Printed electrodes structures as capacitive humidity sensors: A comparison

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Abstract- This work discusses about four planar printed capacitive sensors with different geometrical layouts, fabricated by inkjet printing on a flexible substrate and used as humidity sensors. In particular, we show a comparison among interdigitated electrodes, meandered electrodes, spiral electrodes, and serpentine electrodes in terms of fabrication yields, sensitivities to relative humidity as well as thermal drift taking into account frequency dependencies. In addition, numerical simulations have been performed to further investigate the characteristics of these sensors. All sensors present similar behavior in frequency with humidity. Taking into account sensitivity within the same sensing area, the highest value is achieved by serpentine electrodes, followed by spiral electrodes, interdigitated and meandered electrodes, in this order. The best configuration will be dependent on the specific application and its requirements.

Keywords: *interdigitated electrodes; serpentine electrodes; spiral electrodes; meandered electrodes; inkjet printing; capacitive humidity sensors; comparison*

1. Introduction

Capacitive structures are widely used in electronics to address many different applications [1]; they are especially interesting in the field of sensors due to their characteristics, such as low energy consumption, non-intrusive and non-invasive, no radiation and fast response [2, 3]. The most common sensing capacitive structures are the parallel plate (PP) and the interdigitated electrode (IDE). PP is characterized by the simplicity of its geometry and the ease of calculation and modelling.

IDE sensors are a particular case of planar capacitive structures where the sensor electrodes are placed in a co-planar plane [4]. The planar structure allows to access the device from only one side [5], which is particularly useful when the access to an material under test (MUT) is limited or the other side should be open to the ambient. The advantage of this kind of structures is the fact that they can be fabricated on a substrate by deposition of a layer without including any step of micromaching. That allows compatibility with any kind of technology. Furthermore, this geometry has been used to fabricate with multiple materials and following different manufacturing process, from integration in semiconductor dices to printing on flexible substrates [6-8].This additional feature makes planar capacitive sensors an attractive option for applications in material characterization [5], non-destructive testing (NDT) [9], proximity/displacement measurement [10], intelligent human interfacing [11], and imaging [12, 13]. Many efforts have been devoted to the theoretical modelling of the planar capacitive structures. Igreja et al. [14] presented a theoretical model of the capacitance of IDE structure. These capacitors have also been simulated using different simulations tools [15-17]. Some authors have analysed other designs such as spiral electrodes and concentric rings in order to improve the performance of this design [7, 18, 19]. Other structures such as rectangular-shaped [20] and comb-shaped sensor arrays [21] were studied, showing that desired linearity and sensitivity can be achieved by the optimal selection of a set of structural parameters. Zeothout et al. [22] analyzed a rectangular shaped planar sensor using a numerical method and also compared its performance according to different material properties and boundary conditions.

On the other hand, flexible electronic devices manufactured by printing techniques have become increasingly attractive thanks to their feasibility of large scale processing, potential low-cost per surface area and mechanical flexibility. Great advances have been achieved in the design of flexible and printed humidity sensors [23-26] as well as sensors for other gases and vapours [27-29]. The classical transduction mechanism of these humidity sensors is capacitive due to the requirement of low energy consumption, in particular, through changes in the electrical permittivity of some structural layer of the capacitor or the dielectric thickness. Different printed techniques have been used to manufacture capacitive printed devices and different approaches have been followed to add the sensing capability into the capacitor. The most common strategy has been to deposit a sensing layer over the electrodes [16, 30, 31] such as cellulose acetate butyrate (CAB), poly(methyl methacrylate) (PMMA) or polyvinylchloride (PVC), among others.

Another possibility has been to select a flexible substrate as sensing element saving fabrication steps, in this sense, polyimide [26] and photographic paper [25] have already been used. One of the main interfering factors to obtain an accurate humidity measurement is temperature. In order to compensate this dependence, several strategies have been already described such as differential measurements with reference capacitors (not sensitive to humidity) [32], including additional temperature sensors [33, 34] or using a sensitive layer with very low thermal drift [35, 36]. This latter alternative does not require additional devices, and therefore, less area and energy is consumed.

In this work, we will show the design, fabrication and characterization as humidity sensor of four different coplanar electrodes comparing and contrasting their characteristics. In this regard, we present the design, fabrication and characterization of capacitive humidity sensors which uses the flexible substrate as sensitive element. These capacitors have been printed with silver nanoparticles ink by inkjet-printing on a polyimide thin film. In the context, this paper discusses planar printed capacitive sensors in terms of fabrication yields, sensitivity to relative humidity as well as thermal drift taking into account frequency dependencies. Further investigations of the sensor designs have been carried out using a numerical method. The differences among these planar capacitive sensors are pointed out as well as their advantages and disadvantages of the different electrodes designs.

