Pneumatic Conveying Printing Based on Superhydrophobic Surface

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In today’s era, the inkjet printing (IJP) technique plays important roles in the fabrication of mechanical, electronic, and even biological devices. However, the current IJP techniques are incapable of handling viscous inks, which greatly hinder the extensive industrial application. Here, it is found that utilizing the superhydrophobic materials on the end surface of a nozzle combined with the dragging and shearing effects of an airflow, micrometer-sized droplets can be generated under a quite low ejection/extrusion pressure. This variation of the traditional IJP technique is called pneumatic conveying printing (PCP) and is capable of handling viscous inks. Two PCP prototypes using micropumps and micropiezoelectric as the ink exuders, respectively, are homemade to demonstrate the printing performance. For the PCP using micropump inks with viscosities as high as several thousand mPa s can be printed. For the PCP using a micropiezoelectric, the maximum allowed viscosity is about 700 mPa s. Furthermore, the pressure within the pipeline or ink chamber and the shear rate at the orifice during PCP are much smaller than those of the traditional IJP due to the slower ink extrusion rate. Both experiment and simulation are carried out to reveal the mechanisms of the proposed PCP technique.

1. Introduction

The last several decades have witnessed the great success of the inkjet printing (IJP) technique. Beyond the daily office use, inkjet printing also seeks their applications in industry, such as direct fabrication of electrical,[1] optical,[2] mechanical[3] and even biological devices[4] due to its capacity of efficiently, flexibly and cost-effectively patterning of soft materials.[5] The IJP is the non-contact printing techniques, and is an important branch of the family tree of printing technologies which also comprise the contact printing techniques, such as roll-to-roll printing,[6] microcontact printing,[7] nanoimprint[8] technique, and so on. Traditional IJP techniques, such as piezoelectric IJP and thermal-bubble IJP, are typical drop-on-demand (DOD) printing techniques. The principle of IJP is based on the utilization of the transient high pressure, generated either by the rapid transform of the micropiezoelectric crystal attached to the ink chamber (piezoelectric IJP) or by the rapid expansion of a microbubble within the chamber (thermal-bubble IJP), to eject ink drops out of a small orifice. For both traditional IJP techniques, the viscosity of the inks should not exceed several tens mPa s to ensure printability.[9]

Currently, three strategies are normally adopted by IJP for printing viscous inks. The first strategy reduces the viscosity of the ink to a value accepted by the traditional IJP techniques (normally by heating the ink during printing). Now, inkjet nozzles integrated with heating component are commercially available. This strategy is limited since many inks cannot be heated, for instance, inks containing temperature sensitive materials which might be destroyed irreversibly by high temperature or the viscosity cannot be further reduced below the acceptable value even after heating. The second strategy is based on an increase of the transient pressure within the chamber in order to fiercely eject the viscous ink and give the ink sufficient momentum for the detachment of a drop and its subsequent flying. A typical method is the valve-based printing technique,[10] in which a piston, driven by piezostack,[11–13] electromagnetic coil[14] or pressure[15] is used to generate the extremely high pressure in the chamber. More often, viscosity reducing and pressure increasing are adopted at the same time[16,17] for handling inks with extremely high viscosity, for instance, silica gel, adhesive, sealant and so on.[18] The third strategy is utilizing external forces generated by other mechanisms and devices outside the ink chamber/pipeline, to print/dispense the viscous inks without reducing the viscosity or increasing the pressure within the chamber/pipeline. This strategy is represented by the well-known electro-hydrodynamic printing (also known as e-jet printing).[19] and the recently developed acoustophoretic printing technique.[20] For the e-jet printing, high voltage is applied between the conductive nozzle and the opposite conductive printed surface...
(or a conductive substrate beneath the insulating printed surface). Electrostatic force is generated on the meniscus at the tip of the nozzle and drop ejection occurs once the electrostatic force overcomes the resultant of capillary and the viscous force. In the case of acoustophoretic printing, acoustic force generated by the sound wave is utilized for the drops pinch-off from nozzle tip and subsequent drops conveying. For both e-jet printing and acoustophoretic printing, viscous inks (viscosity higher than several thousand mPa s) can be printed. However, the use of high voltage (normally in the range of kilovolt) and complicated acoustic devices increase the risk and difficulty of operation, as well as the cost of the printing device. Here, we proposed a novel DOD printing technique, pneumatic conveying printing (PCP), capable of printing viscous inks by replacing the dangerous high voltage and complicated acoustic devices by a laminar air stream.

