JOURNAL OF NANO- AND ELECTRONIC PHYSICS
Vol. 18 No 2, 02026(5pp) (2026)

ЖУРНАЛ НАНО- ТА ЕЛЕКТРОННОЇ ФІЗИКИ
Том 18 № 2, 02026(5cc) (2026)

REGULAR ARTICLE

Field-Effect Transistor Based on Reduced Graphene Oxide Film with ZnO and Porous Silicon Absorbing Layers for Ionizing Radiation Detection

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(Received 02 September 2025; revised manuscript received 17 April 2026; published online 29 April 2026)

The graphene field-effect transistor based on the reduced graphene oxide – zinc oxide – porous silicon – silicon substrate sandwich-like structure has been created for ionizing radiation detection. The hybrid structure was obtained as a result of sequential technological processes of photoelectrochemical formation of the nanostructured porous silicon, the electrochemical deposition of zinc oxide, application of a film-forming suspension of reduced graphene oxide nanoparticles, and subsequent drying at room temperature. Dependencies of the drain current on the drain-source voltage and gate voltage of the obtained field-effect transistor were analyzed. An increase in the resistance of the reduced graphene oxide film near the charge neutrality point caused by irradiation with the ^{226}Ra isotope has been found. In addition, a displacement of the charge neutrality point in the direction of a lower gate voltage was observed. It has been established that ionizing radiation has a greater effect on the electronic component of the conductivity of the reduced graphene oxide film than on the hole component. An increase in the sensitivity of the created ionizing radiation detector due to the use of additional zinc oxide and porous silicon absorbing layers was established. Mechanisms of the influence of alpha and beta particles and gamma quanta on the electrical characteristics of the proposed sensor are discussed based on the analysis of the frequency dependences of the internal resistance and electrical capacitance, as well as the capacitance-voltage characteristics of the sandwich-like structure. The obtained results have a high potential to create a new type of small-sized dosimetric devices based on the graphene field-effect transistors using simple and low-cost techniques and materials.

Keywords: Reduced graphene oxide, Field-effect transistor, Porous silicon, Zinc oxide, Ionizing radiation sensor.

DOI: 10.21272/jnep.18(2).02026 PACS numbers: 73.63. – b, 78.70. – g

1. INTRODUCTION

Modern equipment for detecting, identifying, and measuring ionizing radiation is a key component of the safe and responsible development of nuclear science, space exploration, and related fields. The implementation of new technologies and materials is the basis for improving radiation monitoring systems, which provides an increase in their sensitivity and a reduction in size and power consumption. In particular, MOS field-effect transistors (FETs) and PIN diodes with an optimized structure are already used as ionizing radiation sensors [1, 2]. In general, the detection of ionizing radiation is based on various effects that cause changes in the optical, luminescent, or electrical characteristics of radiation-absorbing materials [3-6].

Today, the attention of researchers is focused on nanostructures and composites based on them to create portable radiation sensors [7-9]. However, the sensitivity of such sensors is directly related to the efficiency of radiation absorption. Due to the small volume of nanomaterials, there is a need to register single high-energy particles. Among the main strategies for increasing the efficiency of small-sized ionizing radiation detectors is a hybrid approach that combines an adsorber and a material, the parameter change of which is being recorded. The use of graphene FETs as radiation sensors is a promising solution for this approach [10, 11]. Here, silicon is a radiation absorber, and graphene is a sensitive material. The dependence of the ambipolar conductivity of graphene on the local electric field provides high sensitivity of FETs to radiation-induced charges in the silicon substrate.

The graphene field-effect sensors demonstrate maximum sensitivity near the charge neutrality point on the conductivity profile (i.e., on the dependence of conductivity on the gate voltage). The high mobility of charge carriers and low electrical noise of the carbon monolayer are additional advantages of the graphene-based sensors [12, 13]. Given that the electrical characteristics of graphene FETs depend significantly on the nature and quality of the dielectric layer [14, 15], the use of different supporting layers makes it possible to tune the device parameters for specific practical applications. For example, high-κ dielectric layers or an additional layer that passivates the surface of the carbon monolayer are used to increase the efficiency of graphene FETs.

