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Development of a Reduced Graphene Oxide-Based X-Ray Detector for Space Applications

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Abstract- In this work, a light weight, reduced Graphene Oxide Field Effect Transistor based radiation detector is developed which detects X-rays at room temperature. Graphene Oxide is synthesized from Graphite flakes using modified Hummer's method which is then reduced by Hydroiodic acid fumes followed by low temperature treatment. Material characterization using X-ray diffraction technique and Raman spectra confirmed the reduction of Graphene oxide to reduced graphene oxide. Synthesized reduced graphene oxide is then put in back gate Field Effect Transistor architecture. The electrodes of the Field Effect transistor are made using thin film deposition technique. The detector is housed inside a simple packaging setup. The device showed promising response with X-rays of energy 20-40 KeV at room temperature at various incoming flus of X-ray photons. Response curve of device showed linear response with increasing X-ray energy and current. The device demonstrated a rise time of 0.25 s and fall time of 0.15s

***Keywords*:** Graphene, reduced Graphene oxide, X-ray detector, Field effect Transistor

1. Introduction

Radiation environment in space is a very relevant area of continuous study both from the science and technology point of view. Space radiation environment is dynamic and knowing this or measuring this is of paramount importance in space missions. Thus, a light weight, portable radiation detector which can detect radiations like X-rays and Gamma rays in high flux environment can find significant application in space missions including scientific exploratory missions. Graphene is a two-dimensional sheet of carbon atom in a honeycomb lattice [1]. Graphene and its derivatives like reduced graphene oxide (rGO) show excellent mechanical and electrical properties They are extremely light weight which makes them suitable for fabricating any light weight portable device [2]. Reports are found on the area of radiation detection with Graphene Field Effect Transistors (GFET) using pristine graphene [3]. NASA is leading the league, in this context by bagging a patent in this area [4]. The invention relates to the use of GFETs for radiation sensing applications. But so far, the Graphene based radiation detectors have been reported are made up of exfoliated pristine graphene [4] or made using chemical vapor deposition technique [3]. But controlling the device size with exfoliated graphene is challenging, while CVD made graphene requires sophisticated instruments and trained manpower. Also, the bandgap of graphene is zero, which limits the detection efficiency of pure graphene-based photodetectors [5,6]. This is because a single layer of graphene can only absorb 2.3% of incident light [7]. In addition, since the recombination time in graphene is few picoseconds, very few charge carriers are detected before they recombine. To circumvent this, graphene derivatives with a larger band gap, such as reduced graphene oxide (rGO), graphene quantum dots, and graphene nano ribbons, are being investigated [8,9,10,11,12] for use in photon detectors.

In this work, a thin film of reduced graphene oxide (rGO) is used for the development of a prototype for an X-ray detector. The main advantage of using rGO is that it is synthesized by simple hybrid technic which involves both

chemical and thermal treatment of Graphene oxide. [14,15] The use of rGO in this device makes the synthesis of the sensing material very easy and economic as it does not require trained manpower and sophisticated instruments. Also, the device fabrication can be done with controlled dimensions and the mass production of rGO is easy and achievable. The technical approach is to take advantage of rGO's sensitive dependence on the local electric field, which can be rapidly modified by carriers created by absorbed radiation in the underlying absorber material. This dependence, which is visible even at room temperature, offers the distinct advantages of room temperature operation of this rGO based X ray detector.

2. Material and method

In this work, a reduced graphene oxide-based X-ray detector is fabricated and studied. The detector has a Field Effect transistor architecture. It consists of a 1000 ohm-cm resistivity silicon substrate acting as radiation absorber which is covered by a 100 nm thin layer of silicon dioxide. Figure 1 depicts a schematic of the fabricated bottom gate architecture rGOFET. The silicon dioxide layer that serves as the dielectric layer is covered by a 10 µm-thick layer of rGO. This thin layer of rGO serves as the sensing layer in the X-ray detector. Graphene-based materials possesses the property that its resistance varies with the local electric field [16,17,18] as shown in Fig 2. And this property is utilized in detection of X-rays. Inside the absorber, in this instance silicon, electrons and holes are created when X-ray photons strike the absorber. The gate voltage supplied between the source and gate electrodes generates an electric field across the silicon substrate, which drives charge carriers to the rGO layer. The thin dielectric layer, here the silicon dioxide layer beneath the rGO layer stops charge carriers from entering the rGO layer. The oxide layer has a thickness of 100nm and is produced by the dry oxidation of silicon.

