Micro Sized Interdigital Capacitor For Gases Detection Based On Graphene Oxide Coating

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Abstract. A micro sized interdigital capacitor sensible to CO_2 and NO is studied in this work. The photolithography technique enables to obtain fingers with dimensions of 10 × 500 μm and separated 7 μm between them. The deposition of a film composed of graphene oxide particles as the dielectrics of the capacitor allows to measure the gas concentration of CO_2 and NO mixed with N_2. The sensors were characterized in a gas chamber with a constant flow, obtaining promising results in changes of capacitance at 100 Hz. The sensors have a good linearity and sensitivity with a $R^2 = 0.996$ and $5.026 \cdot 10^{-1}$ pF/ % v/v for CO_2 and R^2=0.972 and $1.433 \cdot 10^{-1}$ pF/ppb for NO.

Keywords: Interdigital, Gas Sensor, Graphene Oxide, Photolithography

1 Introduction

The increase in popularity of trends such as internet of things and smart cities, has open up a path of new types of sensors [1,2]. Among them, the interdigital capacitor sensors are one of the most employed due to its advantageous characteristics such as small size, robustness and easy readout circuit [3]. Furthermore, the photolithography technique enables the miniaturization of the interdigital sensors, improving the performance of them [4] with smaller size and higher capacitance. Another advantage of this type of sensors is the incorporation of a great variety of dielectrics to the capacitor. These dielectrics are chosen for its sensitive properties to different factors such as temperature, humidity, presence of contaminants or gas detection, enabling the fabrication of sensors that cover a wide variety of fields [5]. Particularly, gas sensors play an important role in our daily live, employed in applications such as indoor air quality monitorization, city contamination, diagnosing diseases or controlling production processes among others [6].

Graphene oxide, (GO) has been proposed as the dielectric of the interdigital sensor. Nanomaterials based in graphenoids structures, with a single atomic layer of sp^2 hybridized carbon atoms offer a unique 2-D structure, thermal, mechanical, optical, and

excellent electronic properties [7]. GO creates an intrinsic dipole moment, is displaced by lattice vibrations and creates very high permittivity [8]. It is well known that the homogeneous dispersion of GO can satisfactorily increase the dielectric permittivity [9] .
In this work, the fabrication of interdigital sensors by the photolithography technique is explained. The incorporation of a film with a thin film of GO particles allows using the interdigital capacitor as a gas sensor, producing changes in the capacitance at different gas concentrations. The performance of the sensor to CO_2 and NO gases is studied since previous GO sensors have obtained a good response [10,11]. The results show a novel micro sized interdigital sensor with an extremely high sensitivity to NO at room temperature.

2 Materials and methods

2.1 Interdigital capacitor fabrication

The photolithography technique is employed in the fabrication of the interdigital capacitors. This technique enables the deposition of a metallic thin film in top of a substrate with a desired pattern design. In this case, a polypropylene substrate with a thickness of 250 μm has chosen due to its flexible property, but other substrates such as glass, silicon or polymers could be used.

The fabrication of the capacitor is composed of various steps that are summarized in Fig.1. First, the surface's substrate is cleaned with propanol and Kimtech™ wipes to remove dust and particles from it (Fig. 1.1). Cleanness is an important factor during all the fabrication process, since undesired particles can affect negatively in the results, causing short-circuits or open-circuits in the future electrodes. Therefore, the fabrication of the devices was conducted in a clean room.

On top of the cleaned substrate, an adhesive is deposited to obtain a good adherence for the next film. The Ti-Prime adhesive from the company MicroChemical GmbH was employed using the spin coating technique. In this technique, the centrifugal forces produced during the spinning process spread uniformly the solution forming a thin film. Few drops of the adhesive were placed to cover the substrate with the help of a pipette. The substrate was spined at 2,000 rpm during 30 seconds in the WS-650SZ-6NPP/Lite equipment from Laurell. A curing process was needed to fix the film into the surface, so the device was placed on a hot plate at 60ºC for 60 seconds.

In the next step, a film is deposited with the photoresist resin using the spin coating technique. The employed resin was AZ® MIR 701 14 CP from Merck at 100% concentration. The spinning process was first 10 seconds at 300 rpm and later 40 seconds at 2,500 rpm producing a yellowish color film (see Fig. 1.2). Also, the same curing process is repeated as it was done in the previous film. The resin is sensitive to UV light, so during the fabrication process the room was illuminated with a red light

Once the resin is deposited, the device is ready for the engraving of the desired design using the PicoMaster 100-4PICO Litho BV equipment (Fig. 1.3). The configuration parameters for the process are a 900 nm resolution, an exposure energy of 250 mJ/cm2 and a red power laser of 220 μW. After the engraving, the device is subjected again to

the curing process to fix and stabilize the film. In this part of the process the resin parts that were attacked by the laser lose the yellowish color and become transparent, exposing the engraving design to the naked eye.

