UHF Electromagnetic Structures Inkjet Printed on Temperature Sensitive Substrates: a Comparative Study of Conductive Inks and Sintering Methods to Enable Low Cost Manufacture

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Abstract

In this paper we demonstrate the use of inkjet printing as a facile digital fabrication tool for the cost effective manufacture of UHF RFID transfer tattoo tags and Frequency Selective Surfaces on low-cost flexible and porous substrates. Electrical and morphological properties of conductive features obtained from a range of metal nanoparticle inks and low temperature sintering methods, such as argon plasma and photonic flash, are evaluated. Large scale potential is addressed.

Introduction

With the emergence of distributed and wireless sensor technology readable tags will be able to collect a vast set of data that can be processed to provide new information. Such information could be extremely important for security, health care and location applications. Some examples include mission critical environments such as power plants, airports, military bases and depots, refineries, and access restricted areas to provide the highest quality of security to record trends and provide immediate required actions. In these environments as well as health care, identifying, tracking and monitoring people is vital to interface different services to create a more resilient system.

The widespread adoption of passive UHF RFID tag in asset tagging is well documented and its use is also emerging as particularly useful for monitoring, identifying and tracking people particularly in work environments [1-3]. Current human tagging, external to the body, is usually based on wrist bands or ID badges which can be removed and given to other people. The human body is an especially challenging platform for RFID owing to its high conductivity [4]. We recently reported A platform insensitive UHF RFID tag for mounting directly onto the skin surface in the form of a transfer tattoo [5,6]. Skin mounted electronics, such as UHF RFID tags, could provide an effective solution to many of the issues described above. However, in order to make this technology wide spread and commercially viable tags must be cost effective, reliable and small in size to appeal to the end user.

Another electromagnetic structure is frequency selective surfaces (FSSs) use to potentially enhance the properties of buildings in radio communications. For example, incorporation of band selective filters into the structure of commercial buildings enhances interference suppression to secure communication and offers frequency reuse, leading to improved channel capacities [7]. This can be achieved through the incorporation of FSSs into laminates, foils or wallpaper for use on and in office walls ceilings and windows. FSSs are either arrays of conductive elements (band stop) printed on a suitable substrate, or alternatively they are slots (band pass) etched into a conductor. This etching process is not suited to manufacture large panels required at mobile and WLAN frequencies.

To date the electronic industry largely relies on PBC fabrication methods such as conventional analogue printing, etched circuit methodologies and vapor phase deposition. These technologies do not provide sufficient design flexibility, limit substrate of choice, produce large amount of hazardous waste hence hindering customization at the appropriate cost. In contrast, additive digital technologies such as inkjet printing of metal precursor inks provide a more versatile, eco-friendly, highly scalable technology to prototype and mass produce electronics. For a cost-effective production of printed electronics, the ink and the substrate should be inexpensive and compatible with roll-to-roll production capabilities.

Metal nanoparticles show a reduced melting temperature compare to the bulk material [8]. Metal nanoparticle inks that can be processed by inkjet printing have been intensively researched for many years. Subsequently, some inks are already commercially available [9]. However, typically in order to obtain conductive features a heating process of over 200 ^{o}C for more than 30 minutes is required [10,11]. Typically, high temperature processing is not suited for low-cost foils and paper substrates as it causes deformation and degradation of these temperature sensitive substrates.

Aiming at solving this, in recent years, researchers have reported various methods to reduce the sintering temperature of metal precursor inks following two novel approaches. By tailoring the organics content in the ink conductive silver features with 20% the conductivity of bulk silver have been achieved without heating [12,13]. Alternatively, energy sources such as focused LASER beam [14], a low pressure argon plasma exposure [15], photonic flash sintering [16] and microwave radiation [17] have successfully been employed to sinter metal nanoparticles on low *Tg* foils due to localized heat generation rather than heating a complete sample.