2. Principle of PCP

The principle of the PCP is illustrated in Figure 1. The orifice opens on a superhydrophobic surface which can prevent the spreading of the inks drops after the ink being extruded out of the orifice. An airstream parallel to the superhydrophobic surface and perpendicular to the printed surface is used to detach drops after extrusion through the orifice and to convey the drops to the printed surface. In this work, two kinds of PCP devices were homemade to demonstrate the printing performance of this novel technique. The first PCP device uses a micropump controlled by computer for supplying and extruding the ink out of the orifice (Figure 1a); whereas the second device uses a micropiezo near the orifice for ink extrusion (Figure 1b). A vertical gas pipe, mounted just above the orifice, with inner diameters of 1 and 0.2 mm, were used to supply the gas flow for the device shown in Figures 1a,b, respectively. The flow rate was regulated by gas flow control system. The two PCP setups share almost the same printing mechanism and the only difference is the ink extrusion method. Their printing performance is discussed below.

3. PCP Using Micropump for Ink Extrusion

Without air stream and in the case of a relatively small volume extrusion rate \( Q \), the ink extruded out of the orifice will form a growing spherical drop attached to the superhydrophobic surface vertically aligned to the gravity field. The drop grows until its gravity overcomes the capillary force between the drop and the surface (Figure 2a and Video S1, Supporting Information). The volume \( V_D \) and the radius \( R_0 \) of the drop at the moment of detachment depend on the size of the orifice, on the hydrophobicity of the surface and on the physical properties of the ink. Without air stream, experimental results showed that \( R_0 \) can be about ten times larger than the radius of the orifice \( R_0 \) even on superhydrophobic surfaces with a contact angle larger than 160°. The significant difference between \( R_0 \) and \( R_0 \) can be explained by the weak influence of gravity in the case of small drops and expansion of the contact circle between the drop and the superhydrophobic surface during the volume growth of the drop (Video S1, Supporting Information).

However, once the air stream is introduced, both the shape and the size of the drops will be significantly affected. The dynamic drop generation process is shown in Figure 2b and Video S2 (Supporting Information). The entire printing process consists of the drop formation as the first step and the conveying to the printed surface. They are demarcated by the moment of pinch-off. The time needed for the first step, \( t_1 \), is exactly the time between two adjacent pinch-off. \( t_2 \) is the time between pinch-off and the drop landing on the substrate. Experimental results showed that \( t_1 \sim V_D/Q \) and \( t_2 \sim d/v_A \), where \( d \) is the distance between the orifice and the substrate, \( v_A \) is the average air stream velocity at the outlet of the gas pipe.

Drop pinch-off occurs when the dragging force \( F_D \) acting on the drop and generated by the air stream overcomes the vertical component of capillary force \( F_{c,v} \) of the neck connecting the drop and the orifice (Figure 3a). \( F_D \) can be expressed as \( F_D = \zeta \rho D v^2 \), where \( \zeta = 30/\text{Re}^{0.625} \) is the coefficient with the Reynolds number \( \text{Re} \) between 1 and 1000, \( A = \pi R_0^2 \) is the windward area, \( v \) is the relative speed between the drop and the air stream, \( \mu \) is the viscosity of air\(^{[21]} \), \( F_{c,v} \) can be expressed as \( F_{c,v} = F_c \sin \theta = 2\pi R_0 \sigma \sin \theta \), where \( R_0 \) is the minimum radius.
of the neck, $\sigma$ is the surface tension of the ink, $\theta$ is the angle between the axis of the neck and the horizontal direction. $F_D$ increases with increasing drop size whereas $F_C$ decreases with the thinning of the neck. When $F_D$ is close to, but still smaller than, $F_{CV}$, the random vibration of the drop (see Videos S2 and S3, Supporting Information) will result in an unsteady state of the neck. Rapid thinning and breakup of the neck will occur once the drag force overcomes the capillary force, which will lead to a free drop.

