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ORIGINAL RESEARCH

The effect of ink drop spreading and coalescing on the image quality of printed cotton fabric

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Abstract Cotton fabric has been extensively used as the substrate of inkjet printing to manufacture traditional garments as well as emerging e-textiles due to its comfort, renewability, good dyeability, biodegradability and relatively low cost. In present work, the spreading and coalescence of ink drops on a cotton fabric as well as their effects on the image quality were investigated. A reactive orange 13 dye was selected as the colorant to make it convenient to observe the depositing morphologies of ink drops. The impacting and wetting processes of an ink drop on a cotton fiber were observed through a high-speed camera. Depositing morphologies of an ink drop, coalescing structures of ink drops and patterns printed with different drop spacings were observed through a microscope. The results show that the ink drop stably deposited on the cotton fabric and formed a long strip pattern after wetting. That indicates the inkjet printing pattern on a cotton fabric should be composed of "line segments" instead of round points. The edges of the pattern printed with a small drop spacing appeared bleeding phenomenon due to the ink drops excessively accumulated on the gaps between cotton fibers. Ink drops could not coalesce at a large drop spacing resulting in the printed pattern being discontinuous. The ideal pattern was printed at an intermediate drop spacing, which was 20 μm in this experiment.

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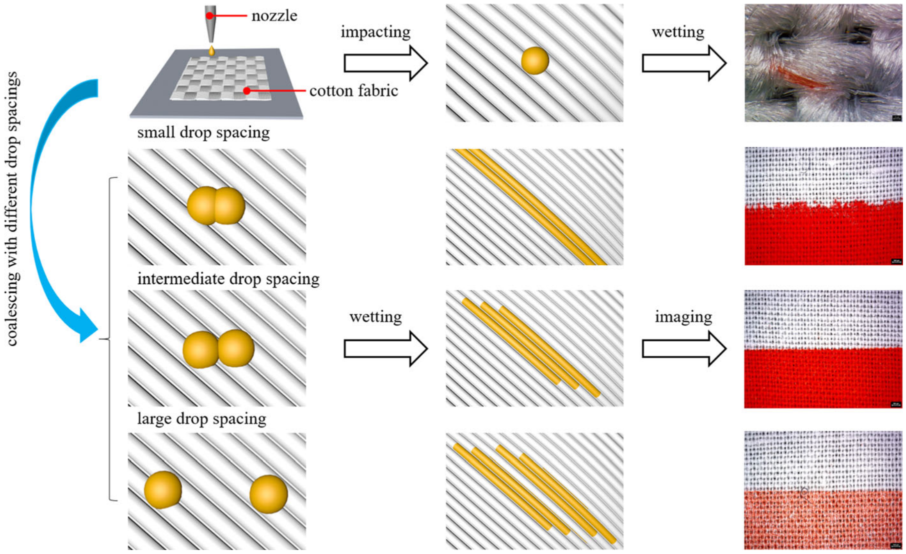
Graphic abstract



Keywords Cotton fabric · Inkjet printing · Spreading and coalescence · Image quality

Introduction

As a major textile fiber, cotton has been dominant for centuries due to its unique combination of properties, including wearing comfort, renewability, good dyeability, biodegradability and relatively low cost (Bao et al. 2019; Wang et al. 2017; Xu et al. 2015). With the development of scientific technology, cotton fabric has been extensively used as the substrate for inkjet printing to fabricate not only traditional garments, but also emerging electronic textiles, such as OLEDs, conductive electrodes, capacitors, sensors, and thin film transistors (Kao et al. 2019; Kim et al. 2013, 2019; Nechyporchuk et al. 2017; Agate et al. 2018; Ren et al. 2017; Li et al. 2017; Shahariar et al. 2019; Zhang et al. 2018; Castro et al. 2015). Inkjet printing is a non-contact printing technology with advantages of simplicity, low costs, high resolution and saving materials (Singh et al. 2010; Song et al. 2018; Wang et al. 2019; Zhang et al. 2019; Carey et al. 2017; Chinga-Carrasco et al. 2012; Kao et al. 2019; Li et al. 2018). Since it is a non-contact printing, the spreading and coalescing of ink drops on substrates affect the quality of printed products directly. Therefore, it is of primary importance to understand the mechanism of ink drop spreading and coalescence on a substrate for improving the quality of printed patterns (Hoath 2016; Carter et al. 2012; Derby 2010).

