Conductivity and Stretchability of Inkjet-Printed Silver Nanoparticle Patterns: Effect of the Number of Printed Layers

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***Abstract*— The development and fabrication of stretchable printed electronics have been thoroughly investigated in several research studies; as they present an attractive solution for low-cost electronics. A special interest has been given to the implementation of these electronics in healthcare applications. This is due to their ability to sustain relatively high strain. Inkjet printing is an emerging technique which is used for the fabrication of printed electronics. Several research efforts have been invested in investigating the effect of varying several inkjet printing parameters on the performance of the fabricated stretchable circuits. In this paper, we particularly investigate the effect of the number of inkjet-printed silver nanoparticle layers on the axial breakdown strain of the stretchable circuits. That is the strain at which the circuit loses its electrical conductivity. Moreover, this study investigates the effect of the shape of the conductive pattern on the correlation between the number of layers and the breakdown strain. Two common shapes are examined: straight-line and horseshoe patterns. Results indicate that increasing the number of layers has an inverse effect on the maximum strain that the stretchable circuit can sustain. The same result is obtained for both investigated patterns.**

***Index Terms*— stretchable circuits, inkjet printing, silver nanoparticles, thin film, silicon rubber.**

I. Introduction

In the last decade, the field of stretchable printed electronics has attracted significant attention from scholars and scientists [1]–[9]. According to [1], [10], the market of printed electronics will be worth $12.1B by 2022 and will rise to $73B by 2027, which highlights the importance of this field. Nowadays, stretchable electronics are implemented in human health monitoring devices; as it is a technology that enhances the wearability of rigid medical devices and reduces their psychological effects on the measured parameters [6], [11]–[17]. Moreover, compared to conventional rigid electronics, wearable sensors conform to the two-dimensional curvature of the human body; hence reducing motion artifacts. Additionally, wearable sensors provide precise, noninvasive, cost effective solutions with the ability of continuous monitoring [1]–[5].

Scholars have extensively investigated the fabrication of stretchable printed electronics using different techniques [10], which are mainly categorized as contact and non-contact methods. Contact techniques such as photolithography [18], [19] and screen printing [10], [20], [21] are the predominant techniques nowadays. On the other hand, non-contact techniques such as inkjet printing are gaining significant interest [1], [10], [22]; due to their advantages compared with the contact methods. These advantages include reduced complexity of fabrication steps, compatibility with various substrates, and reduced material waste [23].

In literature, several studies have investigated the development of stretchable and wearable sensors using printed electronics for biomedical applications [8], [19], [20], [24]–[27]. For instance, Koch and Dietzel [19] developed a stretchable and wearable respiration rate sensor using lithography techniques. Moreover, Chung et al. [27] developed a stretchable and wearable multifunctional sensor for infants for measuring their respiration rate, cardiac signal, and skin temperature. The fabricated sensors were tested for their sustainability at different strains. Each application, and depending on the measurement location, requires a maximum strain which the sensor should sustain [2]. Hence, breakdown strain is one of the most important characteristics of stretchable circuits. It is defined as the maximum amount of strain which the stretchable circuit can sustain before losing its conductivity [2].

Several studies in literature [1], [2], [4], [28]–[33] present different methodologies to increase the stretchability of the printed electronics. For instance, Abu-Khalaf et al. [1] and Kwang et al. [29] presented the use of wavy patterns (horseshoe or sinusoidal) to increase the stretchability of the printed circuits by forming in-plane wavy structures. While other studies [2], [4], [31], [32] presented the development of out-of-plane wavy patterns to increase the stretchability of the printed circuits. Furthermore, Abu-Khalaf et al. [1] and Kwang et

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al. [29] analyzed the effect of several parameters of the printed patterns on the stretchability of the circuits like the pattern line width and length. Commonly, polydimethylsiloxane (PDMS) is the used elastomer for the fabrication of stretchable and wearable sensors; due to its biocompatibility and low cost [18].

