INKJET PRINTING ON PAPER FOR THE REALIZATION OF HUMIDITY AND TEMPERATURE SENSORS

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ABSTRACT

We report on inkjet printing of silver nanoparticles on paper for the design of resistive temperature and capacitive humidity sensors. Temperature and humidity have shown to have an influence on the electrical and mechanical properties of the Ag printed structures on paper. The passivation of the paper substrate and the Ag structures with parylene was investigated to reduce these effects and improve the stability of the structures. The lines were printed with an average thickness of 0.8 µm and a resistivity of 30 µΩ·cm. The Ag thermometer coated with parylene showed a good linearity with a TCR of 0.0011°C^{-1}, while the capacitive humidity sensor exhibited an exponential response. This study foresees the possibility of using paper as substrate with inkjet printing of silver for the design of low-cost environmental sensors.

KEYWORDS

Inkjet printing, silver ink, paper, humidity sensor, temperature sensor, parylene, passivation.

INTRODUCTION

Lately, flexible substrates were identified for manufacturing sensors at lower cost than silicon. The combination of additive fabrication techniques (inkjet, gravure, flexo printing) and roll-to-roll fabrication can lead to the production of inexpensive devices. To further reduce the fabrication cost, paper combined with printing has recently been used instead of plastic [1]. A force sensor with carbon and silver inks screen printed on paper [2] and the fabrication of PCB on paper [3] were already presented. Chemical sensing was also demonstrated with the measurements of ethanol on glossy paper with ITO as sensing layer and carbon-based electrodes [4], and a capacitive humidity sensor fabricated on polymer-coated cellulose films [5].

Here, we report on the evaluation of inkjet printing of silver nanoparticles on paper for the design of temperature and humidity sensors. The inkjet printing parameters were studied to reach reproducible and well defined patterns. The electrical properties of the Ag film were examined. Due to instability of the Ag lines on paper, notably in presence of humidity, passivation of the paper substrate itself and of the printed structures using a thin layer of parylene was evaluated. The inkjet printing of silver was then used to pattern resistors and capacitive structures for sensing temperature and humidity, respectively. For capacitive humidity sensing, a differential measurement method was implemented around a low-power microcontroller. It aimed at reducing the drift and response time as reported in our previous work on polyimide foil [6]. It involved two capacitors, one reference left uncoated and a polymer-coated capacitor for humidity sensing.

Besides lowering the cost, by processing the sensors with large scale compatible techniques, one can imagine the direct integration of the sensors on different media and packaging items made of paper and cardboard.

INKJET PRINTING OF SILVER ON PAPER

Material and method

The substrate used was a paper for printed electronics, the *pe:smart paper type 2* from *Felix Schoeller*. Its thickness and density were 205 µm and 200 g/m^2, respectively. It has a nanoporous coating made of an oxide film to improve inkjet printing. The roughness of the paper was measured by white light interferometry (*Wyko NT1100* from *Veeco*) and was of 3.3 nm. Prior to printing, the paper was dehydrated at 110°C for 2 h and no surface pretreatment was performed.

Silver nanoparticle ink (*DGP 40LT-15C* from *Anapro*) was inkjet printed with a commercial drop-on-demand *Dimatix DMP-2831* printer with a 10 pL cartridge. Test patterns included 100, 200 and 300 µm wide lines for both dimensional and electrical characterization. Different curing times were carried out to determine its impact on Ag resistivity. It was performed at 150°C as recommended by the ink manufacturer and corresponded to the highest temperature specified by the paper supplier. The resistance of the Ag lines was measured at different temperature and humidity levels.

Results on inkjet printed silver structures

The optimum printing parameters were a jetting frequency of 5 kHz and a meniscus vacuum set to 3 in. of H_2O. The voltages applied to the piezoelements of the printhead were tuned to obtain a drop velocity of 8 m/s. The printhead temperature was set to 40°C and the stage to 30°C to avoid coffee ring effect [7]. These parameters led to a printed drop on paper with a diameter of 40 µm. A drop spacing set to 20 µm gave the best printed lines as shown in Figure 1, where smooth edges were obtained. One Ag layer was printed. Due to wettability of the ink on the paper substrate, wider printed lines than designed were obtained. Their width was increased by 40 µm compared to the layout. Minimum line width of 140 µm and gap in between of about 150 µm were achieved. Figure 2 shows the line profile measured by white light interferometry. It had an average thickness of 0.8 µm with no coffee ring effect observed. Ag showed a very good adhesion to paper with a scotch tape test (green *Scotch*

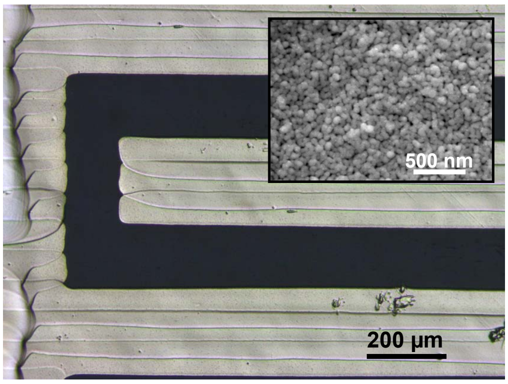

Figure 1: Optical picture of inkjet printed lines of Ag nanoparticles on paper. Inset: SEM picture of the printed silver.

