Article Type | DOI:10.5281/zenodo.8007889
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NANOSAM Journal of NanoScience in Advanced Materials

# High Sensitive pH Sensor with Graphene based Dual-Gate ISFET

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**Electrochemical sensors are devices that use electrochemical reactions to detect and measure the concentration of analytes in a sample. As an electrochemical sensor, an ion-sensitive field-effect transistor, ISFET, is the basic structure of a biosensor. This work presents a high-sensitive pH sensor with a graphene-based dual-gate ISFET. Design and simulations of the proposed device are performed on SILVACO TCAD software. Graphene and aluminum oxide are used as channel and sensing film, respectively. The channel thickness is 10 nm, while the thickness of the device is 800 nm. The transfer and output characteristic curves of the device are obtained through simulations. The effects of the back gate, channel material, and sensitive layer on device performance are evaluated by assessing the electrical characteristics, threshold voltage, and drain current. The sensitivity of the proposed device is achieved, approximately 18.06 times the Nernst limit, as 1066 mv/pH, which is the best performance score in the literature. The proposed device offers a novel ISFET structure with reliable and higher detection sensitivity.**

## 1. Introduction

ISFET, Ion Sensitive Field Effect Transistor, is a type of sensor that can measure the concentration of ions in a liquid. The sensor consists of a tiny semiconductor chip covered with a thin layer of material sensitive to ions. When ions in the liquid come into contact with this layer, they change the electrical properties of the chip, which can be measured and translated into a numerical value indicating the concentration of ions present [1].

ISFET sensors are commonly used in medical and environmental applications, such as monitoring the pH levels of blood or measuring the acidity of water samples. They are preferred over traditional glass pH sensors because they are more durable, less prone to damage, and can be made smaller for use in compact devices.

A graphene-based ISFET typically consists of a thin layer of graphene that is deposited onto a substrate, such as silicon dioxide. The graphene layer is then coated with a material that is sensitive to ions, such as a polymer or metal oxide. On top of the sensing layer, there is a gate electrode, which is separated from the graphene layer by a thin insulating layer, such as silicon dioxide. The gate electrode is used to control the flow of electrical current through the graphene layer, and the insulating layer prevents direct contact between the gate electrode and the sensing layer.

When ions in a liquid come into contact with the sensing layer, they change the electrical properties of the graphene layer, which can be detected and measured by the transistor. By controlling the voltage applied to the gate electrode, the electrical properties of the graphene layer can be modulated, allowing for precise control and measurement of ion concentrations in the liquid.

Graphene-based ISFETs offer several advantages over traditional ISFETs. Graphene is an excellent electrical conductor, which allows for faster and more accurate detection of ion concentrations [2]. Additionally, graphene is extremely thin and flexible, which makes it suitable for use in a variety of applications, including wearable devices and sensors that can be integrated into clothing or other materials.

Overall, graphene-based ISFETs have the potential to improve the accuracy, sensitivity, and durability of ion sensors, leading to new opportunities for sensing applications in fields such as biomedical and medicine.

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Keywords: Graphene; ISFET; pH sensing; TCAD; Sensitivity.

Received: 24 March 2023 | Accepted: 24 May 2023 | Published online: 25 June 2023

Characteristics of materials to be used and their structural properties determine the device performance. Dimensions of the gate terminal and the sensing film affect the device's performance. Remarkably, researchers in their studies concentrate on the sensing layer of the ISFET including silicon dioxide ($SiO_2$), tantalum pentoxide ($Ta_2O_5$), aluminum oxide ($Al_2O_3$), and silicon nitride ($Si_3N_4$).

The electrolyte model and device are designed and simulated using Silvaco TCAD tool, as described in the study of Choksi et. al. [3]. In the related work, the ($Al_2O_3$)-based ISFET has a sensitivity of 59.172 mV/pH, slightly higher than the Nernstian limit (59 mV/pH). Compared to ($SiO_2$) and ($Si_3N_4$), ISFETs with ($Al_2O_3$) sensing films have greater sensitivity. Changing the sensing films lead to variations in threshold voltage and sensitivity for a specific pH value. These changes are due to variations in electrochemical parameters like surface site density and dissociation constants. Sensitivity also varies according to the thickness of the sensing film, with thinner films leading to increased sensitivity.

