

Low Cost Design Methods to Enhance Resolution and Dimensions for Printed Electrodes

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Abstract—In the paper, we demonstrate the feasibility of interdigital electrodes fabrication with the usage of inkjet printing technology. The emphasis was put to obtain better shape quality and lower spacing between electrodes with respect to typical printing process. The paper presents an analysis of the main factors that have an influence on the dimension and quality of printed structures and proposes two methods that allow eliminating the main problems. The first proposed method is based on controlling the time between patterning of successive drops. While the second method is based on changing the design methods considering printing orientation. Both methods do not require any additional technological processes or the use of any special surface preparation methods. Finally, the obtained results and conclusions were presented and discussed.

Keywords—technology, inkjet printing, resolution, design method, interdigital electrodes

I. INTRODUCTION

FOR many years, printed technologies have been developed quite dynamically and are used in various fields related to widely understood electronics. One of the most commonly used printing technologies is inkjet printing (IJP). Due to the specificity of such technology, it is difficult to perceive it as a rival to the well-known and mature silicon technology that has been used continuously for many years in manufacturing of electronic devices. Silicon technology is still unrivaled, especially when device miniaturization is the most important feature. However, the printing technology enables efficient and inexpensive testing, as well as the production of various types of devices that do not require significant miniaturization. Nevertheless, it is expected to produce devices that are easily integrated with the final product, flexible, transparent and many other extraordinary features. In printed technology, various types of elements are made, ranging from simple passive structures, such as conductive traces, passive electronic components (resistors, capacitors, coils), antennas, sensors components and sensors, (e.g. electrodes), etc. It is also possible to fabricate more complex devices that require sequential execution of appropriate processes and the application of materials with different physical, chemical, or electrical properties. In this way, could be created, for example, active elements such as diodes or transistors. But regardless of the final functionality of the printed devices, each of them are made of independent simple geometric shapes, and the obtained

functional parameters largely depend on the precision of the printed figures.

In the printing process, different requirements and functional criteria are set for the individual components of the final device. Typical parameters that characterize these structures are: shape precision, a spacing between adjacent objects, conductivity, adhesion, morphological structure and mechanical resistance.

For example, contact pads are the least demanding structures for shape definition. As a rule, these are areas with a fairly large surface area, requiring relatively high mechanical resistance to abrasion. From a technological point of view, the challenge is to choose the right materials (ink and substrate) rather than the parameters of a printer.

The other simple elements are the lead traces. Typically, this are structures of medium width, at least a few hundred of μm , but usually of considerable length. To increase the packaging density of the elements on the substrate, a small distance between traces is also required. In this case, the emphasis is put on the proper behaviour of the ink on the substrate, so that the ink does not reflow unpredictably and made traces without accidental electrical shorts.

The third type of typically printed item is electrode-like shapes. Most often is an interdigitated comb configuration. They are used both in various types of sensors [1]–[6] and in active elements (like transistors) to create the source and drain electrodes [7]–[9]. In this case, the key parameters are both the width of the electrodes and the spacing between them. These parameters have a significant impact on the sensitivity of sensors or transistors electrical performance. In a typical transistor structure, the spacing between the electrodes is considered as the length of the channel, and the length of the electrode is related to the width of the channel. Such dimensions are crucial from point of view of the functionality of transistors and devices made of them. The drain current is proportional to the width and inversely proportional to the length of the channel. In the case of sensors, the geometric dimensions of the electrodes affect the sensitivity and measuring range of the sensors [4], [10], [11].

The paper focuses only on the issues related to the geometric dimensions of printed electrodes, their precision and the parameters of printing processes, which are responsible for improving the line resolution and the edge definition of printed shapes.

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As mentioned above, one of the major challenges of IJP technology is to achieve the smallest possible critical dimensions. In the case of transistors, this has a direct impact on key parameters such as threshold voltage, max drain current and operating frequency. Features achieved directly by inkjet printing in additive mode are often characterized by poor resolution. Typically, it is tens of micrometers. Such a low resolution is the main disadvantage of the IJP, which makes it difficult to use from that point of view in practical and advanced applications.