In the next step, the transparent parts of the resin are removed from the device (Fig. 1.4). A solution is prepared combining the AZ 400K developer with deionize water in a 1:5 ratio and mixing in a magnetic stirrer for 1 hour. The device is immersed in the solution for 150 s, cleaned with deionize water and dried with compressed air. The immersed time of the device in the solution is a critical parameter since the attacked resin will not totally be removed with less time and with more time, the no-attacked resin will start to disintegrate. Again, the curing procedure is repeated (60º for 60 s).

Another film is deposited to build the electrodes, see Fig. 1.5. In this work silver was chosen as the material due its electrical characteristics and good adhesion to different substrates, although other metals could have been employed. The sputtering DC equipment K675XD from Quorum technologies was used for the deposition of the film with a thickness between 200 and 400 nm. The parameters of the procedure were 150 s at 35 mA and 7·10-3 bar.

After the metallic deposition, the resin layer is removed with the stripping process, see Fig. 1.6. The device is immersed in the solutions AZ100 from the company Merck for 10 minutes. Later the device is cleaned in soapy water for 2 minutes, and again in ultrapure water for 2 minutes. During the three immersions, the fluids and device are agitated in an ultrasonic bath. Later the device is cleaned with propanol and dried with a compressed air gun.

In the next step, two wires are soldered on each electrode of the device (Fig. 1.7). The conductive epoxy adhesive CW2400 from Chemtronics is employed. A curing process of 50 ºC for 10 minutes is needed to harden the adhesive. The capacitor is checked with a multimeter searching for short circuits. The presence of traces of silver that have remained between the electrodes can be removed connecting the capacitor to a high-power supply. The high currents passing trough the defects will evaporate them. Also, the device was observed trough an optical microscope DM 2500 M from Leica to detect visual mistakes in the fabrication process.

Finally, the GO layer is added. Graphene oxide (GO, 4-10% edge-oxidized) was purchased from Sigma-Aldrich (Barcelona, Spain) and used without further purification. GO (3.8 mg) was suspended in deionized H_2O (1 ml), filtered through a 200 μm filter to homogenize the solution. 500 μL of this solution was deposited on the interdigital capacitor (Fig. 1.8) to form a film of GO nanoparticles.

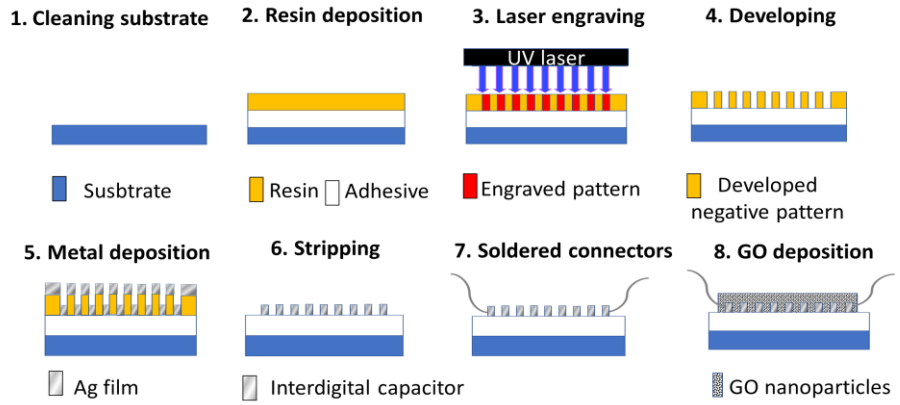

Fig. 1. Diagram of the sensor's fabrication process

2.2 Experimental setup

Fig. 2 shows the setup employed for the characterization of the sensor response to gases. The sensor is placed inside an ad-hoc gas chamber made in stainless steel and with an inner dimension of 14x2x3 cm. It has two tubes for the gas input and output that enable to maintain a constant flow through the chamber. A sealing gasket closes the chamber and prevent gases from scaping.

A gas panel is used to obtain the desired composition of gases. Various mass flow controllers of different gases and gas mixers lead to a single output connected to the gas chamber. The different flows of each controller enable to obtain different concentrations of gases.

The capacitance of the sensor is measured with the impedance analyzer E4990A from KeySight. The studied frequency range is between 10 and 100,000 Hz. The sensor is connected to the impedance analyzer using pass-trough cables into the chamber.

