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A printed capacitive–resistive double sensor for toluene and moisture sensing

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ABSTRACT

This paper presents a flexible printed sensor to detect two different magnitudes: toluene content and relative humidity. This device has been manufactured by using two printing techniques: inkjet printing to define the electrodes and screen printing to deposit the sensitive material. The combination in the same area of two different sensors enables the monitoring of two different gaseous chemicals without interference between them. On one hand, moisture content is associated with changes in the electrical permittivity of the chosen substrate (capacitive effect). On the other hand, toluene concentration is measured through variations in volume of the developed composite (resistive effect). Changes to relative humidity and toluene content have been measured in both capacitive and resistive parts as well as their responses to temperature variations.

1. Introduction

Different electrical transduction mechanisms have been used to develop chemical sensors, such as resistive and capacitive sensors [1]. Chemoresistive sensors rely on the change in the conductivity due to the chemisorption of gas molecules at the sensitive layer surface [2], whereas chemocapacitive sensors are based on variations of the dielectric constant of the sensitive layer in presence of the analyte studied [3].

Many chemical sensors are limited in terms of analyte discrimination or sensor selectivity [4]. Different approaches have been followed to face this drawback, for example, the use of higher-order sensing systems that results in sensor arrays covered with different sensitive films [5,6] or configuring proper algorithms for pattern recognition [7]. Another alternative is to measure different properties of a sensitive layer, known as multifunctional sensors [8,9]. In this respect, a combined Seebeck and resistive sensor was presented by Ionescy for SnO_2 detection [10]; Quintero et al. developed a resistive and capacitive sensor in order to measure temperature and humidity, respectively [11]. A pressure and temperature sensor was introduced by Bulter using capacitive and resistive mechanisms, respectively [12].

The majority of the chemical sensors have been manufactured by semiconductor device technologies [13], such as deposition of material layers, patterning by photolithography or etching to produce the required shapes [14]. Advances in printed electronics sensors on polymeric substrates are being recently reported by several authors [15–18]. The interest in this technology is the large-area manufacturing, low-cost materials, flexibility and biodegradability [19]. Regarding printed capacitive sensors, several moisture detectors have been described. Different fabrication processes have been used to develop this kind of sensors, such as gravure, screen printing and inkjet-printing; and different strategies have been applied to include the sensing capability in the capacitor. The most common approach has been to deposit a sensing layer over the capacitor [20–22]. In this sense, some frequently used polymers are cellulose acetate butyrate (CAB), polymethylmethacrylate (PMMA) and polyvinylchoride (PVC), among others. Another possibility is the direct use of the flexible substrate as sensing element. In this case, polyimide [23] and photographic paper [24] have already been described, saving fabrication steps compared with the former approach. In general, these capacitive sensors have a high thermal drift as one of the main

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challenges to be overcome in order to obtain an accurate humidity measurement but Rivadeneyra et al. found a low-thermal drift in capacitive sensors fabricated on polyimide [25,26].

With respect to printed resistive sensors, different temperature detectors have been reported [11,27–29] as well as gas sensors [30–32]. Chemiresistors have been broadly used in the fabrication of sensor arrays. At first attempt, some authors have used conducting organic polymers such as polypyrroles [33,34] or others [35]; but the need of a set of stable polymers brings in subtle differences in their answers against different chemicals. For this reason, their implementation is not trivial [30]. Alternatively, the use of a set of composites offers better results due to the fact that the signal transduction is achieved by a conventional conducting material and the chemical diversity in the array is addressed by a conventional organic polymer. As conducting materials, different polymers have been used from polypyrrole [36], with the cons of lack of stability, to carbon black [30] or different types of carbon nanotubes, such as multi-walled [37], single-walled [38] or functionalized single-walled nanotubes [39]. Many different types of conventional organic polymers have been employed; working as modifiable insulating phase, typically by swelling, upon contact by analyte vapour that modifies the resistance of the sensing film.

Different techniques have been described for composite films fabrication, mainly by dip coating, used by Lonergan et al. for the construction of a broadly responsive vapour sensor [30], or by printing techniques such as inkjet printing. In this regard, Lorwongtragool et al. developed chemiresistive vapour sensors whose electrical resistance changed in presence of selected volatile organic compounds (VOCs), using three fully inkjet-printed layers of silver interdigitated electrodes, multi-walled carbon nanotubes (MWCNTs) and poly(styrene-co-maleic acid) partial isobutyl/methyl mixed ester (PSE) [40]. Another example of this kind of composites was presented by Shinar et al. [41]. They presented a gas sensor based on graphite microparticles prepared by coating technique.