The study of Wang and Tang [4] introduced a novel ISFET device and investigated how using $GaAs$ as a substrate affects its sensitivity. $a - IGZO$ material was added to enhance the characteristic curve further and increase the sensitivity from three to four and a half times the Nernst limit. The sensitivity is boosted to seven times the Nernst limit by incorporating $SiC$ materials. The study also examines the effect of different insulating layer materials on the device's sensitivity. Overall, the new ISFET structure achieved a sensitivity of approximately 12 times the Nernst limit.

In [5], an $AlN$-gate ISFET pH sensor design, modeling, fabrication, and characterization are reported. The related study optimized the $AlN$ sensing film deposition process to achieve the desired stoichiometry. The effects of various process variables on the development of the film were carefully examined. The stoichiometry and surface morphology of the sensing film were determined using FESEM, EDS, and XRD techniques. The sensitivity of the manufactured device was obtained as 33 mV/pH. In order to validate the experimental findings, an ISFET behavioral macro model was also constructed in the SPICE environment, and the impact of various electrochemical parameters on device performance was discussed. Finally, it was discovered that the dissociation constant and primary amine site density for $AlN$ sensing film significantly affects the sensitivity of the ISFET.

In our previous study [6], the front gate sweeping analysis was performed for the proposed ISFET device and 55 mV/pH of sensitivity which is less than the Nernst limit was achieved. In the related study, influence of channel parameters and the back gate voltage on the device performance are also analyzed.

This work proposes a graphene-based, dual-gate, ultra-sensitive ISFET device for pH sensing. Structure materials and device dimensions are modified to achieve better performance, and then electrical characteristics, performance, and sensitivity analysis are performed. SILVACO TCAD environment is used to run the simulations. The chemical reactions occurring at the electrolyte-insulator interface cannot currently be modeled by TCAD software. However, transport equations for the carriers in a semiconductor and the ions in an equilibrium electrolyte solution are similar. This similarity provides a solution for modeling electrolytes in the simulation environment. Therefore, a monovalent electrolyte can be represented as a semiconductor [3]. This study uses the electrolyte, defined as a semiconductor-like layer, to test the proposed ISFET structure. Finally, results are compared with the literature, and high sensitivity and performance are observed, which validates the proposed model.

## 2. Results and Discussion

The performance of the proposed dual-gate, graphene based ISFET device is examined using the SILVACO tools with specific parameters. Consequently, the optimum device parameters and structure are obtained.

### 2.1 Structure of the Proposed ISFET Device:

The proposed graphene-based ISFET device is constructed in the SILVACO environment. Figure 1.a and 1.b illustrate the block diagram and the constructed structure in the simulation environment, respectively. As shown in the figures GaAs, $Al_2O_3$, graphene, and aluminum have been used as the substrate, sensitive layer, channel, and source/drain material, respectively. The gold region in Figure 1.b is used as a reference electrode. Some layer parameters are shown in Table 1, where channel thickness is 10 nm, the length and thickness of the device are 600 nm and 800 nm respectively.

### 2.2 Electrical Characteristics

In order to obtain the output characteristics of the device, as the behavior of the electrical characteristics is same for all pH values, an acidic target electrolyte with pH level 3 is set to examine electrical characteristics with smaller threshold value [4]. A constant voltage is applied to the gate terminal. In this case, the drain current is increased as the drain-source voltage increases. The characteristic output curve of the proposed device is given in Figure 2(a) for various gate voltages (0-3 V) with 1 V step size and drain-source voltages in the range of 0-2 V with 10 mV step size. It is demonstrated that large gate voltages yield large drain currents. This is consistent with the saturation condition of the drain current in a classical MOSFET. The drain current can reach 0.33 $mA$ at $V_{GS} = 3\ V$.

