

High sensitivity and fast response graphene oxide capacitive humidity sensor with computer-aided design



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ABSTRACT

A COMSOL model of interdigital capacitive humidity sensor based on graphene oxide (GO) as sensing material is presented. The diffusion process of water molecules and the effects on the capacitance value are demonstrated in theory. The capacitance value of the model is shown in simulation under different relative humidity, and also the influences of structural parameters are summarized, which can be used to construct electrode structure for increasing the sensitivity of humidity sensor. Furthermore, response time of the module is tested through dynamic process simulations, the results show more advanced performance compared with another module. The process of argumentation suggests that this model can also be used for the optimization of other gases capacitive sensors.

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1. Introduction

Humidity sensors with intensive attention due to its application in various fields, including meteorology, medical instruments, food quality monitoring, health, agriculture industry [1,2]. Over the years, various types of humidity sensors have been developed. Considerable time and effort have been dedicated to the investigations of various transduction techniques of developing humidity sensors with good sensing characteristics, such as capacitance [3], resistance [4], optical fiber [5], quartz crystal microbalance (QCM), field effect transistor (FET) [6] and surface acoustic wave (SAW) [7].

Due to their high sensitivity, low power consumption and fabrication costs, capacitive humidity sensors are the most commonly available humidity sensors in commercial field [8,9]. Capacitive-type sensors having high response time, low temperature dependence, hysteresis, and low drift with good performance in harsh environment are desirable in various applications. So, choosing appropriate material as the sensing material for capacitive humidity sensors is very important. Aside from the basic needs of high sensitivity, and wide detection range additional demands for shorter recovery time and quicker response exist for capacitive humidity sensors to meet commercial field requirements [8,9].

Graphene, a two-dimensional monolayer of sp²-bonded carbon atoms exhibiting exceptional mechanical thermal and electrical properties, holds great potential for ultrasensitive detection.

Sensors based on graphene prepared via the micromechanical cleavage of graphite or via the reduction of graphene oxide (GO) have attracted significant interests because of their high sensitivities to gases.

Graphene oxide is a graphene derivative covered by oxygen-containing functional group such as epoxy [10], carboxyl and hydroxyl groups which turn graphene oxide into an electrical insulator. Although GO has been known since the year 1855 [11], a heightened interest in GO was triggered by the explosion in graphene research started ago [12,13]. During the past five years, many research journals have reported the favorable properties of GO in various sensing applications with different designs such as direct measurement of capacitance between two parallel plates or in a slightly more complex CMOS architecture where the GO acts as insulating layers in the capacitor [14–22].

To achieve these goals, considerable attention has been directed to the practical application of GO. GO with inherent electrical and mechanical properties, potentially great for ultrasensitive detection, has drawn increased attention as well as two-dimensional network of sp² and sp³ hybridized carbon atoms [16,17].

As an alternative graphene precursor, GO presents considerable advantage as a material for sensing applications, especially for water detection. In this paper, we focus on capacitive humidity sensors with interdigital electrodes and graphene oxide sensitive film. Here we present a computer-aided design to predict the transient response and steady response of these capacitive sensors. We find that humidity sensors based on GO can overcome the existing problems reported in literature [23–30], and exhibit ultrahigh humidity sensitivity, as well as short response time. The sensing

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properties of the presented sensor are investigated over a wide range of 0–100% relative humidity at room temperature. In addition, the relationship between comb electrode parameters and capacitor are explored, and the sensitivity of the sensor is improved by optimizing the parameters.

2. Material and methods

The model setup of the interdigital humidity sensor is shown in Fig. 1. The experiments conducted by Borini et al. [17,21,22] have shown that the humidity can be detected using graphene oxide devices at rather low relative humidity by monitoring the variation of capacitance upon adsorption of the molecules on G–O, suggesting that G–O can be used as humidity sensor with significantly high precision and ultrafast response time. The experiment is verified by the method of optimization and simulation of sensors is accomplished. The simulations reported here are based on finite-element simulation on structure with a 10 μm thick and 20 μm long film, the electrode is made up of 10 and 10 μm wide and 2 μm spacing (Table 1, Fig. 1). The simulation software is COMSOL Multiphysics® 4.4. Material of two coil-like electrodes is platinum. The simulation is quit ideal which based on the assumption that diffusion in the G–O film, and the diffusion of water molecule is free as suggested by Henry's law. The diffusion is described using Fick's law, which indicates how the concentration filed changes over time caused by diffusion. The total amount of water in the film causes variation of permittivity through a variant of the Clausius–Mossotti equation [31,32]. The temperature is set to 25 $^{\circ}\text{C}$, which is normal atmospheric temperature. The structure is simulated with relative humidity from 0% to 100% to analyze the sensitivity of the structure and transient response. This process can be divided into dynamic and static. The static process refers to the variety of capacitance after sensitive material fully absorbs the water molecule, and that mainly concentrate on the influence of electrode structure and parameters. The dynamic process simulation focuses on response time. It gets the shortest required response time when relative humidity change to different degree.