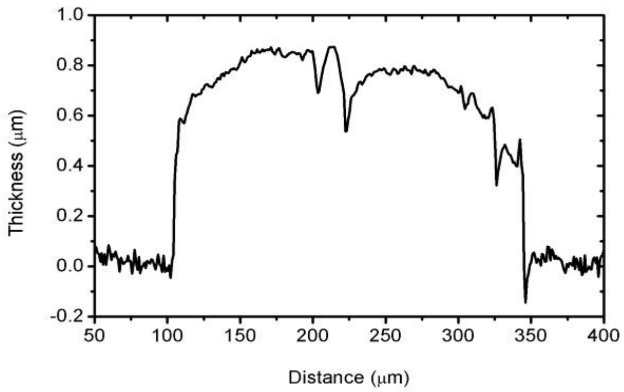

Figure 2: Cross-section of a printed Ag line obtained by white light interferometry. The average thickness of the silver line was 0.8 µm.

tape from *3M*).

The resistivity of Ag for curing times between 5 and 60 min was measured. The results are presented in Figure 3. The resistivity decreased with curing time. However, at a time higher than 30 min at 150°C, the paper started to turn yellow. We therefore defined the curing time to 30 min for which a resistivity and a sheet resistance at room temperature of 30 µΩ·cm and 0.3 Ω/□ were obtained, respectively. SEM observations revealed a porous structure of the sintered Ag ink (Fig. 1). The size of the grain was about 70 nm.

Figure 4 shows the behavior of printed Ag on paper with temperature. It was varied from 20°C to 100°C at 30% RH (relative humidity). One observed a strong degradation of the resistance with an increase for temperature higher than 60°C.

The resistance variation of printed silver lines was then tested between -20°C and 60°C at 30% and 70%RH. The results are presented in Figure 5. A slight hysteresis was observed at low humidity level and it increased drastically with the humidity going up. The inkjet printed Ag on paper has shown to be more stable for temperatures in the range of -20°C to 60°C and at low humidity levels.

Figure 6 shows the response to humidity of a silver capacitive structure printed on paper for relative humidity

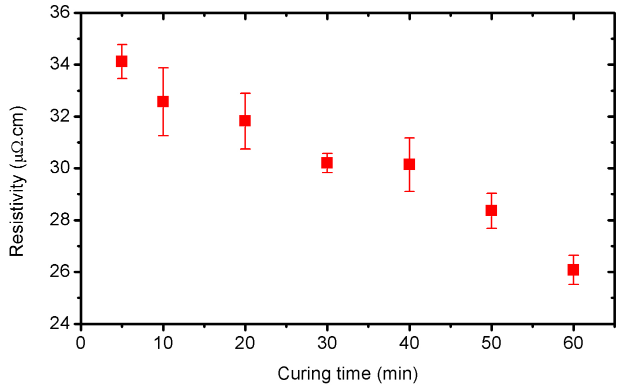

Figure 3: Resistivity of the inkjet printed Ag on paper as a function of the curing time.

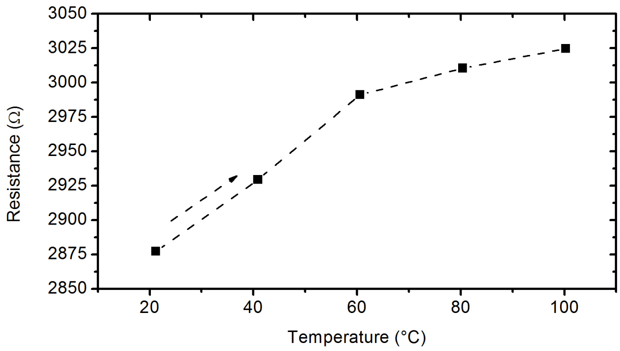

Figure 4: Resistance of a line of Ag printed on paper for temperature between 20 and 100°C at 30% RH. TCR changed for temperature higher than 60°C.