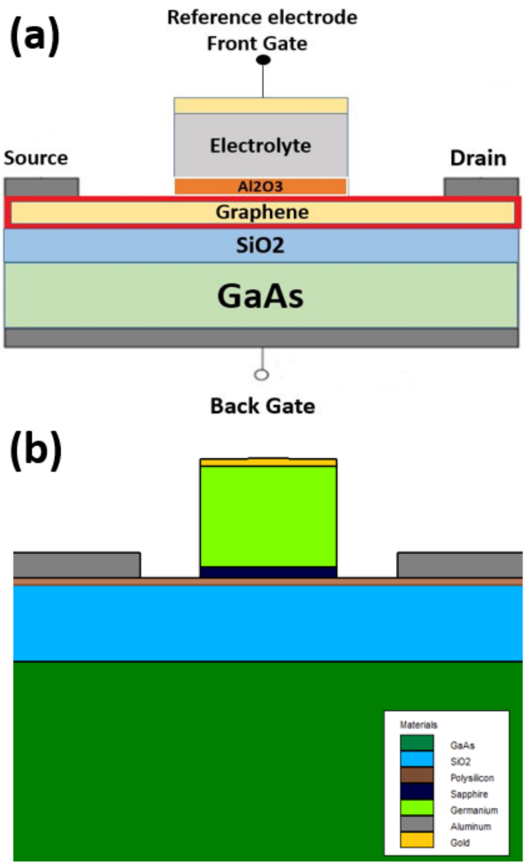


**Figure 1.** (a) Block diagram and (b) constructed structure of graphene based ISFET in Silvaco.

Figure 2(b) illustrates the transfer characteristics of the device, demonstrating the gate voltage's capability to control the drain current. The transfer characteristics curve are obtained by setting the source-drain voltage to 0.5 V, 1 V, and 1.5 V, respectively, and sweeping the gate voltage from 1 to 2 V. The threshold voltage ($V_{th}$) is acquired as 0.5 V.

## 2.3 Sensitivity Analysis

The proposed device is analyzed regarding several sensitive layers. ISFETs with ($Al_2O_3$) sensitive layer provide better sensitivity than those of ($SiO_2$) and ($Si_3N_4$). The sensitivity of the front-gate scanning method for the proposed ISFET with aluminum oxide sensing material is 59.5 mV/pH which is slightly higher than Nernst limit. Table 2 also gives the sensitivity simulation results for different sensing layers.

