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Graphene-based Radiofrequency resonator for noninvasive glucose detection

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Abstract— This paper presents a radio-frequency (RF) sensor to detect glucose oxidase. At the core of the proposed approach is a graphene film deposited on a gap connected to a split ring resonator. The graphene film is doctor bladed on the gap. The film is then properly chemically functionalized in order to detect the presence of glucose. In this paper, we validate the proof-of-concept operation of glucose concentration detection by measuring the frequency shift of the transmission coefficient of the sensor.

Index Terms—microwave sensors, graphene films, glucose sensors.

I. INTRODUCTION

The development of radiofrequency biosensors for biomolecular detection is a very recent trend [1-3]. The introduction of carbon based nanomaterials films [4] seems promising not only for the detection of biomolecules but also for the realization of biosensors for diagnosis of various type of cancers (breast, prostate) [5,6] as well as routine clinical analysis (detection of glucose levels in the serum or drug detection and monitoring).

For the detection of various biomolecules, many invasive techniques such as electro-impedance spectroscopy, enzyme oxidation, time domain reflectometer, and surface plasma resonance exist [7]. Recently, the use of radio-frequency (RF) biosensors based on passive and/or active devices and circuits has been investigated [8-10]. The performance of these biosensors can be enhanced by the introduction of nanomaterials. Therefore, these biosensors possess high potential to modulate their sensitivity and selectivity using tailored chemical functionalization to adsorb particular molecules. Multidisciplinary research capabilities are needed for the realization of biosensors with high sensitivity and low concentration limits.

The goal of this paper is to demonstrate the use of chemically functionalized graphene films for the detection of concentration of glucose from the variation in electrical properties. We focus on passive RF biosensors (split ring resonator). The sensor is designed and realized using standard PCB etching techniques. The graphene film is deposited on the sensor and the film is properly Varying functionalized for detection of glucose. concentrations of glucose are introduced on the film, varying its impedance and resulting in a shift in the resonant frequency of the resonator. The levels of glucose

concentration considered are low and corresponds to the variation of glucose in bodily fluids other than blood (saliva, tears, sweat etc) [11]. A future device designed based on this sensor will hence be a non-invasive glucose testing device.

II. DESIGN AND REALIZATION

A. Graphene film

Tunable passive microwave components based on graphene have been widely studied [12-15]. In these cases the resistance of graphene is varied by the application of a DC bias voltage. The variation of impedance of graphene can also be correlated with a change in its surface properties. This property will be exploited in the following.

The sensor is fabricated by the help of a photolithographic process. The active area (transducer) of the sensor is covered by a graphene film. The film is deposited by the help of a mask that is round with a diameter of 5mm and has a thickness of 500 um. The film is composed of a filler and a binder. The filler is graphene nanoplatelets acquired from Nanoinnova (Spain) and has a surface area to weight ratio of 45 m²/g with a carbon content of 98.9 wt%. The filler to binder ratio in the film is 9:1. The binder is polyvinylidene fluoride (PVdF). The binder helps in retaining the filler together in the matrix and thus gives the film its mechanical stability. The binder is first mixed in N-methyl-2-pyrrolidone (NMP) which acts as a solvent and helps in acquiring a homogeneous mixture of the binder and the filler. The filler (graphene), binder (PVdF) and solvent (NMP) together form the slurry.

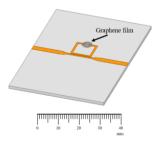


Fig. 1. Geometry of the sensor.

The slurry is mixed overnight for a well dispersed slurry, which is then doctor bladed onto the mask placed in the designated position on the resonator. This is followed by a drying process which result in a film that is void of any solvent and humidity. The drying process lasts over several days and is performed under hoof convection. Finally, the mask is removed and the film is read to be functionalized.

B. Sensor fabrication and measurement setup

The sensor is designed on the FR4 dielectric substrate that has a dielectric constant, $\epsilon r=4.4$ and a loss tangent, tan δ of 0.02. It composed of a high impedance microstrip line fed split ring resonator as shown in Fig. 1. A 50-ohm characteristic impedance microstrip line is tapered into the high impedance section of that excites the resonator. The high impedance section of the microstrip line has a characteristic impedance of 85 Ω . The width of the microstrip line is 3.9 mm and that of the high impedance section is 1 mm respectively. The resonator is a square type with internal length of the side equal to 9 mm. The width of the resonator is 1 mm whereas the gap at the split region is 2mm. The active area of the resonator is based on a graphene film which has a round shape and has a diameter of 5 mm. It is located at the split section of the resonator. The resonator without the film is designed to resonate at a frequency of 2.5 GHz (see Fig. 2).

In order for a sensor to be selective and sensitive to particular molecules, its surface needs to be functionalized. Once the surface is functionalized, specific molecules attach to its surface, ensuring a unique variation in the electrical and frequency response of the sensor [12]. In this case the surface of the film of graphene is functionalized with glucose oxidase. In this way glucose molecules attach to the surface of the film making a signature electrical response. Varying concentrations of glucose introduced to the surface of the film shows a variation in the response of the sensor.

Measurements of the two port scattering parameters of the sensor are performed by the help of a vector network analyzer (VNA, P9371A by Keysight) in the frequency range of 2.5-4.5 GHz. Drops of glucose with of 20 uL volume with concentrations varying from 0 to 20 mg/dl are deposited over the functionalized film and measurements are performed for each concentration. Before the introduction of a drop of different concentration, the film surface is washed with a buffer solution and the resonant frequency returns to a set value.

III. RESULTS

A. Simulation and measurements

The resonator is simulated by the help of the full wave simulator Ansys HFSS. Upon the introduction of the film the resonator resonates at the frequency of 3.6 GHz as shown in Fig. 3. A prototype of the resonator is fabricated and measured. The measured transmission coefficient of the resonator is also shown in Fig. 3. It can be seen that the measured and simulated transmission coefficient are in good agreement with each other.

The introduction of varying concentration of glucose over the film varies its impedance. This change in impedance results in a shift of the resonant frequency of the resonator. The simulated and measured frequency shift of the resonator correlated to the glucose concentration of the drop is shown in Fig. 4. For acquiring corresponding simulations for each glucose concentration value, appropriate real and imaginary part of sheet impedance of the film are assigned to make the resonator resonate at the measured resonant frequency.



Fig. 2. Prototype realization.

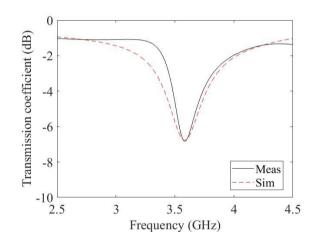


Fig. 3. Resonator with functionalized glucose film. Measurements (solid line) and simulation(dashed line).

IV. CONCLUSIONS

A split ring resonator based non-invasive glucose testing sensor is proposed in this proof-of-concept paper. The resonator has an active area near the split with a graphene based film. The film is functionlized for glucose molecule. The introduction of varying concentration of glucose tends to vary the impedance of the film resulting in a shift in the resonant frequency of the resonator.

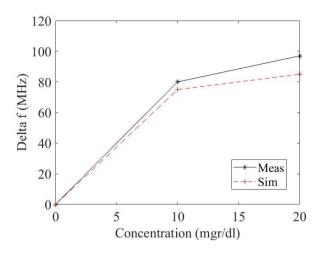


Fig. 4. Frequency shift for different concentrations of glucose. Measurements (solid line), simulations (dashed line).

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