One of the methods for improving the physical resolution of the printouts is the use of printers with higher precision of the cartridge positioning during printing or the use of cartridges which can fire out finer ink droplets. This can be achieved by generating droplets of smaller volumes. The typical minimum mechanical resolution of printers is around 5 μm . However, it does not mean that the obtained dimension resolution of the figures or the distances between them will be characterized by such a number. Typically, tens or even hundreds of micrometres are common characteristic dimensions of figures. In the case of drops volume, this parameter has a direct impact on the critical dimensions of the printed shapes. However, there are some limitations here and the size of the nozzles cannot be reduced arbitrarily. Nozzles with too small orifices are prone to faster clogging and, additionally, they are not suitable for printing inks containing metal particles. Currently, most of the inks available on the market that are characterized by high conductivity are based on a metal nanoparticles such as silver, gold or copper. Besides, low volume droplets do not guarantee that the size of the printed pixel will have a suitably small diameter. This parameter depends of course, on the volume of the drop, but also on the behaviour of the ink drop during the flight and after settling it on the substrate. Here, the main role is played by the interaction between the ink and the substrate. These issues will be described in more detail in the next section.

To improve the resolution of printouts and the definition of shapes, research laboratories use various tricks and procedures adopted from silicon technologies. However, these procedures are much more expensive, require specialized equipment and cannot be used in large-scale applications.

For example Siringhaus et al., (2000) [12] use a special hydrophilic layer separating both electrodes in order to reduce the source-drain distance in printed transistors. Conducting ink droplets deposited on the top of hydrophilic layer repelling each other, remaining distance around 5 μm and create in this way two separate electrodes. However, that procedure requires photolithography and O_2 plasma etching. Another laboratory used so-called coffee-ring lithography (Zhang et al., 2012) [13]. They use local etching process of a mask layer using IJP by applying special solvent on selected places on the mask. Then spin-coating was used to deposit a conductive material into obtained concavity. Another method to gain a micro or even sub-micrometer gaps between adjacent electrodes is self-aligning techniques [14]–[16]. Here, the first printed conductive electrode (typically golden) is functionalized with a self-assembled monolayer. It makes this layer repulsive to the next printed second electrode. As a consequence, the ink droplets flow off the first conductive electrode and dry in close distance to the first electrode. All aforementioned methods offer spectacular effects, however, they need specialised facilities (aside inkjet printers) and they are not rather compatible with printing processes.

II. PRINTOUTS RESOLUTION CONTROLS

One of the first steps to be taken when printing a new ink is testing the action of a single drop of this ink on a selected substrate. It is important because the final resolution of the printout is determined by the properties of the used ink and the substrate, but also by mechanical parameters of the printer. Of course, the user has no influence on the printer parameters, however, appropriate procedures and parameters of the printing process can be easily applied and control.

Fig. 1 shows a photo of a test printout with a matrix of single drops applied on a substrate. The measurement results of droplet sizes and their form and general shape can be used to determine pre-define print resolution.

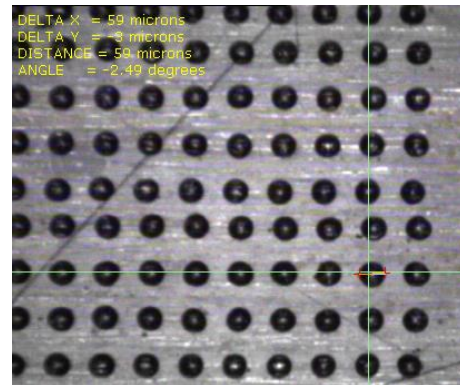


Fig. 1. Test matrix of single ink drops on a selected substrate

Unfortunately, the information about the size of individual spots is not clear unequivocal, as the printed elements are almost never single drop. Typically, the printed shape consists of many spots overlapping each other. And in this arrangement, the ink behaves differently. Various properties of the substrates and inks make it necessary to check individually by experiment the behaviour of each ink on a given substrate. Many parameters to be examined makes it impossible to provide universal guidelines ensuring good print quality for any ink and/or any substrate. Some of the main parameters are:

- physical properties of a substrate,
- rheological properties of an ink,
- method of preparing a substrate (e.g. type of cleaning),
- printout orientation,
- a substrate and ink temperatures,
- frequency of the droplet generation (i.e. a time resolution between two drops jet)
- geometry print resolution.

