Design and fabrication of multilayer inkjet-printed passive components for printed electronics circuit development

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Abstract

Printing electronic passive components suffer from the lack of a wide variety of appropriate materials for developing components with specific characteristics, for specific dimensions. This paper introduces a multilayer approach for the inkjet printing of resistors, inductors and capacitors, showing that it is possible to manufacture tailored passive circuit elements and therefore their implementation into functional printed electronics. The materials and process steps for the manufacturing, the individual component characteristics and the equivalent circuit is provided for all passive devices.

KEYWORDS: Inkjet printing; passive components; multilayer; printed electronics.

Introduction

Modern society is entering a novel technological paradigm based on the interconnections of objects in our surrounding environment – referred to as the Internet-of-Things (IoT) [1,2].

This new concept consists of the connection of everyday objects by networks and databases, allowing the possibility to detect changes in the environment in which they are inserted, and the implementation of the corresponding response [3–6]. Thus, the distributed computing capability and the access to the large quantity of produced information [7] enables to act into

the environment in a preventive or reactive way ensuring better quality of life, energy saving and security, among others.

Although strong benefits are expected from this new paradigm [8], the introduction of its full potential needs the development of new techniques and production methods [9]. In particular, this new concept implies the implementation of active areas (e.g. sensors and actuators) from very small to large scale, on flexible and/or irregular surfaces [10] and they will have to withstand the harsh and varying conditions of the surrounding environment.

As a consequence, a new concept for electronics able to keep up with new sensor and actuator systems is required, being the development of flexible electronics one of the most promising approaches for this new paradigm.

There is an increasing interest to exploit graphic printing techniques for the manufacturing of flexible electronics. Printing technologies allow the deposition of functional materials not compatible with conventional electronic manufacturing processes [11] on large areas and on rigid as well as flexible or irregular surfaces.

There are several printing techniques that can be classified into contact printing and non-contact printing techniques [12]. In the case of contact printing, the substrate is in direct contact with intermediate forms where the ink is located before its transfer to the substrate. There is a risk of contamination and damage of the substrate due to the contact. However, non-contact printing methods can transfer inks to substrate without any contamination and damage risk. Thus, they enable materials deposition on sensitive and irregular surfaces. Contact printing techniques such as flexography [13,14], gravure [15,16] and screen [15] printing are usually characterized by high processing speed and low-cost due to the

possibility to implement it to a roll to roll (R2R) manufacturing process, which can be considered as indicator for mass production.

Non-contact printing methods are e.g. inkjet [15], laser-induced forward transfer (LIFT) [17] and aerosol-jet [18–20] printing. They are typically based on the digital direct-writing approach without any need for physical masks to enable patterning. Therefore, pattern change is possible without associated costs making the process attractive for highly individualized products and/or products with short run length.

Inkjet printing is considered as one of the most promising techniques for non-contact and low-cost printed electronics production. It is applicable to large areas and is compatible with flexible or irregular substrates [15,21–23]. It represents an additive and non-contact process without the need of high temperatures [24], photolithography, high vacuum [24] or etching steps which typically occur in traditional micro fabrications techniques.

Thus, examples of inkjet-printed devices include thin film transistors [25–27], organic memories [28], antennas and RFID tags [29,30], light-emitting devices [31,32], solar cells [33,34], electrical components [35] and sensors [22,36–38], among others.

In any case, the up-to-date examples of inkjet-printed active devices indicate that they cannot compete to the traditional semiconductor production in terms of integration, performance and cost [39]. In the field of passive devices it is considered to be possible to fabricate competitive components by printing technologies. Passive devices are still the most voluminous parts of the current electronic circuits, even considering the surface-mounted-devices (SMD) solutions.

Despite current electronics is increasingly becoming digital, passive components are still essential for electronic development, particularly in the area of sensing. Most of the sensors still have an analogue response [22,37,38], as well as in the wireless communication area [40], where modulation and demodulation of the signal through passive components is necessary.

According to the needs of electronic circuits, it is necessary to be able to fabricate components with specific functional characteristics appropriate to the circuit and the function to be performed. The fabrication of those components is only possible using materials with a broad range of functional characteristics (such as dielectric constant or electrical conductivity), or through process engineering to tailor the few materials that currently exist for printing to the component characteristics to be obtained.

The actual development in the printing of passive components has focused on the improvement of the performance per unit of area. This goal can be achieved by three ways: a) through the use of new printing technologies with higher resolution, higher repeatability and guarantee of thinner and uniform layers; b) the use of new inks with higher performance and stability and c) the use of new manufacture strategies and methods using existing technologies and inks. In this way, and resembling the traditional passive components fabrication method evolution, the use of multilayer architectures can be considered a suitable solution both, for increasing the ratio of performance per unit area and to tailor the characteristics of passive components to accomplish with the specificity of circuit fabrication demands.

