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Analytical Modeling of Bilayer Graphene Based Biosensor

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Abstract

Recently, great attention has been devoted to the graphene because of its unique properties, such as high charge carrier mobility even at a high charge carrier concentration at room temperature, the existence of mass-less Dirac fermions, Quantum Hall effect at room temperature, gas sensing at the single molecule level and gate controlled transport (electron or hole) properties. Gas concentration effect on electrical conductivity of graphene by Green function method has been modeled however sensor analytical modeling needs to be done. In this paper injected carriers by Prostate Specific Antigen (PSA) concentration are simulated and carrier controlling parameters (F, pH) are suggested. Injected carriers from PSA to the graphene surface are monitored and their effect on the capacitance is modeled. Finally comparison with experimental data which illustrates good agreement between them is considered.

Keywords: Biosensor model; PSA detection; Bilayer graphene sensor; Sensor analytical model

Introduction

Graphene is a single layer of sp2-bonded carbon atoms which arranged in a two-dimensional honeycomb lattice [1-3]. Recently numerous excellent properties of graphene such as gas sensing at the single molecule level and long spin relaxation length up to micrometer scale at room temperature direct researchers to the graphene material application [4-9]. In order to prepare single or multi-layer graphene films some methods such as thermal decomposition of commercial Silicon Carbide (SiC) substrates in vacuum, Micromechanical Cleavage (MCP) or chemical exfoliation from bulk graphite powders have been used [10-14]. Graphene has been applied in biological systems, such as detection of DNA and metal ion, protein and pathogen, design of cell/bacterial nanodevice and drug delivery carrier [15-18]. Graphene-modified electrode would facilitate electron transfer and provide a non-cytotoxic, larger surface for biomolecules immobilization because of its amazing electron transport property and high surface area [19,20]. Graphene-based sensors have the potential to detect several types of molecules and ions by itself as well as by logical, physical and chemical modification [21,22]. Cancer marker has been used as a great medical instrument in the diagnosis, monitoring and disease prediction [23,24]. General methods such as surface plasma resonance, microcantilevers and specific enzyme linked immunosorbent assay have been used to detect cancer marker [25-27]. Really PSA is a protein which produced by regular prostate cells. This enzyme plays an important role in the fertility and the dissolution of the seminal fluid coagulum [28,29]. The clinical utility of protein biomarker requires the ability to measure very low concentration proteins to diagnose health and disease [30]. The early diagnosis of many diseases such as, cancer and HIV is vital [31-33].

Since many of methods have some limitation such as ultra low detection, large detection ranges, extremely expensive and complex to realize. It is essential to provide effective approaches to design novel biosensors with nanomaterials, such as carbon nanotube and silicon nanowire with superior performance [24,31,34,35]. For example, through the binding of these biomolecules to the surface of nanostructure and based on electrical altering, carbon based nanomaterials with high sensitivity and selectivity applied to detect small amounts of target biomolecules [22,35]. Therefore, CNTs and graphene based FETs biosensors have attracted much attention for detection of biomarker protein with high sensitivity [24,31,36]. Because of the limitations of CNTs, such as variations in electrical properties of CNT-based devices and the limited surface area of CNTs and important characteristics of graphene used in FET based biosensors is becoming more and more attractive [16-24,31,35,37]. Nevertheless, there aren't any reports on the improvement of graphene FET-based biosensors and their potential as biosensors has not been completely explored [24,37]. Consequently, it is essential to develop nanoscopic graphene based biosensors that is important criterion for biosensors because of small in size, simple in the device structure, allow label-free detection and real-time monitoring of biomarkers [24,35,37,38].

As shown in figure 1 detection method of the biosensor is demonstrated schematically. Based on the assumed detection method bilayer graphene on Polyethylene Terephthalate (PET) substrate as a sensor has been suggested. In addition low-cost, flexible and high sensitive cancer marker for real detection of PSA has been synthesized [35,39,40]. Also conductivity variation by gas molecule adsorption on graphene has been reported [41].

Model

The quantum capacitance characteristic of a BG in nanoscale is applied to evaluate the detectable PSA on graphene based biosensor. Capacitance formed between the channel and gate as most important characteristics of FET device is employed in the modeling of PSA sensor [42-45]. From classical approach, capacitance determined by physical dimensions and the dielectric constant which is between the

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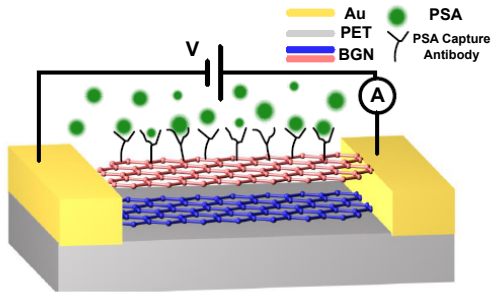

Figure 1: Schematic illustration of bilayer graphene cancer sensor on a flexible PET substrate.