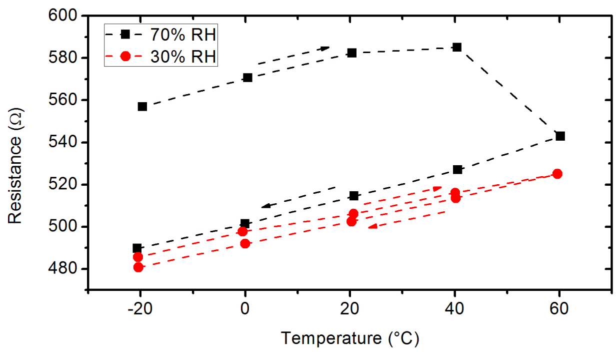

Figure 5: Resistance of printed Ag on paper for temperature between -20°C and 60°C at 30 and 70% RH. An increase in RH caused a strong variation in resistance.

levels between 20 and 80%. An exponential response was obtained with a strong increase of the capacitance for humidity above 60%, very likely due to the highly porous nature of the substrate. The capacitance increased 23 folds between 20 and 80% RH. When exposed at high humidity level (> 60%RH), the paper irreversibly bent.

These results show that there are significant interactions between humidity and the paper substrate especially at high humidity levels. This suggests the need for coating layers to passivate either the substrate or the printed silver structures, or both.

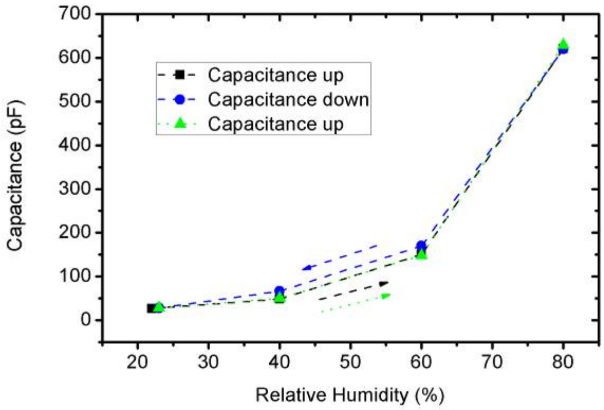

Figure 6: Response of the uncoated printed capacitor to relative humidity. An exponential response was obtained.

SENSORS ON PAPER SUBSTRATE

Design

Once the printing parameters and properties of the printed silver were defined, sensing platforms were designed with the inclusion of a parylene coating, 2 µm thick, as a passivation on the substrate and over the printed silver structures.

They consisted in one resistive thermometer and two capacitors, one as a reference and one functionalized for humidity sensing (Fig. 7). Each sensor covered an area of 16 x 16 mm^2. The designed line width and gap in between of the electrical conductors were 200 µm. These dimensions showed to be highly reproducible and avoided short circuits between parallel lines. The targeted resistance of the thermometer was between 500 Ω and 1 kΩ and the capacitance of the capacitors was about 20 pF.

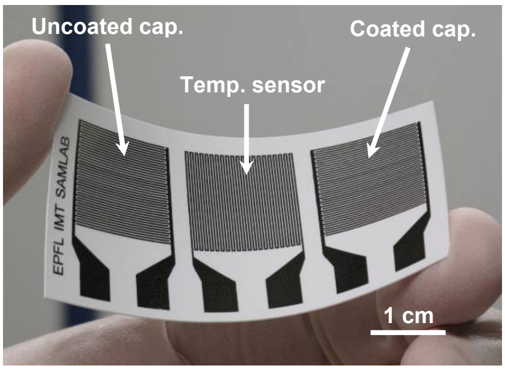

Figure 7: Optical picture of the inkjet printed capacitors and resistor on paper.

Characterization

The operation of the resistive thermometer and the capacitive humidity sensors were evaluated in a climatic chamber (*Espec*). The temperature was varied between -20°C and 60°C and the RH between 20% and 80%. During all experiments, the temperature and relative humidity were monitored with a *SHT15* sensor from *Sensirion AG*. The resistances were measured with an *Agilent 34411A* multimeter and the capacitances with an *Agilent E4980A* LCR meter. They were performed at 30 kHz, which was proven to be suitable for such sensors [6,8].

RESULTS

Resistors as thermometer

The response of the thermometers is shown in Figure 8. The resistor printed on paper had a nominal resistance of 670 Ω at 20°C. The TCR was measured at 30% RH. Its value was 0.0011°C^{-1} with a coefficient of determination to a linear fit of 0.992. The deposition of the parylene coating increased its nominal resistance to 740 Ω, very likely because of the penetration of parylene within the porous Ag. The passivation improved the linearity of the thermometer to 0.9999, while the TCR remained the same. Furthermore, no hysteresis to temperature cycles was observed for the passivated device.