The spreading of an ink drop on a substrate mainly includes two processes (Hoath 2016). First one is the drop impacting on the substrate. In this case, according to different impacting conditions, six different phenomena have been observed including deposition, prompt splash, corona splash, receding breakup, partial rebound and complete rebound. (Josserand and Thoroddsen 2016; Rioboo et al. 2002). The other process is the liquid wetting the substrate. Both the two stages are closely related with the nature of the substrate, such as the surface texture, chemically homogeneous or heterogeneous, hydrophobic or hydrophilic and planar or nonplanar. Surface wettability of the substrate plays a central role on the wetting process and the liquid lamella rupture behavior (Dhiman and Chandra 2010; Roisman et al. 2002; Bartolo et al. 2005). When the contact angle exceeds the static advancing contact angle, the liquid starts to spread. Correspondingly, a contact angle, which is lower than the receding contact angle results in retraction of the liquid (Marengo et al. 2011). Reynolds number is one of the dimensionless numbers to quantify the impact process of ink drops, which is calculated according to Eq. (1):

$$Re = \frac{\rho DV}{\mu} \tag{1}$$

where Re is the Reynolds number, ρ is the density of the liquid, D and V is the diameter and the velocity of the ink drop, respectively, and μ is the surface tension of the liquid (Yarin 2006). During impacting process, liquid lamella breakup occurs when the actual Re value is above the threshold Re value, which is determined by the receding contact angle (Dhiman and Chandra 2010). It is interesting the value of the threshold Reynolds number is the smallest, when the contact angle is a middle value. In contrast, low or very high contact angles correspond to a high threshold Reynolds number, which contributes to stabilizing the liquid film. Spreading and splashing thresholds are usually affected by the surface morphology of the substrate (Roisman et al. 2015; Antonini et al. 2014; Kim et al. 2014; Latka et al. 2012; Pittoni et al. 2015). Under Wenzel state, when the intrinsic contact angle is lower than 90°, the apparent contact angle decreases with the increase of the surface roughness. Correspondingly, increase of the surface roughness causes the apparent contact angle increasing at hydrophobic substrates. The effect of surface microstructures on the deformation of alumina micro-drops (35–55 µm in diameter) has been investigated. The impacting velocity was 90 m/s, which corresponded to the condition of plasma spraying. It has been shown that the surface structure of the substrate influences the splat diameter and the shape stability of the micro drops (Shinoda et al. 2007). Surface curvatures of the substrate influence the spreading diameter of the drop (Hung and Yao 1999; Bordbar et al. 2018). When a drop impacts on a curved surface, the maximum spreading diameter typically increases with the increase of the curvature. The uniform disintegration is mainly observed at a moderate impacting velocity, and the spherical diameter of the substrate is smaller than the droplet diameter.

Designing droplet depositing patterns and ink formulations according to the drop coalescing structures is a necessary condition for obtaining stable droplet patterns on the substrate (Carter et al. 2012). Therefore, more time should be emphasized on understanding the mechanism of the coalescence of ink drops. Good coalescence should form a continuous, straight and uniform line. Many studies have shown that drop spacing is a key factor to determine the drop coalesced structure. Therefore, a suitable drop spacing has a crucial importance in inkjet printing. Soltman et al. used a 60 µm diameter nozzle to investigate the drop coalescing structures at different drop spacings. They found a uniform line formed at a 50 µm drops spacing (Soltman and Subramanian 2008). Stringer et al. found that a stable line width is shown to be bounded by two limits. The lower bound (minimum line width) is determined by the maximum drop spacing for stable coalescence while the upper bound is determined by the minimum drop spacing below which a bulging occurs. The maximum stable track width is also a function of the velocity at which an inkjet printhead traverses the substrate (Stringer and Derby 2010).

The objective of this work is to investigate the spreading and coalescence of inkjet drops on a cotton fabric, focusing on improving the printing quality through adjusting the printing process. The impacting and wetting processes of an ink drop on the cotton fabric were investigated through taking photos by a high-speed camera. Depositing morphologies and coalescing structures of ink drops on the cotton fabric were observed by a super depth of field microscope. Besides, six square patterns with different drop spacings were designed and printed to validate the effect of ink drop spreading and coalescence on the printing quality.

Experimental part

Materials

A desizing, scouring and bleaching plain-woven pure cotton fabric was provided by Yuyue Home Textile Co., Ltd. (Shandong, China). The densities of warp and weft yarns were 110 · 75, respectively. Reactive orange 13 dye purchased from Taiwan Yongguang Co., Ltd. (Taiwan, China) was used as received. Ultrapure water was prepared by a Direct-Q8 device purchased from Millipore Co., Ltd. (America).

Characterization of the cotton fabric

The contact angle of the cotton fiber tested by a DSA30M contact angle tester (KRUSS Co., Ltd, German) was approximately 90°, as shown in Fig. S1. The diameter of the cotton fiber was measured by a Phenom scanning electron microscope (Netherlands).