The use of commercial metal nanoparticles, inkjet printing technology combined with novel sintering techniques compatible with temperature sensitive substrates such a low-cost foils and paper is expected to result in a cost effective manufacturing of low-cost electronics and disposable products such the RFID transfer tattoo tags and wall mounted FSSs structures here proposed.

Prior art

Driven by the interest in "green" and wireless technologies, work in the field of design and fabrication of RFID antennas and tags exploiting paper substrates and inkjet printing technology has recently been published by G. Orcchini *et al* [18]. However, to date, very little work has been reported in the field of electronics mounted directly on skin. Recent published work from Dae-Hyeong Kim *et al* [19] demonstrated the possibility to attach high-performance electronic functionalities to the surface of the skin. Functionalities were produced by conventional PBC deposition techniques on silicone and Polyvinyl alcohol (PVA) substrates. RFID antennas and tags mounted directly onto skin are particularly challenging owed to the intrinsic electrical characteristics of the human body which can interfere with RF components.

To our knowledge, last year we reported the first inkjet printed UHF RFID transfer tattoo tags mounted on skin [20]. A performance and price comparison between tags produced with silver nanoparticles, thermal sintering processing on transfer paper by inkjet means and copper and silver tags produced by conventional PBC technologies was addressed. Similarly, our earlier encouraging work in the field of Frequency Selective Surfaces (FSS) on low-cost foils demonstrated that inkjet printing can be used to produce dipoles (antennas) of performance similar to dipoles obtained by conventional etch processing [21].

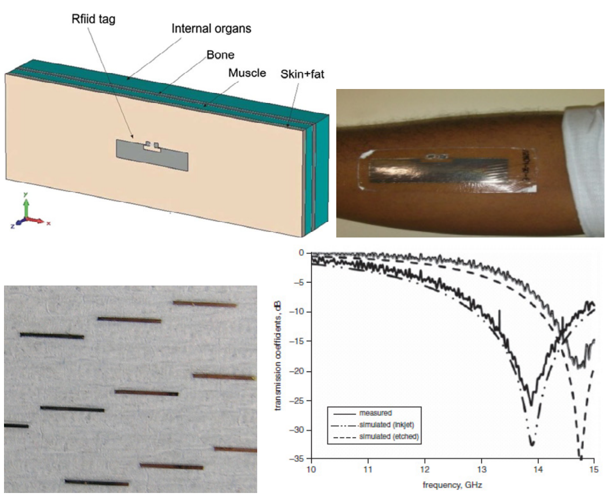


Figure 1. *a) RFID transfer tag mounted on multilayer human model, b) inkjet printed RFID tattoo transfer tag on volunteer arm, c) Inkjet printed FSS on PEN, and d) comparison of measured and simulated transmission curves for inkjet printed and etched frequency selective surfaces [20,21].*

However, although some developments in the field of on-skin electronics and inkjet printing of RFID tags on paper have recently emerged, to date no research has been reported combining both fields. Furthermore, metal nanoparticle sintering processes emerging as an alternative to thermal sintering methods have been investigated on temperature sensitive foils but, to our knowledge, not on paper substrates. In addition to the reduced sintering temperature these alternative sintering methods require shorter sintering times compare to thermal sintering. Therefore, the work presented provides the first study combining the use of metal nanoparticle inks, inkjet printing technology, paper substrates and emerging sintering processes to investigate the producing low-cost UHF RFID transfer tattoo tags and FSSs.

Methodology

In this contribution, we focus on the inkjet printing of silver metal nanoparticle inks to obtain conductive features for two UHF electromagnetic structures: i) thin UHF RFID tags onto temporary tattoo paper, which can be transferred onto the surface of the skin by soft contact, and ii) FSSs on paper and plastic substrates which can be mounted on walls.. We compare four inks, three commercially available inks and one bespoke ink. We compare the effect of three nanoparticle sintering techniques (thermal, argon plasma and photonic flash sintering) in the electrical properties and morphology of simple silver inkjet printed features. This preliminary study provides the basis to obtain the printing and sintering conditions to obtain functional RFIDs and FSSs structures on paper substrates. A comparison of the performance of on-skin mounted RFID tags and FSSs obtained with various inks and sintering techniques is presented. Advantages and disadvantages of each ink, sintering technique for potential low cost roll-to-roll production are discussed.