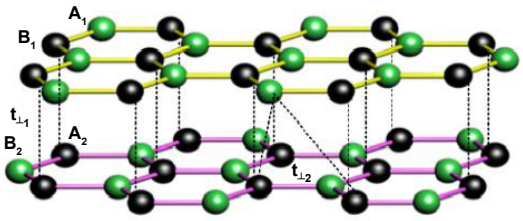

Figure 2: Schematic of AB bilayer graphene lattice structure.

electrodes [46,47]. The conductance of graphene as a function of carrier density and mobility experimentally have been reported which means changes in density and/or charge carriers by adsorption of molecules or ions in graphene will affect the conductance [35]. In the proposed model PSA adsorption as a source of attached carriers is assumed. As shown in figure 1 two Au electrodes of the sensor can be suggested as the source and drain and Dc voltage (V) is applied to them. Also carrier concentration can be modeled by the carrier concentration of BG (n) plus the carrier concentration (n') injected from PSA.

$$\partial Q = e\partial(n+n') \tag{1}$$

Where ∂Q is the change in charge per unit length, n is the intrinsic carrier concentration and n' is the inject carriers by PSA.

Bilayer graphene as a channel material has been explored [44]. Two different stacking configurations (AA and AB) are obtained for (BGs) with armchair edge. The AA-stacked which is metallic and the AB-stacked whereas having a band gap of 0.02 eV, is a semiconductor that is in our focus. In AB structure, atoms on top layer of BG called (A2, B2) and bottom layer are (A1, B1) with hexagonal carbon lattice in figure 2. Near to the K point the electronic dispersion of BG can be written as [44,45]:

$$E(k) \approx (V/2) - \alpha k^2 + \beta k^2 \tag{2}$$

Where $\beta = \upsilon_F^4 / V_{t\perp}^2, \alpha = (V/t_\perp^2)\upsilon_F^2, V$ is bias voltage and Fermi velocity is $\upsilon_F = 3ta_{c-c}/2$. In-plan hopping t is about equivalent to 3.1 (eV), $t\perp$ is an interlayer hopping (the experimental value for bulk graphite is approximately equal to 0.39 eV). Density of state (DOS) as basic parameters on BG shows available energy states. It is notable that electrical property of materials from metal to semiconducting is changing by the gradient of DOS near the Dirac point.

$$D(E) = \frac{1}{2\pi(4\beta k^3 - 2\alpha k)} \tag{3}$$

Carrier concentration in a band can be achieved by integrating the distribution function over energy band.

$$n = \int_0^\infty \left(2\pi \left(4\beta \left[\frac{\alpha \pm \sqrt{\alpha^2 - 4(E-V/2)\beta}}{2\beta} \right]^{\frac{3}{2}} - 2\alpha \left[\frac{\alpha \pm \sqrt{\alpha^2 - 4(E-V/2)\beta}}{2\beta} \right]^{\frac{1}{2}} \times (1+\eta) \right] \right)^{-1} dE \tag{4}$$

Where, $\eta = e^{\frac{E-E_F}{K_B T}}$ is normalized Fermi energy. In order to model the effect of PSA adsorption by graphene we have monitored capacitance variation as:

$$C_q = e^2 \frac{\partial n}{\partial E} + e^2 \frac{\partial n'}{\partial E} \tag{5}$$

It has been demonstrated that the conductance of biosensor depends on the concentration of PSA and different local pH values. Carrier concentration as a function of PSA concentration and different local pH values is considered, which are presented in the form of PSA and PH factors (f,p). The carriers injected from the PSA and localized pH value into the bilayer graphene is added to the conventional carrier concentration as:

$$n' = fF_{PSA} + pP_H \tag{6}$$

So the capacitance based on the proposed model of BG carriers and adsorbing carriers (PSA) by the surface of the biosensor is modified as:

$$C_q = e^2 \left(2\pi \left(4\beta \left[\frac{\alpha \pm \sqrt{\alpha^2 - 4(E-V/2)\beta}}{2\beta} \right]^{\frac{3}{2}} - 2\alpha \left[\frac{\alpha \pm \sqrt{\alpha^2 - 4(E-V/2)\beta}}{2\beta} \right]^{\frac{1}{2}} \times (1+\eta \right) \right)^{-1} \tag{7}$$
$$+ e^2 f \frac{\partial F}{\partial t}\frac{\partial t}{\partial E} + e^2 p \frac{\partial P_H}{\partial t}\frac{\partial t}{\partial E}$$

