

# Inkjet printing for the fabrication of flexible/stretchable wearable electronic devices and sensors

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## Abstract

**Purpose** – This review paper aims to introduce the inkjet printing as a tool for fabrication of flexible/wearable sensors. It summarizes inkjet printing techniques including various modes of operation, commonly used substrates and inks, commercially available inkjet printers and variables affecting the printing process. More focus is on the drop-on-demand printing mode, a strongly considered printing technique for patterning conductive lines on flexible and stretchable substrates. As inkjet-printed patterns are influenced by various variables related to its conductivity, resistivity, durability and dimensions of printed patterns, the main printing parameters (e.g. printing multilayers, inks sintering, surface treatment, cartridge specifications and printing process parameters) are reported. The embedded approaches of adding electronic components (e.g. surface-mounted and optoelectronic devices) to the stretchable circuit are also included.

**Design/methodology/approach** – In this paper, inkjet printing techniques for fabrication of flexible/stretchable circuits will be reviewed. Specifically, the various modes of operation, commonly used substrates and inks and variables affecting the printing process will be presented. Next, examples of inkjet-printed electronic devices will be demonstrated. These devices will be compared to their rigid counterpart in terms of ease of implementation and electrical behavior for wearable sensor applications. Finally, a summary of key findings and future research opportunities will be presented.

**Findings** – In conclusion, it is evident that the technology of inkjet printing is becoming a competitor to traditional lithography fabrication techniques, as it has the advantage of being low cost and less complex. In particular, this technique has demonstrated great capabilities in the area of flexible/stretchable electronics and sensors. Various inkjet printing methods have been presented with emphasis on their principle of operation and their commercial availability. In addition, the components of a general inkjet printing process have been discussed in details. Several factors affect the resulting printed patterns in terms of conductivity, resistivity, durability and geometry.

**Originality/value** – The paper focuses on flexible/stretchable optoelectronic devices which could be implemented in stretchable circuits. Furthermore, the importance and challenges related to printing highly conductive and highly stretchable lines, as well as reliable electronic devices, and interfacing them with external circuitry for power transmission, data acquisition and signal conditioning have been highlighted and discussed. Although several fabrication techniques have been recently developed to allow patterning conductive lines on a rubber substrate, the fabrication of fully stretchable wearable sensors remains limited which needs future research in this area for the advancement of wearable sensors.

**Keywords** Sensors, Wearable, Electronic devices, Inkjet printing, Conducting, Stretchable

**Paper type** General review

## 1. Introduction

Wearable sensing is an emerging research area which has evolved significantly in the past few years, principally due to its crucial role in the continuous monitoring of human physiological data. In particular, wearable sensors have been integrated into clothing garments, which allow for monitoring patients for extended periods of time at their homes, keeping record of athletic performance during training, as well as monitoring the vitals of soldiers in combat. This implementation came into existence because to the recent advances in fabrication of miniaturized devices,

electronics, wireless communication, battery technology and material science (Mukhopadhyay, 2014; Martinez-Tabares *et al.*, 2016).

It is desired to enhance the wearability of physiological sensors in a manner which reduces errors due to motion artifacts between the sensor and the human body curvature. This includes measurement of parameters such as body temperature, orientation and performed activity. Hence, a flexible, and in some cases, a stretchable, version of the sensor is required where a circuit must conform to the arbitrary two-dimensional human body curvature.

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Fabrication of stretchable circuits creates an intellectually rich set of challenges; specifically, it entails patterning conductive lines on a stretchable platform, adding electronic components to the resulting stretchable circuit while maintaining electrical conductivity and interfacing the sensor with external circuitry for power transmission, data acquisition and signal conditioning. Although there are several fabrication techniques which have been recently developed to allow patterning of conductive lines on a rubber substrate, fabrication of fully stretchable wearable sensors is still rather limited.

Conventional lithography techniques have demonstrated their capabilities in making thin films on elastomeric substrates which maintain integrity when stretched (Gray *et al.*, 2004; Lacour *et al.*, 2004; Xiao *et al.*, 2008; Cho *et al.*, 2009); however, they require both costly and complicated fabrication steps. In recent years, inkjet printing has become an attractive alternative to be used in several patterning applications. The advantages of this low-cost technique include reduction of complexity of fabrication steps, compatibility with various substrates, reduction of wasted material and having no requirement for mask patterning (Singh *et al.*, 2010; Haghdoost *et al.*, 2015; El-Molla *et al.*, 2016). It is for the aforementioned reasons that inkjet printing can be strongly considered for producing stretchable circuits by patterning conductive lines on a stretchable substrate.

The second challenge which needs to be addressed, in relation to the fabrication of stretchable sensors, is the insertion of electronic components into the stretchable sensor substrate, while maintaining electrical conductivity. In the case of using rigid surface mount devices and for the sensor to be fully stretchable, bonded connections between the stretchable conductive lines and the rigid electronic components need to be avoided. This involves innovatively embedding the rigid components on top of the previously patterned conductive lines. Alternatively, flexible forms of the electronic devices could be printed in a second rubber layer on top of the conductive lines. The electro-mechanical performance of the sensor is used as a measure of the effectiveness of the implemented solution.

In this paper, inkjet printing techniques for fabrication of flexible/stretchable circuits will be reviewed. Specifically, the various modes of operation, commonly used substrates and inks and variables affecting the printing process will be presented. Next, examples of inkjet-printed electronic devices will be demonstrated. These devices will be compared to their rigid counterparts in terms of ease of implementation and electrical behavior for wearable sensor applications. Finally, a summary of key findings and future research prospects will be presented.

## 2. Inkjet printing techniques

Inkjet printing has recently emerged as a new method for printing flexible and wearable sensors. This is due to the wide range of advantages offered when using this method, such as fast printing, cost-effectiveness and low temperature fabrication (Yin *et al.*, 2010). In the case of pressure-sensitive substrates and substrates of rough or curved non-planar surfaces (flexible/stretchable), even of deep trenches (Wu *et al.*, 2015), inkjet printing is possible. It offers noncontact

patterning with minimized contamination and also provides ease of modification through direct user-friendly graphical user interface. Hence, if any modification is needed, it can be changed using a computer-aided design (CAD) software without the need to modify the hardware properties or assembly (Jensen *et al.*, 2011).

