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Ionizing Radiation Detection Using the Field Effect Transistor Based on Reduced Graphene Oxide

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In this study, the graphene field-effect transistor (FET) has been created by depositing a film-forming suspension of reduced graphene oxide (RGO) on the silicon substrate with a SiO₂ layer and air-drying at room temperature. The possibility of using the obtained FET for ionizing radiation detection was investigated. The electrical characteristics of the radiation sensor based on the RGO film were studied in modes of direct current and alternating current. The dependencies of the drain current and the resistance of the RGO film on the gate voltage of the obtained FET were analyzed. Linear sections on the dependence of the drain current on the gate voltage, the positions of which are determined by the sign of the drain-source voltage, were detected. A decrease in the conductivity of the FET based on the RGO film near the point of charge neutrality due to irradiation from the 226Ra isotope was established. An increase in resistance and a decrease in capacitance of the investigated FET have been found in the frequency range of 25 Hz - 1 MHz caused by the joint action of alpha and beta particles and gamma quanta. A linear dependence of the electrical characteristics of the proposed sensor on the adsorbed dose determined by the irradiation duration was discovered. Possible mechanisms of the ionizing radiation influence on the conductivity of the obtained structure based on the RGO film are considered. The generation of electron-hole pairs in the silicon substrate and the formation of radiation defects in the insulator layer are probably the main factors affecting the electrical characteristics of the RGObased FET under the action of ionizing radiation. The obtained results expand the perspective of using graphene FETs to create a new type of radiation detectors and dosimetric devices.

Keywords: Reduced Graphene Oxide, Field-Effect Transistor, Dirac Point, Ionizing Radiation, Radiation Sensor.

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1. INTRODUCTION

The detection of ionizing radiation and the quantification of absorbed dose are essential for many applications related to safety, medicine, scientific and cosmic research. The development of numerous sensors for various types of radiation is based on different physical principles, which ensures the measurement of radiation dose in a wide range [1]. However, many of the radiation detectors have such disadvantages as large size, low compatibility with traditional microelectronic technology, etc. Meanwhile, modern paradigms like IoT or embedded systems require portable, low-cost and low-power sensing elements that can be used in dosimetric devices [2-4]. These requirements are met by dosimeters using a semiconductor element base. In particular, PIN diodes and MOS field-effect transistors (FETs) with an optimized structure are sensitive to ionizing radiation. The operation of the diode-type radiation sensor at high reverse bias is related to the generation of charges in the depletion region, which causes an increase in the leakage current. The MOSFET-based sensors use the gate dielectric degradation induced by ionizing radiation. Accumulation of charge on the formed electrically active defects leads to changes in the FET drain current. Increasing the PINdiode intrinsic region volume or gate oxide thickness of the MOSFET provides high sensitivity to ionizing radiation. The CMOS active pixel sensors are also used as the alpha, beta and gamma radiation detectors [5].

Another approach to creating compact and low-power sensors for ionizing radiation is based on the use of nanostructures and nanocomposites [6-8]. Radiation is registered by changes in the electrical, mechanical or optical-luminescent properties of nanostructured materials. However, the difficulty of such approaches is the small volume of radiation-absorbing materials, which requires the possibility of detecting single high-energy particles or quanta. Considering this, graphene FETs have a high potential for the radiation-sensing application [9-11].

The prospect of using graphene in sensor electronics is due to the outstanding electronic properties of the 2D carbon nanosheet as a gapless semiconductor. The coneshaped energy structure of graphene causes high mobility of charge carriers and the possibility of controlling the position of the Fermi level by an external electrical field [12-14]. As a result, the ability to inject both positive and negative charge carriers into graphene is used in the graphene FETs [15]. Low electrical noise and high sensitivity of the ambipolar conductivity of graphene near its charge neutrality point (Dirac point) to the local electric field ensure effective registration of radiation-induced charges. In this case, the silicon substrate serves as both

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the FET gate and the absorber of ionizing radiation [11].

