. Elliptical droplet can be observed in the moving direction of the substrate. In this paper, we consider the short axis of the ellipse as the diameter of the droplet.

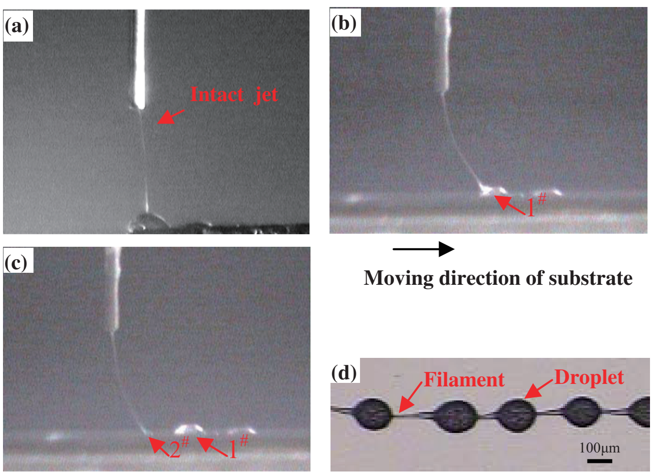

Fig.2. The formation process of the B-S structure. (a) the intact jet between the nozzle and the substrate. (b) the jet connecting with 1# droplet. (c) the jet connecting with 2# droplet. (d) the B-S structure deposited on the substrate when the flow rate, the applied voltage, the substrate speed and the concentration of the solution is 50μl/h, 3.2kV, 5mm/s, and 15% respectively.

2. Moving Speed of the Substrate

The B-S structures printed with 8wt% solution are shown in Fig. 3(a). The moving speed of the substrate is 10, 20, 40, 60, and 80mm/s for each image in Fig. 3(a) respectively, and flow rate is 100μl/h and applied voltage is 4.5kV. There are two connecting ways between droplet and filament. In Fig. 3(b), a detachment can be observed between droplet and filament which is caused by the shrinking effect of the solution subjected to the surface tension. Figure 3(c) shows that although filament still connects with droplet, but the shrinking phenomenon can be observed at the connecting point. From all the B-S structures two obvious features can be observed. One is that the diameter of the droplet decreases with the moving speed of substrate increasing. Another one is that small droplet, which is called satellite droplet, appears when the moving speed of substrate is high .

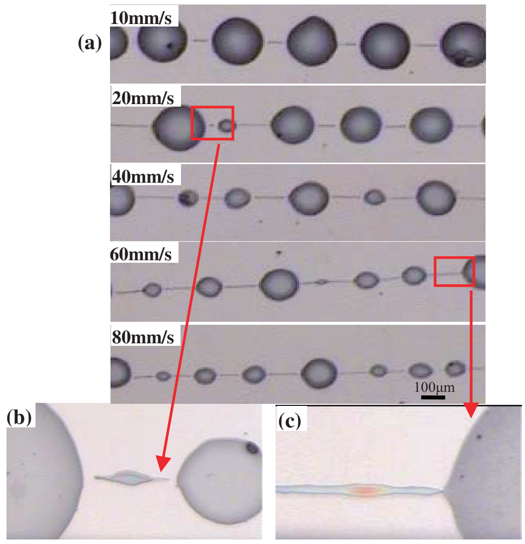

Fig.3. The B-S structures deposited on the silicon substrate. (a) the B-S structures formed at various substrate speed. (b) detachment between the droplet and the filament. (c) connection between the droplet and the filament. Experimental conditions: Applied voltage is 4.5kV, flow rate is 100μl/h and the concentration of PEO solution is 8wt%.

The size of the droplet is determined by the substrate moving speed and the jetting speed. For a given flow rate *Q*, the jetting speed *V* can be estimated by [9]:

$$V = Q / \pi * r^2 \qquad (1)$$

where *r* is the radius of the intact jet and about ~50μm in this experiment. When *Q* is equal to 100μl/h, the calculated value of the jetting speed is about ~14mm/s. The effect of electrical force is neglected in equation (1).