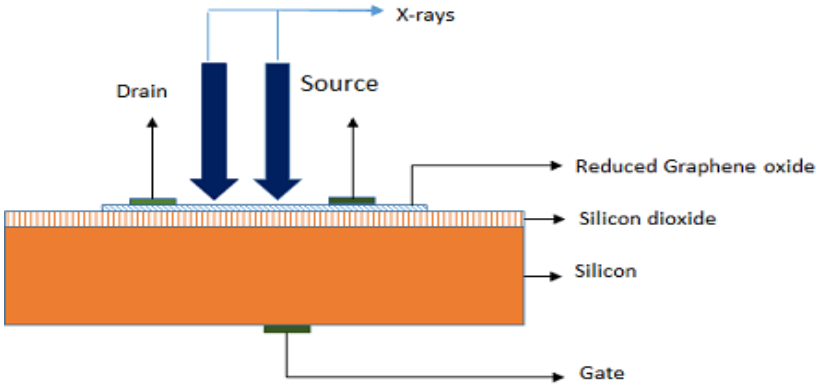

Fig.1 Schematic of rGOFET as X-ray detector.

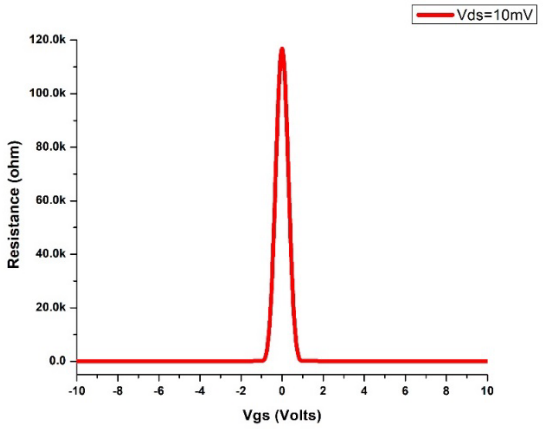

Fig 2: Variation of resistance of rGO with electric field. Vgs represent gate source voltage and Vds represents drain source voltage.

2.1. Synthesis of Graphene Oxide

The process of producing reduced graphene oxide (rGO) starts by utilizing graphite flakes (300 m, 99.9% purity). Modified Hummer Method [19] was employed for the synthesis of graphene oxide (GO). In order to enhance conductivity, a suspension of graphene oxide (GO) in N-Methyl-2-pyrrolidone (10mg / 10 ml) was prepared and subjected to ultrasonication for a duration of three hours to achieve a uniformly dispersed GO solution.

2.2. The Reduction of Graphene Oxide to Reduced Graphene Oxide on a Silicon Substrate

A small piece (1cmx1cm) high resistivity silicon wafer (1000-ohm cm) with a thickness of 500 µm was used as the radiation absorbing substrate for the X-ray detector. The top surface of the wafer is oxidized to produce a 100 nm oxide layer. Over the oxide layer, a square area deposited with Graphene oxide (GO). In order to get a uniform coating of GO, the GO solution was applied onto the oxidized substrate using a drop casting technique, followed by drying at a temperature of 120°C. The square area was produced using shadow mask made up of stainless steel. The GO was subjected to reduction through the utilization of hydroiodic acid and low temperature thermal treatment for a duration of 6 hours [14]. This process was carried out under a mild vacuum condition, maintaining a low temperature of 120°C.The thickness of thin film of rGO produced is ~10 µm.

Fabrication of rGO based X-ray detector.

Source and drain electrodes deposited over the rGO layer using thin film of aluminium. The back side of entire silicon wafer is coated with thin film of aluminium which serves as gate electrode. The sensor is housed in a simple package with a removable cap, attached to a PCB board inside a 3D-printed casing. The cap has a window that can be fitted with a mylar sheet for X-ray testing. Figure 1 shows the schematic of a X-ray sensor. Figure 3(a) shows the image of the fabricated sensor. Figure 3(b) shows the CAD model for the packaging. Detailed description of the experimental setup is given in section 3.2.