2. Materials and methods

A. Fabrication process

The DMP-2831™ Dimatix printer (Fujifilm Dimatix Inc, Santa Clara, USA) was used for inkjet printing. The selected materials were an ink of silver nanoparticles (U5603 SunTronic Technology, San Diego, USA) on a polyimide substrate (Kapton® HN with 75 µm of thickness, Dupont^{TM}). Table 1 shows the main properties of the used ink and substrate, respectively.

According to the manufacturer of the substrate, the relationship between the relative permittivity (ε_r) and the relative humidity (RH) is given by:

$$\varepsilon_r = 3.05 + 0.008 \cdot RH(\%) \quad at\ 1\ kHz, 23\ ℃ \qquad (1)$$

The first step before printing was to prepare the substrate removing all traces of particles with a cleaning process to ensure the best quality and to avoid failed printings. First, the substrate was immersed in acetone for 2 min to remove dust on the surface, and then was submerged in propanol about 2 min to remove the acetone. After that, the substrate was washed with purified water to eliminate the propanol and finally was dried at 120ºC for 5 min. During the printing process the substrate temperature was fixed at 40ºC and a drop space of 25 μm was settled for 50 μm landed diameter drops. Finally, a sintering step is carried out at 120 ºC for 60 min.

In order to know the thickness of the printed layer, the theoretical model given in [37] has been used. According to this model, the amount of used ink is 22.7 nl for one printed layer with a thickness of 460 nm. With that printing and curing conditions, the resistivity of the conductive electrodes were 23 ± 2 μΩ·cm [37]. The fabrication time is much shorter than in the case of other sensors because no other sensing layer was needed [16, 30, 31]. The fabrication process is also simplified because it only required printing one layer on one side of the substrate. In a previous work, we demonstrated that thickness layer does not contribute significantly to capacitance and therefore the definition of electrodes by only one printed layer is enough for a proper sensor performance [35]. A matrix of twenty capacitors of each type was fabricated in order to test the reproducibility of the process.

B. Physical and electrical characterization

The geometrical characterization, the roughness of printed patterns as well as the thickness of the patterns have been measured using a Wyko NT1100 Optical Profiling System (VEECO, Tucson, AZ, USA), and a Dektak XT™ Stimulus Surface Profiling System (Bruker Corporation, Conventry, UK).

The AC electrical characterization for the different fabricated capacitors has been performed by measuring their capacitance and dissipation factor, using the four-wire measurement technique with a precision Impedance Analyser 4294A and an impedance probe kit (4294A1) (Agilent Tech., Santa Clara, CA, USA). The excitation voltage applied in all measurements was $V_{DC} = 0$ and $V_{AC} = 500$ mV. The frequency sweep of analysis was from 100 kHz to 10 MHz. In all prototypes, one of the end sides has been enlarged to facilitate its connection to any analyser. A SMA (SubMiniature version A) male connector has been glued to these end~~s~~ points using silver-filled epoxy EPO-TEK® H20E (Epoxy Technology, Inc., Billerica, USA). A complete compensation method has been implemented to eliminate the contribution of parasitic capacitances by measuring several commercial capacitances placed in the same configuration as the devices under test with the 16034G surface mount device (SMD) Test Fixture (Agilent Tech., Santa Clara, CA, USA).

The stationary humidity and temperature responses of the sensors have been measured in a climatic chamber VCL 4006 (Vötsch Industrietechnik GmbH, Germany). The humidity deviation in time was ± 1% to ± 3%, whereas the temperature deviation in time was ± 0.3 °C to ± 0.5 °C. The data acquisition and analysis have been automated using Labview 2012 software (National Instruments Corporation, Texas, USA). Due to the slow response of the mentioned climatic chamber, the dynamic response has been measured in a customized humidity measurements set-up (9 x 3 cm^2), which automatically controls wet and dry airflow inside a small gas cell at room temperature. Two LCR-meters (HP 4284L and Agilent E4980A, Agilent Tech., Santa Clara, CA, USA) have been used to measure the corresponding capacitance values at a frequency of 100 kHz every 5 s. Capacitance and time measurements of the printed sensors have been controlled and recorded using the software Labview 2012. RH and temperature measurements have been also registered by a commercial sensor (SHT15, Sensirion AG, Switzerland) in order to verify the data given by the chambers' displays. In all cases, the capacitors have been placed in the middle of the climatic chambers allowing the atmosphere interaction in both faces of the sensors, printed and non-printed.