To control the definition of a printed shape, it is necessary to recognize and understand the physical and chemical principles that determine the behaviour of the ink on the substrate. As mentioned above, the IJP pattern formed on the substrate is made up of individual droplets fired out from the nozzles of the printhead that are settled on the substrate surface and stick to each other. To create a continuous line pattern, the distances between the spots must be smaller than its diameter. Typical IJP printers allow print resolution settings from 5 μm to hundreds of μm .

A single drop of ink applied to a substrate forms a segment of the spherical cap on it. The spot size is dependent on the fluid to substrate interaction. On the sphere formed at the surface acts

forces of surface tension. They are divided into three components: solid-air, solid-liquid and liquid-air. These components are, respectively, two surface tensions σ_{SA} , σ_{SL} and interfacial tension σ_{LA} . The angle formed by the plane surface of the flat substrate and the tangent plane of the ink surface is referred to as the "contact angle" θ . Fig. 2 shows described phenomena. The observed contact angle is a measure of the wettability of the substrate by the liquid applied to the surface. It strongly determines how the drop spreads off on the substrate and, consequently, what the precision of the printed pattern may be. The dependence of the contact angle θ on the individual surface tensions is described by the Young's equation (1) [17].

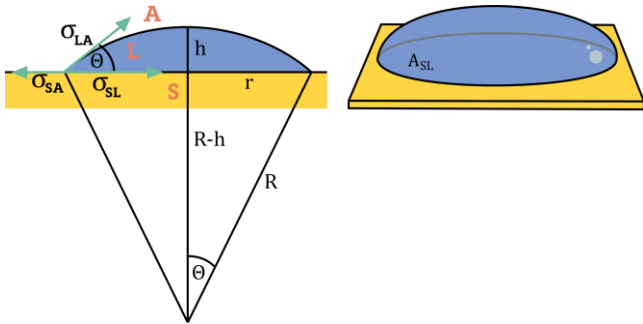


Fig. 2. Relationship of surface tensions with the contact angle and dimensions of the spherical cap on the substrate. Where: L - liquid, A - air, S - solid

$$\cos\theta = \frac{\sigma_{SG} - \sigma_{SL}}{\sigma_{LG}} \quad (1)$$

$$r = R \sin\theta \quad (2)$$

where:

$$R = V^{\frac{1}{3}} \left(\frac{\pi}{3} (2 - 3 \cos\theta + \cos^3\theta) \right)^{-\frac{1}{3}} \quad (3)$$

$$A_{SL} = \pi V^{\frac{2}{3}} \left(\frac{\pi}{3} (2 - 3 \cos\theta + \cos^3\theta) \right)^{-\frac{2}{3}} \sin^2\theta \quad (4)$$

The principles that describe the behaviour of a single drop seems to be quite simple. The size of the spherical cap depends on the volume V of the drop and the contact angle determined by the surface tension of the ink and substrate. During printing, the volume of the drops is roughly constant and in the case of the Dimatix printer can be 1 or 10 pL which is determined by used cartridge. The radius and surface area of a spherical cap with a known contact angle and a drop volume can be found using equations 2 and 4 [18]. In fact, the equations represent only a simplified model. In practice, during printing, the surface tension of both the ink and the substrate varies as a function of temperature. For this reason, the temperature of the printer cartridge and the substrate must be controlled for the best print quality. Besides, the surface tension of the substrate also depends on physicochemical properties of the surface and its morphological structure (e.g. roughness). These are the factors that change the geometry of the spherical cap formed by a single ink drop. Therefore, the right surface preparation is another important aspect of the printing process that should be analysed and considered. It can be achieved by applying the appropriate cleaning agents, methods of their application, or applying especial intermediate layers. The above factors introduce variation in the dimensions of the ink spherical cap and cause hysteresis of the contact angle. This phenomenon manifests

itself in different dimensions of the spherical cap formed by the same ink volume, on the same substrate and under the same conditions. In fact, the hysteresis describes the allowed range of contact angles that an ink drop can form on a particular substrate. The minimum contact angle is called the receding contact angle and the maximum contact angle is the advancing contact angle.

The presence of both angles and the difference between them is one of the problems of maintaining the appropriate precision of printouts. The angles can be determined by increasing or decreasing the volume of the liquid drop respectively until its diameter becomes larger and smaller.