Moreover, inkjet printing is made accessible due to the availability of commercial inkjet printers with affordable prices. This method also has the ability to print highly conductive traces and patterns (Kawahara *et al.*, 2013) with excellent compatibility with both organic and inorganic materials, distinguishing it from other printing techniques. It is also implementable in low resource settings where low-cost fabrication is mandatory to overcome the need for costly masks and clean room facilities (Medina-Sánchez *et al.*, 2012), as well as the use of low cost substrates such as polydimethylsiloxane (PDMS), polyethylene terephthalate (PET) and paper. The process of inkjet printing requires an inkjet printer, which could be operated under different modes and actuation methods, as well as inks and substrates which are selected according to the required application.

### 2.1 Inkjet printing modes of operation

There are two modes of operation for inkjet printing: drop-on-demand (DoD) and continuous inkjet printing (CIJ). In most DoD printers, the droplets are thermally generated (thermal-DoD) or induced by a piezoelectric actuator. In CIJ printers, a continuous electro-conductive stream of fluid is delivered through a nozzle because of piezoelectric actuator vibrations, regulating the breakup of the stream into individual uniform droplets with uniform spacing (Castrejon-Pita and Baxter, 2013). In the following subsections, we will compare both modes and identify their dissimilarities.

#### 2.1.1 Drop-on-Demand

In the DoD method, single drops of ink are released onto the substrate in response to a digital signal or waveform. It allows for the control and calibration of the ejection of ink droplets from each nozzle (Basiricò *et al.*, 2011). This method is widely used due to its high placement accuracy, controllability and efficient material usage (Yin *et al.*, 2010), where no excess ink is wasted, as it is only dropped where and when needed.

In the thermal-DoD mode, a thin layer of the liquid, present in the ink chamber, is heated by a thin film heater. This forms vapor bubbles in a few microseconds, which rapidly expand to eject ink droplets through the nozzle. The initial actuation pressure is close to the saturated vapor pressure of the liquid at the superheat limit (about 4 MPa for water and 1 MPa for organic solvents). The minimum printable droplet size of a thermal bubble inkjet device is about 0.5  $\mu\text{m}$  for water and 1  $\mu\text{m}$  for methanol and toluene (Wu *et al.*, 2015). The temperature raises to about 300°C, accordingly clogging of the nozzle is a main issue (Kim *et al.*, 2014). Thermal-DoD uses mostly water as a solvent and may therefore be restricted to specific applications, such as desktop printing. It is also not suitable for printing biological fluids and polymers (Ko *et al.*, 2007).

In the piezoelectric DoD, the waveform of the piezoelectric actuator controls the expelling of smaller droplets from the nozzles, which allows for the printing of both aqueous and UV

curable inks on a wide range of substrates. As a result, this method is frequently implemented in research and in industrial applications. In comparison to thermal DoD, piezoelectric DoD is found to be more adaptable to printing of water-based inks containing conjugated polymers, as it uses lower temperatures. High temperatures cause water to evaporate in the nozzle surface, resulting in nozzle clogging and possible modification of the structure and properties of organic molecules (Basiricò *et al.*, 2011). Instead, piezoelectric inkjet printing relies on the mechanical action of a piezoelectric membrane to generate a pulse (Ko *et al.*, 2007). Nozzle sizes are in the range of 20–30  $\mu\text{m}$ , and droplets sizes are in the range of 10–20 pL (Kim *et al.*, 2014). The diameter of an ink drop on a substrate is about twice the size of the released drop (Yin *et al.*, 2010).

**2.1.1.1 Piezoelectric DoD modes of operation** Piezoelectric DoD has several modes of operation (Cummins and Desmulliez, 2012), such as:

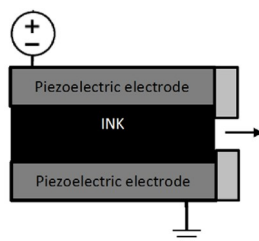
- squeeze-mode print heads;
- bend-mode print heads;
- push-mode piezoelectric print head; and
- shear-mode print head.

Each of the aforementioned modes are discussed below.

**2.1.1.1.1 Squeeze-mode print heads.** In this mode of operation, the print head consists of a hollow tube of piezoelectric material (Zoltan, 1972). The piezoelectric electrodes contract due to the applied voltage, which results in squeezing of the ink chamber and a droplet being forced out of the nozzle, as shown in Figure 1.

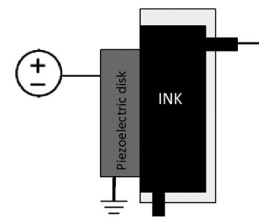
**2.1.1.1.2 Bend-mode print heads.** In this mode, droplet ejection is induced by the bending of the ink chamber wall (Kyser and Sears, 1976). The bend-mode DoD print head (described in Stemme, 1972; Kyser and Sears, 1976) consists of a small rectangular chamber with a supply tube, which provides ink from the ink reservoir, and an exit for the nozzle on the opposite side, as shown in Figure 2. A 0.7-cm-diameter piezoelectric disk is mounted on one side of the chamber. When applying voltage to the piezoelectric disk, it causes the chamber to flex inwardly, which reduces the volume of the chamber, generating the pressure required to force the ink droplet out of the nozzle. Spot sizes between 0.1 and 0.65 mm can be produced on paper at a jetting frequency of approximately 700 Hz (Cummins and Desmulliez, 2012). The bend-mode

**Figure 1** Squeeze-mode print head



**Note:** Redrawn based on the work presented in Cummins and Desmulliez (2012)

**Figure 2** Bend-mode print head



**Note:** Redrawn based on the work presented in Cummins and Desmulliez (2012)

print heads are sold by Tektronix, Xerox, Kyocera and Epson (Cummins and Desmulliez, 2012).