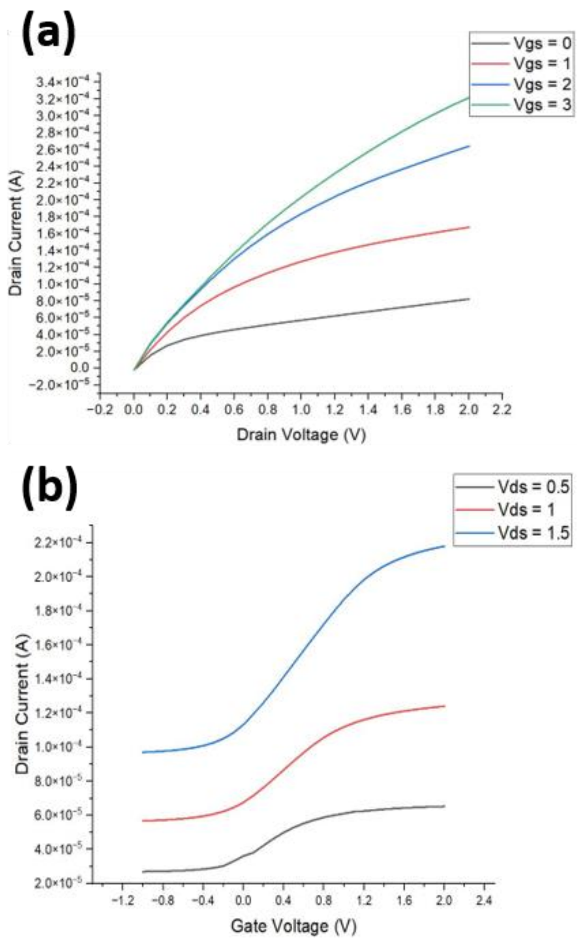


**Figure 2.** a) Output and b) Transfer characteristics of the proposed device.

**Table 1.** Table caption must be located just above the table and written Cambria 10 font.

| Parameters | Graphene | GaAs | $Al_2O_3$ | Fluid Gate |
|---|---|---|---|---|
| Permittivity | 80 | - | 7.5 | 80 |
| Thickness (nm) | 10 | 400 | 10 | 200 |
| Length (nm) | 600 | 600 | 600 | 160 |
| Affinity | - | - | - | 3.9 |

**Table 2.** Sensitivity of front-gate scanning for different sensing films.

| Sensing Film | Sensitivity (mV/pH) |
|---|---|
| Aluminum Oxide ($Al_2O_3$) | 59.5 |
| Silicon Nitride ($Si_3N_4$) | 56.8 |
| Silicon Dioxide ($SiO_2$) | 56.1 |

$$sensitivity = \frac{V_{th}(pH_2) - V_{th}(pH_1)}{pH_2 - pH_1} \quad (1)$$

In the simulation, the drain to source and the front-gate ($V_{ref}$) voltages are set to 2V and -1V, respectively. Then, the back-gate voltage is swept. To validate the proposed model's accuracy, the sensitivity of the device with $Al_2O_3$ as the insulating sensitive layer is tested with different pH values (4, 7, and 10), as shown in Figure 3. It is clear that raising the pH level raises the threshold voltage and changes the direction of the overall trend to the right. In this study, the calculated sensitivity of the device reached a value of 1066 $mV/pH$, which is better than the studies in the literature.

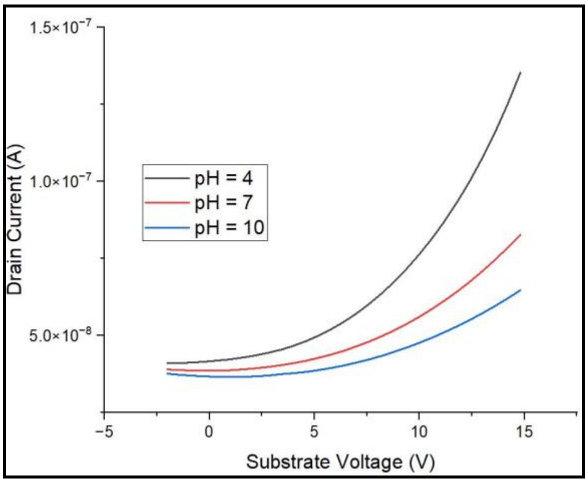


**Figure 3.** ISFET transfer characteristics with different pH values.

Table 3 summarizes the previous state of art studies of ISFET in terms of sensitivity and sensitive layer. The proposed graphene-based device presents the best sensitivity score compared to the literature.

**Table 3.** Sensitivity of front-gate scanning with different sensing films.

| **Channel material** | **Sensitive layer** | **Sensitivity (mV/pH)** | **Ref.** |
|---|---|---|---|
| Graphene | Nano-graph. | 140 | [7] |
| SiGe | $Al_2O_3$ | 360 | [8] |
| IGZO | $Ta_2O_5$ | 585.3 | [4] |
| Inp | Indium oxide | 58.3 | [9] |
| $MoS_2$ | $Al_2O_3$ | 50 | [10] |
| Si | $Al_2O_3$ | 51.2 | [11] |
| Si | $SiO_2$ | 152.1 | [12] |
| Si | $Si_3N_4$ | 57.14 | [3] |
| **Graphene** | **$Al_2O_3$** | **1066** | **In this study** |