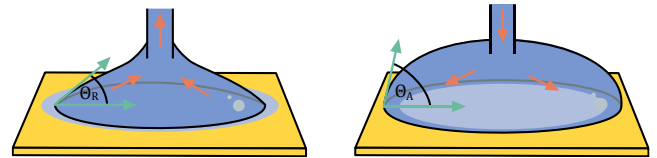


Fig. 3. Scheme for measuring the receding and advancing angle

The angle at which the spherical cap created by the ink increases in diameter is the receding angle θ_R , while the angle at which a droplet decreases in diameter is the advancing angle θ_A . Fig. 3 shows the measurement procedure how the both angles are determine.

In the case of the IJP, the contact angle of the spherical cap is usually closer to the receding contact angle. It is because the drop is fired out onto the substrate at a high speed. The kinetic force causes the spot to occupy a larger diameter in the initial phase and then, it reduces to the final diameter [19]. This effect has consequences when combining more drops, which occurs during printing traces. When combining multiple droplets with a contact angle close to the receding contact angle, they tend to form a larger spot. As a result, the ink located on the edges of the just printed spot surface is sucked into the centre of the previous printed drop [20]. In the case where the hysteresis of the contact angle is small, ink spreading across the shape may also occur due to the increase in the volume of the spherical cap.

These properties are particularly evident when printing very narrow lines. This is due to a significant disproportion between the dimensions of the printed pattern. The length is much greater than the width, which can cause periodic gaps or line widening. The mechanism of line widening, and in the extreme case the local discontinuities, is shown in the diagram in Fig. 4.

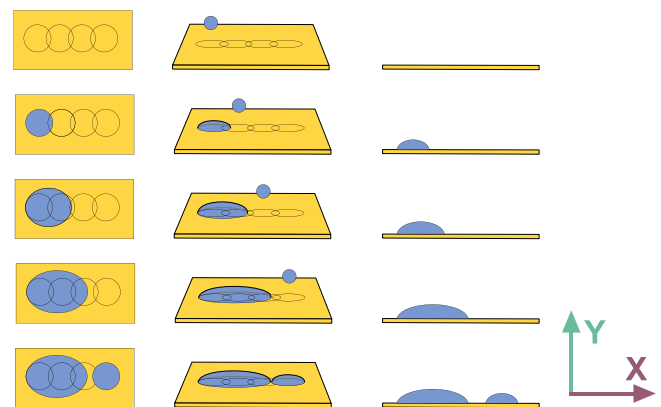


Fig. 4. Diagram of the formation of excessive spots and discontinuities in printed narrow traces

The ink from successively printed drops is drawn into drops already deposited, so they create large asymmetric spherical cap. Figure 5 shows an example of this effect observed with Sicrys I30EG-1 ink and Kapton substrate. In most cases, a well-chosen resolution of the printout allows to eliminate ink spreading around, but an appropriate value of the hysteresis of the contact angle is necessary. In case when the resolution is too high, the intermolecular interaction forces of the ink are not compensated by the adhesion forces of the ink to the substrate. Therefore, during printing progress, the energy value of the imbalance states increases until it exceeds the energy value needed to increase the area occupied by the ink. To prevent the effects of excessive widening of the traces, the print resolution should be selected in a way that the amount of ink per printed area should be as low as possible [21].

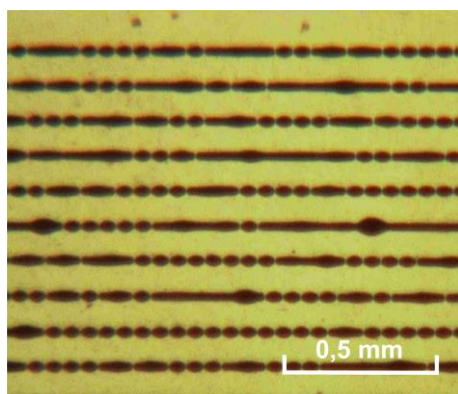


Fig. 5. Periodic breaks and widenings of printed traces

As outlined in the introduction, one of the main challenges for research laboratories is to reduce the width of the traces and the distance between them. If the optimal print parameters are selected, it is possible to obtain a continuous line consisting of a single row of spots. However, its width is usually greater than the diameter of a single spot. For Sicrys I30EG-1 ink, the size of a single spot on the Kapton substrate is approximately 30 μm . However, it is extremely difficult to obtain a continuous and uniform line of these dimensions. The width of the trace created from single drops for the aforementioned materials is inconstant and varies from 30 up to even 100 μm , which can be seen in Fig. 5. The coefficient of increasing the dimensions of the printed shapes in relation to the spot diameter is difficult to predict and depends on not only the selected ink and substrate, but also on environmental conditions. This makes difficult to control the spacing between adjacent parallel lines. And thus, avoiding unwanted accidental short circuits between neighbouring lines.