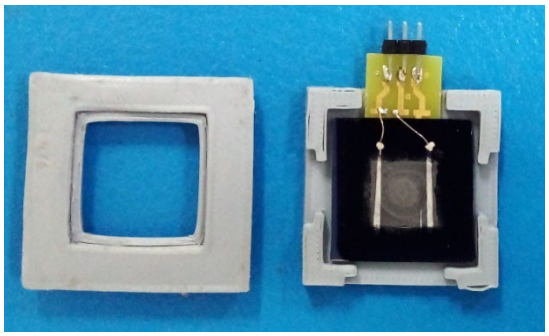

Fig 3(a) Actual picture of the fabricated rGOFET with separate top cap

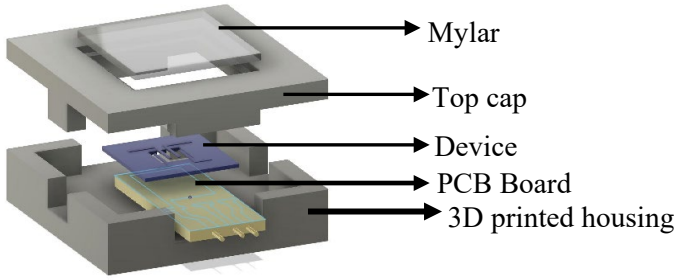

Fig 3(b): CAD MODEL for the packaged device

3.Results and Discussion

3.1. Material Characterization of Graphene Oxide and Reduced Graphene Oxide

The produced graphene oxide (GO) and reduced graphene oxide (rGO) were subjected to material characterization using X-ray Diffraction (XRD) and Raman spectroscopy techniques. In Figure 4(a), it is seen that the peak corresponding to graphene oxide (GO) occurs at an angle of $2\Theta = 11.90°$, while for reduced graphene oxide (rGO), it is observed approximately at $2\Theta = 24.6°$. This observation provides evidence supporting the reduction of GO to rGO. Figure 4(b) displays the Raman spectra of graphene oxide (GO) and reduced graphene oxide (rGO). The D band of graphene oxide (GO) has a characteristic peak at a wavenumber of 1350 cm-1, while the G band is detected at 1591 cm-1. In the case of reduced graphene oxide (rGO), the D band is detected at a wavenumber of 1355 cm-1, whereas the G band is discovered at a wavenumber of 1593 cm-1. The ratio of the intensities of the D and G peaks, ID/IG, serves as an indicator of the degree of reduction. The intensity ratio of ID/IG for reduced graphene oxide (rGO) is 1.39, which is greater than the corresponding ratio of 0.92 for graphene oxide (GO), as depicted in Figure 4(b). Thus, it is proved that Go has been reduced properly to rGO.

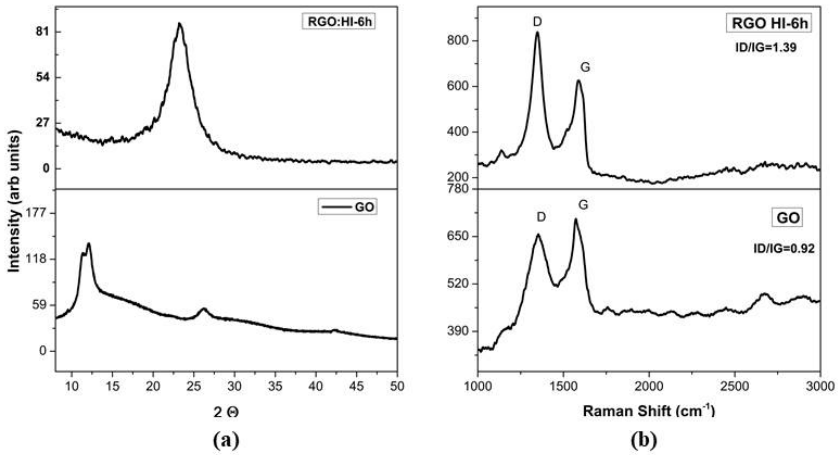

Fig. 4(a) X ray diffraction spectrum of synthesized GO and rGO. (b) Raman spectra of synthesized GO and rGO.

3.2 Performance study

X-ray testing was performed on the fabricated detector to determine the responsivity of rGO material to X-rays. Experiments were done for various energies and flux of incoming X-ray photons ranging from 20 KeV to 40 KeV. The test was performed in a cyclic ON-OFF procedure to estimate the responsivity, response time and the recovery time of the device. The photocurrent flowing through the device was measured using Keithley 2450 source meter. Responsivity of a detector is defined as the change in photocurrent (current produced due to incoming X-ray photons/light photons falling on the detector) per unit change in power of the incoming radiation.

X-rays were made to fall on rGOFET array from X-ray gun (mini AMPTEK X-Ray tube) which emits X-rays up to 40 KeV energy. The detector was placed 15 cm away from the X-ray gun. It was kept inside a black box for the test while testing to cut off the interference of ambient light. A small light shielded aperture was used through which radiation entered the black box and falls on the detector. Unless otherwise specified, the device is operated at 5 V and 4V gate source voltage. The device responded to the X-rays at room temperature. Fig 5(a, b) shows the results of the ON OFF cycle for 40 kV for different current values. From the graphs it is seen that there is slightly some charge build up after every On-OFF cycle for 60,80 and 99 µA current. The effect is more pronounced at 80 µA and 99 µA. This effect is not seen at low flux (20 µA, 40 µA current). This can be explained as follows. When the incoming photon flux is high, there is some charge accumulation happening near the rGO layer. This accumulated charge is showing up in the drain source current even when the irradiation is switched off. But this is not the case for current value till 40 µA. The rise time and the fall time of the sensor is 0.25 sec and 0.15 sec respectively. The rise time and recovery time study reveals that the detectors responses very fast. This is because, it does not require any external mechanism to come back to initial level once the source is switched OFF as observed with other graphene-based detectors and hence it shows an instant response to switching ON and OFF the X-ray source at room temperature.