One of the promising methods of obtaining graphene is the oxidation and exfoliation of graphite to obtain graphene oxide with its subsequent chemical reduction [16]. This approach does not require complex, long-term, and energy-consuming technological processes and ensures a low cost of reduced graphene oxide (RGO) nanosheets. In addition, the RGO film also demonstrates the dependence of resistance on the external electric field and can be used as a conducting channel of the FET [17]. Therefore, the possibility of using the RGO-based FET as a sensor of ionizing radiation was studied in the work.

2. EXPERIMENT

The weakly doped silicon wafer with a thickness of $400\,\mu\mathrm{m}$ and a specific resistance of $10~\mathrm{Ohm}\,^{\bullet}\mathrm{cm}$ was used as a substrate for the radiation detector based on the RGO film. The dielectric layer on the silicon substrate surface was obtained by evaporating silicon powder using a VUP-5M vacuum system with a residual air pressure of $\sim 10^{-3}$ mm Hg and subsequent depositing non-stoichiometric $\mathrm{SiO}_x~(x<2)$ formed as a result of the reaction of vaporous silicon with residual oxygen. The SiO_x layer thickness measured using the MII-4 microinterferometer was about $100~\mathrm{nm}$. Additional heat treatment in air at the $1050^{\circ}\mathrm{C}$ temperature for 2 hours ensured the formation of the silicon dioxide dielectric layer for RGO-based FET.

Graphene oxide aqueous suspension produced by Sigma-Aldrich (USA) with a concentration of 2 mg/ml was used to obtain RGO nanosheets. Graphene oxide nanoparticles were reduced by the joint action of hydrazine monohydrate and ultrasonic treatment for 20 min. The obtained suspension was mixed with the 0.2 M aqueous solution of sodium dodecylbenzene sulfonate for hindering aggregation of the RGO nanosheets and deposited on the SiO₂ layer surface. The RGO film obtained after drying suspension in the air at room temperature was used as the FET conducting channel. The source and drain silver contacts at a distance of 1 mm from each other were formed on the RGO film surface by thermal deposition technique. The silicon substrate was the gate of the created FET. The inset of Fig. 1 shows a schematic representation of the RGO-based FET for ionizing radiation sensing.

The electrical characteristics of the obtained FET were studied in both DC and AC modes at room temperature. The influence of ionizing radiation on the resistance and capacitance of the RGO-based FET was studied in the 25 Hz -1 MHz frequency range using an RLC measuring device. The radium isotope $^{226}\mathrm{Ra}$ with a radiation intensity of 0.1 mKi was used as a radiation source for experimental studies. As a result of the decay of the $^{226}\mathrm{Ra}$, the most probable emission is γ -radiation with energy of 0.19 MeV. The average energy of α - and β -particles is 4.78 and 0.17 MeV, respectively. Since the RGO-based FET was placed directly near the radioactive source, the sensor response depended on the joint effect of alpha, beta and gamma radiation. The exposure duration determined the dosage of absorbed radiation.

3. RESULTS AND DISCUSSION

One of the main parameters of the FETs is the switching characteristic, which is determined by the dependence of the drain current I_D on the gate voltage V_G and reflects the ratio of the current in the on state to the current in the off state. In general, the switching characteristic is a weak point of graphene FETs due to the zero band gap of the graphene monolayer. On the other hand, exactly the region near the Dirac point on the ambipolar conductivity profile of graphene is the most important for sensor applications [18]. In this case, the electrical properties of graphene are the highest sensitive to the local electric field. The dependence of the drain current on the gate voltage of the RGO-based FET for the bias voltage $V_D = \pm 1.5$ V is shown in Fig. 1.