Preparation of dye solution

Reactive dye a kind of water soluble molecular dye can move with water molecules in solution (Wang et al. 2020; Xie et al. 2020). Therefore, a reactive orange 13 dye was selected as the colorant to make it convenient to observe the depositing morphologies of ink drops on the cotton fabric. The dye solution used in this experiment was composed of 5 wt%reactive orange 13 dye and 95 wt% water. The solution was filtered by a 0.22 μm filter film before used in inkjet printing. Surface tension and viscosity of the dye solution were measured by a BP-100 surface tension device provided by KRUSS Co., Ltd. (German) and a Fluidicam RHECO instrument purchased from Formulaction Co., Ltd. (France). The surface tension and viscosity were approximately 72.5 mN/m and 1.15 mPa s, respectively, as shown in Fig. S2a and S2b.

Inkjet printing and imaging system

The inkjet printing and imaging system used in this work was composed of a custom-built research inkjet printer supplied by Shanghai Ruidu Optoelectronics Technology Co., Ltd. (Shanghai, China) and an i-SPEED 7 high speed camera purchased IX cameras Co., Ltd. (England) as shown in Fig. 1. The frame rate of the camera was 100,000 frames per second. A Microfab piezoelectric drop-on-demand dispensing printhead with a 30 μm orifice was used in this experiment. During inkjet printing, the nozzle can move in up-down directions while the platform can move in front-back and right-left directions. The cotton fabric was attached to the platform so that it could move with the platform to form patterns.

The waveform was adjusted according to the surface tension and viscosity of the dye solution to obtain a stable droplet formation, as shown in Fig. S2c. The droplet formation during observing the impacting and wetting processes was shown in Fig. S2d. The droplet diameter was 30 μm, which was same with the nozzle diameter. Falling velocity of the droplet at different dropping moments were calculated as shown in Fig. S2e. It illustrates that the droplet falling can be regarded as a uniform linear motion. The average velocity approximated to 2.4 m/s.

In the experiment, the moving velocity of the platform was constant 15,000 μm/s. Different drop spacings were achieved through varying the jetting frequency. The drop spacing was calculated through the Eq. (2), as follow:

$$d = \frac{V}{F} \tag{2}$$

where d was the drop center spacing, V was the moving velocity of the platform, and F was the jetting frequency. When the drop center spacings were 10 μm, 20 μm, 30 μm, 40 μm, 50 μm and 60 μm, the corresponding jetting frequency was 1500 Hz, 750 Hz, 500 Hz, 375 Hz, 300 Hz and 250 Hz, respectively. The droplet formations of different jetting frequencies were almost same, as shown in Fig. S3.

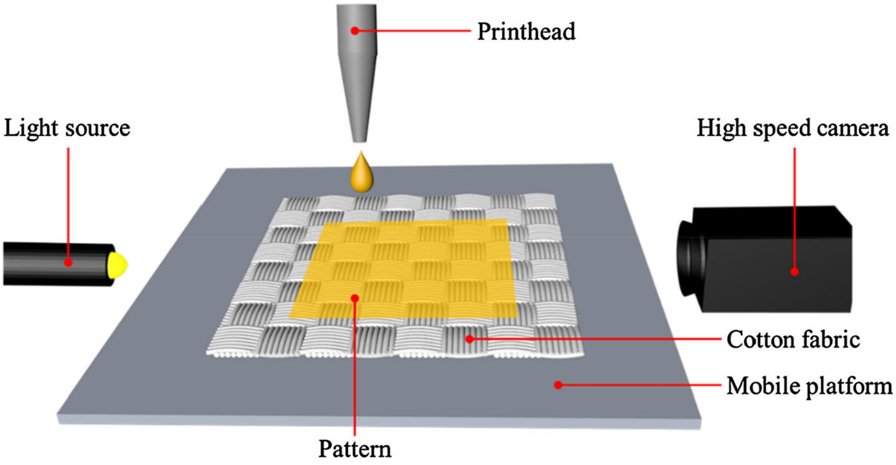


Fig. 1 Schematic graph of the inkjet printing system

Observation of the depositing morphologies

The depositing morphologies and the coalescence structures of ink drops on the cotton fabric were observed by a DVM6M Microscope (Leica Microsystem Co, Ltd., German). The printed patterns were scanned by a Kyocera scanner (Japan).