There are several different ways to obtain graphene. The reduction of graphene oxide (GO) nanoparticles is a

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Cite this article as: I.B Olenych et al., *J. Nano- Electron. Phys.* 18 No 2, 02026 (2026) https://doi.org/10.21272/jnep.18(2).02026

simpler and lower-cost method of producing 2D carbon material compared to epitaxial growth or chemical vapor deposition [16-18]. Although the structure of reduced graphene oxide (RGO) usually contains residual functional groups and defects that decrease the mobility of charge carriers, FETs based on RGO films can effectively detect alpha, beta, and gamma radiation [19]. To increase the sensitivity of RGO-based ionizing radiation sensors, an innovative solution may be to use supporting layers other than a traditional gate dielectric. In particular, nanostructured layers of porous silicon (PS) and ZnO have a high potential for use in radiation detectors as absorbing materials [7, 20]. Besides, PS and ZnO nanostructures are characterized by low electrical conductivity due to the increased band gap. Therefore, we can expect an increase in the radiation sensitivity of field-effect sensors with additional absorbing layers of PS and ZnO, as was observed in photosensitive FETs based on the RGO-PS-Si structure [21]. In this work, the possibility of using the RGO-ZnO-PS-Si sandwich-like structure for ionizing radiation sensing was studied.

2. EXPERIMENT

A low-alloyed silicon wafer with a thickness of 400 μm was the substrate and gate of the RGO-based FET. A thin film of gold thermally deposited on the back surface of the wafer and annealed at a 600 °C temperature for 30 min was used as the gate contact. The PS layer was formed on the opposite side of the wafer. Photoelectrochemical etching of silicon in an ethanolic solution of hydrofluoric acid for 10 min at an anodic current density of 30 mA/cm^2 was used to obtain the porous layer. After washing in distilled water, a layer of nanostructured zinc oxide was deposited on the PS surface from an aqueous solution of 0.05 M $Zn(NO_3)_2 \cdot 6H_2O$ and 0.1 M $NaNO_3$ at a potential of – 1.4 V and a temperature of 65 °C for 2 min. This method demonstrated the effective growth of ZnO nanostructures, the morphology of which depends on the deposition parameters [22].

To form the RGO film on the ZnO layer surface, an aqueous suspension of GO manufactured by Sigma-Aldrich (USA) with a concentration of 2 mg/ml was used. GO was reduced with hydrazine monohydrate under the influence of ultrasound for 20 min. A 0.2 M solution of sodium dodecylbenzene sulfonate in water was mixed with the suspension to prevent aggregation of the RGO nanoparticles. The resulting film-forming mix was deposited onto the surface of the ZnO layer and dried in air at room temperature. Two silver contacts at a distance of 1 mm from each other, which served as the source and drain of the FET, were thermally deposited onto the surface of the formed RGO film. A schematic representation of the obtained FET based on the RGO-ZnO-PS-Si sandwich-like structure is shown in Fig. 1.

The electrical characteristics of the created FET were studied in both DC and AC modes. In particular, the dependencies of the drain current and resistance on the gate voltage were measured using a Siglent SDM 3055 multimeter and a Hantek 1833C RLC-measuring device, respectively. The effect of ionizing radiation on the resistive and capacitive properties of the RGO-ZnO-PS-Si structure was studied when current passed through the structure perpendicular to the surface.

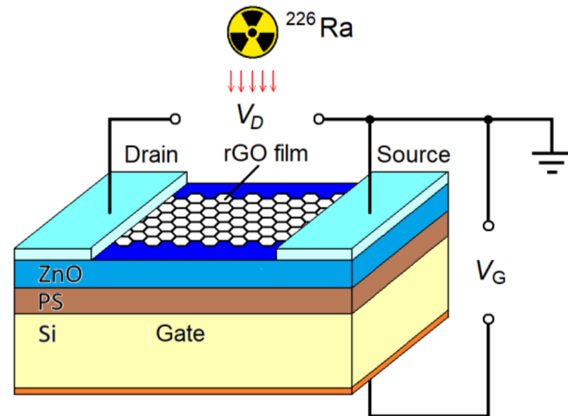

Fig. 1 – Schematic representation of the FET based on the RGO-ZnO-PS-Si structure

The isotope of radium ^{226}Ra with 0.1 mCi activity was used to irradiate the experimental sample with alpha and beta particles and gamma quanta as described in [19].