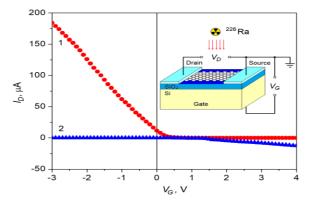


Fig. 1 – The dependencies of the RGO-based FET drain current on the gate voltage for $V_D = 1.5$ V (1) and $V_D = -1.5$ V (2). Inset: schematic representation of the RGO-based FET

The measured I_D – V_G dependencies have linear sections, the position of which depends on the sign of the drain-source voltage V_D . A larger change in the drain current I_D associated with the change in gate voltage V_G was observed at $V_D = 1.5$ V than at $V_D = -1.5$ V. The obtained dependencies of the drain current I_D on the gate voltage V_G are correlated with the resistance R profile of the RGO-based FET in the AC mode at a frequency of 1 kHz (see Fig. 2).

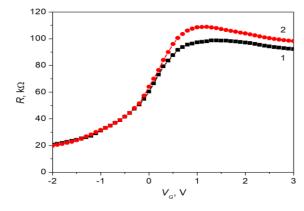


Fig. 2 – Dependence of the RGO film resistance at a frequency of 1 kHz on the gate voltage before (1) and after (2) irradiation from 226 Ra isotope for 90 min

It is worth noting that there was no pronounced Dirac point (i.e., the minimum of the RGO film conductivity in the case where the Fermi level passes through the point of contact of the cone-shaped conduction and valence bands of graphene). The RGO film resistance gentle maximum associated with the point of charge neu-

trality was observed near the gate voltage $V_G = 1.2 \text{ V}$. The revealed features of the RGO-based FET electrical properties can be due to the inhomogeneity of the RGO film formed from graphene nanosheets. Besides, electrically active defects in the SiO_2 layer on the silicon substrate can have different effects on the transfer of charge carriers of various signs in graphene.

As can be seen in Fig. 2, the RGO film demonstrates an increase in the resistance under the influence of the joint action of alpha and beta particles and gamma quanta from the ²²⁶Ra source. The greatest impact of ionizing radiation on the RGO-based FET resistance profile was observed near the Dirac point. Besides, the response of the electronic component of the RGO conductivity to radiation was greater than that of the hole component.

To obtain additional information about the mechanisms of ionizing radiation influence on charge transfer processes in the RGO-based FETs, the frequency dependences of resistance and capacitance in the range of $25~{\rm Hz}-1~{\rm MHz}$ were studied. Impedance spectra were measured using the source and drain contacts. A decrease in internal resistance and capacitance of the experimental sample was observed with increasing frequency (see Fig. 3 and Fig. 4, respectively).

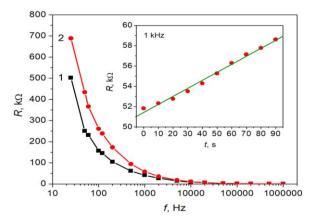


Fig. 3 – Frequency dependence of the RGO-based FET resistance before (1) and after (2) irradiation from $^{226}\mathrm{Ra}$ isotope for 90 min. Inset: the RGO-based FET resistance at a frequency of 1 kHz as a function of irradiation time

Irradiation of the FET from the RGO film side with the $^{226}\mathrm{Ra}$ isotope for 90 min causes increasing its resistance and decreasing the capacitance. Radiation-induced changes in the electrical characteristics of the RGO-based FET were higher in the low-frequency range. In the case of increasing the dose of α -, β - and γ -radiation, which corresponds to a longer duration of exposure, the obtained sensor of ionizing radiation demonstrates a linear increase in resistance and a linear decrease in capacitance, as shown in insets of Fig. 3 and Fig. 4.