III. THE METHODS OF IMPROVING THE DEFINITIONS AND DIMENSIONS OF TRACES

Printing with Sicrys I30EG-1 ink is challenging not only on the Kapton substrate. The widening of the traces, despite the selection of the optimal print resolution and appropriate preparation of the substrate, also appears when printing on other polymer substrates like pure PET or PEN, but also as well as on other substrates covered by polymer dielectric film of SU-8. On these materials, undesirable effects of a trace widening are also caused by the same phenomena. This means that in the case of the ink and test substrates, the maximum contact angle is

inappropriate and uniform trace cannot be obtained. One of the simpler methods of changing the wettability (contact angle) is regulating the temperature of the substrate or by specific chemical cleaning. However, this does not always give the expected results. Temperature control normally allows for a small adjustment, while cleaning, and in particular plasma cleaning, usually gives too strong and uncontrolled effect. After plasma treatment, the contact angle can be so small that the ink freely flows over the surface and begins to create extremely wide traces with completely irregular shapes. Fig. 6 shows an example of a photo of a trace printed by Sicrys I30EG-1 ink on PEN substrate treated with oxygen plasma. The photo shows that the trace, although it consists of only two drops of ink in width, has a considerable size and an irregular trace definition. For this reason, plasma cleaning works well when there is a need for printing shapes with large surfaces such as contact pads, large area dielectric layers [22], or when using inks with a very high contact angle.

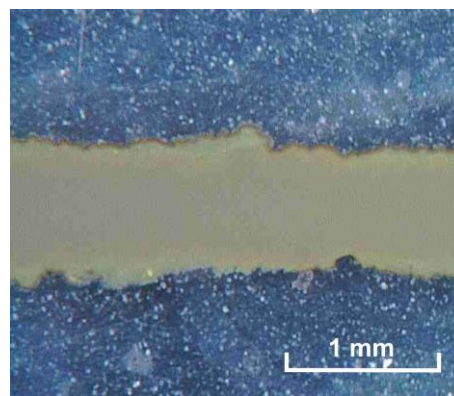


Fig. 6. A piece of trace of two-pixel width, printed with a resolution of 25 μm by Sicrys I30EG-1 ink on PEN substrate after oxygen plasma treatment

Adjusting the contact angle with the temperature of the substrate also causes faster evaporation of solvents, which may result in the poor connection of successive drops. However, it prevents the landing drop from being sucked in by the previously applied ink. An effective way to eliminate the effect of drawing the ink into single spots may be as fast as possible evaporation of the solvents contained in the ink. Solvent evaporation can be easily achieved by reducing the printing frequency, and thus increasing the time between deposition successive drops. The evaporation time of solvents from the inks can be very different, so the adjustment of the printing frequency does not always give satisfactory results.

Functional limitations of printers often do not allow to control of print parameters in a sufficiently wide range. For example, the Dimatix printer DMP 2831 allows to set the minimum printing frequency to 1 kHz, and the maximum substrate temperature up to 60°C. Therefore, in the paper we propose two methods that allow for an additional extension of the time between the application of successive drops. This ensures control of the reflowing of the ink on the substrate, and thus the final improvement of the resolution of the prints. The first option is to change the orientation of the print direction, and the second option is to divide the printout into appropriately selected stages and print them at the required time intervals.

In typical IJP printers, the substrate is attached to a worktable that can move on one direction only, for example on the Y-axis

referring to the print plane. During printing, the cartridge moves in the direction of the X-axis, i.e. orthogonally to the movement of the table. For the mechanical reasons, the printing process is much faster on the X-axis than on the Y-axis. It is due to the fact, that once the printing of a pattern fragment along the X-axis is finished, the cartridge must return to its initial position and the substrate must be shifted by a certain distance along the Y-axis. Additionally, this cycle quite often also includes a cleaning process of the printer head.