3. RESULTS AND DISCUSSION

To clarify the features of charge transport in the obtained FET, dependencies of the drain current I_D on the drain V_D and gate V_G voltages were measured. The I_D-V_D curve at the gate voltage $V_G = 0$ is shown in the inset of Fig. 2. The non-linear nature of the measured I_D-V_D dependence indicates complex charge transfer processes in the FET conductive channel that may be related to the inhomogeneity of the RGO film formed by individual carbon nanoparticles. Besides, surfactant added to the film-forming suspension can cause electrical barriers between RGO nanosheets. Finally, the possibility of injecting carriers from the ZnO supporting layer into the RGO film should not be neglected.

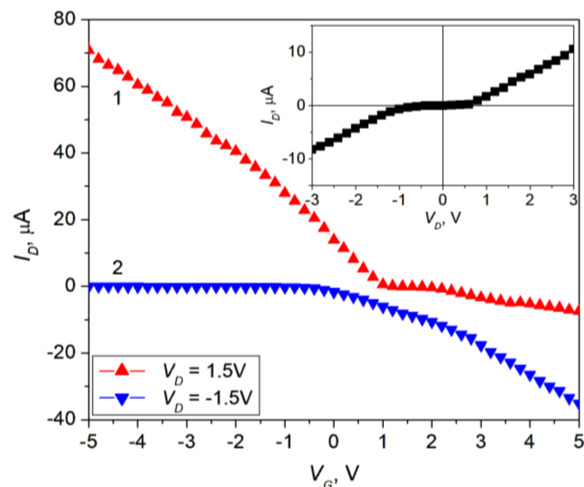

Fig. 2 – The dependencies of drain current I_D on gate voltage V_G of the FET based on the RGO-ZnO-PS-Si structure at drain-source voltages of V_D = 1.5 V (1) and V_D = – 1.5 V (2). Inset: the dependence of the drain current I_D on the drain-source V_D voltage

The dependence of the conductivity of the RGO-based FET on the gate voltage V_G also has features due to the conical shape of the electronic structure of the sp^2-bonded carbon atoms. In particular, a linear in-

crease in drain current was observed when the gate voltage changed from about 0.5 to – 5 V for the bias voltage V_D = 1.5 V (see Fig. 2). Similarly, the conductivity of the RGO film increased when the gate voltage increased from 0.5 to 5 V for V_D = – 1.5 V.

The dependence of the RGO-channel resistance on the gate voltage at a frequency of 1 kHz is consistent with the I_D-V_G curves (Fig. 3). Measurement in the AC mode makes it possible to obtain the value of the differential resistance R of the RGO film regardless of the sign of the voltage V_D. A non-monotonic dependence of the differential resistance on the gate voltage with a maximum at V_G = 0.6 V was observed. The maximum resistance (i.e., the minimum conductivity) is associated with the charge neutrality point, the position of which depends on the quality of the supporting layer and the local electric field caused by various factors, such as the adsorption of polar molecules or radiation-induced charges. In addition, it was found that the electronic conductivity of the RGO film (i.e., the right part of the resistance profile) is lower than the hole conductivity.

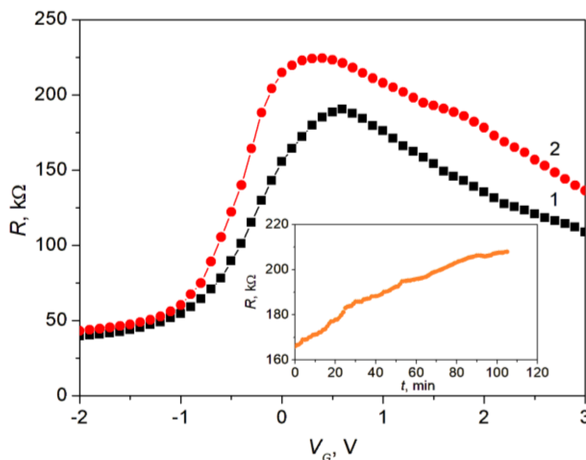

Fig. 3 – Dependencies of electrical resistance of the FET based on the RGO-ZnO-PS-Si structure at a frequency of 1 kHz on the gate voltage before (1) and after (2) irradiation from the ^{226}Ra isotope for 120 min. Inset: the FET resistance at a frequency of 1 kHz as a function of irradiation time

Although the drain current dependence on the gate voltage is usually a weak point of graphene FETs as switching devices, it can be effectively used for sensor applications. In particular, the highest sensitivity of graphene FETs to a local change in the external electric field is in the region of the charge neutrality point (Dirac point) [23]. An increase in the resistance of the FET based on the RGO-ZnO-PS-Si structure and a displacement of the charge neutrality point in the direction of a lower gate voltage were observed as a result of the joint action of alpha and beta particles and gamma radiation for 120 min. In addition, the effect of ionizing radiation on the electronic component of the RGO film conductivity was more significant than on the hole component.