The observed dependencies may be due to several factors. In particular, the electron-hole pairs generated by radiation in the silicon substrate are separated by the

electric field of the gate in such a way that holes accumulate near the $\mathrm{SiO_2}$ layer [10]. As a result, the field of localized holes causes a change in the conductivity of the RGO film. The formation of radiation defects in the insulator layer is probably the main factor affecting the electrical characteristics of the RGO-based FET after the cessation of the action of ionizing radiation. The formed defects cause the trap of charges in the $\mathrm{SiO_2}$ layer. The total ionizing dose effect in MOSFET structures is usually explained by trapped charges [19]. In addition, the formation of structural changes in the RGO film under the action of alpha particles, as was found in graphene oxide nanosheets [20], should not be neglected. In turn, structural defects can be centers of scattering of charge carriers in the RGO film.

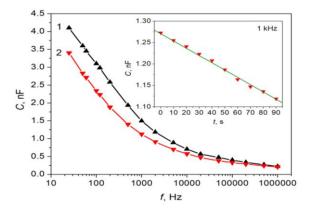


Fig. 4 – Frequency dependence of the RGO-based FET capacitance before (1) and after (2) irradiation from 226 Ra isotope for 90 min. Inset: the RGO-based FET capacitance at a frequency of 1 kHz as a function of irradiation time

4. CONCLUSIONS

A new technical solution for creating ionizing radiation detectors based on graphene FETs is proposed in the work. The RGO film deposited on the surface of the SiO₂ layer on the silicon substrate was used as the conducting channel of the FET. Based on the analysis of the dependencies of the drain current on the gate voltage and resistance profiles, it was found that the position of the Dirac point in the obtained RGO-based FET was near the 1.2 V gate voltage. Irradiation by alpha and beta particles and gamma quanta from the ${}^{226}\mathrm{Ra}$ isotope causes a change in the dependence of the RGO film resistance on the gate voltage. The obtained sensors demonstrate the highest sensitivity to ionizing radiation in the region near the charge neutrality point. A linear increase in the internal resistance and a linear decrease in the capacitance of the RGO-based FET with increasing the irradiation duration have been established in the AC mode. The ionizing radiation influence on the electrical characteristics of the sensor was higher in the low-frequency range.

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Детектування іонізуючого випромінювання за допомогою польового транзистора на основі відновленого оксиду графену

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У роботі створено графеновий польовий транзистор шляхом нанесення плівкоутворювальної суспензії відновленого оксиду графену (RGO) на кремнієву підкладку з шаром SiO2 і подальшого висушування на повітрі за кімнатної температури. Досліджено можливість використання отриманого польового транзистора для детектування іонізуючого випромінювання. Вивчено електричні характеристики сенсора радіації на основі плівки RGO у режимах постійного та змінного струму. Проаналізовано залежності струму стоку та опору плівки RGO від напруги на затворі отриманого польового транзистора. На залежності струму стоку від напруги на затворі виявлено лінійні ділянки, положення яких визначаються знаком напруги зміщення. Встановлено зменшення провідності польового транзистора на основі RGO поблизу точки нейтральності заряду внаслідок опромінення ізотопом ²²⁶Ra. Заресстровано збільшення опору та зменшення ємності досліджуваного польового транзистора у частотному діапазоні $25~\Gamma \text{ц} - 1~\text{М}\Gamma \text{ц}$, зумовлене спільною дією альфа- та бета-частинок і гамма-квантів. Виявлено лінійну залежність електричних характеристик запропонованого сенсора від поглинутої дози, яка визначається тривалістю опромінення. Розглянуто можливі механізми впливу іонізуючого випромінювання на провідність отриманої структури на основі плівки РГО. Генерація електронно-діркових пар у кремнієвій підкладці та утворення радіаційних дефектів в ізоляційному шарі SiO₂ є, ймовірно, основними факторами, що впливають на електричні характеристики польового транзистора на основі плівки RGO під дією іонізуючого випромінювання. Отримані результати розширюють перспективи використання графенових польових транзисторів для створення нового типу детекторів радіації та дозиметричних пристроїв.

Ключові слова: Відновлений оксид графену, Польовий транзистор, Точка Дірака, Іонізуюче випромінювання, Сенсор випромінювання.