Additionally, inkjet printing offers a relatively low-cost and a less-complex technique for the fabrication of stretchable sensors compared to other techniques such as photolithography [2], [18] and screen printing [20], [21], [29]. As aforementioned, inkjet printing presents a contactless method for the deposition of conductive inks for the fabrication of printed electronics. Usually, inkjet printers are categorized based on their deposition mode; that is continuous deposition (continuous fluid stream) or drop on demand mode. In the continuous mode, a stream of conductive ink is continuously deposited on the substrate through a nozzle with huge amounts of waste material. While for inkjet printers with drop on demand mode, the ink is deposited on the substrate only when it is needed. In this mode, the printer usually contains several piezoelectric nozzles that control the deposition electronically which would significantly reduce the amount of wasted material [1], [22].

Inkjet printers have the ability to deposit various types of inks or fluids with specific viscosity [1], [23], [34], [35]. For instance, inkjet printers could print conductive inks with suspended nanoparticles of silver, copper and gold [1], [35]–[38]. Also, the ink could contain metallic (silver, gold, copper) nanowires, metal-organic materials such as silver β-ketocarboxylate or even polymer-based conductive materials such as PEDOT/PSS ink.

Inkjet printing has several printing parameters that affect the conductivity, as well as the overall dimensions of the printed patterns, such as drop spacing (DS), cartridge/nozzle temperature, platen (substrate holder) temperature, sintering temperature, and the number of printed layers [1], [33]. The effect of the latter on the stretchability of inkjet-printed circuits is not extensively investigated in literature. Hence, in this study, the effect of the number of inkjet-printed layers on the stretchability of wearable circuits is analyzed. Also, the effect of drop spacing on the resistance and the width of straight-line patterns is inspected. Additionally, straight-line and horseshoe conductive patterns are inkjet-printed on PDMS substrates and the resulting circuits are examined.

II. METHODOLOGY

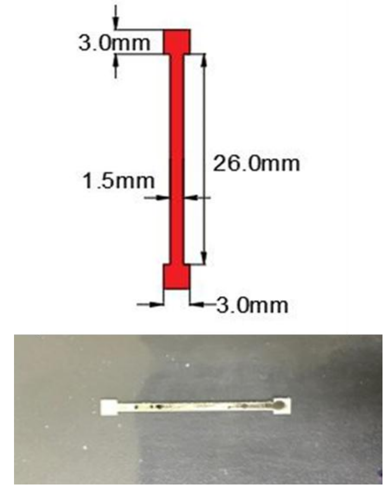
In this study, PDMS substrates are prepared using rectangular acrylic molds. Specifically, 4 ml of liquid PDMS mixture are poured in a 40 mm × 100 mm rectangular mold. The PDMS mixture is prepared by mixing the PDMS base with a curing agent from Dow Corning® Sylgard with a volume ratio of 10:1 (base: curing agent). The filled mold is then placed in a vacuum oven at room temperature for gasification for one hour. Next, it is cured for two hours at 70 ^{o} C. The PDMS surface is hydrophobic which hinders the formation of conductive traces. However, the wettability of the PDMS surface can be enhanced using either chemical or physical techniques. In this work, plasma barrel etching technology is used for PDMS surface treatment. In particular, the PDMS sample is treated for 15 minutes at full power (50 Watt) in the ZEPTO Diener plasma etcher (ZEPTO Diener, Germany), where the plasma is generated using O_2 gas.

Subsequently, the conductive patterns are printed by depositing Silver Nanoparticles (Silverjet DGP-40LT-15C from Sigma Aldrich) on 40 mm × 100 mm × 1 mm PDMS substrates after applying the proper surface treatment. Fujifilm Dimatix Material Printer DMP-2831 is used for the deposition of the silver nanoparticles where the printer has 16 printing piezoelectric nozzles and has the ability to operate on Drop on Demand mode to reduce the amount of wasted ink. The desired patterns are fed to the inkjet printer as images with specific resolution depending on the desired drop spacing (DS). The drop spacing is defined as the distance between the centers of the droplets and it is one of the parameters that affect the conductivity of the printed patterns. A 10 pl cartridge print head is used for ink deposition with DS of 30 μm as recommended in literature [1], [2], where the resolution of the pattern's image is found using the correlation in (1).