Results and discussion

Spreading of the ink drop on the cotton fabric

The impacting process of an ink drop on the cotton fabric is shown in Fig. 2a. The moment when the ink drop was about to hit the cotton fabric was assumed as the 0 s. The drop began to impact the cotton fabric at 0.01 ms and then deformed into a cap-like shape at 0.02 ms. Immediately, this cap shaped liquid spread rapidly from 0.03 to 0.05 ms and then there was no obvious variation from 0.06 to 0.08 ms. The liquid disappeared from the cotton fabric surface at 0.3 ms, which might be resulted from the penetration of the liquid on the gap between cotton fibers. Obviously, the impacting process of the ink drop on the cotton fabric was a stable depostion, which usually happened when a droplet with a low Weber number hits a hydrophilic substrate. Weber number is the ratio of kinetic energy to surface energy, which can be calculated from the Eq. (3):

$$We = \frac{\rho V^2 D}{\sigma} \tag{3}$$

where We was the Weber number, ρ was the density of the liquid, V was the droplet falling velocity, D was the droplet diameter, and σ was the surface tension of the liquid. Low Weber number means the kinetic energy has little effect on the surface energy, which results in too little increase of the liquid surface area to splash. After the drop landing on the substrate,

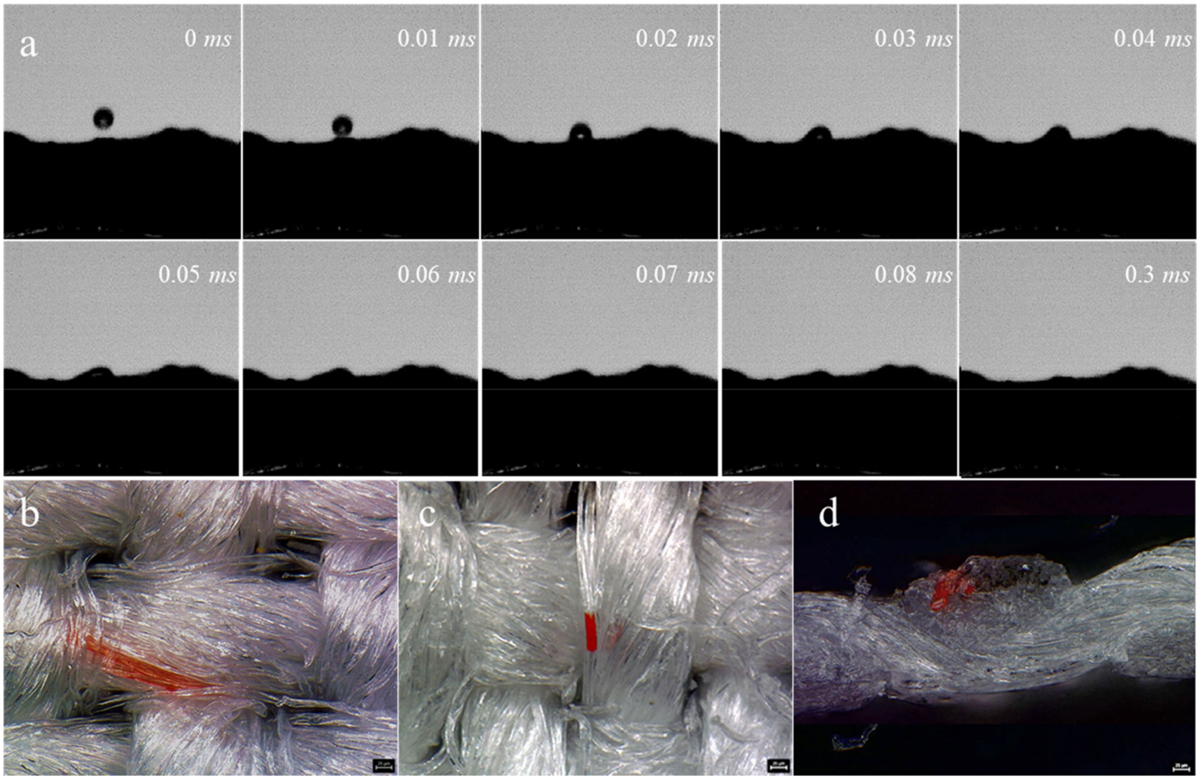


Fig. 2 **a** Impacting process of an ink droplet on the cotton fabric. **b**, **c** depositing morphologies of an ink drop on the cotton fabric surface. **d** Cross section of the cotton fabric printed with an ink drop. The ink drop fell on the cotton fabric surface from the direction above the cotton fabric. In order to maintain the initial structure of the cross section, the cotton fabric was fixed by paraffin before slicing

rebound of the liquid must overcome the adhesion work (W_A), which increases with the increasing of surface energy of the substrate, as shown in Eq. (4):