After pinch-off, the dragging force can be experimentally determined by the mass and vertical acceleration of the drops obtained by analyzing high-speed images. In order to differentiate $F_D$ obtained by theoretical calculation, $F_{D_{Ex}}$ is used to represent the dragging force obtained by image analysis. The results showed that $F_{D_{Ex}}$ is very close to $F_D$, justifying the reasonability of the above theoretical calculation. It should be noted that the air stream velocity around the drop is asymmetric due to the existence of the superhydrophobic surface. There is a pressure difference between the inner side (close to the superhydrophobic surface) and outer side (far from the superhydrophobic surface) of the drop. This pressure difference will generate a lateral force, $F_P$, acting on the drop, which will drive the drop away from the superhydrophobic surface. The magnitude of $F_P = ma_L$ was determined by the droplet mass $m$ and lateral acceleration $a_L$ of the drops, which can be obtained by analysis of the high-speed images. Figure 3b indicates a value of $F_D$ of no more than 1 $\mu$N for a drop with radius of about 58 $\mu$m and $v_A$ of 16.2 m s$^{-1}$. Gravity can be neglected since the Bond number $Bo = \rho g R_D^2/\sigma$ is always smaller than 0.1 in our experiments, where $\rho$ is the density of the ink and $g$ is the acceleration of gravity.

As shown in Figure 3c, the size of the drops is comparable to the thickness of the boundary layer of thickness $\delta$, which can be expressed as $\delta \approx 5.0 x / \sqrt{Re}$ for laminar flow,[22] where $x$ is the position along the semi-infinite plate, and $Re = \rho v_{\infty} x / \mu$ is the Reynolds number ($\rho$ and $\mu$ is the density and dynamic viscosity of the gas). The boundary layer is defined as the region close to the surface where the velocity is less than 99% of the far-field velocity $v_{\infty}$.

A series of experimental results revealed that the radii $R_D$ of the droplets were affected significantly by the radius of the orifice $R_O$ and the air stream velocity $v_A$. For a given $R_O$, the droplet radii will decrease significantly with increasing $v_A$ (Figure 4a). As stated above, pinch-off occurs once the dragging force of the airstream $F_D$ is close to the vertical component of the capillary force $F_C$, which scales as $F_C \sim R_N$. For a given nozzle, $R_N$ is proportional to the radius of the nozzle, $R_N \sim R_O$. From $F_D = \zeta A \rho v_{\infty}^2$ and $\zeta = \frac{30}{Re^{0.625}} = \frac{30}{(\rho v_{\infty} d / \mu)^{0.625}}$, we can...
It should be noted that approximately according to \( D \) relationship between \( R \) and \( v_\lambda \) yielding a good linear relationship between them. It was found. Figure S1 and Video S3 (Supporting Information) illustrate the droplet formation and transportation process at different \( Q \). Figure S2 (Supporting Information) demonstrates that a printing frequency of several kHz can be achieved. In our experiment, the smallest \( R_D \) \((V_{30})\) of 33 \( \mu \text{m}\) (150 \( \mu \text{L}\)) is achieved by using an orifice with \( R_O = 30 \mu \text{m}\) and a strong air stream with \( v_A = 16.2 \text{ m s}^{-1}\). A further increase of the air stream velocity will blur the printed pattern due to the lateral movement of the droplet on the printed surface before its spreading on the surface or its uptake by the substrate material (Figure S3, Supporting Information).