Experimental

Materials

Three commercial and one bespoke silver nanoparticle dispersions were used in this work. Ink A, was a dispersion in water from Novacentrix (Metalon JS-B25HV) containing 25 wt% of silver nanoparticles. Ink B, was a dispersion in ethanol/ethylene glycol mixture from Sigma-Aldrich (Sun Chemicals, 719048) containing 20 wt% of silver nanoparticles. Ink C, Harima Chemicals NPS-JL containing 40 wt% silver nanoparticles. And Ink D, Huji-005 a bespoke ink with 15 wt% silver nanoparticles.

Inkjet tattoo paper was supplied by www.craftycomputerpaper.co.uk and purged with a flow of air to remove dust particles. Conventional microscope glass slides (2.54x7.62 cm; Sailing Boat, Cat. No. 7101, China) were used and ultra-sonicated for 10 minutes in an acetone bath, rinsed with deionized water and placed in a convection oven at 60 ^{o}C for 10 minutes prior to use.

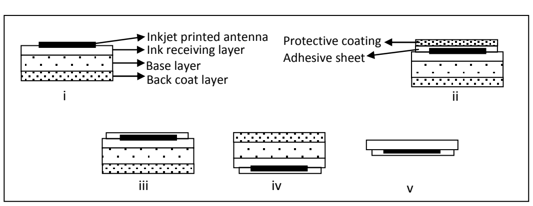
The RFID integrated circuit chip used on the all the tag designs was an NXP RFID ASIC (Application Specific Integrated Circuit) mounted on copper straps for contact with the RFID metal antenna. The NXP chip (NXP Semiconductors, Stockport, UK) had a high frequency input impedance consisting of a resistive part, 15 Ohms, and a capacitive component equal to 128 Ohms at the RFID frequency. The RFID integrated circuit was connected to the metal antenna by direct ohmic contact and mechanically attached to the antenna by proprietary adhesive tape. The ohmic contact was achieved by applying point pressure to the copper straps and the underlying printed conductor.