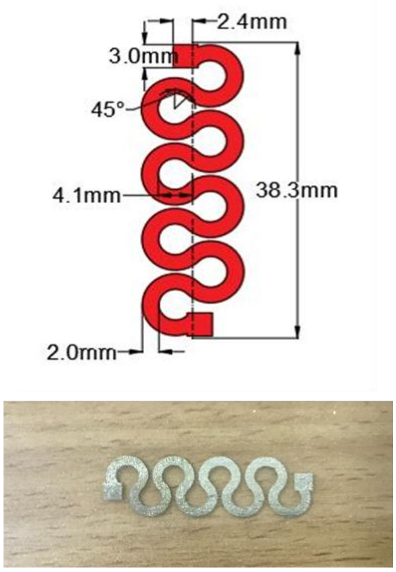
$$Rs = 25400/ DS \quad (1)$$

where Rs is the pattern resolution [dpi] and DS is the drop spacing [μm]. In addition, the used inkjet printer has the ability to print several layers, where each layer has a thickness of 600 nm as reported in [1], [3]. In order to maintain a constant line width while printing several layers, the platen temperature of the inkjet printer is set at 60 ^{o} C. This allows for the sintering of the layers with 30 minutes delay between the layers' deposition [22]. Once the printing process is completed, the fabricated circuit is placed in the oven for sintering at 150 ^{o} C for one hour.

Straight line and horseshoe patterns, shown in Fig. 1, are printed on PDMS substrates with different thicknesses; in order to highlight the effect of the thickness (number of layers) of the printed pattern on the breakdown strain of the circuit. Fig. 2 illustrates the fabrication procedure of the stretchable circuits. The stretchability of the printed circuits is examined under axial strain, where an in-lab automated stretcher, shown in Fig. 3, is used to apply external strain on the substrates. The desired amount of strain is entered to a microcontroller which controls the rotation of a stepper motor resulting in elongating the substrate under examination axially. While stretching the substrate, the electrical resistance is continuously sampled by the microcontroller to be able to determine the breakdown strain at which conductivity is lost. This breakdown strain is used as a measure of the stretchability of the circuit, where a larger breakdown strain indicates enhanced stretchability. It should be noted that the automated stretcher was built based on the design introduced in [39].



(a)



(b)

Figure 1. Inkjet-printed patterns with their dimension: a) straight-line and b) horseshoe.

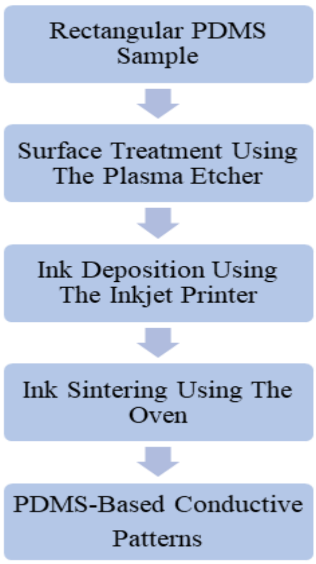


Figure 2. The fabrication process of the stretchable PDMS circuits.

III. Results

A. The Effect of Drop Spacing

Drop spacing affects the resistance of the printed patterns as it represents the distance between the ink droplets. Increasing the distance between the silver nanoparticles reduces line connectivity and accordingly increases the resistance of the printed patterns. Fig. 4 shows the relationship between drop spacing and the resistance of the inkjet-printed straight line.

As can be depicted from Fig. 4, the resistances of the printed pattern with drop spacing between 15 and 30 µm are almost the same with no significant difference. While beyond 30 µm there is a significant increase in the resistance of the patterns. Furthermore, the drop spacing affects the actual width of the printed traces as shown in Fig. 5.