When the substrate moving speed is slower than the jetting speed, the diameter of the droplet does not only depend on the spreading due to the buckling at the impacting point [10], but also on the local accumulation of the solution. The large droplet will appear in the B-S structure. If the substrate moving speed is slow enough, the large droplet in the B-S structure will merge into [9] a line, as shown in Fig. 4.

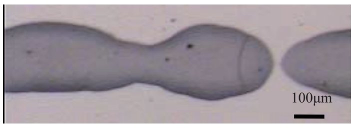

Fig.4. The line deposited on the substrate with the substrate speed of 2mm/s. Experimental conditions: Applied voltage is 4.5kV, flow rate is 100μl/h and the concentration of PEO solution is 8wt%.

When the substrate moving speed is higher than the jetting speed, the droplet diameter is mainly determined by the local spreading and influences a little by the local accumulation, thus the diameter of the droplet decreases. Meanwhile, since the substrate speed is larger than the flow speed, the liquid can not be supplied in time. The intact jet will be elongated and becomes thinner. The thinner jet would "jump" to the next position to form satellite droplet. Before the incoming of liquid supply the "jump" action always produces satellite droplets. The faster the substrate speed is, the more the number of satellite droplets is. This can be observed in Fig. 3.

We increased the flow rate to 250μl/h and repeated the above experiment. The similar results can be found, but the diameter of the droplet is larger than those in 100μl /h case. The higher jetting speed due to the larger flow rate results in more spreading and accumulation of the solution, thus the size of the deposited droplet increases. Figure 5 shows the relationship between the droplet diameter and the substrate speed when the flow rate is 100μl/h and 250μl/h, respectively. The diameter of the satellite droplet also decreases slightly with the increasing of the substrate speed, as shown in Fig. 6.

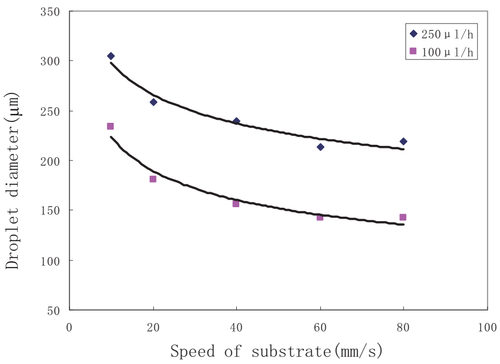

Fig.5. Variation of droplet diameter as the speed of substrate increasing.

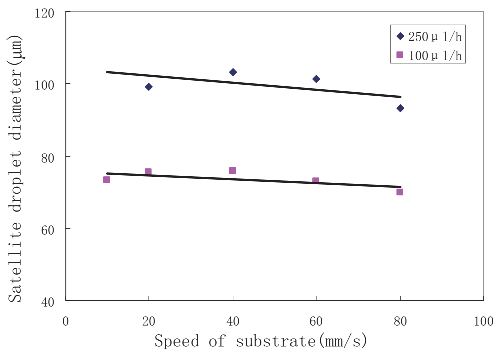

Fig.6. Variation of satellite droplet diameter as the speed of substrate increasing

3. Concentration of the Solution

The concentration of the solution is a key factor for the generation of the B-S structure. At the flow rate of 50μl/h, the applied voltage of 3.2kV and the moving speed of the substrate of 40mm/s, solutions with different concentration are used to print. For 3wt% PEO solution, a continuous line with the mean width of ~300μm was produced, as shown in Fig. 7(a). The line that exhibits rivulet fluctuation [9] owing to the good fluidity is printed in the contact mode [7]. No cone-jet mode is generated due to the low viscosity [11] of 3wt% solution.

When the concentration of the solution is in the range of 5-15 wt%, the B-S structures similar to those shown in Fig. 3 can be generated. Moreover, the diameter of the droplet decreases with the increasing of the concentration, as shown in Fig. 8. We assumed that it is due to the more radial spreading of the low concentration of solution.