The proposed field-effect sensor demonstrates an almost linear increase in resistance with irradiation time, which determines the dose of absorbed radiation (see inset of Fig. 3). It is worth noting that the radiation-induced relative change in the resistance of the FET based on the RGO-ZnO-PS-Si structure was larger than that for the RGO-SiO_2-Si structure [19]. This fact is probably related to the additional absorbing layers of the PS and ZnO. In particular, ionizing radiation can cause the formation of defects as charge capture centers in these absorbers. As a consequence, the electric field of the charges accumulated in the PS and ZnO nanostructures enhances the response of the RGO film resistance to the action of α-, β-, and γ-radiation.

To obtain additional information about the mechanisms of the influence of ionizing radiation on charge transfer processes in the FET based on the RGO-ZnO-PS-Si structure, the frequency dependencies of the resistance and capacitance between the source and gate contacts were investigated in the range of 10^2-10^5 Hz. A decrease in internal resistance and capacitance of the sandwich-like structure was observed with increasing frequency (Fig. 4).

As a result of radiation exposure for 120 min, an increase in resistance and a decrease in gate capacitance of the experimental FET were observed in the entire frequency range. The change in the resistive and capacitive characteristics of the RGO-ZnO-PS-Si structure is likely due to the formation of radiation defects. Since no noticeable recovery of the conductivity of the RGO film was detected after the cessation of the ionizing action, radiation defects are probably the main factor in the electrical response of the field-effect detector of ionizing radiation. Moreover, the greater thickness of the PS and ZnO layers compared to the nanometer-sized SiO_2 layer in graphene FETs can reduce the influence of radiation-generated electron-hole pairs in the silicon substrate on the conductivity of the RGO film. However, the possibility of the formation of GO-like structural defects in the RGO film due to the alpha particle irradiation should not be neglected [24]. The revealed features of the radiation effect on the conductivity of the FET based on the RGO-ZnO-PS-Si structure can be used to create dosimetric devices.

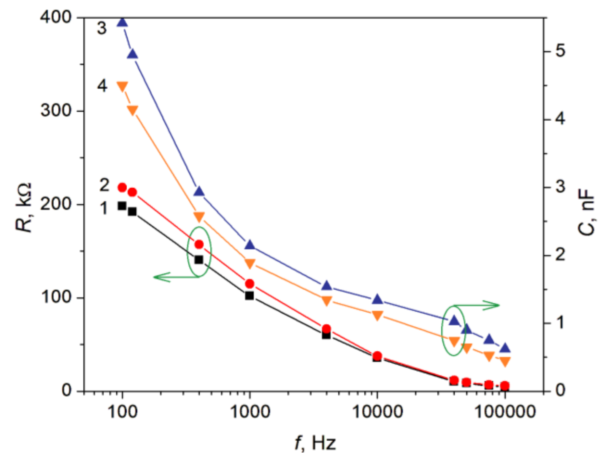

Fig. 4 – Frequency dependencies of the resistance (1, 2) and capacitance (3, 4) of the RGO-ZnO-PS-Si sandwich-like structure before (1, 3) and after (2, 4) irradiation from the ^{226}Ra isotope for 120 min

An additional argument in favor of the hypothesis regarding the formation of radiation defects in the high-resistance layers of the nanostructured PS and

ZnO can be the results of capacitance–voltage profiling. Normalized capacitance–voltage characteristics of the RGO-ZnO-PS-Si structure before and after irradiation for 120 min are shown in Fig. 5. The ratio of minimum to maximum capacitance C/C_{max} is usually used to evaluate the concentration of charged impurities in a semiconductor and the quality of the dielectric layer of MOS structures. Since the dopant concentration remains unchanged, the C/C_{max} ratio depends on the surface charge associated with electrically active defects in the PS and ZnO layers.

