Ultraviolet illumination effect on monolayer graphene-based resistive sensor for acetone detection

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ABSTRACT

In this study, a room-temperature acetone gas sensor based on monolayer graphene is proposed with the improvement by ultraviolet (UV) illumination. The monolayer graphene film was grown by low pressure chemical vapor deposition and then transferred to the interdigital gold electrode by the standard transferred method. To test the acetone sensing properties, the concentration of 300, 600 and 900 ppb acetone provided by a gas standards generator is applied in the testing chamber. The resistance changes following a doping type modification of graphene between illumination and acetone gas. A 1.9% change in short time illumination on graphene could be obtained at 600 ppb and room temperature, which could make this a candidate for biomedical application in the future. In the meantime, a mechanism is proposed to explain whole behavior based on photo-induced molecular desorption.

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1. Introduction

Currently millions of people suffer from diabetes around the world. The current practice of diagnosing diabetes involves blood sampling, which is cumbersome, invasive and painful. In order to reduce the pain of patients by using other chemical markers for detecting glucose level of diabetes, acetone gas concentration in the human breath is a potential marker for noninvasive diagnosis of diabetes. The range of acetone concentration in breath could be from 0.3 to 0.9 ppm for common people. However, in the exhaled air of diabetic patients, the acetone concentration is reported to be more than 1.8 ppm [1].

To have a home-care monitor method for glucose concentration,

an acetone gas sensor could be a candidate with advantages of easy and non-invasive testing. In the past few decades, the development of acetone gas sensors based on nanotechnology and semiconductor materials has received considerable attention. Various metal oxides have been investigated for acetone gas sensors such as ZnO [2], indium oxide [3], WO_3 [4], copper oxide [5], titanium dioxide [6]. An acetone gas sensor based on semiconductor oxide has a high response and a short reaction time at higher operation temperature such as 340 °C. Normally, the response with unit of percentage is defined as $(R_{gas} - R_{min})/R_{min} \cdot 100$, where R_{gas} and R_{min} are the resistances of the sensor at different time and the minimum value at the beginning, respectively [2–6]. For example, the output response of 1.0 wt% La-doped ZnO sensor is 48% at 340 °C in acetone concentration of 100 ppm [2]. However, these devices have high operation temperature and high power consumption which makes difficulties of use for medical and portable application as a glucose monitor. In order to detect lower acetone concentration at room temperature, some nanostructure and two dimensional materials including graphene [7] [8], reduced graphene oxide (rGO) [9] [10], carbon nanotube [11] [12] and MoS_2

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[13] have been developed so that the sensor could detect acetone at room temperature due to a high surface-to-volume ratio. The development of graphene-based sensors due to its 2D crystal structure offers every carbon atom to be a surface atom so that electron transport through graphene can be highly sensitive to adsorbed molecules and that can be demonstrated down to a single molecular level [14]. The ability of gas sensing with graphene is due to the absorption or desorption of gaseous molecules that act as donors or acceptors on the graphene surface. In particular, F. Liu et al. reported that a sensor based on graphene with $ZnFe_2O_4$ exhibits a good selectivity and reproducibility to 10 ppm acetone vapor at 275 °C [8]. For acetone sensing at room temperature, S.J. Choi et al. showed that a 0.1 wt% graphene-WO_3 resistive sensor has 3.9% response to 2 ppm of gas. With the decoration in rGO, the detection limit could be further reduced to 200 ppb [15]. However, the effect of using a graphene gas sensor with UV illumination has not been reported and is discussed for acetone gas sensing performance in this study.

In this paper, the modification of time period of UV illumination on a monolayer graphene-based resistive sensor is presented for acetone sensing properties. Three different sets of experiments were carried out: no UV illumination on graphene, 5 min irradiation and continuous radiation and the acetone sensing performance was measured in each case. Detailed results and discussion are presented to explain the improvement in acetone gas sensing and to establish the mechanism.