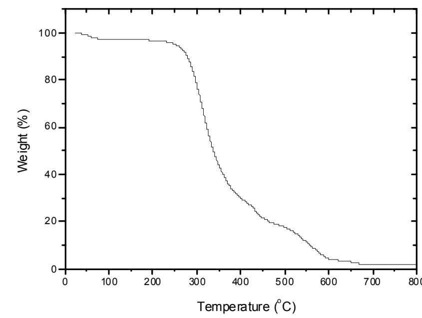
Equipment

Inkjet printing was performed using a piezoelectric Dimatix DMP-2800 system (Dimatix-Fujifilm Inc., USA), equipped with a 10 pL cartridge (DMC-11610). The nozzle plate consists of a single raw of 16 nozzles of 23 μm diameter spaced 254 μm. The print head contains a row of 16 nozzles. Printhead height was set to 1 mm and the printer platen temperature 50 ^oC unless otherwise stated. Thermal sintering was carried out on a hot plate. Sintering temperature was 135 ^oC unless otherwise stated. Sintering times between 15 and 120 minutes were used to study resistance as function of sintering time. Photonic flash sintering was carried out in our custom-made apparatus (the details are disclosed in [22]). The inner surface of the elliptic reflector was covered by a highly reflective foil (reflectivity >98%). The Xenon flash lamps (XOP-15, Philips, The Netherlands) have a maximum power of 1000 W, an emission spectrum ranging from 350-900 nm. Low pressure argon plasma sintering was performed using a low-pressure argon plasma instrument form Diener Electronic (Nagold, Germany). The power was wet at 300 W. Thermogravimetric analysis (TGA) was performed under ambient atmosphere in the range from room temperature to 600 ^oC with heating rate of 10 ^oC min^{-1} using a TGAQ5000 system from TA Instruments. Differential scanning calorimetry (DSC) measurements were recorded using a Perkin Elmer Jade DSC instrument under nitrogen atmosphere, 5–10 mg of the sample was sealed in an aluminum pan with a crimping tool. The sample was heated from 25 ˚C to 200 ˚C at a heating rate of 10 ˚C/min, held for 5 minutes at 200 ˚C and then cooled to 25 ˚C at a rate of 10 ˚C/min. This cycle was repeated three times. The electrical resistance of sintered silver features was measure by the 4-point probe technique. A Jandel multi-position wafer probe system (Jandel Engineering Ltd., Leighton Buzzard, UK) mounted with a cylindrical probe head (solid tungsten carbide needles of 0.40 mm diameter spaced 1.0 mm) was employed. Surface topography, thickness, and cross-sectional areas of the printed silver tracks were measured with an optical profilometer (Dektak Veecko32). Scanning electron microscopy (SEM) images were taken using a EVO®LS 15 system (Carl Zeiss Microscopy, Germany) operating at an accelerating voltage of 20 kV. Tag read distance was measured using a Voyantic Tagformance lite RFID measurement system (Voyantic Ltd., Finland). After calibrating the system at 35cm the RFID tag was transferred to the volunteers arm and the read range was extrapolated by measurement at the global RFID UHF frequency bands within permitted transmission power levels.

Results

This study was started with the analysis of the structure and thermal stability of the paper used in this work. A schematic of the various layers of tattoo paper and the steps to produce and transfer an RFID tattoo tag on human body are described in Figure 2. As shown in Fig. 2b, the thermogravimetric analysis (TGA) of the ink receiving layer of tattoo paper shows an initial mass loss at approximately 100 ^oC attributed to the loss of adsorbed moisture, with no further mass loss observed until between 225 and 685 ^oC. The various mass loss steps between 225 and 685 ^oC are attributed to the decomposition of the various components within the ink





Figgure 2 (a) Schematic of the formation and transfer of an inkjet printed RFID tag onto the body. i) The RFID antenna pattern is inkjet printed onto the ink-receiving layer. Upon sintering, the IC chip is attached to the ports of the antenna in order to form the RFID tag. Then, ii) the adhesive sheet with protective coating is applied to the printed RFID tag, iii) the protective coating is removed, iv) the tattoo is flipped and applied to the skin and moistened with water, and v) the base and back coat paper layers are removed.(b) thermogravimetric analysis of transfer paper.

receiving layer. This suggests that after an initial dehydration step of 8 wt-%, decomposition does not begin until 225 ^oC. Transfer tattoo paper shape and color changes were visually assessed as a function of temperature. At 150 ^oC within less than 15 minutes tattoo paper changed from white to a yellow-brownish colour, however at 140 ^oC to the naked eye there was no change in color or in shape during an 8 hours period. Based on these results, unless otherwise stated, 135 ^oC was adopted as the sintering temperature in this work.

Upon optimizing inkjet printing conditions to obtain good image quality of the silver nanoparticle inks on tattoo paper their electrical performance was investigated quantitatively, and for comparison, inkjet printing onto glass substrate was also carried out. It was found that the resolution and electrical performance of simple printed features is dependent on the ink, printing parameters and sintering technique. Optimal parameters to obtain conductivy features for each ink-printing condition-sintering technique combination were obtained.

To demonstrate the potential applicability of inkjet printing as a powerful tool to fabricate low cost passive UHF RFID tags on tattoo paper, RFID antennas of the structure depicted in Figure 3 were inkjet printed. Nominal antenna dimensions (in mm) are: L = 65, W = 20, l = 14.5, w = 3 and t = 0.5. Details of fabrication by etching and stencil printing means, design considerations, simulated electrical flow within the antenna and their read range performance upon transfer onto skin have been reported somewhere else [5]. An image of an inkjet printed RFID tattoo tag upon transfer onto a volunteer's arm is shown in Figure 3 (right).