## 3. Conclusion

This paper analyzes the effect of the graphene layer, as a channel material, on the performance of the ISFET device. The device was tested using the electrolyte model implemented in the Silvaco environment. The acquired characteristic curves were also physically examined. The effects of the back gate, channel material, and sensitive layer on device performance are considered by assessing the electrical characteristics, threshold voltage, and drain current.The proposed device was constructed using GaAs as a substrate, graphene as the channel material, and $Al_2O_3$ (sapphire) as the sensitive film. Graphene-based ISFETs provide better sensitivity than conventional types, and the threshold voltage shows variations upon c8hanging the electrolyte pH level. The channel thickness in the proposed structure is 10nm. Consequently, the proposed ISFET device has a pH detection sensitivity of 1066 $mV/pH$.

## Method

In this work, as a preliminary study, the fabrication of the graphene-channel-ISFET based on silicon on insulator (SOI) is designed and simulated using the Silvaco TCAD environment. The GaAs substrate of the ISFET is doped with boron. A thick and lengthy SiO2 layer is grown over a GaAs wafer by dry oxidation at a temperature and pressure of 1000°C and 1 atm, respectively. Since the graphene material is undefined in SILVACO

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TCAD, software tools cannot simulate graphene. Instead, a polysilicon layer mimics the graphene by changing its properties, such as bandgap, the density of states, mobility, and permittivity. Over the channel region, a thin gate oxide ($SiO_2$) is formed as a top gate. Aluminum and gold are used for the source, drain, and gate terminals.

Transport equations for the holes-electrons in a semiconductor material and the cations-anions in an equilibrium electrolyte solution are similar [13]. Therefore, a monovalent electrolyte, with a bandgap of 1 eV and a dielectric constant equal to that of water, can be represented as a semiconductor with the density of states given by equation (2).

$$N_C = N_V = \quad 10^{-3} N_{av}(c_0 + c_{HB}) \qquad pH \leq 7$$
$$= 10^{-3} N_{av}(c_0 + 10^{-14}/c_{HB}) \qquad pH > 7 \quad (2)$$

where, $c_0$ and $c_{HB}$ are the molar salt ion and hydrogen concentration in mol/l in the solution, respectively. $N_C$ and $N_V$ are expressed in $cm^{-3}$. $N_C$ and $N_V$ are 6.625310 $cm^{-3}$, while $N_{av}$ is Avogadro's number. The bulk electrolyte can be defined as a region with constant potential and zero net electrolyte charge since the total concentrations of positive (p) and negative (n) ions are equal. Figure 4 illustrates the relationship between density of states and different pH values [14].

$$n \cong N_C e^{-\frac{E_C - E_f}{kT}}$$
$$p \cong N_v e^{-\frac{E_f - E_v}{kT}} \qquad (3)$$

where $E_V, E_C, E_f$ are the upper energy level of the valance band, lower energy level of the conduction band, and the Fermi energy level, respectively.

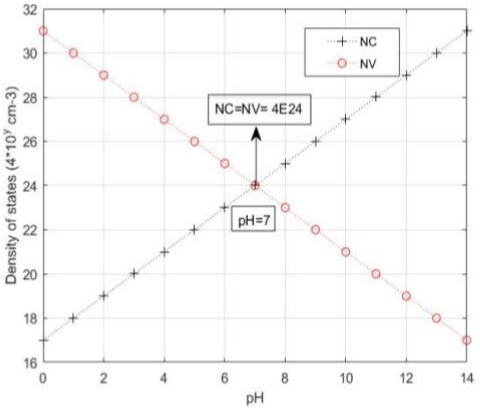


**Figure 4.** Density of states $N_C$ and $N_V$ versus pH values.

## Acknowledgements

This work is supported by Scientific Research Projects Unit of Çukurova University, in project FYL-2022-15581.

## Authors' contributions:

A.H.: Design and simulations, writing the original draft of the manuscript; M.I.: Methodology, review and editing, supervision.

## Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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