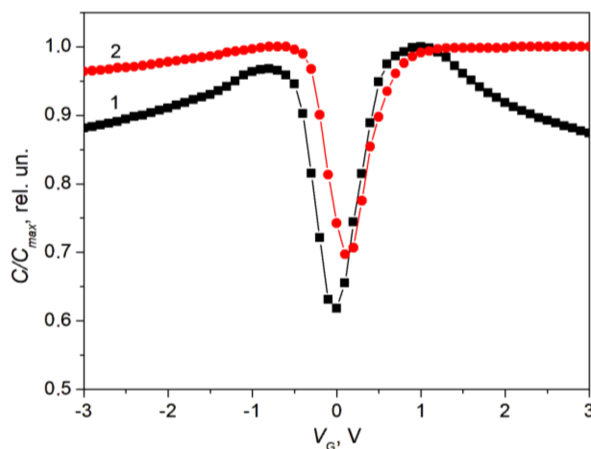

Fig. 4 – Normalized capacitance–voltage characteristics of the RGO-ZnO-PS-Si sandwich-like structure before (1) and after (2) irradiation from the ^{226}Ra isotope for 120 min

The α-, β- and γ-radiation joint action from the ^{226}Ra isotope causes the transformation of the C-V_G curves, as can be seen in Fig. 5. In particular, an increase in the C/C_{max} ratio and the displacement of the minimum capacity of the RGO-ZnO-PS-Si structure were observed, which indicated an increase in the concentration of electrically active defects in the high-resistance PS and ZnO layers.

4. CONCLUSIONS

In this study, we proposed the FET based on the RGO-ZnO-PS-Si sandwich-like structure as a sensor of ionizing radiation. Peculiarities of the process of charge carrier transfer in the created FET have been investigated using the analysis of the I_D-V_D and I_D-V_G curves and resistive and capacitive properties of the hybrid structure in AC mode. It was found that the maximum of the RGO film resistance associated with the charge neutrality point on the conductivity profile of the FET was at the gate voltage of 0.6 V and shifted in the direction of a lower gate voltage due to the influence of ionizing radiation. In addition, the joint action of the α-, β- and γ-radiation from the radium isotope causes not only an increase in the RGO film resistance but an increase in the internal resistance and a decrease in the electrical capacity of the RGO-ZnO-PS-Si sandwich-like structure as well as a transformation of its capacitance–voltage characteristic. The formation of electrically active defects in high-resistance layers of the nanostructured PS and ZnO is considered as the main mechanism of the electrical response of the proposed field-effect sensor to the action of ionizing radiation.

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Польовий транзистор на основі плівки відновленого оксиду графену з поглинаючими шарами ZnO та поруватого кремнію для виявлення іонізуючого випромінювання

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Для виявлення іонізуючого випромінювання створено графеновий польовий транзистор на основі сендвіч-структури відновлений оксид графену – оксид цинку – поруватий кремній – кремнієва підкладка. Гібридну структуру було отримано в результаті послідовних технологічних процесів фотоелектрохімічного формування наноструктурованого поруватого кремнію, електрохімічного осадження оксиду цинку, нанесення плівкоутворювальної суспензії наночастинок відновленого оксиду графену та подальшого висушування за кімнатної температури. Проаналізовано залежності струму стоку від напруги стік-витік та напруги затвора отриманого польового транзистора. Виявлено збільшення опору плівки відновленого оксиду графену поблизу точки нейтральності заряду, спричинене опроміненням ізотопом ^{226}Ra. Крім того, спостерігалося зміщення точки нейтральності заряду в сторону нижчої напруги на затворі. Встановлено, що іонізуюче випромінювання здійснює більший вплив на електронну складову провідності плівки відновленого оксиду графену, ніж на діркову. Встановлено підвищення чутливості створеного детектора іонізуючого випромінювання завдяки використанню додаткових поглинаючих шарів оксиду цинку та поруватого кремнію. На основі аналізу частотних залежностей внутрішнього опору та електричної ємності, а також вольт-фарадних характеристик сандвіч-структури обговорюються механізми впливу α- та β-частинок і γ-квантів на електричні характеристики запропонованого сенсора радіації. Отримані результати мають високий потенціал для створення нового типу малогабаритних дозиметричних приладів на основі графенових польових транзисторів з використанням простих та недорогих методів і матеріалів.

Ключові слова: Відновлений оксид графену, Польовий транзистор, Поруватий кремній, Оксид цинку, Сенсор іонізуючого випромінювання.