$$W_A = \sigma_{sg} + \sigma_{gl} - \sigma_{sl} \quad (4)$$

where σ_{sg}, σ_{gl} and σ_{sl} were the surface energy of solid-gas interface, gas-liquid interface and solid-liquid interface, respectively. The higher the surface energy of a solid is, the more easily it is wetted. The hydrophilic surface, therefore, generally means high surface energy (Liu and Lu 2006). That is why the rebound of a drop is more difficult on the hydrophilic substrate. In this experiment, the *We* was 2.4, which was a very low Weber number as a *We* below 30 is considered as a low Weber number (Bordbar et al. 2018). The contact angle of cotton fabric was 73° has been reported (Simončič et al. 2008). However, in present work, the contact angle of water on the cotton fiber was measured as 90°. That may be caused by the different pretreatment process and the contact angle hysteresis. As the low *We* number of the ink drop and the hydrophilicity of the cotton fabric, the impacting process of the ink drop on the cotton fabric should be a stable deposition.

Depositing morphologies of an ink drop on the cotton fabric were observed to assist to understand the wetting process of an ink drop on the cotton fabric. In this experiment, the diameter of the ink drop was 30 μm and the diameters of the cotton fibers were range from 8.03 to 14.9 μm, as shown in Fig. S4. Thus, an ink drop should fall on two to three cotton fibers. Figure 2b shows that an ink drop deposited on three cotton fibers and formed a long strip pattern. It can be seen the length of dropped dye solution along the cotton fibers is far longer than the length perpendicular to the cotton fibers. That is caused by the groove-textured surface of the cotton fabric. The surface of cotton fabric is composed of cotton fibers in certain arrangement. There are gaps between the cotton fibers. These gaps can be looked as space barriers preventing the ink drop diffusion along the direction perpendicular to cotton fibers (Vaikuntanathan and Sivakumar 2016). However, when the ink drop diffused along the cotton fibers, these gaps can be regarded as capillaries to promote the movement of the ink drop. Hence, the ink drop diffused a far longer distance along the cotton fibers than perpendicular to the cotton fibers.

During weaving process, it can’t be avoided that a few of cotton fibers protrude from the yarn surface. When the ink drop fell on a raised cotton fiber, it diffused a far shorter distance along the cotton fiber than along several cotton fibers, as shown in Fig. 2c, which should be resulted from the different wetting types. As mentioned above, the gaps between cotton fibers can be regarded as capillaries. Hence, the diffusion of the ink drop on several cotton fibers should include the spreading on cotton fiber surface and the penetration along the gaps simultaneously. However, it only spread on the cotton fiber surface, when an ink drop fell on a single cotton fiber. It indicates the ink drop penetrated a far longer distance along cotton fibers than it spread. From dynamics perspective, spreading of an ink drop on a fiber surface was driven by the contact angle, which is related with the interaction between water molecules and the substrate. The schematic diagram of a dye aqueous solution drop spreading on a cotton fiber was shown in Fig. 3a. When the drop deposited on the cotton fiber, the actual contact angle was larger than the equilibrium contact angle. As the water molecules bonded to the hydroxyl group on the fiber through hydrogen bonds, the droplet spread on the fiber surface and the contact angle decreased until it is equal to the equilibrium contact angle. Besides contact angle, the penetration was also driven by the additional pressure generated from curved liquid surface. When the ink drop penetrated on the gaps between cotton fibers, it diffused along the cotton fibers until reaching the equilibrium contact angle and then it continued to move along the fibers under the additional pressure as shown in Fig. 3b. The phenomenon can also be explained from the energy perspective. The areas of gas-liquid interface and solid-liquid interface increased simultaneously as the area of solid-gas interface decreased during the spreading process. When changing in a unit area, the work of ink drop spreading (W_s) is shown in Eq. (5):

$$W_s = \sigma_{sg} - \sigma_{gl} - \sigma_{sl} \quad (5)$$

Unlike spreading, only the area of solid-liquid interface increased with the decreasing of the area of solid-gas interface during drop infiltration. The area of gas-liquid interface was fixed, which was related with

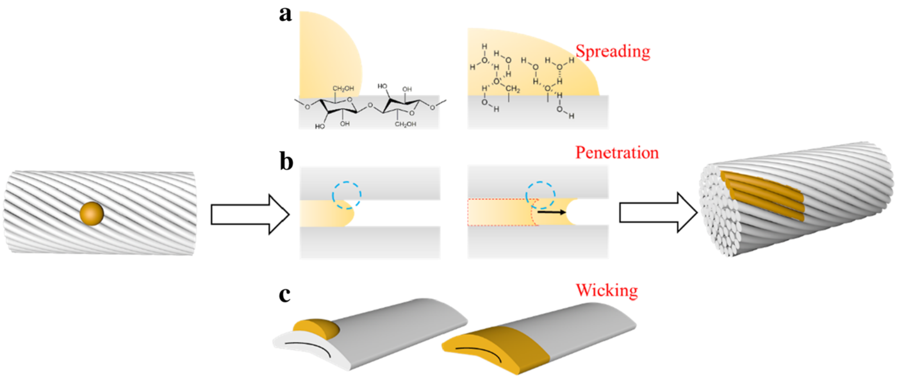


Fig. 3 Schematic diagrams of the dye aqueous solution **a** spreading on the surface of cotton fiber, **b** penetrating between the fibers and **c** wicking into the fiber