C. Electrodes designs

We have analysed four different electrodes configurations as capacitive sensor. In this section, we present the geometry of each device, pointing out the dimensions and areas of the fabricated capacitors.

Interdigitated electrodes

First, interdigitated electrodes (Figure 1a) have been studied. As we mentioned before, this structure is the most common one to develop planar capacitive sensors thanks to its compromise between ease of manufacturing and performance. Table 2 presents the dimensions of the modelled IDE sensor, resulting in a designed area of 11.65 mm^2 (L = 1.85 mm × W = 6.3 mm) and a capacitance of 1.97 pF at dry atmosphere calculated by numerical simulation.

Meandered electrodes

Meandered electrodes have been developed as capacitive sensor as shown in Figure 1b. Although this geometry shows at first glance lower contact area than the previous ones, it could result interesting in some applications, as we will describe in the following section. The designed sensor structure area is 89.0 mm^2 (L = 4.00 mm x W = 22.25 mm) following specifications shown in Table 3 and the numerical capacitance is 3.14 pF at dry atmosphere.

Spiral electrodes

Another studied structure has been the spiral electrodes. This structure has been previously reported by [18, 38] to achieve a high compactness. In this case, the electrodes are placed as a coil inductor (Figure 1c). Table 4 shows the physical dimensions of the modelled spiral capacitor with a total area of 5.52 mm^2 (L = 2.35 mm x W = 2.35 mm) and numerical capacitance of 0.91 pF at dry atmosphere.

Serpentine electrodes

Finally, this periodic electrode structure consists of a combination of meandering and interdigitated electrodes in a serpentine-shaped geometry [36] (Figure 1d). Serpentine electrodes (SRE) were previously used as electrode guard of IDE structures [39], impedance sensor for conformal skin hydration monitoring [40] or a three phase electrode array for AC electro-osmotic flow pumping [41]. SRE geometry is describe in Table 5, covering a total area of 11.65 mm^2 (L = 1.85 mm × W = 6.3 mm). The numerical capacitance of this structure is 2.26 pF at dry atmosphere.

3. Results and discussion

A. Simulation results

With the aim of structure optimizations and after different simulations varying the fundamental geometrical parameters, we have used COMSOL Multiphysics 4.2a (www.comsol.com, COMSOL, Inc. USA). This is a powerful interactive tool for solving problems based on partial differential equations with the finite element method. This software has previously been used to calculate distributions of potential field in this kind of structures [17, 34]. The equations solved in the determination of the capacitance in each finite element are:

$$-\nabla\varepsilon_0\varepsilon_r\nabla V = \rho \qquad \text{(Eq. 2)}$$

$$\boldsymbol{D} = \varepsilon_0\varepsilon_r\mathbf{E} \qquad \text{(Eq. 3)}$$

where ρ is the charge density, D is the electric displacement, and E is the electric field. The electrostatic energy density needed to charge a capacitor is equal to that energy of the electrostatic field, and is given by:

$$W_e = \int_{\Omega} DEd\Omega \qquad \text{(Eq. 4)}$$

Then, the total capacitance is determined by the integral of the electric energy density through the relation:

$$C = \frac{2}{{V_{port}}^2}\int_{\Omega} W_e d\Omega \qquad \text{(Eq. 5)}$$

where V_{port} is the value of the applied voltage in the port of the sensor. The other electrode is connected to ground. The electrical parameters of the substrate given by the manufacturer and the printed and cured conductive silver ink according to our characterization were included in the numerical simulator [37].

The main results obtained are summarized as follow:

- If the sensing strategy is based on a sensitive substrate, the influence of the electrode thickness is virtually negligible in comparison with the other geometrical features. This result has been appreciated in all simulated structures.
- Keeping fixed gap between electrodes and their width, the bigger the structure, the bigger its capacitance and therefore, the sensitivity is enhanced.
- Regarding the capacitance in the same substrate area, simulations show that SRE presents the greatest value followed by spiral electrodes, IDE and meandered electrodes.

B. Physical characterization

Twenty samples of each type of electrode configuration were fabricated with just one printed layer. To experimentally check the simulated layouts, the replicas of each structure were physically characterized. According to a previously developed physical model of thickness layer by inkjet printing [16, 37], the calculated thicknesses are 430 nm for one printed layer. The measured dimensions of the structures are presented in Table 6 for IDE and SRE, Table 7 for spiral electrodes and Table 8 for meandered electrodes, showing the differences between measured and modelled dimensions. Uncertainties were calculated as one standard deviation of the experimental data. R_a is the arithmetic average of the absolute values, both of the surface roughness height and R_q is the root mean square (RMS).

