Influences of Encapsulated HfO$_2$ Film on the Performance of Graphene Field Effect Transistors

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Abstract. Although the sensitivity of the traditional electrolyte-gated graphene FET is high, the graphene channel is easy to be polluted because of the direct contact between graphene and electrolyte, which leads to the poor stability of the device and not reusable. Ion sensitive field-effect transistors based on graphene (G-ISFET) with top dielectric layer could address the problems. However, depositing dielectric layer on graphene channel is a kind of doping process which debase the performance of G-ISFET. Therefore, the influences from the top dielectric layer in G-ISFET are important and should be studied. In this work, a bottom gate G-ISFET is designed and fabricated, which uses graphene as channel and insulating layer HfO$_2$ as the protective film of graphene. Besides, through a series of electrical characterization, the influence of the deposited HfO$_2$ top layer on graphene FET is analyzed, and the device’s characters as pH sensor is further explored.

1. Introduction

In the past decade, the biochemical sensors based on field-effect transistors (FET) have made great progress for its superior performance, like high sensitivity, fast response and the compatible fabrication processes with traditional CMOS for mass production [1].

Graphene, composed of single-layer carbon atoms, is one of the most promising materials for high-performance sensors [2], since it has excellent electrical properties. For example, electron mobility of graphene at room temperature can reach 15000 cm$^2/(V·s)$ [3]. In addition, high-quality, large area and uniform graphene could be fabricated by chemical vapor deposition technology [4]. Therefore, it is an ideal material for electronic devices in the future. Therefore, graphene FET is widely used in pH detection [5], biological diagnosis [6,7].

In the research of graphene FET sensor, most of the current researches are based on the changes of carrier concentration caused by the adsorption of target molecules [8]. In recent years, graphene sensor based on solution gate FET with graphene as channel has been widely used in pH [9] DNA and protein detection [10, 11]. However, the traditional graphene sensor is easily affected by the external environment when it is not working and directly exposed to the external environment [12], because of the bare sensitive materials. The problem cannot be ignored for high reliability and sensitivity graphene FET biosensor, because even a small doping from external will induce large impact on the signal of the circuit inducing a reduction of the signal-to-noise ratio, thereby greatly reducing the advantages of graphene and affecting the stability of FET.
Ion sensitive field effect transistor (G-ISFET) sensors based on Graphene is one of the most promising platforms to address the above problems. By depositing dielectric and sensitive layer on the surface of graphene channel, graphene is hardly affected by the external environment when it is directly exposed to the external environment own to the protection of the dielectric layer, thereby stabilising the characters of the sensor. In 2013, Wang et al. [13] deposited HfO$_2$/SiO$_2$ on the surface of graphene, and fabricated a top gate G-ISFET, which was used to detect the pH values of solution. However, depositing the dielectric layer on the surface of graphene channel is a kind of adoption for graphene intrinsically. Because of the adoption, the electricity characters of graphene is dased and thus the performance of G-ISFET. Therefore, the influences from the top dielectric layer in G-ISFET should be investigated.

This paper reports the construction of a sensor based on graphene field effect transistor. HfO$_2$ with higher dielectric constant (~25) is deposited on the surface of graphene. The influence of the deposited insulating layer on graphene FET is analyzed, and the responses to pH is further explored. This work is instructive for the future study of G-ISFET.

2. Experiment

The fabrication process of G-ISFET encapsulated with HfO$_2$ layers is shown in figure 1. Firstly, a 30-nm thick silicon dioxide (SiO$_2$) layer was deposited by PECVD on a 500-μm-thick silicon wafer (Figure 1a), to form the dielectric layer (Figure 1b). After this step, the 4-inch wafer was diced into small chips with the dimension of 15 mm × 15 mm. Secondly, monolayer CVD-grown graphene was transferred onto the SiO$_2$ layer and patterned using oxygen plasma etching (Figure 1c). Thirdly, source and drain electrodes consisting of 10-nm-thick Cr and 50-nm-thick gold (Au) were deposited on the graphene layer by electron beam evaporation, and traditional graphene FETs were obtained (Figure 1d). Finally, the top HfO$_2$ layer with the thickness of 10 nm was deposited on the open-face G-ISFETs by PECVD, and the G-ISFET encapsulated with HfO$_2$ layers is fabricated.