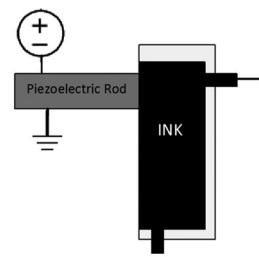
**2.1.1.1.3 Push-mode piezoelectric print head.** The push-mode print heads consist of a piezoelectric rod, which is placed next to a membrane connected to the ink chamber. On applying voltage to the piezoelectric rod, the rod expands and pushes against the chamber wall, thereby ejecting a droplet out of the nozzle, as shown in Figure 3. It was patented in 1984 by Stuart Howkins of the Exxon Company (Hertz and Welinder, 1986). The push mode print heads are also called bump-mode print heads. Advancements to push mode piezoelectric print heads have been made by Trident, Brother, Hitachi and Epson (Cummins and Desmulliez, 2012).

**2.1.1.1.4 Shear-mode print head.** The shear mode was patented by Kenneth Fischbeck and Allen Wright of the Xerox Corporation. In shear-mode print, the voltage is applied to the piezoelectric elements, causing a deformation in the upper half of the chambers, which is mirrored to the lower half of the channels. This deformation forces the channels into a chevron shape (Stringer and Derby, 2009). Flexing of the channel induces droplet ejection as shown in Figure 4. The shear-mode print heads are typically sold by Xaar and Fujifilm Dimatix (Kawahara *et al.*, 2013).

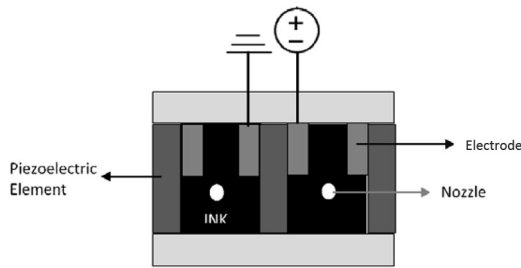
**2.1.1.2 Piezoelectric DoD inkjet printing process** The general piezoelectric DoD inkjet printing process (Cummins and Desmulliez, 2012) consists of three main stages:

- 1 drop ejection and flight;
- 2 drop impact and spreading; and
- 3 drop solidification.

**Figure 3** Push mode piezoelectric print head



**Note:** Redrawn based on the work presented in Cummins and Desmulliez (2012)

**Figure 4** Shear-mode print head

**Note:** Redrawn based on the work presented in Cummins and Desmulliez (2012)

Each stage is described in details as follows.

**2.1.1.2.1 Drop ejection and flight.** Droplet ejection involves the generation and breaking away of drops at the print head. Droplet formation occurs when an electrical signal is applied to the piezoelectric element contained within the ink cartridge. The flexing of the electrically induced piezoelectric element causes a sudden change in volume of the ink chamber. Subsequently, the ink is forced out of the nozzle head as a droplet. The droplet deforms to form a thin neck between the droplet and the nozzle head. If the kinetic energy, produced by the pressure wave, is sufficient to overcome the surface tension, the droplet breaks away from the nozzle plate keeping it attached to the nozzle (Cummins and Desmulliez, 2012).

**2.1.1.2.2 Drop impact and spreading.** During the drop impact period, which is less than 1 ms in duration, the kinetic energy of the impact is partially dissipated by viscous forces. Next, spreading occurs, where the rest of this energy is converted into surface energy. This causes the droplet to spread to a diameter determined by the relative surface energy between the ink and the substrate during the deposition process (Yarin, 2006; Stringer and Derby, 2009).

**2.1.1.2.3 Drop solidification.** Two mechanisms, namely, evaporation and polymerization, dictate the phase change of the ink producing a solid deposit. The occurring mechanism is determined by the ink type (Le, 1998), and whether the deposition process is an impact-driven (solidification) or surface energy-driven (evaporation) process (Schiaffino and Sonin, 1997; Deegan et al., 2000). Volume reduction often accompanies droplet solidification. This reduction in volume can be significant if solidification is obtained due to solvent evaporation (Cummins and Desmulliez, 2012).

### 2.1.2 Continuous Inkjet Printing

Continuous inkjet technology differs from DoD in the way the ink is deposited from the cartridge. Specifically, CIJ produces a continuous stream of ink, where drops break up from the print head nozzle by applying harmonic modulation. The separation of the drops is determined by the modulation frequency and the speed of the jet. Continuous inkjet systems typically produce 80- to 100- $\mu\text{m}$  droplets travelling at speeds of 20 m/s with drop frequencies that can exceed 250 kHz. In the most common implementation of CIJ printing, electrostatic charging and deflection are used to select and steer individual drops to define the final printed pattern (Yin et al., 2010).

On the other hand, CIJ can operate with fluids of lower viscosity and at a higher drop velocity than DoD (Castrejon-Pita and Baxter, 2013). However, CIJ printing requires a larger amount of fluid. This demonstrates the advantage of DoD systems over CIJ in the area of inkjet-printed electronics, as it consumes less ink and accordingly cuts down the overall fabrication cost.

## 2.2 Inkjet printing components

In any inkjet printing process, three main components are required: the inkjet printer, the ink and the substrate. The following sections discuss each component in details.

### 2.2.1 Commercially available inkjet printers

There are several commercially available inkjet printers which are widely used for research purposes. Among these are the Dimatix printer which has been reported by Van Osch et al. (2008), Rida et al. (2009), Wang et al. (2010), Briand et al. (2011), Vyas et al. (2011), Li et al. (2012), Molina-Lopez et al. (2012), Alcácer et al. (2013), Francisco (2014), Kim et al. (2014), Gao et al. (2014), Lessing et al. (2014), Medina-Sánchez et al. (2014), Tao et al. (2015); the PIXDRO LP50 reported by Lange et al., 2013); and the Epson Stylus Photo R230 reported by Cummins and Desmulliez (2012).

Nowadays, Dimatix Fujifilm printers (DMP-2831 and the newer DMP-2850) are among the most commonly used types of inkjet printers in research. They are bench top material deposition systems, which are designed for micro-precision jetting of a variety of functional fluids onto virtually any surface. These surfaces include plastic, glass (Dearden et al., 2005), ceramics and silicon, as well as flexible and stretchable substrates from membranes, gels, elastomers and paper products (Abadi et al., 2014).