The most common used structure in both capacitive and resistive modes is the interdigitated electrodes [20,40,42,43] due to its high contact area rate and ease of manufacturing [44] but other designs have been also adopted [45,46].

Here, we present a sensor structure for simultaneously determination of relative humidity and toluene concentration in a non-interacting manner. Since graphite composite is highly responsive to volatile organic compounds (VOCs) and within the model of chemicals that can be released from a variety of products into indoor air at room condition, we have selected toluene as target vapour [30].

The proposed design is based on four meandered electrodes printed on polyimide substrate. Two of these electrodes are covered with a resistive graphite-based composite to sense toluene concentration by volume changes of the deposited film. The other half is directly used as capacitive humidity sensor by variations in the electrical permittivity of the substrate. To verify its response, we have measured both capacitance and resistance changes as function of relative humidity (RH) and toluene concentration. Temperature drifts have also been analyzed as a possible interfering magnitude. The main advantage of this double sensor is the compactness of this design, allowing the introduction of more sensors in the same area without interfering between their measurements.

2. Materials and methods

2.1. Sensor design

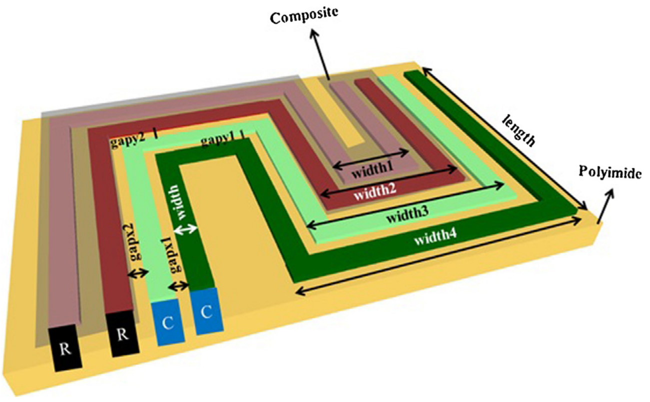

Fig. 1. Layout of the designed capacitive–resistive sensor indicating the notation of the dimensions. R corresponds to resistive terminals, C form capacitive sensor.

The device analyzed in this study is a planar meandered interdigitated electrode structure which provides a direct interaction between the sensor and the surrounding environment. Although there are other geometries that offer higher nominal capacitance per surface area [20,26] than the one displayed in Fig. 1, we have chosen this design because it allows us to increase the functionality of the platform by including in the same area both a resistive sensor and a capacitive one with the same footprint. The usual approach for providing humidity or, in general, gas sensitivity is to deposit a sensing layer on the electrode layout with some electrical property dependent upon humidity or chemical [20,21]; but another possibility is to directly use the substrate as sensing element [23,25,26]. We have combined both strategies in a single device; therefore we have two different sensors in virtually the same area.

In order to functionalize this double or mixed sensor, the two selected magnitudes to be sensed are relative humidity and toluene concentration. The relative humidity is measured through changes in the electrical permittivity of the substrate, so that, the capacitance of these electrodes changes with the humidity. The toluene concentration is derived from changes in the resistivity of a deposited composite onto the other two electrodes of this sensor; therefore, this part of the sensor is resistive whereas the other part is capacitive.

A three dimensional view of the simulated structure including the notation of its geometrical parameters is depicted in Fig. 1. This combined sensor consists of four meandered electrodes depicted in different colours (see Fig. 1) in the web version of this paper, and four terminals. The two top electrodes with the deposited composite constitute the resistive sensor for toluene determination. Whereas the two bottom electrodes create the capacitive sensor with the substrate as sensitive layer. As shown in Fig. 1, each part (resistive or capacitive) of this structure has two terminals for external connection.

This sensor structure allows the introduction of more sensing capabilities into the devices, saving manufacturing materials and area. For this purpose, we have used COMSOL Multiphysics 4.2a (www.comsol.com, COMSOL, Inc., USA) to optimize the design by partial differential equations through the finite element method. This software has been previously used to deeply study similar structures [47,48]. Several parametric analyses have been performed varying the fundamental geometrical parameters of the meandered structure such as the number of fingers, the gap width between consecutive fingers from different sensors and their dimensions (width, length and thickness of each finger). In order to optimize the area, we have fixed the finger width to the minimum secured diameter of landed drop by inkjet printing (in our case 50 μm) and the gap between fingers of the same sensor also to 50 μm. This gap could be reduced below the drop diameter value but this reduction will lead to a strong possibility of short-circuit