A typical approach in printing long and narrow shapes (such as electrodes) is to orientate them in the project in such a way that they are printed when the cartridge move along the X-axis (Fig. 7a). This is a very time-effective method, however, it often leads to the previously discussed problems with an uncontrolled flow of ink.

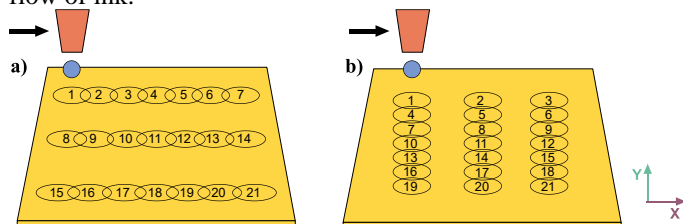


Fig. 7 Diagram illustrating the printing process of electrodes in a) horizontal and b) vertical orientation.

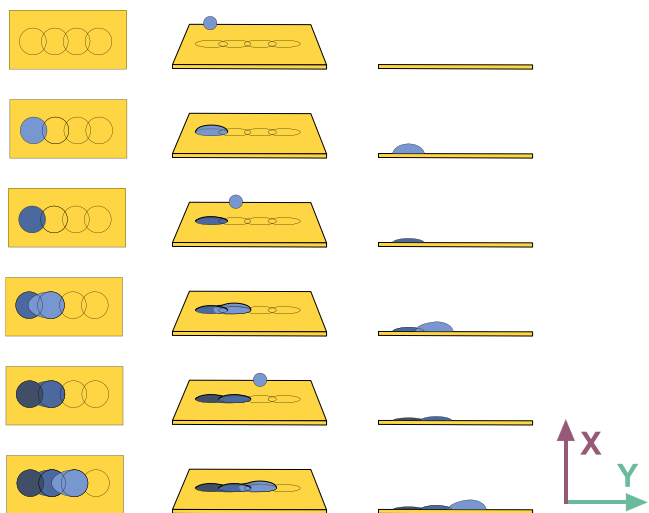


Fig. 8. Diagram illustrating the process of printing traces when plotting in vertical orientation. The coordinate system is rotated by 90° to make the graph more readable

Changing the orientation of the printout allows for a significant delay in printing subsequent drops. When electrodes are printed in the vertical orientation (perpendicular to the movement of the printer head), successive drops that form single electrode are printed with a much longer time interval (Fig. 7b). In this case, this time is usually not less than 1 s.

Vertical electrode printout works well with inks of moderate evaporation time. The process of printing electrodes using this method is shown in Fig. 8. Unfortunately, quite often popular inks have a very long evaporation time. In that case, the method of changing the print orientation to vertical may not be enough effective. To manage this situation, the usage of the second method of dividing the printout into smaller stages is strongly recommended. A diagram illustrating the idea of this printing concept is shown in Fig. 9. It shows a situation where the

electrode pattern has been divided into two sections consisting of even and odd pixels. In this case, all sections can be printed individually in sequential processes. The time interval between all stages can be adjusted and may include processes such as preheating or drying the ink. In this method, free spaces in the pattern are successively filled in and new ink drops are deposited adjacent to the fully or partially cured spots. This method is very effective and gives positive results even with very troublesome substrates and inks. However, its main drawback is the requirement of high precision in positioning and repeatability of printing. Thanks to the application of the method, it is possible to achieve the smaller gaps between adjacent traces and to precisely control the dimensions of the printed patterns.