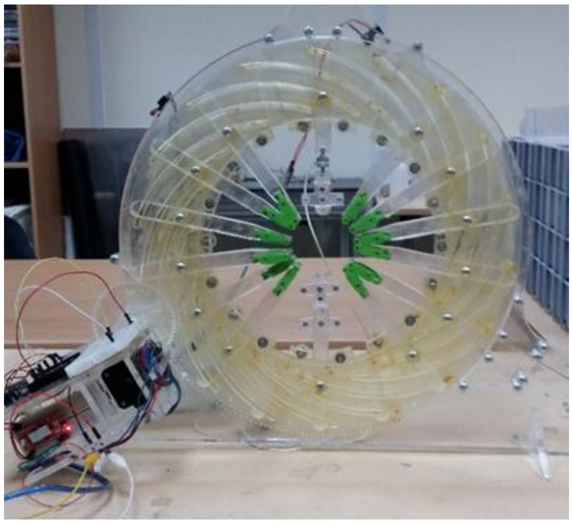


Figure 3. Automated stretcher used to apply axial strain of stretchable circuits.

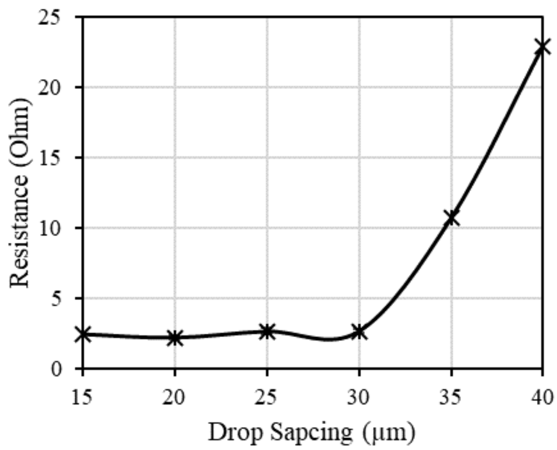


Figure 4. The relationship between the resistance of the inkjet-printed straight-line pattern and drop spacing.

The error curve in Fig. 5 shows how the line width differs, from a desired value of 1.5 mm, as the DS is varied. The obtained results are in agreement with the results reported in literature, which recommend the use of DS of 30 µm. At this specific DS, the width of the printed pattern is the closest to the desired line width at approximately 1.59 mm. On the other hand, increasing the drop spacing is expected to distort the shape of the printed pattern and increases the width of it.

B. The Effect of The Number of Layers

The electrical resistance of the printed conductive pattern is measured at 3 different values of the number of printed layers (1, 3, and 5). The general trend, shown in Fig. 7, establishes that resistance decreases as the number of layers increases; where the resistance has a nearly linear relationship with the number of layers. Increasing the number of layers from 1 to 5 resulted in a decreased resistance by a factor of 5 (from 2.5 ohms to 0.5 ohms). The electrical resistance of a uniform printed line can be computed from (2).

$$R = \frac{\rho l}{A} \tag{2}$$

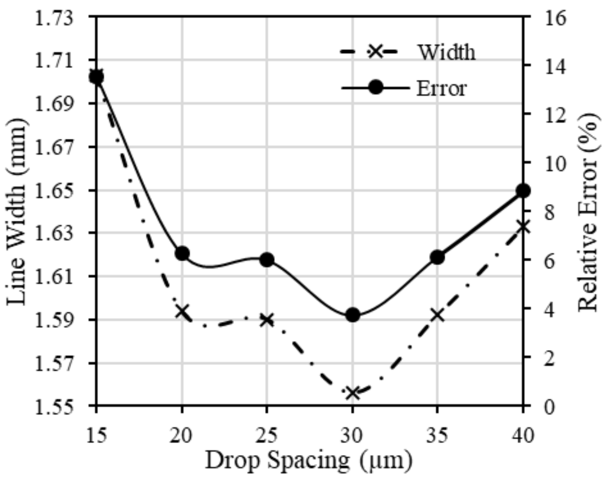


Figure 5. The relationship between drop spacing and the line width of inkjet-printed straight line.

where ρ is the electrical resistivity of the material (silver) [Ohm.m], l is the length of the line [m], and A is the cross-sectional area of the line [m^2]. Printing parameters which affect the cross-sectional area are the drop spacing and the number of printed layers. Specifically, increasing the number of layers increases the thickness of the line which increases the cross-sectional area. An increased area decreases the line's resistance, which agrees with our findings from Fig. 6.