the capillary radius. Hence, the work of drop penetration (W_p) is shown in Eq. (6):

$$W_p = \sigma_{sg} - \sigma_{sl} \tag{6}$$

It is obvious that W_p was smaller than W_s, which indicates the spreading of ink drop on the fiber surface consumes more energy than the penetrating along the cotton fibers. Therefore, when the ink drop fell on several cotton fibers, it diffused a longer distance than on a single cotton fiber.

Besides the spreading and penetration, Fig. 2d shows that the ink drop can wick into the interior of the cotton fibers. That is related with the swelling performance of the cotton fiber. Cotton fiber is composed of cellulose macromolecules including crystalline area and amorphous region. The molecules on the amorphous area show poor degree of orientation, low density and weak intermolecular interaction. Therefore, the liquid can easily diffuse into the amorphous region as shown in Fig. 3c. It is difficult to observe the wicking process of an ink drop on the cotton fabric surface directly due to absence of enough lightness. Therefore, in order to understand the time of an ink drop wicking into the cotton fabric, the wetting process of an ink drop on a single cotton fiber was observed as shown in Fig. 4. It can be seen that impacting and spreading of the ink drop on the cotton fiber were rapidly completed in 0.002 s. The shape of the liquid changed little with the time increasing from 0.002 *s* to 0.003 *s*. And then the liquid volume on the fiber decreased gradually with the time increasing from 0.2 to 1.4 s and the liquid disappeared completely at 1.6 s. Consequently, wetting of the ink drop on the cotton fabric should include the rapid spreading and penetration as well as the slow wicking. The depositing morphology of a single ink drop on the cotton fabric was a long strip shape instead of a round point due to the surface topology of the cotton fabric. That means the pattern inkjet printed on the cotton fabric should be composed of "line segments", which is different from the conventional dot imaging.

Coalescence structures with different drop spacings

Coalescing structures of the ink drops with different drop spacings on the cotton fabric were lines made up of some "line segments", as shown in Fig. 5. It can be seen from Fig. 5a when the drop spacing was 10 μm, the line was continuous and occupied a whole cotton yarn. Although the line with a drop spacing of 20 μm or 30 μm was also continuous, it occupied only part of a yarn in the wide direction as shown in Fig. 5b, c. It indicates the ink drops with a 10 μm drop spacing penetrated a longer distance along the cotton fibers than that with a drop spacing of 20 μm or 30 μm. When the drop spacing was larger than 40 μm, a discontinuous line was formed due to the ink drops couldn't coalesce with each other, and the blank parts in the line segment increase with the increasing of the drop spacing, as shown in Fig. 5d–f. It illustrates there should be a critical drop spacing value, which is in range from 30 to 40 μm in this experiment. When the drop spacing was smaller than this value, ink drops can coalesce to form a continuous line. On the