3. Result and discussion

3.1. Effect of relative humidity

Fig. 2 displays the relationship between capacitance and relative humidity when we chose the graphene oxide as sensing material and the setting relative humidity of the simulation environment is raised from 0% to 100%. It is obvious that the capacitance value increases with increasing of RH levels monotonically. The relationship can be expressed by following equation (1) [25].

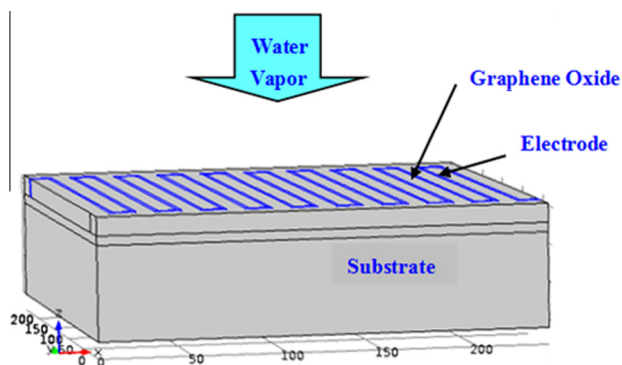


Fig. 1. Configuration of the interdigital capacitive humidity sensor with graphene oxide: three-dimensional model.

$$C = (\epsilon_r - i\gamma/w\epsilon_0)C_0 \quad (1)$$

where γ denotes the conductance. As expression expresses that the capacitance of the sensing material is proportional to γ . Moreover, the physisorption of water molecules on a G–O film influence γ , accurately different sorption processes produce different γ values. As described in the papers [14,25], the increase of RH makes γ increase. Such that the capacitance increases. At low RH, the physisorption process of water molecule on G–O surface which called the first-level adsorption here. Water molecules are primarily physisorbed onto the available active sites of the G–O surface through double hydrogen bonding (Fig. 3). In this process, water molecules cannot move freely because they are usually doubly bonded to two hydroxyls of the surface. In the first-level adsorption, although discontinuous mobile layers constrain the protons in G–O films, they contribute to the leak conduction γ [14,25], that is the essential reason why the capacitance increases at low RH. With the increase of RH, more water molecules are physisorbed through single hydrogen bonding on the hydroxyl groups (Fig. 3). That is called second-level adsorption. After that, the water molecules turn to active, and then they and the bulk liquid gradually become more same. As entering higher levels adsorption progresses, the water molecules can be ionized and to yield a huge amount of hydronium ions (H_3O^+) as charge carriers under an electrostatic field. So, the growing humidity leads to growing γ . The appearance of this trend is due to the facts that the increased humidity makes the physisorbed water layers act liquid-like behavior gradually and the large number of existing epoxy groups in G–O promote proton transfer. And these causes lead to a quite high sensitivity at high, which is consistent with the tendency shown in Fig. 2.

Compared with experimental test, the relative humidity of the environment chamber is raised from 0% to 100%, which makes the measurement of small capacitance easier. The average sensitivity [33] S can be expressed:

$$S = \frac{C_H - C_L}{RH_H - RH_L} \quad (2)$$

The sensor's capacitance is expressed as C_H and C_L , at the highest humidity RH_H and the lowest humidity RH_L , respectively.

The average sensitivity for sensors can be calculated from (2), within the range of 0–100% RH, the sensitivity of the humidity sensor is 19.8 fF/%RH when the size is set to the raw data.

3.2. Effect of comb electrode parameter

Under the same conditions, the parameters of the comb electrode affect the sensitivity of the sensor. So, the test structure is simulated with changing comb electrode parameters under the condition that humidity, temperature, frequency and others remain stable in order to check the linearity. The graphene oxide humidity sensor shows linear response with the variation as shown in Fig. 4. The switching capacitance test is performed through comb electrode for different parameters, including comb number and comb thickness, comb spacing, comb width as well as comb length. The capacitance values linearly increase with comb numbers, thickness, width and length. On the contrary, the capacitance decreases with increasing of comb electrode spacing. At the same RH, the same temperature and frequency, we can get a semi-empirical formula:

$$C = k \frac{ntwl}{s} \quad (3)$$

where k is a constant which is affected by sensitive material, relative humidity, temperature, frequency etc.; n is comb number; t is comb thickness; s is comb spacing; w is comb width; l is comb length. Thus, when the parameters are constant, the capacitance

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