It is also desired to see the effect of the line thickness on the stretchability of the printed patterns. In previous studies including that presented in [1], horseshoe patterns have demonstrated their superior capabilities for stretchable circuits when compared to other shapes (straight lines and sinusoidal patterns). Therefore, we varied the number of layers for the printed horseshoe pattern shown in Fig. 1b and measured the breakdown strain. Fig. 7 illustrates the relationship between the number of inkjet-printed layers and the breakdown strain of the circuit for both a straight line and horseshoe pattern. It can be depicted from Fig. 7 that increasing the number of printed layers decreases the maximum strain which the circuit can sustain regardless of the shape of the pattern.

Fig. 8 shows microscopic images of the inkjet-printed straight-line pattern at various number of layers (1, 4, and 6). It is evident that line cracks increase as the number of layers increases. Accordingly, when the stretchable printed circuits are loaded deformation occurs in the circuit and thicker lines with more initial cracks result in a lower breakdown strain, decreasing the overall stretchability of the circuit. Hence, it is important to find the optimal number of layers to be used in the fabrication of inkjet-printed circuits such that there is a balance between the increased conductivity and increased stretchability.

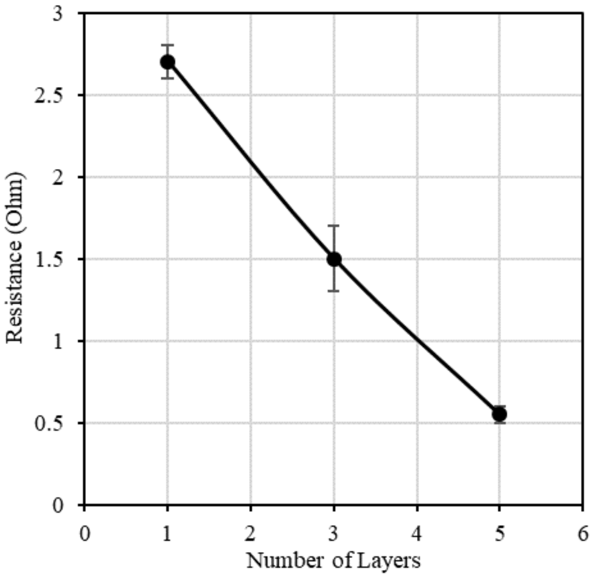


Figure 6. The relationship between the number of layers and the resistance of the inkjet-printed straight-line pattern.

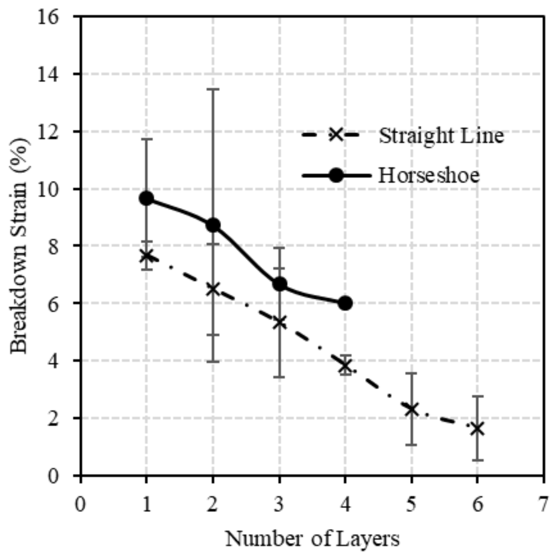
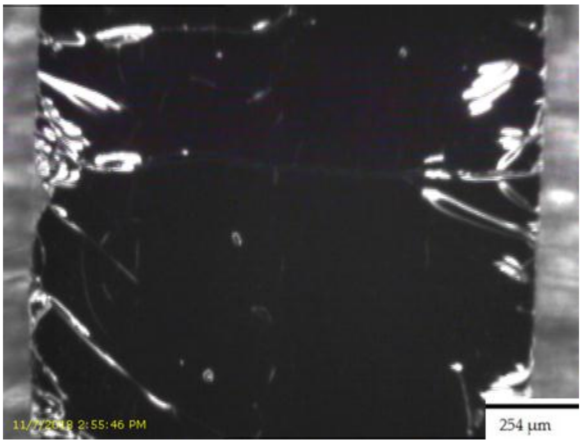


Figure 7. The relationship between the number of layers of the inkjet-printed patterns and their breakdown strain.