DMP is a software-controlled inkjet printer system, which allows the user to deposit colloidal fluids on diverse substrates. This inkjet printing system has a print head, ink cartridges of 1- and 10-pL droplet sizes, a platen (substrate holder), a fiducial camera, a drop-watcher camera, stepper motors for  $x$ -,  $y$ - and  $z$ -directional control and a computer with the Dimatix Drop Manager (DDM) software. The print head has 16 nozzles with a distance of 254  $\mu\text{m}$  between the nozzles. The diameter of each nozzle is related to the size of the drop generated through the nozzle (Shin et al., 2011).

As for Pixdro LP50, it is considered an advanced research inkjet printer, which can be used for the evaluation and development of inkjet materials, processes and applications. Its compact design, high-flexibility, multi-functionality and ease of use make it a powerful tool which is well suited for general laboratory usage. The intuitive graphical user interface (GUI) supplies the operators with various advanced tools, guiding them through the inkjet printing process. The availability of numerous standard modules enables the user to easily configure and setup the system to the desired specifications. Because all these modules are embedded in the open PIXDRO system-architecture, easy scaling into an industrial system is possible. The LP50 is also compatible with a range of inks such as solvent-based (including nanoparticles [NPs] such as silver), water-based (including KOH), acidic, hot-melt and UV curable inks. The Epson Stylus Photo R230 is a six-color inkjet printer, which uses piezoelectric DoD. Its print head has six

rows of orifices, with each row featuring 90 orifices measuring approximately 28  $\mu\text{m}$  in diameter (Shen *et al.*, 2014).

### 2.2.2 Conductive inks

Various types of inks, such as NP-based, organometallic and conductive polymer inks, may be used in printing wearable electronics depending on the application (Park *et al.*, 2016). Inks could either be bought from different manufacturers (Kim *et al.*, 2011c; Jung *et al.*, 2014) or prepared chemically in a lab if specific properties are required (Shen *et al.*, 2014). The most important step in selecting an ink is determining its fluidic properties: surface tension ( $\gamma$ ), viscosity ( $\mu$ ) and density ( $\rho$ ) (Cummins and Desmulliez, 2012). These properties affect the deposition of the ink on the substrate as shown in Table I.

NP inks (Cummins and Desmulliez, 2012) have two common types, namely, silver and gold (Cui *et al.*, 2010), which are widely used due to their high chemical stability, low chemical reactivity and high electrical conductivity. However, copper (Park *et al.*, 2007) and nickel NP inks are also used, but they have a tendency to oxidize which affects the lifetime of the ink. NP-based inks are composed of NPs suspended in a solvent that is usually organic. Owing to the presence of water in inks, the printed pattern requires post treatment known as sintering.

In general, NPs have the advantages of being produced in large quantities, being dispersed in high concentrations and being able to produce relatively good electrical conductivity. However, NP inks are susceptible to the clustering of the suspended particles, which causes an increase in viscosity and surface tension, and may eventually lead to clogging the print head nozzles. Certain concentrations of NP inks can be prepared for printing of conductive patterns (e.g. 5–25 Wt.% of silver) (Shen *et al.*, 2014). As a general rule of thumb, particle size should be less than 1/100 of the size of the nozzle, and particles should be homogeneously distributed through the solution to ensure good jetting.

Silver is one of the most common types of organometallic inks (Smith *et al.*, 2006; Cummins and Desmulliez, 2012; Teichler *et al.*, 2013), as it can form a wide range of compounds soluble in organic solvents. Other types of organometallic inks are platinum (Cummins *et al.*, 2011), gold (Nur *et al.*, 2002), copper (Rozenberg *et al.*, 2002), nickel (Li *et al.*, 2009) and graphene (Secor *et al.*, 2013). One of the main advantages of the organometallic inks is that they are in a form of a solution rather than particle suspension. This eliminates the risk of agglomeration and thus clogging of the nozzles. In addition, higher conductivity can be achieved compared to NP equivalents with lower sintering temperature.

As for conductive polymers, PEDOT: poly (3,4-ethylenedioxythiophene), polypyrrole and polyaniline are common types. This type of ink is not widely used due to its low conductivity compared to metallic inks (Gamerith *et al.*, 2007). Its susceptibility to ambient humidity and reactivity to oxygen dictates that it may require the use of inert atmospheres. Additionally, it can exhibit a non-Newtonian behavior (Meixner *et al.*, 2008).

### 2.2.3 Flexible and stretchable substrate

Flexible substrates are those which can only bend in one dimension at a time, while stretchable ones can conform to a surface with an arbitrary two-dimensional curvature, such as the human body. Paper substrates are the most common flexible type of substrates, which are used in inkjet printing of electronics and sensors (Rida *et al.*, 2009; Kim *et al.*, 2013; Hoppmann *et al.*, 2013; Kim *et al.*, 2013; Määttänen *et al.*, 2013; Lessing *et al.*, 2014; Choi *et al.*, 2016). This is due to their availability, flexibility, recyclability (Määttänen *et al.*, 2013) and the fact that paper is one of the cheapest materials in the world.