This work is focused on the introduction of the multilayer concept for inkjet-printed passive component fabrication, in particular, coils, capacitors and resistors, allowing the fabrication of complete circuits with fewer materials and ensuring, therefore, high reproducibility and success rate. Further, this methodology will allow to suitably tailor the characteristics of the components for specific application needs.

All components have been fabricated entirely by inkjet printing, based on silver nanoparticle, polymeric conductive and dielectric inks. Full details of the fabrication process are included to contribute for the development of the passive component printing area.

The performance of the structures is included as well as the modelling of the different components, allowing to compare the theoretical response curves with the measured ones and therefore the modelling and design of more complex analogic circuits.

Experimental details

a) Equipment

Inkjet printing was performed using a Drop-on-Demand (DoD) Dimatix Material Printer (DMP) 3000 from Fujifilm Dimatix equipped with a DMC-11610 (propylene) cartridge and a print head with 16 nozzles. The print head ejects a nominal drop volume of 10 pL per nozzle. The nozzle-to-nozzle distance is 254 μm and the capacity of the ink cartridge is about 1.5 ml.

The DMP 3000's additive bottom-up fabrication process is capable of jetting a wide range of functional fluids after the definition of a large set of parameters such as waveform, voltage

and drop ejection frequency, print head and substrate temperatures, in order to optimize the drop generation and formation according to the substrate, ink and patterns application.

The manufacturing process ends with the post-processing of the deposited ink e.g. a curing, drying and sintering process. The post-treatment for the inkjet-printed layers were carried out in a Nabertherm TR240 oven.

b) Materials

Heat stabilized Teonex Q65HA (Teijin DuPont films) poly (ethylene naphthalte) (PEN) films having 125 μm thickness were used as a flexible plastic substrates

The PEN substrate is similar to poly(ethylene terephthalate) (PET) [41] substrates (typically used in these applications), except that PEN shows an excellent thermal stability, with a dimensional shrinkage of 0.1% at 150 °C for 30 minutes. It withstands temperatures up to 190 °C and has a surface treated face especially dedicated to printing applications. Further, it shows transparency in the visible range, which facilitates the printing and characterisation of the devices. The surface energy of the PEN substrate was measure with a Kruess Mobile Drop system. It was determined to $\sim 38 \text{ mN/m}$.

All the conductive layers were printed with UTDAgIJ1 [42] silver nanoparticle ink from UT DOTs Inc. which is a commercially available ink formulation.

It was necessary to optimize the deposition parameters for the print head to eject well-defined droplets resulting in well-defined patterns. The control signal applied to the piezo-electric print head is called as waveform which is a certain voltage over time. **Figure 1a)** shows the optimized jetting and non-jetting waveform for the UTDAgIJ1 silver ink and the ejected

droplets with a drop-watcher camera. The non-jetting waveform is applied to the print head in the idle mode avoiding the drying of the nozzle orifice due to a soft movement of the meniscus of the ink.

The maximum drop ejection voltage was 29 V and the drop ejection frequency was limited to 2 kHz for higher repeatability. It was also verified that the best results were obtained with 40 °C print head and 45 °C substrate temperatures. With these well-defined conditions, lines with 60 μm width were obtained. Finally, the nanoparticle silver ink was dried and sintered at a temperature of 150 °C for 30 minutes.

Figure 1. Silver nanoparticles ink, jetting and non-jetting waveform and drop formation image a), dielectric ink, jetting and non-jetting waveform and drop formation image b), conductive polymer ink, jetting and non-jetting waveform and drop formation image c). All drop formation images have the same scale bar, represented in a).

The dielectric layers were printed using a cross-linked poly (4-vinyl phenol) insulating material. The solvent was propylene glycol monomethyl ether acetate (PGMEA) and Poly(melamine-co-formaldehyde) methylated (PMFM) was applied as crosslinking agent.

This ink was filtered (1 μm filter pore size) before placing it in the cartridge. The deposition parameters were optimized to obtain well-defined drops, as shown in **Figure 1b**). In this case, the pattern definition was not particularly important. But the aim was to obtain uniform and as thin as possible layers.

After an optimization procedure, the tested print head nozzles were used to print the desired patterns using the waveform shown in **Figure 1b**). Maximum drop ejection voltage and frequency of 30 V and 5 kHz were used to print the patterns. According to the ink characteristics, printing took place with the print head and the substrate at room temperature.