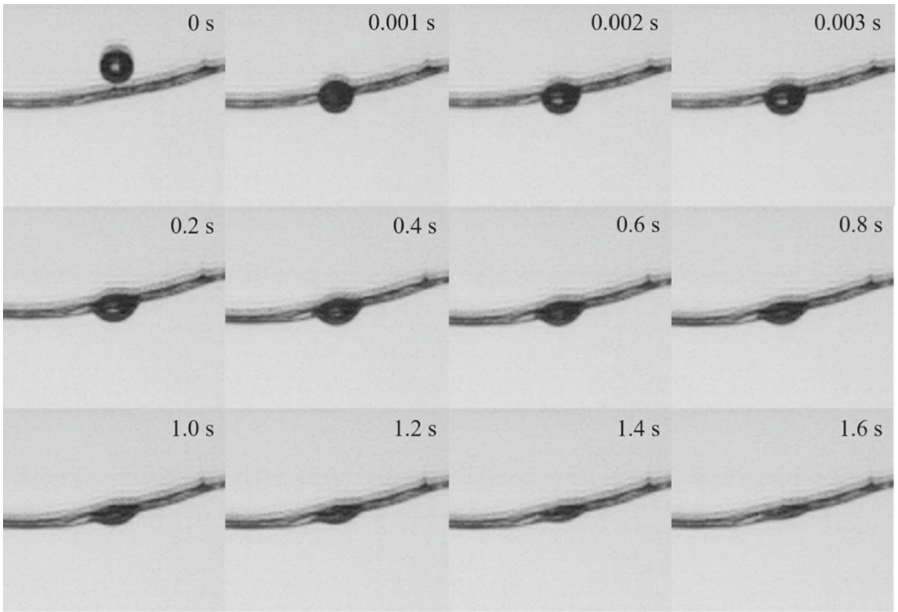


Fig. 4 Wetting process of an ink drop on a cotton fiber

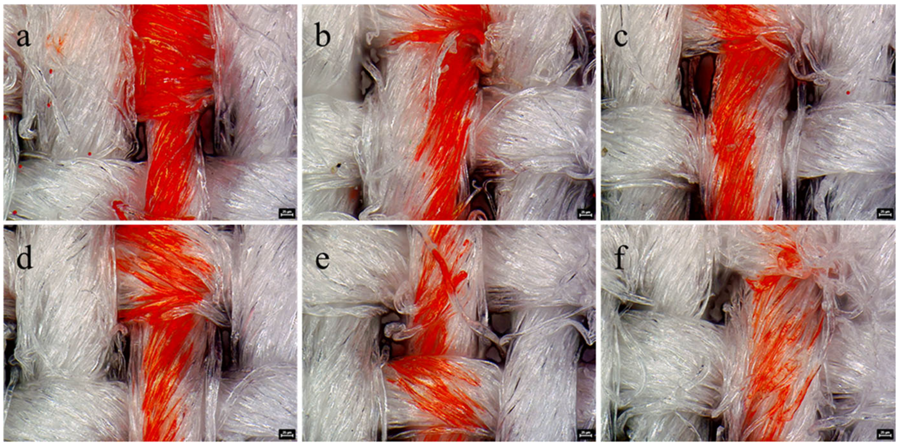


Fig. 5 Depositing morphologies of ink drops with different drop spacings on cotton fabric. The distance between two neighbor drops from a to f was 10 μm, 20μ m, 30 μm, 40 μm, 50 μm and 60 μm, respectively

contrary, the ink drops can't coalesce with others resulting in an intermittent line.

In this work, different drop spacings were accomplished through adjusting the jetting frequency. Corresponding jetting frequency of the line printed with a drop spacing of 10 μm, 20 μm, 30 μm, 40 μm, 50 μm and 60 μm was 1500 Hz, 750 Hz, 500 Hz, 375 Hz, 300 Hz and 250 Hz, respectively. It indicates that when the jetting frequency was 250 Hz, the interval of neighboring two ink drops was the longest, which was 0.004 s. As shown in Fig. 4, an ink drop could reside on a cotton fiber for 1.6 s. Therefore, combining the drop wetting process, the coalescing structures of the drops and the residue time of an ink drop on the cotton fiber surface, the mechanism of ink drop coalescence can be obtained, as shown in Fig. 6. When ink drops coalesced at a small drop spacing, most part of the ink drops overlapped on same gaps between cotton fibers, which resulted in too much dye solution accumulation. In subsequent wetting process, the dye solution would diffuse an excessively long distance along these gaps

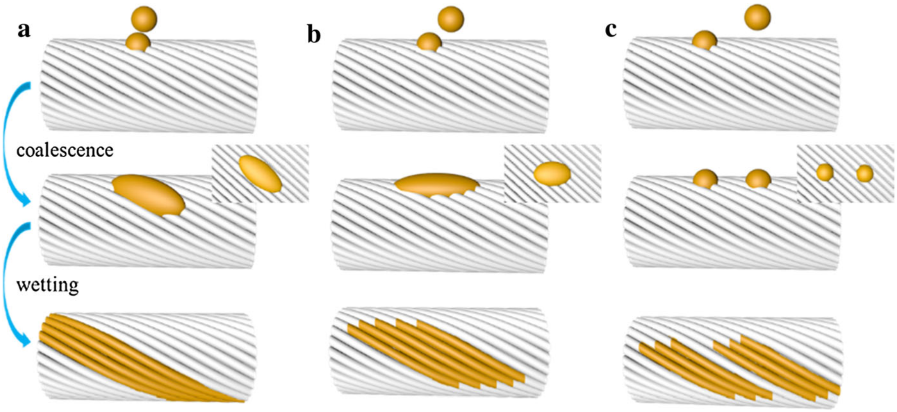


Fig. 6 Schematic diagrams of ink droplets coalescence on a cotton yarn at **a** a small drop spacing, **b** an intermediate drop spacing and **c** a large drop spacing

due to the capillary effect, as shown in Fig. 6a. Figure 6b shows that the ink drops partly coalesced with each other and then formed an ideal coalescence structure after wetting. As the drop spacing increased continuously, the ink drops couldn't coalesce with others resulting in failure to form a continuous line, as shown in Fig. 6c.