In general, predicted and experimental dimensions are quite similar, although the bigger discrepancies are found in electrodes width and interspacing. As explained by Derbi et al. [42], printing electronics techniques present undesirable effects resulting in an inaccuracy on the printed structure, such as wider fingers and narrower inter-spacing. Table V summarizes the main features of each electrode configuration.

The yield rate was estimated from the low frequency capacitance measurements of the several replicas of each type of capacitor. During these tests, a preliminary manufacturing yield of 90% was found for IDE (including samples totally and partially broken) and meandered capacitors whereas lower values where found for spiral and serpentines electrodes, about 80%. This lower manufacturing yield is directly associated with the higher complexity of the latter configurations with respect to the former ones. It should be highlighted that in the case of IDE capacitors the device works when there is any broken finger, being the only effect a decreased in the total capacitance. In the other configuration studied, capacitors are totally useless when there is any printing error. Therefore, the manufacturing yield of IDE capacitors would be about 95% if we admit broken fingers. Comparing SRE and spiral electrodes, the manufacturing yield obtained is virtually the same but taking into account that spiral electrodes are about half area of SRE devices, this yield should be lower in the case of spiral electrodes if we have fabricated these capacitors with double area. An easy way to avoid the increase of failure is to connect in parallel this device, so that the equivalent capacitance is equal to the sum of individual values.

C. RH Sensitivity and Thermal drift

For five capacitors of each electrode configuration, the capacitances have been measured from 100 kHz to 10 MHz as a function of the relative humidity and temperature. In order to gain a better understanding of the whole sensor responses eliminating the area influence, we have defined the area-normalized partial sensitivities of the capacitance with the relative humidity, $S_{RH}(f)$, and the temperature, $S_T(f)$ as a function of the measurement frequency, f, as:

$$S_{RH}(f) \equiv \left. \frac{\partial C\ (RH)}{A \partial\ RH} \right|_{T=cte} \tag{2}$$

$$S_T(f) \equiv \left. \frac{\partial C\ (T)}{A \partial\ T} \right|_{RH=cte} \tag{3}$$

where A corresponds to the structure area. The humidity response has been calculated in a range from 20%RH to 90%RH at constant temperature of 40 ºC, while the thermal drift has been obtained in a range from 20 ºC to 50 ºC at constant moisture content (50%RH). The RH sweeps were performed in both directions in steps of 10%RH while temperature sweeps were performed in both directions in steps of 5°C.

Figure 2 shows the comparison of the sensitivity to relative humidity of each sensor as a function of frequency. All sensors show a similar trend in frequency, a relative higher value at lower frequencies and sensitivity trends to a constant value at higher frequencies. In terms of performance, SRE shows the highest normalized sensitivity values, followed by spiral electrodes and IDE. Meandered electrodes present the lowest value. For example, the normalized RH sensitivity at 1 MHz is 0.44 fF/%RH·mm^2 for SRE, 0.42 fF/%RH·mm^2 for spiral electrodes, 0.36 fF/%RH·mm^2 for IDE and 0.06 fF/%RH·mm^2 for meandered electrodes. Whereas the same sensitivities at 100 kHz is 0.52 fF/%RH·mm^2 for SRE, 0.48 fF/%RH·mm^2 for spiral electrodes, 0.38 fF/%RH·mm^2 for IDE and 0.06 fF/%RH·mm^2 for meandered electrodes. As can be observed, the sensitivity is virtually no affected by frequency in the case of meandered electrodes.

Looking at the displacement vector, the highest values are found in all the structures around the edges regions, with SRE and spiral electrodes being the designs that show more area at highest value due to the fact that they present more edges than the other two configurations (see Figure 3). This result can explain the capacitance values obtained. The electric displacement field varies the charge stored between the sensor electrodes and, therefore, alters the inter-electrode capacitance (see Eq. 3 to 5 above). This can be used to infer the properties of the material under test, in this case, the humidity sensitive substrate. These results agree with the study of the capacitance sensitivity as a function of the structure shape by Hu et al. [4] where meandered shapes (interdigitated and spiral electrodes) showed higher sensitivities.