In particular, photo paper has an advantage of being coated by photographic emulsion, which is a light-sensitive colloid similar to gelatin. Photo paper, compared to other flexible

**Table I** Summary of selected conductive inks, their suppliers and specifications

Ink type	Supplier	Product	Viscosity (mPa.s)	Surface tension (mN/m)	Cure temperature (°C)
Silver	Advanced Nano Products	DGP 40LT-15C	10-17	35-38	120-150
	Colloidal Ink, Co	Drycure Ag-J	8 - 12	30	120
	Harima Electronic Materials	NPS-J	9	–	120
		NPS-JL	11	–	220
	InkTec	TEC-IJ-010	9-15	30-32	100-150
		TEC-IJ-060	5-15	27-32	100-150
	Sigma-Aldrich	Ag nanoparticle ink	10-13	28-31	150-300
		Metalon JS-B25HV	NovaCentrix	8	30-32
	Metalon JS-B40G	NovaCentrix	8-12	28-32	180
	Novacentrix	JS-A102	8-12	25-30	140
Gene's Ink	CSO1121	13 $\pm$ 3	27 $\pm$ 3	150	
Nickel	Applied Nanotech	Ni-IJ70	16-25	26-31	Photosintered
Copper	Novacentrix	ICI-002HV	9-12	–	PulseForge
	Applied Nanotech	Cu-IJ70	10-20	20-30	Photosintered
Gold	Harima Electronic Materials	NPG-J	7	–	250
	Colloidal Ink	DryCure Au-J Solid	10	30	120
Polymer	PEDOT AGFA	Orgacon IJ-1005	7-12	31-34	130
Graphene	Innophene	I-3016	7-12	23-25	100
	Sigma-Aldrich	Graphene nanoparticle	1	30	250
CNTs	Brewer Science	Carbon nanotubes	<10	–	110-150

substrates, is advantageous, as its photographic emulsion manufacturing neither involves harmful chemicals nor releases environmental hormones when heated. In addition, the photo paper is recyclable; wood-free paper is also available. However, durability of printed silver patterns on paper is not efficient, as silver patterns are easily affected by the surrounding environment. For example, moisture could be absorbed by paper causing malfunctioning in printed electronics.

Polymer substrates such as polyether imide, polycarbonate, polyarylate, polyamide, polyethylene and terephthalate are also types of commonly used flexible substrates (Ko *et al.*, 2007; Perelaer *et al.*, 2009; Borghetti *et al.*, 2016). Because polymers are sensitive to high sintering temperatures, which may damage or affect their properties, a one-step inkjet printing process has been introduced by Perelaer *et al.* (2009) to eliminate the need for a sintering stage. In particular, silver organometallic ink was directly printed on a heated polymer substrate at 130°C instead of using NP ink, which would normally require higher sintering temperatures. Yeo *et al.* (2014) investigated the modification of the surface of PET substrates to achieve higher resolution of printed patterns while controlling the spreading of the ink. Modified substrates have a hydrophilic wetting area where the printed pattern and hydrophobic non-wetting barriers are required. The study suggested that a temperature of at least 80°C is required for successful spreading.

PDMS is frequently used a stretchable substrate (Chung *et al.*, 2011; Wu *et al.*, 2015; Jiang *et al.*, 2016; Sun *et al.*, 2016). It is a silicon-containing elastomer, which is also widely used to construct microfluidic devices and lab-on-chip systems (Al-Halhouli *et al.*, 2014, 2015, 2016). Direct printing of conductive solutions on PDMS is challenging owing to its elastomeric and hydrophobic nature. The wettability of the substrate also affects the size and stability of the printed patterns. Therefore, the PDMS substrate can be modified by physical and chemical methods to improve its hydrophilicity (Chung *et al.*, 2011; Shin *et al.*, 2011; Sun *et al.*, 2013). Generally, the surface of PDMS is hydrophobic and does not provide good wettability for polar solvents. Hence, surface modification of PDMS to a hydrophilic surface, prior to printing, is critical for successful printing of silver ink. This could be accomplished by using a simple method, such as air plasma treatment. According to Kim *et al.* (2014), silver lines printed on a plasma-treated PDMS substrate are continuous and do spread well. On the other hand, separated ink dots, which do not spread well, are observed on untreated PDMS surfaces.

### 3. Variables affecting printed patterns

Inkjet printing of conductive patterns and structures is influenced directly by various variables related to their conductivity, resistivity, durability and dimensions of the printed patterns. Printing multilayers of a desired conducting ink helps in increasing the density of the particles for a given area, so that the NPs form a uniform solid structure of a less porous pattern. However, printing multiple layers results in a larger thickness of the conductive traces, and accordingly, the bulk of conductance increases (Kim *et al.*, 2013).

Another variable of interest is ink sintering, where higher sintering temperatures increase electric conductivity by

dissolving more of the solvent and further fusing the silver NPs. However, high sintering temperatures may harm the substrate and change its properties. Sintering, being the last step in an inkjet printing process, is essential for the evaporation of any solvent liquid that might be present in the ink. It can be performed by applying heat to the printed pattern either thermally or by using laser. As stated earlier, the major drawback in this process is the fact that stretchable substrates such as PDMS cannot handle high temperatures (around 150°C), whereas sintering may heat the pattern up to 300°C. This is expected to result in deformation and alteration of substrate properties. More than one study proposed heating the substrate (up to 130°C) prior to printing by using a hot plate. This method saves time, whereas in a typical printing process, sintering is considered a separate step that may last up to 15 min.

When it comes to ink cartridges and print heads, both the nozzle diameter and the space between the nozzle and the substrate affect the diameters of the printed drops. Also, the substrate material and its properties affect the spread of the printed dots. Ink viscosity affects the speed of printing and the refilling of the cartridge. The contact angle depends on the material of the substrate, where the smaller the contact angle, the wider the drops of inks which are formed on the substrate. In addition, higher NP concentrations result in higher conductivity of the printed patterns.

As for the printing process itself, the effect of changing printing parameters, such as jetting temperature, jetting frequency, printing height, number of printing nozzles and jetting waveform, should be analyzed to reach an optimized state of targeted ink printing. One of the main concerns affecting the wide spread of this novel, cost-effective and affordable fabrication technology, is the need for a thorough investigation of the aforementioned variables and their effect on the end result. The first step would be to understand how inkjet printing works. Next, the properties of different inks and substrates need to be studied to understand their effect. Lastly, tests need to be performed on the resulting printed patterns. For instance, it is observable that the printed patterns agglomerate after sintering which causes the overall connectivity to increase. In particular, in the case of silver NP sintering, the silver NP becomes one bulk increasing the electrical connectivity of the printed pattern.