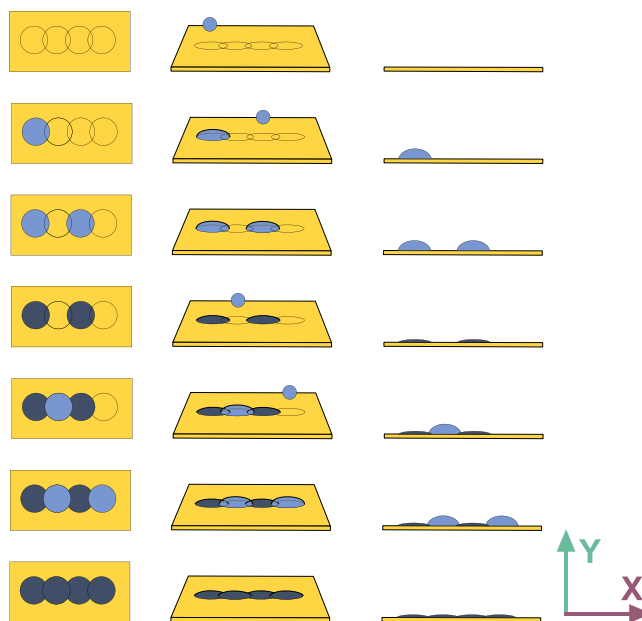


Fig. 9. Diagram illustrating a printing method that divides the printing process into smaller separated stages

IV. EXPERIMENTAL RESULTS.

Both methods: the one with the change of the print orientation and the other one implementing division into sections were tested using Dimatix DMP 2831 printer. A Kapton foil of $125\ \mu\text{m}$ thickness was used as the substrate. Silver nanoparticle conductive ink Sicrys I30EG-1 was used as tested ink and a cartridge with a nominal drop volume of 10 pl. The substrate temperature was set to 60°C to accelerate the solvent evaporation process. The cartridges utilized to the printer enable printing from 16 nozzles simultaneously. However, during printing precise and thin traces only one nozzle should be used. It reduces probability of accidental nozzles clogging and imprecise setting of nozzles, which may lead to discontinuities in the printed pattern.

Prior to printing the test pattern, the size of a single drop was measured on the substrate. The mean diameter of drops was about $30\ \mu\text{m}$. The reference printout made in the horizontal orientation is shown in Fig. 10a. The pattern was printed with the resolution of $25\ \mu\text{m}$, and the 4-pixel gap was used between traces ($100\ \mu\text{m}$). The obtained traces width was from 30 to $100\ \mu\text{m}$. In case of horizontal printing (X-axis) traces have multiple ink spreads. Due to ink spreading, the spacing between

electrodes must be many times larger compared to the case without this effect. Furthermore, the spreads appear irregular and vary in size. For comparison, Fig. 10b presents a printout of the electrodes made in the vertical orientation (Y-axis). Here, the gaps between the electrodes are much smaller, and the electrodes are from 30 to 40 μm wide.

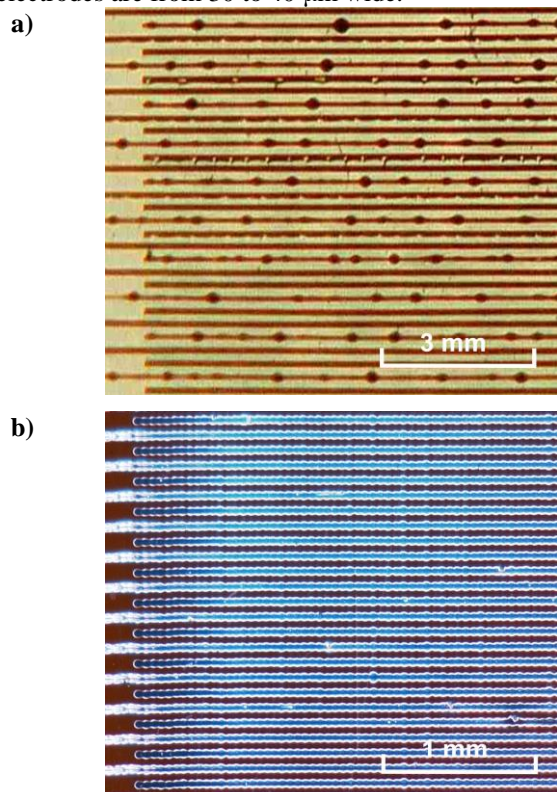


Fig. 10. Interdigitated electrodes on Kapton substrate printed using Sicrys I30EG-1 ink with two different printing orientation: a) horizontal, b) vertical. On traces printed in horizontal orientation local spreading of the ink due to the ink migration occurred. (Different colours of the traces are caused by another backlight type of the microscope)