4. Printing Performance

**Frequency:** For a given \( v_A \), a linear relationship between printing frequency \( f \) and extrusion rate \( Q \) was found. \( Q \) has little effect on the droplet size \( R_D \) (Figure 5a,b). Figure S1 and Video S3 (Supporting Information) illustrate the droplet formation and transportation process at different \( Q \). Figure S2 (Supporting Information) demonstrates that a printing frequency of several kHz can be achieved. In our experiment, the smallest \( R_D \) \((V_{30})\) of 33 \( \mu \text{m}\) (150 \( \mu \text{L}\)) is achieved by using an orifice with \( R_O = 30 \mu \text{m}\) and a strong air stream with \( v_A = 16.2 \text{ m s}^{-1}\). A further increase of the air stream velocity will blur the printed pattern due to the lateral movement of the droplet on the printed surface before its spreading on the surface or its uptake by the substrate material (Figure S3, Supporting Information).

**Ink Viscosity:** Inks with viscosity in the range of 1–4000 mPa s were tested in this work. As shown in Figure 5c, the size of the droplet slightly increases with increasing ink viscosity. This can be explained by the increment of the viscous force during the pinch-off process. In order to break the neck, the drag force generated by the air stream has to overcome the summation of the capillary force and viscous force.

**Pressure within the Pipeline:** For the PCP using plunger pump for ink supply, the pressure inside the pipeline is dependent on the geometric parameters of the pipeline, the ink viscosity and the volume extrusion rate. Figure 5d shows the pressure within the pipeline during printing inks of different viscosity. The pressure within the pipeline increases with increasing ink viscosity and extrusion rate. For the ink with a viscosity of 1400 mPa s at an extrusion rate of 0.1 \( \mu \text{L s}^{-1}\), the pressure inside the pipeline is less than 0.7 \( \times 10^5\) Pa.

5. PCP with Micropiezo for Ink Extrusion

The PCP using micropump cannot be regarded as a real drop-on-demand (DOD) printing technique since the resolution of the pump, i.e., the minimum volume that can be extruded, is much larger than the volume of the printed drops. Here, DOD-PCP was demonstrated by using a micropiezo as the ink extruder. As shown in Figure 1b, a traditional piezo inkjet nozzle was horizontally placed. And the end surface of the nozzle was treated to be superhydrophobic. There is not a fundamental difference about the drops generation and conveying mechanisms between the two PCP devices. However, when micropiezo is used as the extruder, it not only extrudes the ink out of the orifice, but also sucks the drop suspended at the orifice back to the chamber when the piezo restores its original shape (Figure 6a). This is observable as the drop oscillation phenomenon at the orifice.\(^{[23]}\) Once the shear direction air stream is turned on, the suspended drops will be blown away.
instead of being sucked back into the chamber (Figure 6b). Experimental studies have shown that the droplets can be stably and quickly transferred to the destination after the droplets departed from the nozzle. This nozzle can be mounted on three-axis mobile platform for pattern printing (Video S5, Supporting Information).