### 4. Approaches of adding electronic components to the stretchable circuit

In their simplest form, fabricated stretchable sensors use resistive patterned conductive lines, which respond to strain as sensory elements (Paik *et al.*, 2011). This eliminates the need to use rigid electronics within the stretchable sensors and reduces sensor fabrication to stretchable circuit patterning. In a similar fashion, inkjet printing techniques have been used to fabricate various flexible/stretchable resistive and capacitive sensors. Examples include flexible resistive micro Pirani pressure gauge (Sette *et al.*, 2013) and capacitive sensor for liquid-level monitoring (Kisic *et al.*, 2015).

More complex humidity, temperature, gas and chemical sensors could also be fabricated using inkjet printing. This is usually achieved by printing polymer layer(s) sensitive to the

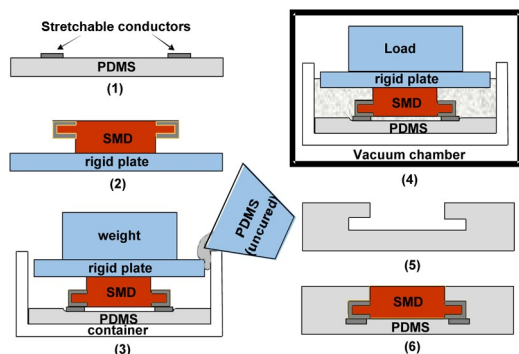
variable to be measured on top of electrodes acting as electric contacts. For example, the fabrication of an ink-jet printed flexible humidity sensor has been reported (Starke *et al.*, 2011). This sensor is fabricated by inkjet printing of conductive finger electrodes on flexible polyimide substrate with a silver ink. Next a humidity-sensitive layer is printed on top of the electrodes by using a polymer particle-based ink. Ultimately, the sensor's capacitance increases with increasing relative humidity.

However, to be able to measure more variables of interest, it is necessary to be able to integrate electronics within stretchable sensors. There have been two main approaches in encountering the challenge of adding electronic components to the stretchable circuit without compromising its stretchability. The first, presented by Mascaro *et al.* (2014), embeds rigid components in a second stretchable layer on top of the patterned conductive lines without any physically bonded connections. The second approach involves creating flexible/stretchable forms of the electronic components. In this section, both approaches will be reviewed and discussed.

#### 4.1 Embedding rigid electronics into stretchable platform

Figure 5 illustrates the fabrication steps of the embedding concept presented by Abu-Khalaf (2012). Cavities for the rigid electronic devices are created on top of the stretchable conductors in a second layer, such that the embedded devices are connected electrically with the underlying conductors. Without rigid bonding, the entire circuit could be stretched for multiple cycles. The embedding technique is specific to the T-shaped surface-mount device (SMD), where an SMD is adhered upside down onto a rigid plate, and then placed on top of the stretchable circuit. Electrical contact between the device and the underlying conductors is made sustainable by pre-loading the SMD during the embedding process. Uncured liquid rubber, commonly PDMS, is poured around the SMD to create the desired cavity. When the added PDMS layer cures, the SMD can be removed, causing the cavity to shrink as the PDMS relaxes. Once a similar functional SMD is inserted in the formed cavity, it will expand to accommodate it. The concave bending of stretchable sensors when applied to a two-dimensional surface, such as the human body, will assist the

**Figure 5** Embedding technique of SMD



**Note:** Redrawn based on the work presented in Abu-Khalaf (2012)

reliability of the formed electrical connection (Abu-Khalaf, 2012).

#### 4.2 Creating flexible and stretchable electronic components

In the second approach, we address the fabrication of flexible/stretchable forms of electronic components which could be implemented in stretchable sensors. We will specifically focus on optoelectronic devices which include organic light-emitting diodes (OLEDs) and photodiodes (PDs), arrays of micro inorganic light emitting diodes, polymer light emitting diodes and photodiodes, flexible and silicon photodetectors and nanowire photodetectors. Different types of stretchable and flexible LEDs and PDs will be reviewed, along with their fabrication techniques. Furthermore, methods for creating stretchable arrays of LEDs or PDs, such as stretchable interconnections on stretchable substrates or stretchable electrodes and recent improvements in their performance, will be discussed.

Sun *et al.* (2006) introduced an innovative technique which laid the pathway for creating stretchable optoelectronics. This method depends on controlling semiconductor nanostructure configurations of the device, such that it can be more stretchable and flexible. Buckling geometries were created in the nanoribbons of GaAs and Si, which was then used to fabricate stretchable metal–semiconductor–metal photodetectors. The electrical current increased when the PD was stretched up to 44.4 per cent and then started decreasing. Furthermore, Koo *et al.* (2010) presented improved current and power efficiencies and an enhanced electro-luminescence spectrum in OLEDs by implementing buckling patterns. Huang *et al.* (2015) demonstrated the ability to fabricate stretchable UV photodetectors, could be stretched up to 160 per cent, by forming buckles in SnO<sub>2</sub> nanowebs. Khang *et al.* (2006) produced a stretchable form of silicon involving *p-n* junction diodes that can be used as photodetectors. This was performed by structuring thin elements of single crystal Si or complete integrated devices and supporting them with elastomeric substrates (PDMS). Lee *et al.* (2011) used silicon PDs to build photodetector arrays on flexible plastic substrates. These flexible and stretchable silicon PD arrays were used to manufacture a hemispherical electronic eye camera (Jung *et al.*, 2010, 2011).

When talking about flexible optoelectronic devices, one of the things that first comes to mind is OLEDs, and organic photodetectors (OPDs). OLEDs and organic light-emitting electrochemical cells (OLECs) can be fabricated using different techniques that are explained in details in the literature (Sun *et al.*, 2006; Koo *et al.*, 2010). Recent advances regarding OLEDs and OPDs include fully transparent OLED displays (Görrn *et al.*, 2006; Song and Li, 2014), fully sprayed flexible OPDs (Falco *et al.*, 2014) and most importantly stretchable OLEDs and stretchable OLED displays (Sekitani *et al.*, 2009; Song and Li, 2014). Most of these advances are achieved by replacing conventional indium tin oxide with transparent conducting electrodes (TCEs) in the OLEDs. TCE materials include carbon nanotubes (CNTs), stretchable carbon films, nanowires, graphene and conductive polymers (Li *et al.*, 2013; Song and Li, 2014; Song and Zeng, 2015). One of the most current and significant applications of organic optoelectronics

is their usage to build a flexible sensor for pulse oximetry (Lochner *et al.*, 2014).