A first iteration of inkjet printed antennas comprising 1, 2 and 3 ink layers (named 1, 2, 3 full antenna) were produced. Sintering was performed when all ink layers had been deposited. After sintering, antenna port-to-port resistance (R_{p2p}) was measured. In order to fabricate the tag, a commercially available integrated circuit (IC) chip of specific impedance (14.8+j128 Ohms) was manually attached to the ports of the antenna. Subsequently, following the procedure described in Fig. 1a, the tag was transferred onto a volunteer's arm, as shown in Figure 3. Maximum read distance of the RFID tag upon transfer was measured (initial read distance). Read distance was evaluated every hour whilst the volunteer engaged in routine office activities for a period of 8 hours, a standard working day. In terms of tag performance over time, the majority of the tags tested on the volunteer's arm did not change performance (read range distance) within an eight hour measurement regime. This result indicates that tags were mechanically robust enough to be used in a real life environment.

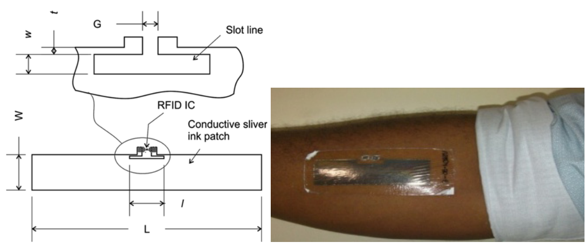


***Figure 3.** RFID tag schematic (left). Inkjet printed RFID tag upon transfer onto the body (right).*

Both, antenna image quality and R_{p2p} have an impact on the read distance. Better image quality (good antenna edge definition) improves energy coupling between the antenna and the RFID integrated circuit chip while lower R_{p2p} of the metal antenna will reduce resistive loss to heat, and hence the tag read distance is expected to increase. Therefore the lower resistivity associated with thicker conducting layers is beneficial due to increased read efficiency and the R_{p2p} of inkjet printed antennas could potentially be decreased by increasing the number of printed layers. However, this will increase the volume of ink employed to fabricate the antenna resulting in more expensive inkjet printed tags. In addition to this, we also aim to minimize tag thickness to make it less intrusive to the end user and facilitate over printing by conventional artwork. Preliminary sintering results show that argon plasma sintering of RFID tags with only one layer of Suntronic ink results in R_{p2p} as low as that obtained for thermal sintering, hence potentially decreasing RFID fabrication costs due to the fewer ink needed to fabricate the tag. On the other hand, preliminary FSS work using transfer paper shows that photonic sintering processing lasting a few seconds can yield FSS dipoles of the same resistance that dipoles thermally sintered for 30 minutes. Furthermore, preliminary FSS photonic sintering work was carried out using a sheet-to-sheet system suitable for large area manufacturing.

Conclusions

In summary, we have demonstrated that by choosing the appropriate ink, printing settings and sintering technique FSSs and RFID transfer tattoo tags functional in a real life scenario can be produced by inkjet printing means. Furthermore, by selectively depositing ink on specific areas of the tag, the read distance of inkjet printed tags was substantially larger than the read distance of stencil tags and approximating that of etched tags. In addition to this, two strategies have been demonstrated to reduce the volume of ink used per tag: i) selective ink addition in specific areas of the tag and ii) optimisation of argon plasma sintering of thin tags. Furthermore, inkjet printed tags are much thinner than etched or stencil printing tags thus, making them less noticeable, more comfortable and therefore more appealing to the end user. Following these initial encouraging results we believe inkjet printing of passive UHF RFID transfer tattoo tags and FSSs can potentially be used to mass-produce tags at a commercially viable price. Further work is now underway to step towards commercialization. This involves modifying tag and FSS designs and optimizing ink, printing and sintering processes in order to improve device performance, make the process compatible with large-scale production and reduce costs to realise commercialization.

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