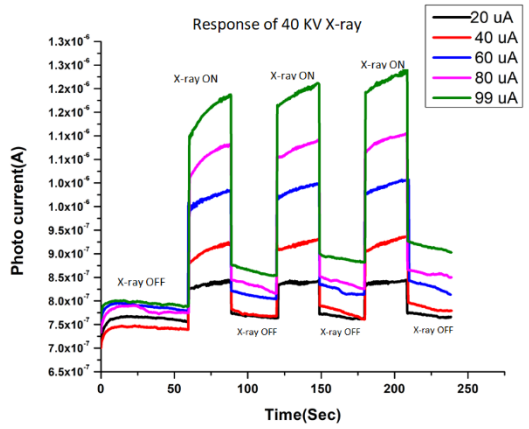

Fig 5(a): Cyclic ON-OFF response for X-rays at 40 KV and different current values at Gate Source Voltage of 5V.

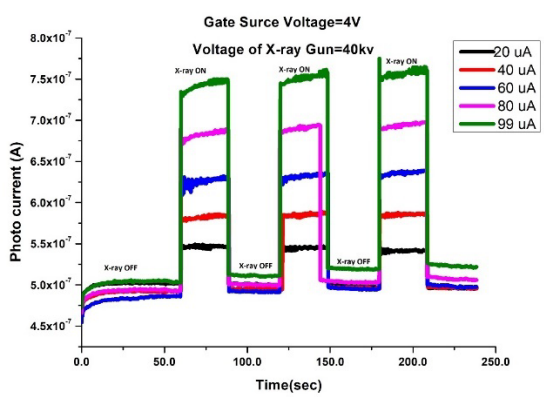

Fig 5(b): Cyclic ON-OFF response for X-rays at 40 KV and different current values for Gate Source Voltage of 4V.

Fig 6 shows the response curve of the sensor for various energies of incoming X-rays and for various flux of incoming ray photons. Higher the current of the X-ray gun, higher is the flux. It is observed that the response is linear with increasing photon flux and the photocurrent increases with increase in the energy of the incoming X-rays. This is as expected as higher energy will produce more electron and hole pairs in the absorber medium leading to increased photocurrent.

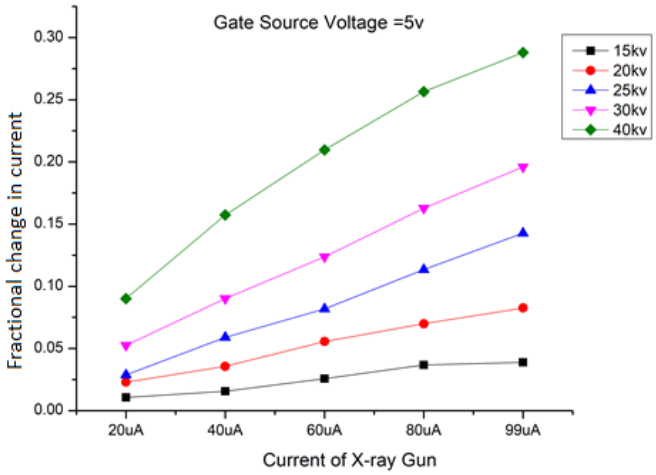

Figure 6 Response for various X-ray gun voltages and currents.

4. Conclusion

rGO has been synthesized through a hybrid process involving Hydroiodic acid fumes and thermal annealing. Graphene oxide is reduced directly on an oxidized Silicon substrate (oxidized at the upper surface), so transferring the sensing medium (rGO film) to the device substrate is not a problem. The rGOFET was then subjected to X-ray testing

to determine the rGO material's sensitivity to X-rays. The X-ray testing was conducted with incoming photon energies and fluxes spanning from 20 to 40 KeV. The test was conducted in a cyclic ON-OFF fashion to determine the rGOFET's responsiveness, response time, and recovery time. At ambient temperature, the device responded to the X-rays. The device's response curve demonstrates a linear relationship between incident X-ray energy and current. The rise and fall times of the sensor are respectively 25 seconds and 0.15 seconds. This work paves the way for investigating the rGO-based X-ray detector's response with other absorbing substrates like silicon carbide which have higher bandgap than silicon.

5. Acknowledgements

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Note: In this paper, both first and second author have contributed equally to the work.

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