Real-time nucleic acid detection via field-effect transistor sensors based on graphite oxide decorated with trimetallic nanocluster of gold, silver, and platinum

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Abstract
Field effect transistor (FET) based sensors are witnessing robust evolutions in the fabrication and characterization for the purpose of biomedical applications. In this article, a graphite oxide FET based sensor is designed, fabricated, and characterized for real-time detection of various concentrations of nucleic acid with a detection limit of 1.28 nM of DNA. This sensor consists of two gold electrodes connected through a channel of graphite oxide. Moreover, the sensor sensitivity is enhanced by decorating the graphite oxide channel with composite trimetallic nanoclusters that include gold, silver, and platinum. The developed sensor is investigated by both simulation and experiment. Both experimental and simulation agree where the current signal is higher for sensors decorated with trimetallic nanoclusters, which indicate higher sensitivity. Moreover, increasing the concentration of DNA results in an increment in the current signal thus the response is proportional to DNA concentration. The results indicate a promising sensor for real-time, reliable, and cost-effective DNA detection.

1. Introduction
Immediate bio-molecule detection is highly critical in several areas such as health and food analysis [1–3] toxicity identification [4], and cancer detection [5]. Traditional detection methods of bio-molecules require labels which provide high sensitivity, but these methods do not provide real time detection and they are costly. Health monitoring and biological detection are vital. Photoelectrochemical (PEC) technique is a new detection method, which has the electrochemical and optical techniques advantage. PEC sensors are highly selective, however they are costly [6]. Moreover, self-powered temperature sensor based on Seebeck effect transduction can open the door for new methods of clinical diagnosis. But this method requires the biomolecule to be labeled with one enzyme for steric reasons [7]. Various studies are aiming to design label free sensors to detect the target molecule, where a number of transduction techniques are being tested by researchers. Surface plasmon resonance method achieved high sensitivity for label free biological detection [8]. Currently, various sensors such as carbon nanotube [9], and silicon nanowire field effect transistors (FETs) are showing high sensitivity [10, 11]. One-dimensional based sensors such as carbon nanotubes and nanowires are being utilized for immediate identification of various bio-molecules with high sensitivity [12–14].
DNA plasmids may be genetically altered and utilized in the recombinant DNA technology. Plasmids are known as replicons, a unit of DNA that is capable of replicating autonomously by an appropriate host. Plasmids can be utilized in gene therapy, genetic vaccination, and DNA recombination technology. The quantification and detection of plasmid DNA allows surveys of the dissemination of plasmid-specific sequences. Moreover, helps in linking the plasmid abundance with environmental factors [15]. The first biosensor is an enzyme electrode sensor, and it was introduced in 1962 by Clark and Lyons [16]. After that, various bio-molecular sensors and mechanism emerged. One type of the most interesting mechanism is FET based sensors due to their novel features such as quick development, cheap production, fast response, and high sensitivity [17]. Moreover, the availability of mature manufacturing techniques offers the advantage of integration with other systems and circuits which is highly important in sensors development. Nanomaterial based FETs are promising candidates for bio-molecular detection by utilizing electronic measurements [18, 19]. Transistor-based sensors consist of three electrodes: source, drain, and gate where the channel connecting the two electrodes works as the biological detection area that interacts with the molecules to sense their electrical measurements, concentrations, and presence [20].