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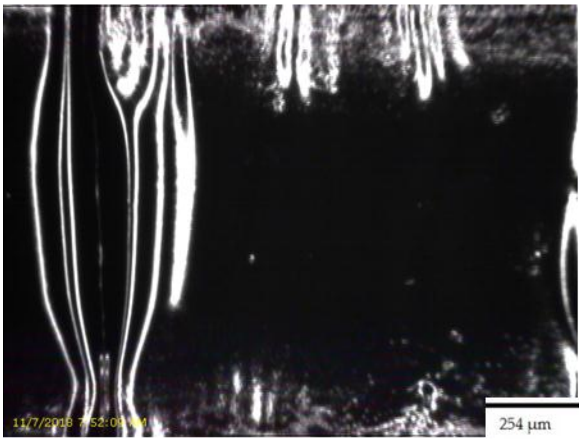

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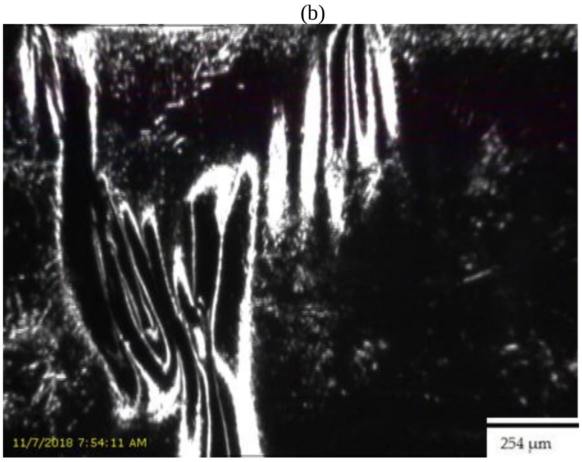

(c)

Figure 8. Microscopic images of the inkjet-printed straight-line patterns at: a) 1 layer, b) 4 layers and c) 6 layers.

IV. Conclusions

In this paper, the conductivity and stretchability of inkjet-printed silver conductive lines on PDMS are examined. In particular, the effect of the number of printed layers on the resistance of the lines is measured. Results indicate that increasing the number of layers decreases the resistance of the lines which is desirable. This agrees with the definition of the electrical resistance of a conductor where increasing the thickness of the conductor results in a smaller resistance.

To measure the stretchability of the printed circuit, each circuit is axially stretched until conductivity is lost and the strain value at that point, which is known as the breakdown strain, is recorded. Straight-line and horseshoe patterns were examined showing that increasing the number of printed layers decreases the maximum strain which the circuit can sustain regardless of the shape of the pattern. This is due to increased crack formation in the lines as their thickness increases.

In conclusion, the thickness of the inkjet-printed lines highly affects the conductivity and stretchability of the printed circuits. In the future, the number of printed layers will be optimized to obtain circuits with lower resistance and higher breakdown strain. Furthermore, it is desired to minimize the amount of used ink to sustain cost-effective fabrication which requires further optimization of the printing parameters.

Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

Conceptualization, J.A.-K. and L.A.-G.; methodology, J.A.-K. and L.A.-G.; software, L.A.-G.; validation, L.A.-G.; formal analysis, L.A.-G.; investigation, L.A.-G. and A.N.; resources, J.A.-K. and A.A.-H.; data curation, L.A.-G.; writing—original draft preparation, L.A.-G. and J.A.-K.; writing—review and editing, L.A.-G., J.A.-K. and A.A.-H.; visualization, L.A.-G.; supervision, A.A.-H., J.A.-K. and L.A.-G.; project administration, A.A.-H., J.A.-K. and L.A.-G.; funding acquisition, J.A.-K. and A.A.-H.

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