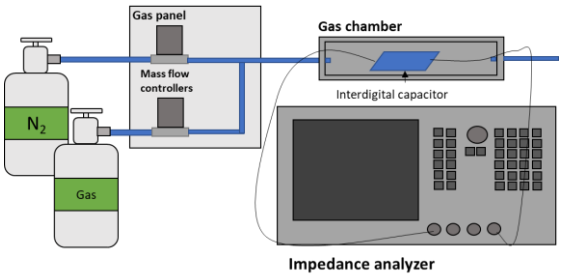

Fig. 2. Experimental setup for the characterization of the sensors to different gases

3 Results

The interdigital capacitor has been designed with a total dimension of 27x15 mm, see Fig. 3. It is composed by 560 interdigital blocks connected in parallel to increase the capacity. A gap of 100 µm was incorporated between the blocks to facilitate the removal of the deposited metal in the fabrication process. Each interdigital block has 16 pairs of fingers (making a total of 8,960 pairs) whose dimensions are 10x500 µm and are separated 7 µm between each other. Fig, 3c shows the clear gap between the metallic fingers that prevents any short-circuits in the capacitor.

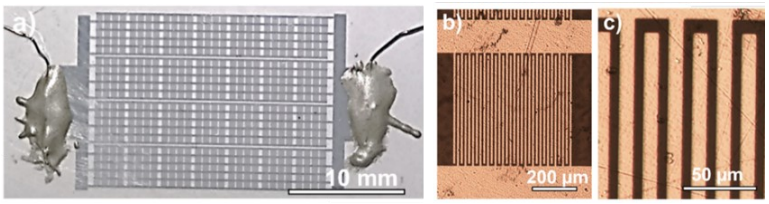

Fig. 3. a) Image of the sensor, b) microscope image of an interdigital block, c) microscope image of the fingers [12]

Once the interdigital capacitor sensor is coated with the sensitive GO it is characterized in the gas chamber trough different gas concentrations. Two gases were measured for this work, carbon dioxide, CO_2, and nitric oxide, NO. During the measurements, the flow of the mix of gases was keep constant at 0.2 L/min with nitrogen, N_2, as the gas carrier.

The different concentrations of CO_2 were done with different percentage between 0-100% of CO_2 in a mix of N_2 and CO_2. Fig. 4a shows the results of the capacitance at different concentration and frequencies. It can be seen that the capacitance of the sensor is linear to the gas concentration with R^2 higher than 0.99. Also, it can be noted that at lower frequencies the sensitivities are higher with a $5.026 \cdot 10^{-1}$ pF/ $\%CO_2$ at 100 Hz. This behavior can be appreciated in Fig. 4b where the capacitance response at two different concentrations (5% and 100% of CO_2) is displayed. The shapes of the curves are similar to an exponential curve with higher capacitances at lower frequencies. Also, the difference between the two concentrations is more accentuated at low frequencies with small differences at higher frequencies (although is not represented in Fig. 4a, the sensitivities are $1.94 \cdot 10^{-2}$, $8.55 \cdot 10^{-3}$ and $6.16 \cdot 10^{-3}$ pF/ $\%CO_2$ at 1,000, 10,000 and 100,000 Hz respectively).