For 18wt% PEO solution, electrospinning process happened and nanofiber with the diameter of ~600nm was collected. Figure 7(b) shows the deposited pattern [12]. Nanofibers prefer to aggregate together to form a cluster periodically along the moving direction of the substrate, as shown in Fig. 7(c). Two adjacent clusters are connected with one single nanofiber, as shown in Fig. 7(d). It can be observed that the deposited pattern is similar to the B-S structure, if we consider the cluster as the droplet and the single nanofiber as the filament.

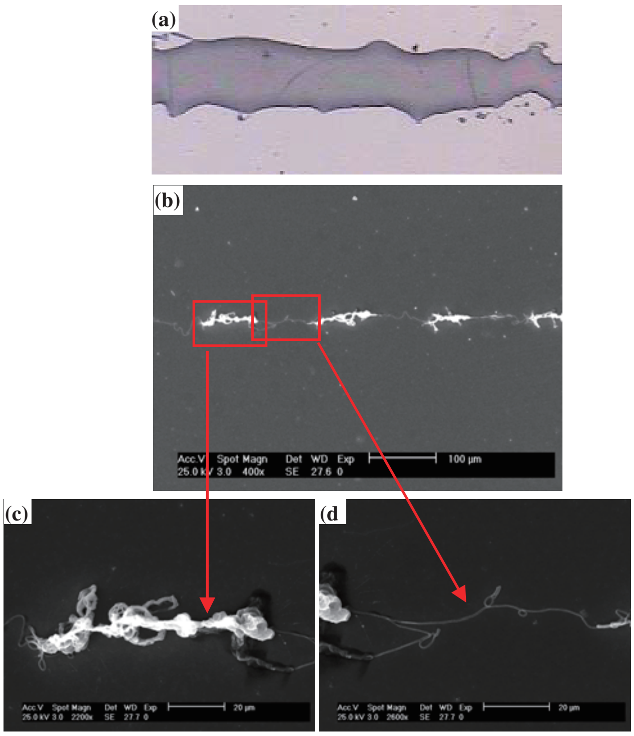

Fig.7. Patterns deposited at different concentration of solution. (a) a line when the concentration of the solution is 3 wt %. (b) nanofibers deposited on the substrate when the concentration of the solution is 18%wt. (c) the cluster of the nanofibers. (d) the single nanofiber which connects two adjacent clusters. Applied voltage is3.2kV, flow rate is 50μl/h and speed of substrate is 40mm/s.

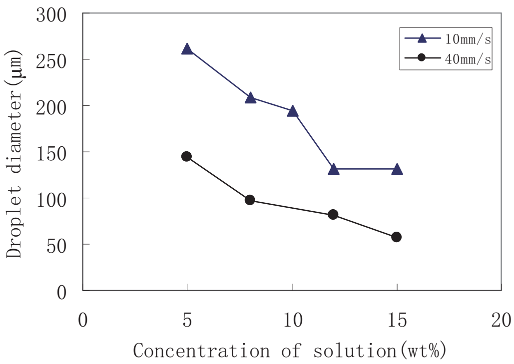

Fig.8. Relationship between the diameter of the droplets and the concentration of PEO solution. Applied voltage is3.2kV, flow rate is 50μl/h and speed of substrate is 40mm/s.

III. CONCLUSION

Based on electrohydrodynamic printing system, the B-S structure is deposited on the silicon wafer. The formation process of the B-S structure is demonstrated and discussed. The substrate moving speed influenced the B-S structure greatly. As the substrate moving speed increasing, the size of the droplet in the B-S structure decreases, while the number of the satellite droplet increases. The effect of the solution concentration on the deposited patterns is studied. Experimental results show that when the concentration of the solution is in the range of 5-15wt%, the B-S structure can be generated. Meanwhile, the size of the droplet decreases with the concentration of the solution increasing. Further experiments are needed to understand the effect of the substrate contact angles on the B-S structure.

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