6. Drop Oscillation at the Orifice

The transform rate (magnitude) of the piezo can be adjusted by the slop (magnitude) of the voltage waveform applied on it. The typical structure of the piezoelectric inkjet nozzle consists of an ink chamber equipped with a micropiezoelectric
actuator, an orifice for ejecting the ink drops and an inlet for supplying the ink. The piezoelectric actuator attached to the ink chamber contracts inward if a voltage is applied to it. This action decreases the volume of the chamber and the ink will be ejected out of the orifice. Then the voltage decreases to zero and the actuator returns to its initial shape. Due to the capillary force and the pressure applied at the inlet, the chamber will be refilled for the next ejection within a very short time. For stable drop ejection, the drop must acquire enough momentum during the ejection process to overcome the surface tension. However, if the ejection volume is too small and/or the ejection speed is too slow, the momentum of the ink drops will be insufficient for detachment from the orifice. It will then be sucked back into the orifice when the actuator returns to its initial shape (Figure 6a and Video S4, Supporting Information). This phenomenon is observable as the drop oscillation with the frequency of the piezoelectric actuator. Experimental results showed that, drop oscillation phenomenon can be achieved by adopting a voltage program with either lower amplitude or lower slope \( \frac{dU}{dt} \) than those needed for drop ejection. Besides the traditional rectangle and the trapezoidal waveform, which is generally used for the traditional piezo inkjet, sinusoidal waveform also results in drop oscillation. The adopting of sinusoidal waveform greatly reduces the pressure inside the chamber.

7. Pressure Inside the Chamber and Shear Rate at the Orifice

Since the pressure inside the nozzle is difficult to be directly measured during the actual printing process, computational fluid dynamic (CFD) simulation was carried out to reveal the fluid dynamics and the pressure inside the chamber for both the ejection and the oscillation process. The results showed that the pressure in the chamber during oscillation is much smaller than that during ejection. For instance, with an ink viscosity of 20 mPa s, the minimum pressure needed for drop ejection is about \( 2.23 \times 10^6 \) Pa, whereas the pressure needed for drop oscillation is only \( 1.51 \times 10^5 \) Pa (Figure 7a,b). The shear rate at the orifice during oscillation is about one order of magnitude smaller than that during ejection due to the lower extrusion rate (Figure 7c,d). When the viscosity of the ink is 500 mPa s, the minimum pressure inside the chamber needed for ejection and oscillation is about \( 1.53 \times 10^7 \) and \( 3.53 \times 10^5 \) Pa, respectively (Figure S4a,b, Supporting Information).

Experimental results show that stable ejection cannot be achieved if the ink viscosity is higher than about 50 mPa s, which is consistent with previous reports about the allowable viscosity of the traditional inkjet.[9] However, our experimental results showed that stable oscillation still can be obtained with ink viscosity as high as 700 mPa s. That means the maximum

![Figure 7. Pressure inside the chamber and shear rate at the orifice during PCP and comparison with traditional IJP. a) Maximum pressure and b) pressure distribution, within chamber during drop ejection and drop oscillation; c) maximum shear rate and d) shear rate distribution, at the orifice during drop ejection and drop oscillation.](image-url)
allowable viscosity of the ink of the traditional inkjet technique can be increased by at least one order of magnitude, just by adopting a novel printing mechanism.

Compared with PCP using a micropump, when a micropiezo is used as the ink extruder, the droplet radius $R_D$ is mainly dependent on the ink volume extruded by one piezo contraction, which in turn depends on the electrical parameter applied to the piezo. As shown in Figure S5 (Supporting Information), the velocity of the air stream $v_A$ still has an influence on the size of the droplets to some extent, but does not have a significant influence on $R_D$. The decisive factor is the amplitude of the driven voltage of the micropiezo, which determines the volume of droplets extruded out of the nozzle. When the extruded liquid is to be sucked back into the orifice rapidly, a thin liquid neck will form at the inner orifice, so the droplet at the orifice can be easily cut by air flow drag force $F_D$ with the cooperation of the oscillation. The size and trajectory of the droplets are relatively uniform and stable under the constraint of the gas flow, which is very important for forming an accurate pattern on the substrate. Figure S6 (Supporting Information) shows the droplet size distribution. During the printing process, the distance between the nozzle and the substrate was kept within 3 mm, and with this method, accurate printing can be performed on the flexible substrate or even the uneven bottom surface. Figure 8 shows some patterns that include straight lines, diagonal lines, curves, and arbitrary lines implemented by the PCP technology. Video S5 (Supporting Information) shows the real-time printing process.