Where ∂F is the PSA concentration, variation and ∂PH is different in local pH values, therefore the current voltage characteristic can be modified by proposing capacitance model as:

$$I = \left(e^2 \left(2\pi \left(4\beta \left[\frac{\alpha \pm \sqrt{\alpha^2 - 4(E-V/2)\beta}}{2\beta} \right]^{\frac{3}{2}} - 2\alpha \left[\frac{\alpha \pm \sqrt{\alpha^2 - 4(E-V/2)\beta}}{2\beta} \right]^{\frac{1}{2}} \times (1+\eta \right) \right)^{-1} \right.$$
$$\left. + e^2 f \frac{\partial F}{\partial t}\frac{\partial t}{\partial E} + e^2 f P_H \right) \times V_{ds}\mu_e E \tag{8}$$

As shown in figure 3 current-voltage characteristic of graphene based PSA biosensor by the quantum capacitance effect is plotted. Based on the experimental data it is explored that Sensor I-V characteristic can be controlled by PSA concentration (as an antibodies parameter), therefore the concentration coefficient by iteration method is estimated as:

$$f = f_1 Ln(F) + f_2 \tag{9}$$

Where $f1=1.84\ 10^{-2}$ and $f2= 0.3991$

As shown in figures 3 and 4, after incubating bio sensor in different concentrations of PSA and labeled by HRP the normalized conductance of graphene not only raises with the increasing of PSA concentration but also conductance of the bilayer graphene changes with the different local pH values. Thus the pH effect on the carrier concentration in the same manner with PSA concentration by iteration method is modeled as:

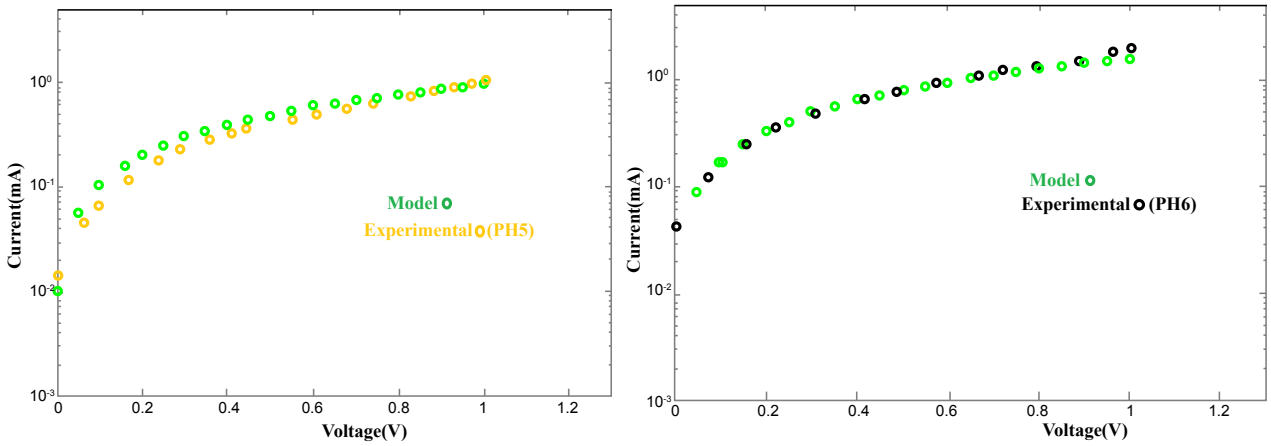

Figure 3: Comparison of current than voltage based on quantum capacitance modeling and experimental data for pH=5 and pH=6 that show a promising agreement between theoretical models and experimental data.