Figure 4 presents the normalized thermal drift of each electrode design. Depending on the specific application and its requirements, we could choose a configuration or another. For example, SRE and spiral electrodes should be selected when sensitivity is a critical factor. In addition, depending on the shape where the sensor in going to be placed could be better the key requirement to select the electrode shape. But these two designs are more complex than the other two and, therefore, their yield rates are lower than the other two configurations.

IDE electrodes show the best compromise between sensitivity, area and manufacturing yield. Although meandered electrodes show the lowest performance, this configuration can be very useful to develop more complex sensor configurations such as double sensor as presented in [43]. Furthermore, the distance between electrodes in this latter design can be reduced and the performance could be closer to the other capacitors. In particular, the simulated capacitance obtained is 0.12 pF/mm^2 when width1 and width2 are 150 μm and 250 μm, respectively. This result improves four times the sensitivity obtained by simulations with the fabricated meandered sensor although lower than the capacitance obtained with the other configurations.

D. Time response and Time stability

The dynamic response of IDE and SRE sensors present a high stability along different measurement cycles and over time. On one hand, the response time can be defined as $t = \tau$, corresponding to the 63% of the maximum value of capacitance (reached at equilibrium) for every increasing step of RH. This time is equal to 356 ± 3 s and 350 ± 3 s for IDE and SRE, respectively. On the other hand, the response time for desorption, τ', can be described as the time associated to the 37% of the maximum value of capacitance for every decreasing step of RH; this value is 367 ± 4 s for IDE and 365 ± 4 s for SRE [35, 36]. These values have been calculated as the mean value of the sensor response to changes in RH from 15% to 45%, 15% to 65% and 15% to 85% at room temperature.

These times are virtually the same for both electrode configurations studied. The explanation to these long response times can found in the difference between diffusion rates in the solid substrate used compared to the deposited layers in other similar structures [16, 30]. Although these printed sensors can hardly compete in terms of response time with commercial CMOS-based humidity sensors (SHT15, Sensirion AG, Switzerland) with times smaller than 10 s, this lower performance in response time should not be a problem to use of these printed humidity sensors since humidity is an environmental property that often changes gradually. Furthermore, these times can be easily reduced by using a thinner substrate. Harrey et al. [44] presented a parallel-plate capacitive sensors with several humidity sensitive polymers including polyimide (Kapton HN). Indeed, they showed that the times responses achieved varied from 5 min to 10.5 min for Kapton HN depending on the thickness of the film. Anyway, all these sensors as they are presented here could be applied for environmental humidity monitoring, where variations are usually gradual and slow.

Regarding time stability, the IDE and SRE sensors have been measured once a week for more than 6 months and the variation between data has been less than 10 fF in both cases. In order to determine the aging drift, we performed a stability test 5 months after their fabrication. After the fifth month, sensors were tested for 10 days at fixed relative humidity (30%) and controlled temperature (30°C) every 6 hours. The aging drift was less than 3%RH for both sensors, which is within the time drift specification of the used climatic chamber.

For the other two designs, we have not measured their dynamic responses and either their time stabilities. As we have previously studied, the response time is directly related to the square value of the thickness of the sensitive layer [45]; as we have used the same substrate, the time response should be around 350 s. The time stability is directly associated with the sensing materials. Then, as all the materials are exactly the same, we can assume that the other sensors are expected to have a long lifetime.

4. Conclusions

In this work, we present the comparison between four different electrodes layouts: interdigitated electrodes (IDE), meandered electrodes, spiral electrodes, serpentine electrodes (SRE). These designs are planar electrodes developed in only one surface of the substrate. All these structures have been characterized as capacitive sensors, in particular, as humidity sensors. We show a comparison in terms of numerical simulations as well as experimental results. In order to measure the response of all the sensors to moisture content, we have fabricated them by inkjet printing using a silver ink on a polyimide substrate whose electrical permittivity is directly related to the relative humidity.

We have studied their responses to RH in a frequency range from 100 kHz to 10 MHz. Furthermore, we have looked into the thermal drift of each structure. In order to compare and contrast each response, we have normalized the values found taking into account the area occupied by each structure.

In terms of sensitivity to RH, all sensors show similar trends in frequency but the highest value is achieved by SRE, followed by spiral electrodes, IDE and meandered electrodes, in this order. Depending on the specific application and its requirements, we could choose a different configuration. For example, SRE and spiral electrodes should be selected when sensitivity is a critical factor. But these two designs are more complex than the other two and, therefore, their yield rates are lower than the other two configurations. Regarding IDE electrodes, it shows the best compromise between sensitivity a manufacturing yield. Finally, meandered electrodes can be easily adapted to more complex designs [43].

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