Nanotechnology evolution triggers the utilization of nanostructures such as nanotubes, nanowires, and nanoclusters in the bio-molecular detection field. It is highly important to take into consideration the material used to fabricate the bio-molecular transistor-based sensor. Graphene is made of 2D honeycomb lattice of carbon atoms [21]. Graphene has outstanding characteristics such as high mobility [22, 23], very large area [24], ultra-high mechanical strength [21], and unique electrical properties [25] which made it the perfect material in bio-molecular transistor applications. Numerous experimental and theoretical research have been conducted targeting graphene properties in various applications [26]. Graphene is considered the ideal membrane for DNA identification [27–34] since transistor based sensors with graphene channel offer quick, accurate, real-time, and label-free detection. But it is inefficient to produce large amounts of graphene membranes. Therefore, graphite oxide is being used as an alternative. Graphite oxide can be used as a promising alternative of graphene since it is easier to produce, more economical, and has the required sensing features. Graphite oxide is attracting researchers’ interest since it can be easily diluted in water and used in various applications such as nano-electronic devices [35]. Graphite and its derivatives such as graphene, graphite oxide, and graphite oxide provide novel opportunities and approaches for DNA detection. Graphene oxide is an oxidized form of graphene, while graphite oxide is multilayer of graphene oxide. Graphite oxide exfoliation results in stabilized individual graphene oxide layers. In this work, DNA sensors were built experimentally using graphite oxide, while one layer of graphene oxide was used to build the sensor in simulation to reduce the computational cost in simulation. Decorating the fabricated sensor with metallic nanoclusters improves the sensor sensitivity and response time [36, 37]. These nanoclusters change the surface to volume ratio of the sensor [38]. Nanoclusters can be generated using an inert gas aggregation technique inside an ultra-high vacuum compatible system (UHV) [39] which has some advantages over other chemical techniques such as high purity, and size selection [40, 41].

Platinum nanozymes have been massively used in various research areas, such as pressure-based biosensors [42]. Single-atom catalysts such as platinum combined with DNA nanotechnology opens the door for new methods for DNA detection [42]. Single-atom biosensor could massively enhance the application of different sensing platform and provide the opportunity to build a new type of biosensor by integrating with other sophisticated nanotechnology [42]. Moreover, the PEC sensing has become a hot research topic of electrochemical sensing due to its advantages such as high sensitivity, low cost, and easy miniaturization [43]. The photoactive materials are critical in designing a high performance PEC sensor [43]. The PEC sensing mechanism has provided the new opportunities in detecting gas molecules, metal ions, small molecule, proteins, and nucleic acid [43]. Furthermore, the piezoresistive sensing method was used to substitute the conventional fluorescence quenching which improves the detection process. Wireless piezoresistive biosensor offers a substitute to develop molecular detection and shows more possibilities in developing wireless bioanalysis [44]. Noble metal nanoclusters have been widely used for various applications such as biomedical ones [45–49] where of the most used noble metals nanoclusters are gold, silver, and platinum. Silver nanoparticles have potential in different chemical/biological applications. Silver nanoparticles can be used in different applications due to the variation in their shape, size and synthesis process such as photosynthesis [50]. Eco-friendly synthesis processes for nanoparticles such as plant-based green synthesis are being more common in nanobiotechnology [50, 51]. Noble bimetallic and trimetallic nanoclusters are attracting researchers’ interest because of the possibility to design their properties by integrating with other sophisticated nanotechnology [52–55], and surface modifications [56, 57]. Bimetallic and trimetallic nanoclusters have diverse morphology [58, 59], improved antimicrobial action [60], enhanced catalytic activity [61–66], very good stability [67, 68], and high sensitivity and selectivity [69–71]. As an example of composite metallic nanoclusters, mixing Au with Ag improves the antibacterial activity and the therapeutic concentration because of the strong electronic ligand impact. Ag/Au/Pt trimetallic composite
have shown impressive outcomes in bio-sensing applications [60]. These promising characteristics are assigned to the multi-functional influence stimulated by two or three metals within the nanocluster. Previous work shows that using trimetallic nanocluster such as Cu/Au/Pt has stronger catalytic activity than bimetallic or monometallic nanoclusters. Therefore, trimetallic nanoclusters can be investigated for biosensing applications such as cancer cell and glucose detection [72, 73].

The aim of this study is to develop, design, and fabricate an FET based sensor made of graphite oxide channel and decorate it with trimetallic nanoclusters of silver, gold, and platinum which are generated by an UHV system. The sensor is utilized to detect various concentrations of DNA. Moreover, the sensor electrical characteristics and performance are examined by simulation to confirm the experimental results. The novelty in this article is based on the addition of new materials (composite nanoclusters of gold, silver, and platinum) that enhanced the sensor performance significantly as shown in simulation and experiment.