The printed ink layers were finally cured in an oven at a temperature of 150 °C for 30 minutes.

For developing the printed resistors a conductive polymeric ink was used: Poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT:PSS) with the commercial ink provider Orgacon IJ-1005 from AGFA [43]. This ink has a viscosity between 7 to 12 cP. For the present application it was necessary to optimize the deposition parameters to obtain well-defined drops (**Figure 1c**), allowing proper pattern definition and a fully uniform layer.

After the optimization procedure, the selected print head nozzles were used to print the pattern using the waveform shown in **Figure 1c**). Maximum drop ejection voltage and frequency for the printing process were 34.5 V and 2 kHz respectively. No elevated temperature for the print head was applied, but the substrate temperature was set at 40 °C,

allowing in this case a faster evaporation process of the solvent to ensure a good control over the printing of patterns. Finally, the printed ink layers were cured in an oven at a temperature of 130 °C for 5 minutes.

c) Process

The components presented in section 3 show different characteristics and formats and, therefore, it was necessary to optimize the various steps of the printing process in order to ensure the best results for each component. The optimization procedure is generically represented in **Figure 2**, the details for each component being provided in the description of the corresponding devices.

Figure 2. Process diagram for the fabrication of the printed passive components

d) Characterization

Passive components including resistors, coils and capacitor were designed and fabricated using the materials and processes described above. Then, the full characterization of the components was performed.

The electrical characterization was carried out using a QuadTech 1920 Precision LCR Meter [44] connected to the control computer via IEEE-488 interface, the LCR meter being fully controlled by an application developed on LabVIEW System Design software from National Instruments [45].

With this application, various parameters in the frequency range between 20Hz and 1 MHz can be measured easily.

The necessary parameters for the characterization are defined according to the equivalent model of each component. The connection between the LCR meter and the components to be characterised was performed using a 4-wire method connected with ball point needles coated with gold.

Results

The fully printed multilayer passive components and corresponding characterization is shown in the following.

a) Capacitors development and characterization

Printed capacitors can be achieved by two approaches, the most common being the use of two layers, where an interdigitated conductive layer is printed, giving rise to the positive and negative capacitor conductive plates. In this case it is critical to ensure the minimum space between the fingers without short-circuit. Then, the second layer consists in a dielectric material filling the gap between the fingers [22,46–48]. At the present moment, this solution

shows a low capacity ratio per unit of area, being this solution particularly used in capacitive sensors solutions.

Another method consists in the use of three different layers, as represented in **Figure 3a**). First, a conductive layer is printed, followed by a dielectric one and finally, a second conductive layer forms the second conductive plane. This is the approach used in the present work.

Figure 3. Schematic representation of the various patterns needed to print a single capacitor a); printed single capacitor b); optical image of multi capacitor, where it is visible the different layers c) schematic of the stack approach for multi capacitor printing d); printed triple capacitor e); cross sectional view of the various layers that forms the multiple capacitor f); total capacitor profilometry d).

Several steps are thus involved in the printing of a multilayered capacitor. A PEN Teonex Q65HA film is used as a substrate, which should be thoroughly cleaned. An initial layer of silver conductive layer is printed on the PEN substrate with a drop space of 40 μm , resulting in lines with a minimum width of approximately 60 μm .

Two active area dimensions were printed in order to verify the influence in capacitance per unit area in different dimensions, so square patterns were printed to form the first capacitor plan (**Figure 3a**) with an active area of 5 x 5 mm^2 and 2.5 x 2.5 mm^2 respectively.

After curing the silver ink layer, condition as mentioned in the previous experimental section, the dielectric layer is printed in the active area using a drop space of 15 μm , to guarantee a perfectly uniform layer and to prevent short circuit between the first and the third conductive layers. The size of this layer is 5.5 x 5.5 mm^2 and 3 x 3 mm^2 respectively, ensuring the perfect isolation between layers.

The capacitance of the capacitor can be also tailored by controlling the thickness of the capacitor.

After the curing procedure of this layer (see experimental section), the capacitor is finished by printing a new silver conductive layer that corresponds to the top conducting electrode. This layer is also subjected to a curing process, and the obtained printed single capacitor can be seen in **Figure 3b**).

To increase the performance per unit area, a stack configuration was implemented. As a proof of concept, three capacitors in stack were developed that correspond to a printing process of seven different layers (**Figure 3d**), with the corresponding printing and curing process for each layer, to obtain the final result presented in detail in **Figure 3f**) and **Figure 3c**).