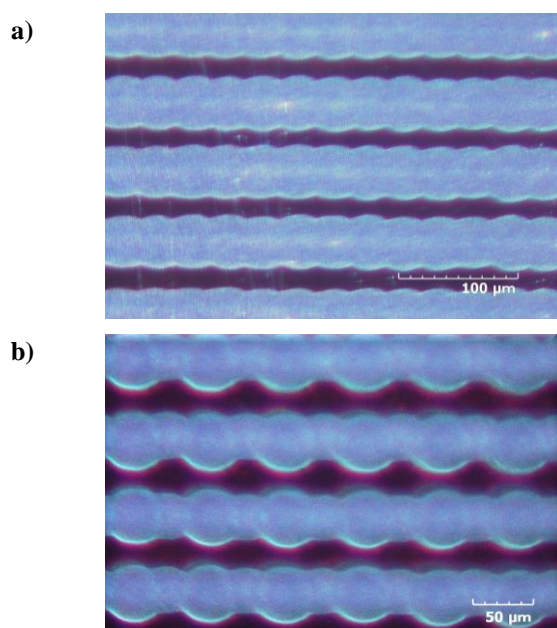


Fig. 11. One-pixel silver traces printed with Sicrys I30EG-1 ink on Kapton substrate. Print was made by two methods: a) vertical orientation print b) horizontal print with splitting the printout into two stages. In both cases, the printhead was shifting horizontally

The shape of the traces made with both proposed methods is shown in Fig. 11. After applying proposed printing methods, the ink spreads are almost unnoticeable. Due to that fact, it is possible to achieve gaps between traces of less than 20 μm . In Fig. 11a we can see local widenings of traces made during vertical printing. These widenings are caused by circular shape of droplets that are being connected and not by ink spreading which was observed with the horizontal traces. The widenings appear in undulating the boundary of the traces. However, the undulating is regular and much slighter than when printing horizontally. Despite the undulations appearing in this method, successive drops merge into a continuous trace. In case of printing using second method you can see the outline of individual ink drops in the traces. Fig. 11b shows that the ink from single drops has already dried and the path consists of single drops overlapping each other.

The disadvantage of second method is uncertainty of spot connection between successive spots and the properties of the trace created this way. On the other hand, the enormous advantage is full control over the dimensions of the trace at every print point. Although the vertical printing method provides a better connection between the drops, it is characterized by a significant widening of the traces at their starting point. Such a widening can be observed in Fig. 12. The occurrence of this widening confirms the migration of ink from the printed drops to the previously printed areas. In this case spreading is much weaker than in horizontal traces, but it still occurs.

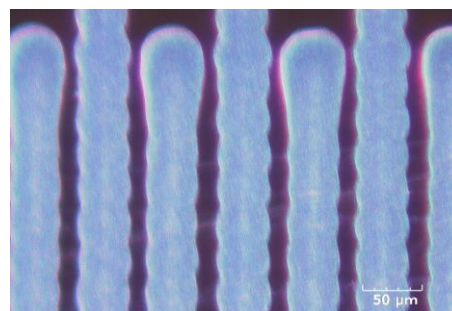


Fig. 12. Widening that occurs at the beginning of thin silver traces. The traces were printed with Sicrys I30EG-1 ink on Kapton substrate

CONCLUSION

This paper describes two methods of counteracting troublesome spreading of the ink and other undesirable defects occurring with printing narrow traces. Both methods are based on partial evaporation of the solvents contained in the printed ink drop before printing the next drop. The mechanisms causing defects when printing traces on mismatched ink-substrate pairs were also discussed and described. Both presented methods effectively prevent unwanted bridges and short circuits from appearing on the printed patterns. As the result, even when using an extremely unfavorable ink-substrate combination, it becomes possible to print precisely conductive traces. What is more, the spacing between traces may be as much as half the diameter of a single spot. A drawback of the methods is its usage mostly for printing narrow structures. When printing at once of different shapes is needed, e.g. electrodes and contact pads, deploying separate processes is recommended.

In the case of interdigitated electrodes used e.g. in sensors and transistors, the conductivity of traces is not crucial, therefore

this topic was not discussed in the paper. However, in each of the proposed printing methods, the volume of the ink drops was the same. So, we can expect that the conductivity of the traces should be comparable. Nevertheless, some differences are possible due to local variations in track thickness and width. Lower conductivity may be also due to a poor drop spot connection in the second method. The effect of these methods on the conductivity of traces will be the subject of future research.

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