Printed patterns

In order to validate the above obtained results, the effect of the drop spreading and coalescence on the quality of printed patterns was explored. Six square shaped patterns with different drop spacings were designed and printed as shown in Fig. 7 and the partially enlarged images can be found in Fig. S5. It can be seen when the drop spacing was in range from 40to 60 μm (Fig. 7d–f), there are many white dots in the patterns due to the ink drops can't coalesce with others. Although ink drop can coalesce with a drop spacing of 30 μm, there are still a few white dots in the pattern as shown in Fig. 7c. As mentioned above, there are a few of raised cotton fibers in the cotton fabric during manufacturing process. When an ink drops fell on a raised fiber, the spreading length and width are shorter than it on several fibers. Hence, these ink drops can't coalesce with other ink drops nearby them resulting in appearance of the white dots. For the patterns, these white spots will affect the aesthetics, and for electronic textiles, these vacancies will affect the performance of the device. Thence, it is necessary to avoid these white dots. When the drop spacing continuously reduced to 20 μm, these white dots can be avoided effectively, as shown in Fig. 7b. However, Fig. 7a shows when the drop spacing was 10 μm, the edges of printed pattern appeared bleeding phenomena, which was harmful to the beauty and performance of printed products and caused waste of expensive materials. The bleeding edges were caused by that the drops coalesced at a small drop spacing causing too long distance of the liquid penetration along cotton fibers. Therefore, in this experiment the best pattern can be printed with a drop spacing of 20 μm.

Conclusion

In conclusion, the ink drop can stably deposit on the cotton fabric and then form a line segment pattern after spreading, penetration and wicking. That indicates the pattern inkjet printed on cotton fabrics is composed of "line segments" instead of round points. Drop spacings affect the coalescing structures greatly. When the drop spacing is large, ink drops can't coalesce with each other resulting in many vacant spots in the printed pattern, which is not allowed to the product. A small drop spacing results in the printed pattern appearing bleeding edges due to much liquid accumulates on the same gaps between cotton fibers. That is harmful to the beauty and performance of printed product and not conducive to saving materials. A continuous pattern with clear edges can be printed at an intermediate drop spacing. Therefore, in order to obtain the best printed pattern, it is of primary importance to adjust the drop

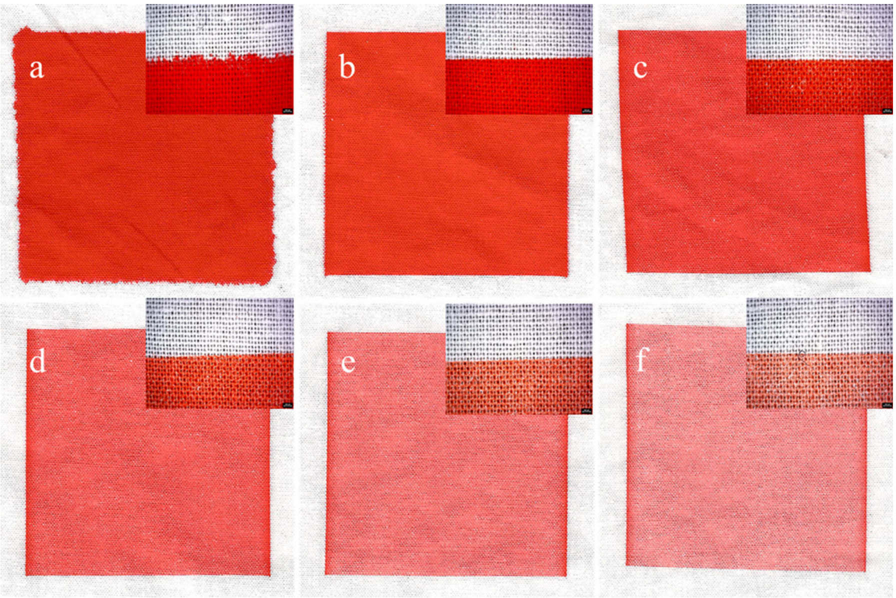


Fig. 7 Printed square patterns on the cotton fabric with different drop center spacings. The magnification of the embedded pictures was 200 times. The drop spacing from a to f was 10 µm, 20 µm, 30 µm, 40 µm, 50 µm, and 60 µm, respectively

spacing according to the spreading and coalescence of ink drops on the cotton fabric.

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