CNTs were also implemented in many applications in flexible and stretchable optoelectronics; one application used single-walled CNTs (SWNTs) to fabricate a stretchable active-matrix OLED display that could be stretched by 30–50 per cent while maintaining its mechanical and electrical function (Sekitani *et al.*, 2009; Park *et al.*, 2013; Cai and Wang, 2015). Other applications which use CNTs include fabrication of flexible field emission devices using SWNT as the conducting electrodes and thin multi-walled CNT/TEOS hybrid films as the emitters (Jeong *et al.*, 2011). In addition, CNTs were used in the form of CNT–polymer composite electrodes to form the anode and cathode of stretchable polymer light emitting electrochemical cells. The cells can undergo up to 45 per cent strain without damaging electroluminescent properties (Yu *et al.*, 2011). Polymer LEDs and PDs will be discussed later in more details.

Graphene is another material that contributed a lot to the field of organic optoelectronics and optoelectronics in general. In particular, many scientists have used graphene electrodes to fabricate improved-performance and enhanced-efficiency OLEDs and PDs (Bonaccorso *et al.*, 2010; Wu *et al.*, 2010; Li *et al.*, 2013; Liu *et al.*, 2015). Graphene has also been used in the fabrication of inorganic LEDs as shown in Lee *et al.* (2010, 2011). Graphene is generally useful for stretchable microscale ILED arrays, as it is used to form transparent stretchable interconnects (Kim *et al.*, 2011a, 2011b). Additionally, perylene/graphene composites were used to build a stretchable photosensor on PDMS substrate; the sensor works by changing resistance according to light and can undergo up to 25 per cent axial strain (Ali *et al.*, 2015).

Metal nanowires and nanowire networks have also contributed to the fabrication of optoelectronic devices, especially flexible and stretchable photodetectors (Zhang *et al.*, 2012; Wang *et al.*, 2014; Yan *et al.*, 2014; Yoo *et al.*, 2015). Recently scientists from Pohang University of Science and Technology in South Korea were able to fabricate high performance nanowire photodetectors. These PDs come with a broad spectral response starting from UV and visible light to near infrared (NIR). The PD array can be stretched up to 100 per cent without affecting the performance of the PDs (Yoo *et al.*, 2015). For further information, a comprehensive review about flexible PDs based on 1D nanostructures has recently been published (Lou and Shen, 2015).

Recently, the use of conductive polymers in electronics and optoelectronic devices has led to the emergence of flexible and stretchable polymer LEDs and photodetectors (Zainelabdin *et al.*, 2010; Yu *et al.*, 2011; Liang *et al.*, 2013, 2014; Liu *et al.*, 2013; White *et al.*, 2013). For example, Liang *et al.* introduced an all-solution-process for fabricating high-performance elastomeric light-emitting electrochemical cells (EPEC), which can emit light at strains up to 120 per cent (Liang *et al.*, 2013). Another considerable progress in this field is the fabrication of ultrathin flexible and stretchable foils of organic polymer light-emitting diodes (PLEDs). The foils could stand mechanical deformation and strains up to 100 per cent (White *et al.*, 2013). Furthermore, Liang *et al.* were recently able to fabricate fully stretchable white PLEDs. They used transparent conductive electrodes (TCEs) based on a silver nanowire

percolation network, which is modified with graphene oxide. The resulting PLEDs can be stretched up to 130 per cent (Liang *et al.*, 2014).

For the purpose of achieving stretchable displays and stretchable LED arrays, micro-scale optoelectronic devices are required. Inorganic LEDs and micro ILEDs have drawn the attention of the scientific community in recent years for their promising prospective and applications. Various fabrication processes of micro-inorganic optoelectronics (LEDs and PDs) have been published and discussed in details (Park *et al.*, 2009; Kim *et al.*, 2010, 2015, 2011a, 2011b). Innovative high-performance stretchable arrays of micro ILEDs and PDs that can undergo different types of deformation and strain while maintaining their function have been achieved in recent years (Park *et al.*, 2009; Kim *et al.*, 2010; Hu *et al.*, 2011; Jang *et al.*, 2014). These arrays have been used in several applications including medical diagnostics, such as balloon catheters, cardiac measurements and robotics (Kim *et al.*, 2010; 2011a, 2011b; Xu *et al.*, 2014). Other approaches for the fabrication of flexible and stretchable optoelectronics include building PDs using stretchable molybdenum disulphide transistors. They also have the potential of being used in fabricating LEDs (Sundaram *et al.*, 2013; Pu *et al.*, 2014; Xu *et al.*, 2014) and building LED strips on an elastomer that could be twisted to 720° and stretched up to 140 per cent (Drack *et al.*, 2014).

It is worth mentioning that other techniques have been used to create stretchable devices by physically connecting SMD devices to conductive interconnects, which are patterned on stretchable platforms such as PDMS. (Ohmae *et al.*, 2015) built a stretchable 45 × 80 red–green–blue (RGB) LED display using this technique. One of the useful applications which resulted from this method is the fabrication of a stretchable pulse oximeter introduced by scientists from the University of Freiburg (Ruh *et al.*, 2014). However, this technique does not result in a completely stretchable sensor, as physical connections still exist between the rigid electronic components and stretchable interconnects.

## 5. Performance of inkjet printing for wearable sensor applications

It is desired to inkjet-print high-performance wearable sensors. This entails high pattern quality, high device flexibility/stretchability and high device durability (Gao *et al.*, 2017). Several strategies, which could be used to achieve wearable devices with a desired performance, have been presented in the literature. Here we introduce a summary of the key considerations.