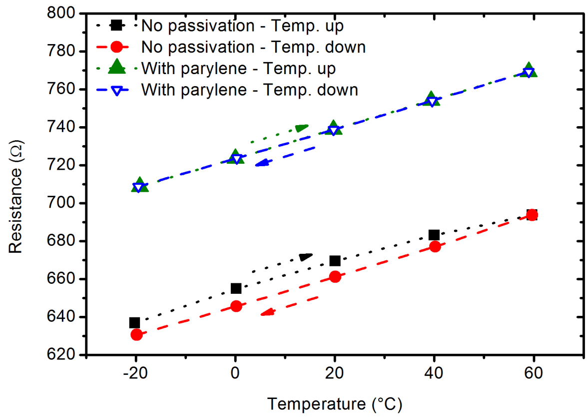

Figure 8: Thermal behavior of the inkjet printed silver resistor between -20 and 60°C with 30%RH. It showed a thermal coefficient of resistance of 0.0011°C^{-1} and a linear coefficient of determination of 0.9999 for the parylene-coated thermometer and 0.992 for the uncoated one.

Capacitors as humidity sensor

The capacitors had a nominal capacitance of 23 pF at room atmosphere, with and without the parylene coating. The functionalized capacitor was drop-coated with a CAB (cellulose acetate butyrate) layer after activating the surface with a corona treatment. The CAB increased its values by about 7%. Figure 9 shows the capacitance difference between the CAB-coated and uncoated capacitor used as reference. It increased from 1.6 pF at 20% RH to 4.3 pF at 80% RH. The capacitive sensors showed a very long response time independently of the parylene coating. The latter had the advantage to limit the mechanical deformation of the device after exposure to high humidity levels.

Differential measurement of the capacitive sensors

A readout system based on time-to-digital conversion [9] was implemented around a *MSP430* microcontroller from

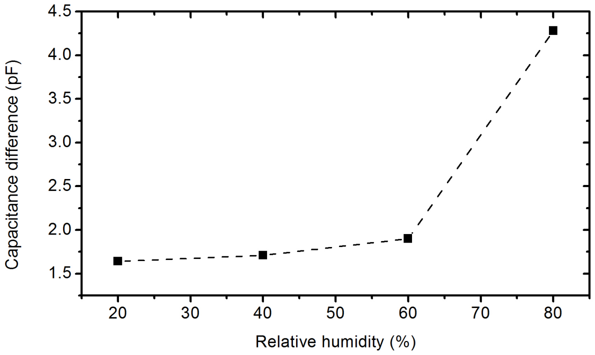

Figure 9: Capacitance difference between the CAB-coated and the uncoated capacitor when exposed to RH levels between 20 and 80%.

TI to measure the capacitance difference between the CAB-coated and the reference capacitors. This technique has several advantages as demonstrated in ref [6]. By subtracting the two humidity sensitive capacitor signals, the influence of the substrate can be removed, the linear response is improved and the response time is significantly reduced.

Figure 8 shows the differential response of the capacitive sensors on paper when interfaced with the electronic circuitry and tested in a custom-made gas mixing system where humidity level can be changed rapidly.

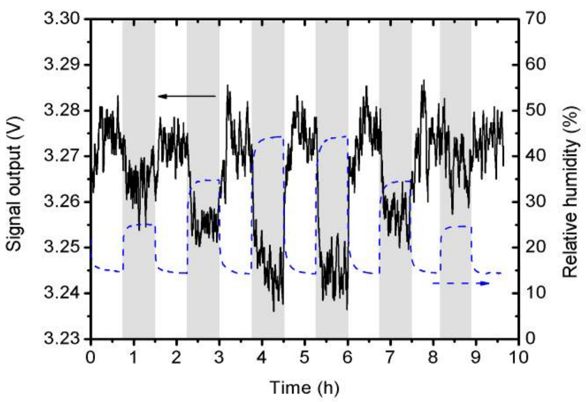

Figure 9: Differential humidity measurement performed with the capacitive sensors on paper. The relative humidity was monitored with a SHT15 sensor from Sensirion AG.

CONCLUSION

Inkjet printing of Ag was performed on paper substrate. Printing parameters were tuned and led to uniform and smooth Ag lines. After sintering at 150°C for 30 min a resistivity of 30 μΩ·cm was measured.

Passivation of the thermometer with parylene improved its linearity and minimized the hysteresis in the range of -20°C to 60°C. The TCR of the resistive sensor was 0.0011°C^{-1}, making suitable for temperature measurement. Capacitive sensors exhibited an exponential response with humidity level. Besides using the differential measurement, the passivation of the paper prior to printing will be investigated in order to reduce the strong response of the substrate to humidity. Additionally, work is still required to optimize the chemical capacitive sensors by optimizing the sensing layer, for humidity but also other gases. Passivating the silver electrodes for an improved stability under the presence of humidity might also need to be considered depending on the long term behavior of the devices, especially when exposed to the higher temperature and humidity levels.

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