The sectional view (**Figure 3f**) shows that the first dielectric layer (C-PVP) has a thickness of approximately $1.4\ \mu\text{m}$ while the upper dielectric layers show a thickness of $\pm 2.6\ \mu\text{m}$, while the thickness of the conductive layer is around $0.15\ \mu\text{m}$. The profilometry graph of the total capacitor is shown in **Figure 3g**, where it can be noted that the layers have low roughness, with a thickness in the order of $6.9\ \mu\text{m}$. The high layer thickness is related to the coffee ring effect, being the layer thickness lower at the center of the device.

Capacitors were built with two connecting pads and measured using the 4-wire probe method. The discrete capacitors deviate from an ideal capacitor according to the equivalent circuit shown in **Figure 4a**). Their characteristics can be specified by a series equivalent circuit composed by an ideal capacitance and additional electrical components which model losses and inductive parameters of the real capacitors. In this case, the parallel equivalent resistance (R_p) represents the sum of the insulation resistance of the dielectric material with the resistance that represents the leakage current. The serial equivalent resistance (R_s) summarizes the ohmic losses of the capacitor, in particular in the contacts and capacitor conductive layers. Finally, the equivalent series inductance (L) corresponds to the effective self-inductance of the capacitor.

Figure 4. Printed capacitor equivalent circuit model a), equivalent circuit parameter results for the single layer capacitors and the stack capacitors b).

Based on the equivalent circuit model, the various parameters of the model were measured both for the single capacitors and the stack capacitors, the results being summarized in **Figure 4b**).

The obtained results demonstrate that the produced capacitors can be approximated to an ideal capacitor, due to the high value of R_p and the low values of the series resistance (R_s) and L .

Furthermore, based on the obtained capacitance results for the stack capacitors, it can be concluded that the multilayer stack option supports a significant increase of the capacitance per unit area of inkjet-printed capacitors. Moreover, it is also verified that increasing the number of layers has an associated trend for the capacitor to operate as a non-ideal capacitor, motivated by the irregularities in the different layers, which gets aggravated with increasing

number of layer and results in larger dielectric capacitor losses, as shown in **Figure 3f**), represented by (R_p) (**Figure 4b**).

b) Inductors development and characterization

Only one conductive layer is required for the fabrication of a printed inductor , although their fabrication in printed electronic circuits implies the printing of two more layers to access the center of the coil by forming a dielectric layer over the windings (bridge).

A coil implementation with two terminals, as the one presented in **Figure 5a**), was fabricated.

Figure 5. Schematic representation of the various patterns needed to print a high density coil a); printed single coil b); detail view of the printed patterns of the coil c); profilometry of the conductive layer d).

In order to obtain coils with high inductance it is necessary to increase the density of the turns. This leads to an increase in the complexity of the printing process due to the need of high printing density in a large area, which does not occur in coils with a low number of turns [40].

High printing density within the coil leads to an assurance of the line continuity along all coil turns for the lines with small width, as can be seen by the final printing result shown in **Figure 5b)**. After a large set of printing tests, coils with dimension of $14 \times 14 \text{ mm}^2$ were fabricated

successfully with an optimum line width of $\sim 100\text{ }\mu\text{m}$ and the line thickness is approximately $1,6\text{ }\mu\text{m}$, being the line spacing of around $75\text{ }\mu\text{m}$, as it is shown in the **Figure 5c)** and **Figure 5d)**. These dimensions allowed printing reproducibility of this component of approximately 70%.

The coil is composed by 25 turns that correspond to a total line length of $\sim 1\text{ m}$. The equivalent circuit of the physical inductor is shown in **Figure 6**. The inductor is composed by a series resistor (R_s), which can be attributed to the resistance of the silver ink layer. In addition, the inductor has a parallel capacitor (C_p), which is a measure of the internal capacitance due to the capacitance between the turn lines.

Figure 6. Printed planar coil equivalent circuit model.

According to the equivalent model, the various parameters of the model were measured, leading to an inductance of $5.42\text{ }\mu\text{H} \pm 13\%$, a serial resistance (R_s) of $12.7\text{ k}\Omega \pm 9\%$, and a parallel capacitance (C_p) of $50\text{ pF} \pm 1\%$.

The printed planar coils are far from ideal coils due to the high value of R_s , as it makes use of a printed line of $\sim 1\text{ m}$ with a layer thickness of only a few hundred nm. On the other hand,

the printed lines behaviour along the vertical printing direction is different from the horizontal (**Figure 5a**), resulting in lines with slightly different dimensions and characteristics (**Figure 5c**) causing the coil resistance to increase.