### 5.1 High pattern quality

The quality of the inkjet-printed lines is characterized by their uniformity and resolution (Gao *et al.*, 2017). These properties ensure the stability and conductivity of the resulting devices. It has been demonstrated that patterns of sub-micrometer resolution can be jet-printed by using electro-hydrodynamically induced fluid flows through fine micro-capillary (Park *et al.*, 2007). This suggests that a controlled droplet formation process enhances the quality of the printed lines. As previously discussed in Section 3, the optimization of printing parameters such as jetting temperature, jetting frequency,



printing height, number and diameter of printing nozzles and jetting waveform will result in optimized ink printing.

### 5.2 High device flexibility/stretchability

Many sensing applications require skin-like sensors which can be bent and stretched on the 2D human body curvature. Hence, flexibility and stretchability have become essential for the manufacturing of wearable sensors. Primarily, printing on elastomer substrates allows for the fabrication of flexible/stretchable circuits. Many flexible circuits use paper and polyimide as substrates. As for stretchable circuits, PDMS substrate is a favorable choice owing to its durability, adjustable stiffness, biocompatibility and commercial availability (Gray *et al.*, 2004). If straight lines are inkjet-printed on a stretchable substrate, it is expected that the lines will crack upon stretching the circuit. Hence, several techniques have been developed to overcome this challenge. Such techniques include printing elastic conductors on stretchable platforms (Sekitani *et al.*, 2009) and printing highly conductive ink on stretchable substrates with wavy structures (Chung *et al.*, 2011).

### 5.3 High device durability

The mechanical durability of the printed wearable circuits is another key consideration for determining the performance of the inkjet printing process. Clearly, a wearable circuit which can sustain prolonged bending or stretching, while maintaining electrical conductivity, is desirable. Typically, circuits are tested for the number of bending/stretching cycles which they can undergo before cracking and/or slipping of the conductive lines occur. The formation of cracks usually occurs during circuit fabrication, as well as when the circuit is bent or stretched. When inkjet printing is used and the circuit is subjected to post-sintering, the solvent in the deposited ink evaporates and the conductive lines, which have a different thermal expansion coefficient from that of the substrate, tend to crack (Gao *et al.*, 2017).

The formation of cracks affects the electrical conductivity of the printed lines. However, this effect can be reduced by printing thicker lines, printing on thin substrates, using flexible inks and substrates and limiting the usage of the circuit to relatively low-stress applications. In addition, techniques such as sandwiching the conductive lines between two elastomer layers has shown a reduction in crack formation (Guo *et al.*, 2013). Slip between the printed conductive lines and the substrate occurs when the adhesion between them is insufficient (Gao *et al.*, 2017). To overcome this issue, it is common to use hydrophilic substrates or treat the surfaces of hydrophobic substrates to enhance their adhesion to the deposited conductive lines. For example, PDMS substrates can be treated chemically or by a technique known as plasma etching prior to jetting the ink on their surfaces.

Once a flexible/stretchable sensor is fabricated using inkjet printing, its overall performance needs to be characterized. This requires evaluating the static and dynamic characteristics of the sensor, such as its accuracy, repeatability, sensitivity, range, response time, etc. Next, the results are compared to similar sensors produced via various fabrication techniques. The inkjet printing process has demonstrated great potential for use in the fabrication of flexible low-cost wearable sensors in different applications. For example, the first micro Pirani gauge

fabricated by inkjet printing technology (Sette *et al.*, 2013) had a sensitivity range of  $10^{-2}$  to  $10^4$  Pa, making it comparable to the best reported gauges. Several studies analyze the performance of inkjet-printed sensors, such as the study conducted by Loffredo *et al.* (2009) where inkjet-printed chemical sensors was compared to a casting sensing devices at low concentrations of acetone and toluene vapors. The inkjet-printed devices were found comparable to the casting devices; however, they require further optimization as their response is affected by the ink deposition method.

It is worth reiterating that several fabrication techniques, including inkjet printing, have been developed to allow patterning conductive lines on a rubber substrate, and yet fabrication of fully stretchable wearable sensors is still rather limited. The potential of inkjet printing for the development of wearable sensors has been demonstrated in this section. Nevertheless, meeting the requirements of printing highly conductive, highly stretchable and highly durable lines requires comprehensive optimization of the printing process. More complex sensors can be fabricated by either implementing inkjet-printed electronics or innovatively embedding commercially available rigid electronic components, where both approaches represent intellectual research endeavors. There is a vast room for research when it comes to enhancing the wearability of a stretchable sensor in terms of powering it, and acquiring data from it. Up until this point, rigid external circuitry has been used in most applications to power these sensors, and acquire and analyze their outputs. The circuitry is usually interfaced with the sensor via physical wire connections soldered to the patterned lines, making the sensor un-stretchable at the connection point. Innovative solutions need to be implemented to overcome this issue. Finally, a wearable sensor would entail an on-board miniature power source and wireless transmission of data.

## 6. Conclusion

In conclusion, it is evident that the technology of inkjet printing is becoming a competitor to traditional lithography fabrication techniques, as it has the advantages of being low cost and less complex. In particular, this technique has demonstrated great capabilities in the area of flexible/stretchable electronics and sensors. Various inkjet printing methods have been presented with emphasis on their principle of operation and their commercial availability. In addition, the components of a general inkjet printing process have been discussed in details. Several factors affect the resulting printed patterns in terms of their conductivity, resistivity and durability, as well as their geometry. This includes the types of used inks and substrates, number of printed layers, ink sintering and printing process variables.

Examples of inkjet-printed electronic devices were provided to show the ease of implementation of this fabrication method and the resulting electromechanical behavior of the produced flexible/stretchable electronics. Specifically, flexible/stretchable optoelectronic devices which could be implemented in stretchable circuits have been thoroughly discussed. Although several fabrication techniques have been recently developed to allow patterning conductive lines on a rubber substrate, fabrication of fully stretchable wearable sensors remains

limited. Future research in this area is crucial for the advancement of wearable sensors. Specifically research advancements are required to print highly conductive and highly stretchable lines, and reliable electronic devices, while interfacing them with external circuitry for power transmission, data acquisition, and signal conditioning.

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