In conclusion, a variation of the traditional inkjet printing technique, pneumatic conveying printing, was proposed and two different solutions for ink extrusions were demonstrated and their performances were characterized. Due to the hydrophobicity of the end surface of the nozzle, drops can be easily generated and conveyed to the printed surface by a laminar air stream. This novel printing mechanism allows PCP capable of handling inks with viscosity up to several hundred mPa s compared to several tens mPa s for a traditional inkjet nozzle. PCP using a micropump as ink extruder even allows ink viscosity as high as several thousand mPa s. Furthermore, the pressure within the ink chamber during PCP is much smaller than those of the traditional IJP. The size of the drop depends on the size of the orifice, the hydrophilicity of the surface and the velocity of the air stream. By adjusting the air stream velocity and liquid supply rate, printing frequencies of several kHz can be achieved for liquids with high viscosity. The results open the door for printing viscous media, especially bioinks in which pressure-sensitive and thermally unstable materials, such as biological cells and proteins are contained.

8. Experimental Section

Materials: The superhydrophobic coating agent (Glaco Mirror Coat “Zero”) was purchased from Soft 99 Co., (Japan). Glycerol was purchased from Sinopharm Chemical Reagent Co., Ltd (China). The color inks were made by deionized water and commercial blue inks for pens. Before mixing, the commercial blue inks were filtered by the mixed fiber resin aqueous phase filter, whose specification was 13 mm × 0.22 μm. The mixture was processed by ultrasonication for 10 min before use. The inks with different viscosity were obtained by mixing different ratios of blue inks and glycerine. PET plate, photo paper, and normal A4 paper without any surface pretreatment were used as the substrate.

Instruments and Experimental Methods: In the case of PCP using a micropump as ink extruder, the nozzle was homemade by traditional manufacture methods. Teflon was used to make the nozzle due to its machinability and chemical stability. The orifices with different size were fabricated by a micromilling cutter. The surface on which the orifice opens was treated by the commercial superhydrophobic coating agent following the instructions of the supplier. During coating, a continuous gas stream was blown through the orifice in order to avoid the blocking of the small orifice. It took 12 h to achieve full consolidation after coating. The conical glass capillary used for constraining the air stream was made by a laboratory micropipette puller. The end surface of the glass capillary was polished by a pipette grinding apparatus to obtain the required size. The inks were supplied by a programmable micropump (Harvard Apparatus, PHD ULTRA, USA). The flow rate of this pump could be adjusted between 0.026 μL s$^{-1}$ and 3.67 mL s$^{-1}$.

In the case of PCP using piezo inkjet nozzle, the nozzle (MJ-AT-01-40) was purchased from MicroFab. The nozzle was horizontally
mounted onto the homemade PMMA base. The glass capillary was perpendicularly mounted on the base just above the nozzle. The gas flow was supplied by the vacuum diaphragm pump (SJ-PUMP-02, purchased from Shanghai Ruidu Photoelectric Technology, China) and flow rate of the gas stream was adjusted by changing the pressure inside the pipeline with a precision pressure controller (CT-PT-21, purchased form MicroFab, USA) with resolution of 2 mm Hg. The back pressure needed for the nozzle during printing was also supplied by the same pressure controller.

The observation of the dynamic formation and movement of the droplet was carried out on the platform of an inverted optical microscope (Leica, DM i8, Germany). And the whole process was monitored and recorded by the high-speed camera (Photon, SA-Z, Japan) connected to the optical microscope. The frame rates of this high-speed camera could be varied from 50 to 100 000 frames s\(^{-1}\) with constant resolution from 1024 \(\times\) 1024 to 572 \(\times\) 260 pixels. The droplet-size parameters were obtained by image analysis by comparing it size to the known size of the image. The nozzle was mounted on a three-axis moving stage driven by three linear motors for printing the patterns. The whole process of ink jet printing was recorded by a CCD camera (SENTECH, STC-MB133USB, Japan).

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

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