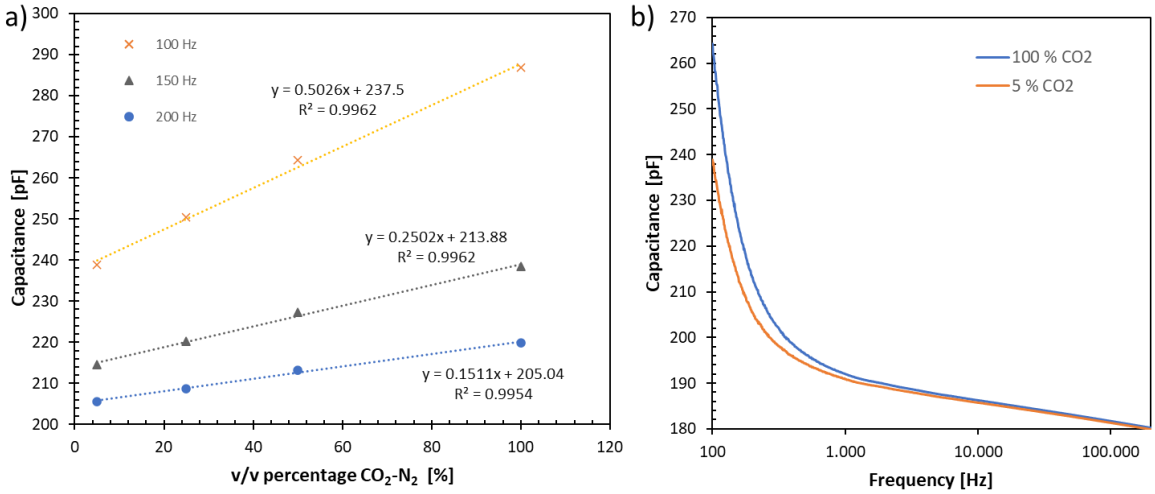

Fig. 4. a) Capacitance of the sensor at different frequencies and gas concentrations of CO_2 with N_2 as carrier gas, b) Capacitance of the sensor at different frequencies at two concentrations of CO_2

The response to NO gases has also been characterized. In this case, due to the high toxicity of the gas, it is used at low concentrations (parts per billion in weight, ppb w/w) in a mix with N_2. The concentrations were modified changing the flow of N_2 and the mix of NO and N_2. In this case, the response to the NO was similar to the previous gas, linear to the concentrations (see Fig. 5a) and with an exponential shape of the curves (see Fig. 5b). The main difference between them was the higher response of the sensor ($1.433 \cdot 10^{-1}$ pF/ppb NO at 100 Hz) since the concentration of NO were seven orders of magnitude smaller than CO_2 and have changes in the capacitance of the same order. Although is not displayed in Fig. 5a, the sensitivities of the sensor at high frequencies were $1.25 \cdot 10^{-2}$, $8.09 \cdot 10^{-3}$ and $6.18 \cdot 10^{-3}$ pF/ ppb of NO at 1, 10 and 100 KHz respectively.

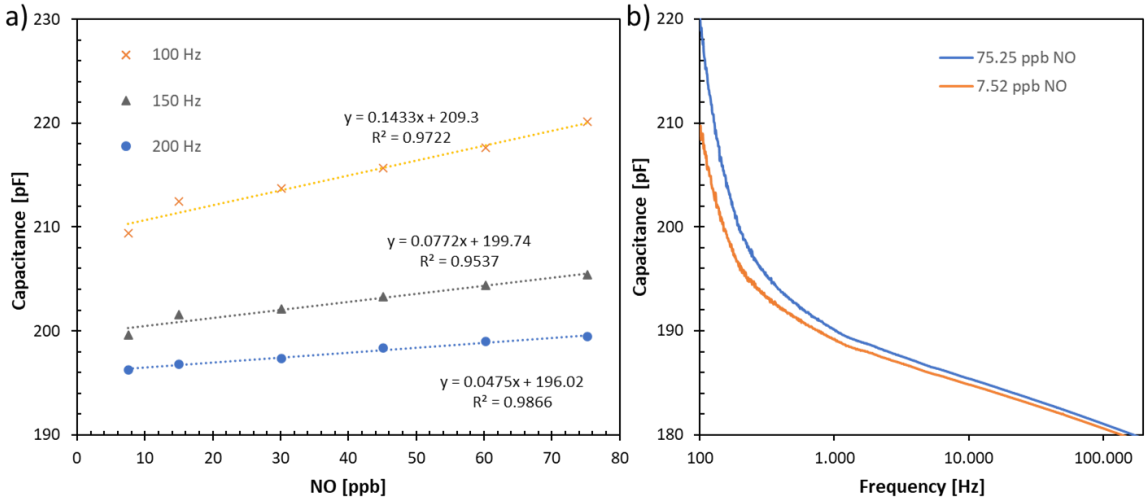

Fig. 5. a) Capacitance of the sensor at different frequencies and gas concentrations of NO with N_2 as carrier gas, b) Capacitance of the sensor at different frequencies at two concentrations of NO.

4 Conclusions

A new micro sized interdigital capacitor has been successfully fabricated. Fingers with dimensions of 10x500 µm and a separation between them of 7 µm has been obtained by the photolithography technique. The reduction of the dimension enables to obtain a great capacitance (approx. 200 pF at 100 Hz) in a small area. The dielectric composed of GO particles allows to use the device as a gas sensor for CO_2 and NO. A good linearity and sensitivity have been found in all cases. The sensitivities at 100Hz were $5.026 \cdot 10^{-1}$ pF/ $\%CO_2$ and $1.433 \cdot 10^{-1}$ pF/ppb of NO.

Improvements in the interdigital design could be implemented in future works to obtain greater capacities. Also, other gases could be studied to obtain the cross sensitivity of the sensor as well as greater sensitivities.

This study opens the door to new micro sized gas capacitor sensors that can be employed in real applications in fields like the internet of things and smart cities.

Acknowledge

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