Finally, as a result of being planar coils, their capacitive component is also relevant, due to the capacitance formed between the turn's lines, being the capacitance higher for larger turn density.

c) Resistors development and characterization

Resistors are one of most used component in current electronic applications, which typically requires a wide range of values to satisfy the circuit's needs. Similar to the traditional resistors production method, printing technologies allow to obtain a wide range of resistors values using only one material, e.g. by varying the active area geometrical conditions to vary the final resistor value (i.e. varying the length, thickness and width of the active area [49,50]).

Inspired by the SMD components manufacturing method, the resistance variation of the PEDOT:PSS active material was evaluated with varying length of the active area from 0.5 mm to 2 mm and the number of material printing layers (without curing process between layers) for 2, 4 and 6 printed layers. The active area width was kept constant at 0.5 mm for all samples.

For the fabrication of printed resistors, one layer is necessary for the active material and a second layer for the contact terminals. Thus, two layers are printed, the first layer, based on silver ink, corresponding to the contacts and the second layer that corresponds to the active material (**Figure 7a**).

Figure 7. Schematic representation of the necessary patterns to print a single resistor a), results for two printed resistors with different lengths b), optical image of the electrode layer (Ag) and the active layer (PEDOT:PPS).

The final result of some printed resistors are shown in **Figure 7b)** and an image with higher detail in order to verify the homogeneity of the active layer is presented in **Figure 7c)**

The printed resistors are represented by an ideal resistor in series with a coil, both in parallel with a capacitor, as represented in **Figure 8a)**. The values of capacitance and inductance are mainly determined by the dimension of the connecting terminals and the geometrical characteristic of the active material layer.

Figure 8. Printed planar resistor equivalent circuit model a), resistance value dependence on the number of active layer and active area length b).

The results for the different printed resistors are presented in **Figure 8b)**, where it is verified that the resistance varies approximately from 25 Ω to 650 Ω depending on the number and length of the active layers, being the tolerance of always below 5%.

The remaining parameters of the equivalent circuit have been also measured, resulting in a C_p from 534 fF for a length of 0.5 mm of an active area and 80fF for a length of 2 mm of an active area. The inductance value can be considered as 0.

Based on the obtained values it can be considered that we are in the domain of ideal resistors, where it is possible to tailor resistance values depending on the application.

Discussion

The obtained results indicate that the stack capacitor implementation is a viable solution to obtain tailored capacitor values in small areas. On the other hand, increasing stack capacitors number of layers, lead to a lower capacity increase due to increased inhomogeneity of the printed layer.

The stack method is also an appropriate solution to increase the coil inductance, but in this case it is more difficult to achieve due to the larger complexity of the pattern to be implemented. By simplifying the complexity (i.e., reducing the density of lines) and increasing the number of coils in the stack, it must be possible to increase the inductance per unit area, as well as to reduce the coil capacitance by optimizing the implemented pattern.

Finally, from the fabrication results of the printed resistors it can be concluded that extrapolating the printing conditions and patterns to different dimensions and different number of active material layers, printed resistor from 10 Ω to over 100 k Ω can be achieved with a single material simultaneously.

The multilayer printing of components is an advantageous solution for printing electronics. The use of this method may be just limited by the non-uniformity of the obtained layers, resulting in faulty components, which may lead to limitations in the maximum number of layers to be printed according with the curing conditions of the various materials.

Thus, this work has proven the ability to build tailored passive components based on a multilayer approach as well as the possibility of fabrication of multilayered complete analogic electronic circuits [51] by the combination of multiple passive components.

Finally, this work was carried out using inkjet printing, but this can be implemented in different mass productions printing methods, which can increase the viability of this new electronic circuit production method.

Conclusions

This paper introduces a multilayer approach for the inkjet printing of capacitors, inductors and resistances, showing that its potentiality to fabricate tailored passive circuit elements and therefore their implementation into functional printed electronics. For all passive components, the fabrication materials and steps, the component characteristics and the equivalent circuit is provided.

This work allowed to prove the advantages associated with the stack components implementation, where in the capacitor case allowed to increase from 2 nF/cm² for the capacitance for a single capacitor implementation to 3.1 nF/cm² for multilayer capacitor implementation. For coils implementation it was demonstrated the possibility to print a high density of turns which enables to fabricate coils with an inductance about 5.5 μ H for an external size of 14 x 14 mm². This value can increase by adopting the same concept for the fabrication of the capacitor.

Finally, for the printed resistors implementation it was verified the possibility to obtain resistors with the desired value in the range from 10 Ω to 100 k Ω using a single material and just changing the pattern format and the number of printed active layers.

It is also noticeable that, despite inkjet printing being a part of the printing techniques, these concepts can easily be applied to the other printing techniques.

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