2. Experiments

2.1. Materials and methods

A resistive gas sensor was fabricated with a monolayer graphene grown by low pressure chemical vapor deposition (LPCVD) transferred on interdigital gold electrodes. As shown in Fig. 1, the width and spacing of interdigitated electrodes (IDE) was designed as 50 μm and 200 μm in the mask layout for pattern transfer, respectively. A lift-off process was chosen by processing a standard photolithography process with this designed mask first. Then a Cr layer with thickness of 10 nm was thermally evaporated on a glass substrate as an adhesion layer. An Au layer with thickness of 150 nm was deposited. The thickness of Cr and Au layer can be controlled by a standard quartz oscillator. The Cr and Au layers deposited on the photoresists can be removed by using a cleaning process with acetone solution. Second, a monolayer of graphene was grown by chemical vapor deposition (CVD) on 25 μm thick Cu foil (Alfa Aesar, No.13382, thickness 25 μm, purity 99.8%). To remove the oxidized layer on the Cu foil, a pre-anneal treatment at 1000 °C in H_2 flow rate of 80 sccm and pressure of 1.3 mbar was performed for 60 min in the center of a quartz tube and furnace (TF55030, Lindberg/Blue/M). Then methane and hydrogen both with flow rates as 20 sccm were injected into a quartz tube with the temperature at 1000 °C. The monolayer graphene can be directly grown on the Cu foil. Before taking samples out, the quartz tube was cooled down to 200 °C with a cooling rate of 5 °C s^{-1}. Afterward, a CVD-grown monolayer graphene was transferred from Cu foil to the fabricated interdigital electrode by using poly-dimethylsiloxane (PMMA) as the supporting layer and the Cu foil was etched by Cu etching solution. Only the monolayer graphene attached on the PMMA can then be transferred to interdigitated electrodes [15]. Then PMMA was removed by dipping in acetone solution at 70 °C followed by a methane and distilled (DI) water clean. Detail process flow of whole graphene-based resistive sensor is shown in [Fig 1].

2.2. Gas sensing measurement setup

Fig. 2 shows the experimental setup with a real-time electrical measurement for the fabricated sensors. The gas sensors were located in a chamber with volume of 3 L. A UV light emission diode (LED) (370 nm, 70 mW, KOODYZ.) is applied as a UV-light source to provide the illumination on monolayer graphene resistive sensor. To have a quantitative analysis with concentrations at ppb level, the 491 M type gas standards generator from KIN-TEK Laboratories Inc. (La Marque, TX, USA) was used to control the flow rate of analytic gas by diluting control with nitrogen gas. A gas concentration of acetone can be achieved by the automatic control on N_2 flow through the permeation tube. To have a lower concentration of acetone, a dilution process is performed by additional N_2 gas flow controlled by this gas standard generator. To check the recovery response of gas sensors, a purge process with pure N_2 gas was carried out to remove residual sensing gas ambience. To study the response of this developed sensor, a HP34401A multi-meter was
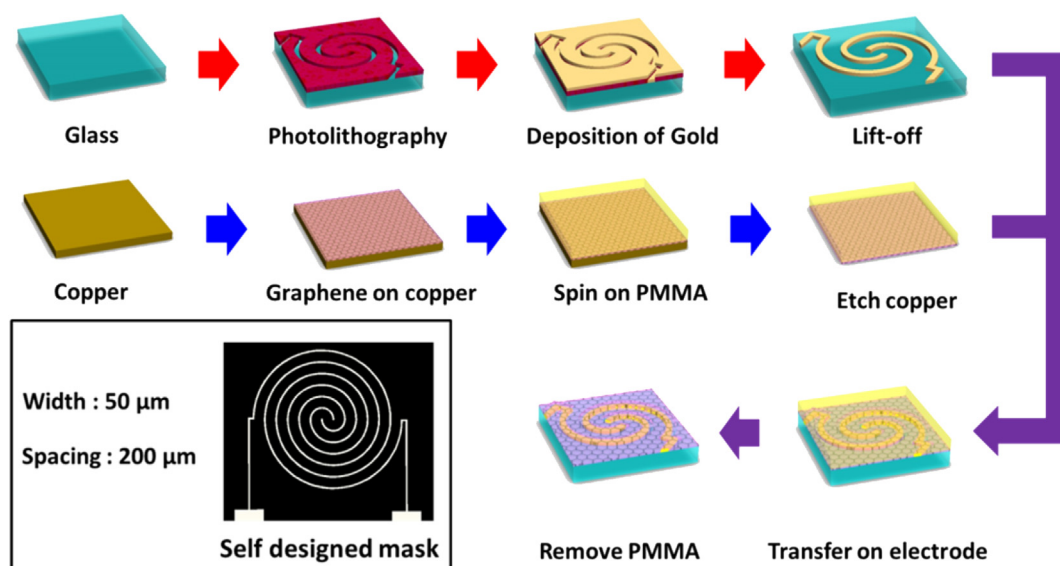

Fig. 1. Detail of process flow of interdigitated electrodes with the transfer of graphene.