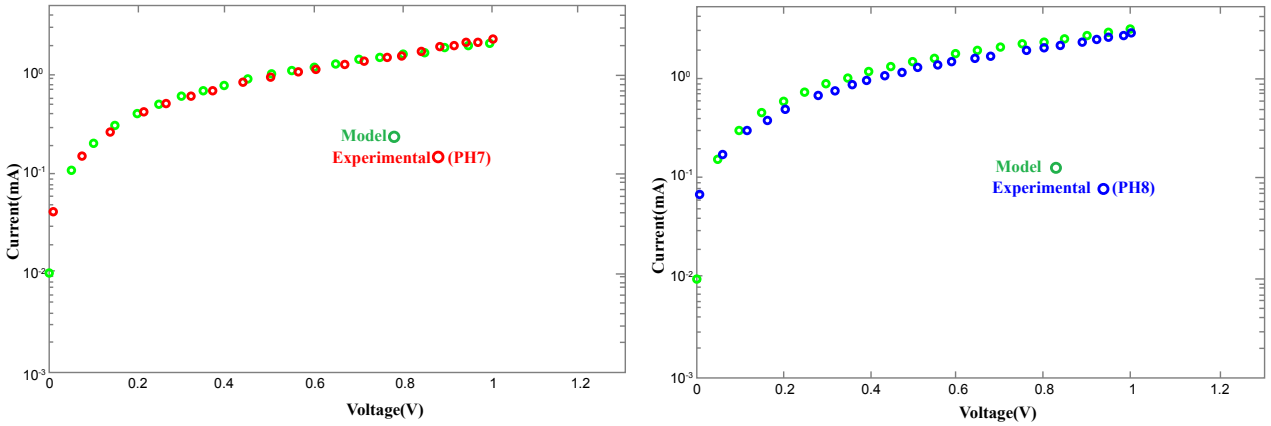

Figure 4: Comparison of current than voltage based on quantum capacitance modeling and experimental data for PH=7 and PH=8 that show a promising agreement between theoretical model and experimental.

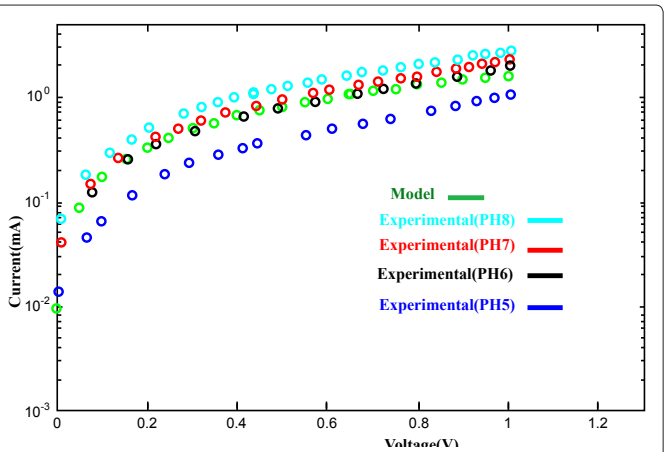

Figure 5: Comparison of current than voltage based on quantum capacitance modeling and experimental data for different PH.

$$p = p_1 P_H^2 - p_2 P_H + p_3 \quad (10)$$

Where p_1=170.31, p_2=15.063 and p_3=5.3251. Also the suggested PSA detection modelling can be employed for almost any target antigen with a known antibody.

$$I = \left(e^2 \left(2\pi \left(4\beta \left[\frac{\alpha \pm \sqrt{\alpha^2 - 4(E - V/2)\beta}}{2\beta} \right]^{\frac{3}{2}} - 2\alpha \left[\frac{\alpha \pm \sqrt{\alpha^2 - 4(E - V/2)\beta}}{2\beta} \right]^{\frac{1}{2}} \right) \times (1 + \eta) \right)^{-1} + e^2 (1.84 \times 10^{-2} Ln(F) + 0.3991) \frac{\partial F}{\partial t} \frac{\partial t}{\partial E} + e^2 (170.31 P_H^2 - 15.063 P_H + 5.3251) P_H \right) \times V_{ds} \mu_e E \quad (11)$$

As shown in figure 5 based on the presented model it is notable that by raising the concentration (as an example for pH value is 6) model is closer to the experimental data (black line). In the same manner we can compare other experimental data's as well.

Current-voltage characteristic altering by carrier concentration and local pH values, also based on the supposed model sensor I-V characteristic which is controlled by PSA concentration parameter and PH parameter.

Conclusion

Graphene based material because of its amazing electron transport

property and high surface area has been employed in biological applications. Green function method in the gas sensor modeling has been used although pH physical based sensor modeling needs to be explored. In this research current voltage characteristic of a BG in terms of capacitance model is applied to evaluate the detectable PSA on the graphene based biosensors. PSA adsorption and local pH value effects on the carrier concentration are monitored and injected carriers as a function of control parameters (f,p) are presented. Finally, good agreement between proposed model and experimental data is reported. We have concluded that sensor characteristics altered by PSA concentration and local pH values, by means of which the proposed model sensor can be controlled.

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