![Figure 1. The fabrication process of G-ISFET encapsulated with HfO$_2$ layers](image)

3. Results and Discussion

Figure 2a shows the comparison of transfer characteristics of the G-ISFET before and after the top HfO$_2$ deposition process. The open-face G-ISFET demonstrates a typical ambipolar characteristic, and the Dirac point, which is defined as the gate voltage at which of the minimum source-drain current, lies at ~11 V, indicating the graphene is highly doped by H$_2$O, O$_2$ or other contamination. The transfer characteristic curve of the HfO$_2$-encapsulated G-ISFET is significantly lower and flatter than that of the open-face G-ISFET. For the HfO$_2$-encapsulated G-ISFET (Figure 2a), an ambipolar characteristic is observed and the Dirac point shifts to ~7.2 V, indicating the overlying HfO$_2$ layer can cause an
effective n-type doping in graphene. Figure 2b shows the comparison of output characteristics of the G-ISFET before and after the top HfO₂ deposition process. Driving current of G-ISFET with top HfO₂ is smaller than that of the G-ISFET without top HfO₂ indicating the higher resistance after depositing the top dielectric layer.

In order to quantitatively investigate the effect that the HfO₂ encapsulation layer has on carrier mobility and contact resistance, we adopt a model proposed by Kim et al. [13] to analyze the resistive behaviour of the G-ISFETs before and after the top HfO₂ deposition process. Figure 2c and d show the resistance profiles of a G-ISFET before and after the deposition of HfO₂ encapsulation layer.

\[
R = R_{contact} + R_{channel} = R_{contact} + \left(\mu e(n_0^2 + n^2)^{1/2}\right)^{-1} \frac{L}{W}
\]

The resistance profiles (R) can be fitted to the equation where \(R_{channel}\) is the resistance of the graphene channel, the contact resistance \(R_{contact}\) consists of the metal/graphene contact resistance and the uncovered graphene section resistance, \(L\) and \(W\) are the length (30 µm) and width (200 µm) of the graphene channel, \(e\) is the electronic charge, \(\mu\) is intrinsic field effect mobility, \(n_0\) is the density of carriers at the minimum conductivity (Dirac point), and \(n\) is the gate voltage modulated carrier concentration. From the fitting curve, \(\mu\) and \(R_{contact}\) can be extracted.

![Figure 2. The electrical characters of G-ISFET with and without HfO₂ layers](image)

For the open-face G-ISFET (Figure 4c), \(R_{contact} = 64\ \Omega\), and \(\mu = 796\ \text{cm}^2/(\text{V} \cdot \text{s})\). The mobility of the open-face G-ISFET is compatible with that of reported G-ISFETs [34]. After depositing the 30-nm-thick HfO₂ encapsulation layer, the contact resistance significantly increases to \(R_{contact} = 133\ \Omega\), and the extracted hole mobility decreases to \(\mu = 619\ \text{cm}^2/(\text{V} \cdot \text{s})\), 78% of the charge mobility of G-ISFET without HfO₂ encapsulation layer, as shown in Figure 2d.
Figure 3 displays the pH-sensing characteristics of the fabricated G-ISFETs. When the value of the pH buffer solution changed from 0.93 to 11, the resistances increased and the current declined. This resistances changes due to the surface-potential change of the sensing membrane and is mainly influenced by the electrolyte pH. Figure 3a shows the curve of $\Delta R/R$ versus pH value, where $R$ is the initial resistance and $\Delta R$ is the difference between $R$ and the resistance at stable current. From the fitting line in figure 4b, it is seen that the pH sensitivity in G-ISFETs in this work is about 55 mV/pH at room temperature (300 K).

![Figure 3. pH-sensing characteristics of the fabricated G-ISFET](image)

4. Conclusion
This paper reports the construction of a biosensor based on graphene field effect transistor. HfO$_2$ with higher dielectric constant, is deposited on the surface of graphene. The pH sensitivity of the G-ISFETs is 55 mV/pH at room temperature. Furthermore, the influence of the deposited insulating layer on graphene FET is analyzed. After encapsulating the HfO$_2$ layer, the resistance of graphene is improved and the mobility is declined to 78%, which is instructive for the future study of G-ISFET.

5. References
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