2. Material and methods

2.1. Experimental

2.1.1. Design and fabrication of DNA detection sensors

DNA sensor was fabricated by utilizing a commercial silicon wafer which has a top layer of silicon dioxide (SiO2). The silicon wafer was divided into small parts of a size of 1.0 cm × 0.5 cm. Then, the silicon wafer was cleaned with acetone, ethanol, and deionized water. After that, the wafers were dried by nitrogen gas. Depositing the electrodes on the silicon wafer started by placing a 5 nm layer of nickel-chrome (NiCr) by thermal evaporation process, then 30 nm layer of gold (Au) was deposited through a shadow mask displayed in figure 1(c). The NiCr was deposited to enhance the adhesion among the gold electrodes and silicon wafer [36, 74]. Commercial graphite oxide (GO) of 4 mg ml⁻¹ was utilized in the fabrication process. A drop of the commercial GO was placed between the fabricated electrodes and left in room temperature for 24 h. Figure 1(a) displays a schematic diagram of the sensor which has been developed and utilized to detect various concentrations of DNA, and figure 1(b) shows the cross-sectional diagram of the sensor.

Keithley 236 source measuring unit that is controlled by a computer was used to study the sensor performance by generating the current–voltage (I(V)) measurements [75]. It was also utilized to measure the difference in electrical current due to DNA placement on the sensor to determine the sensor sensitivity.

2.1.2. Nanocluster deposition

Composite nanoclusters of gold, platinum and silver were dropped on the graphite oxide surface inside the UHV system (Nanogen-50, Mantis Deposition Ltd. Oxfordshire, UK). Magnetron sputtering and inter gas condensation were deployed to generate nanoclusters from composite target of gold, platinum, and silver in ratio of 1:1:1 and 99.99% purity that was fixed on the magnetron sputter head. The UHV has two turbo pumps and a rotary pump to evacuate the system and reduce its pressure to 10⁻⁶ mbar [76]. Argon gas was used to produce plasma inside the chamber to sputter metal from the target by DC discharge power. Nanoclusters were then generated and travel inside the UHV to be placed on the FET. The condensation of the sputtered nanoclusters was facilitated by argon (Ar) gas [76, 77]. The aggregation length (L), which is defined as the interval between the exit nozzle and the target, was set at 70 mm. The argon gas flow rate was fixed at 40 sccm, and the discharge DC power was set at 10.8 W. The nanocluster size can be modified by changing the Ar flow rate, aggregation length, and the DC discharge power [38]. A quadrupole mass filter (QMF) was utilized to measure the size distribution of the nanoclusters within the source chamber by utilizing four parallel metal rods [78, 79]. Each couple of opposite rods is electrically connected together to potentials of −[U + V cos(wt)] and [U + V cos(wt)], where U refers to dc voltage and V cos(wt) refers to ac voltage. U/V ratio represents the resolution of the mass filter and it was fixed at 0.08 while the frequency w was varied during scanning the mass distribution [78, 79]. The nanocluster ion flux of the selected mass/size was measured by a grid placed at the exit of the mass filter. The nanoclusters composition was confirmed by employing energy dispersive x-ray spectroscopy (EDS) technique.

2.1.3. Preparation of the DNA solution

Plasmid DNA preparation was carried out using QIAGEN Midi Kit (QIAGEN) as per the manufacturer’s instructions. Briefly, transformed bacterial were grown overnight in LB media supplemented with ampicillin. Cells were pelleted and resuspended in P1 buffer and then lysed with P2 buffer. Nucleic acid was precipitation by addition of solution P3 followed by incubation on ice for 30 min. The DNA plasmid was then recovered by centrifugation and loaded onto Qiagen tip column and eluted with QF buffer. Eluted Plasmid DNA was next precipitated using isopropanol, pelleted by centrifugation, washed with 70% ethanol, air dried before being resuspend in HyPure molecular biology water (Hyclone).
Figure 1. (a) Schematic diagram of the transistor-based sensor. The Au electrodes are connected through a channel of graphite oxide where the channel is decorated with composite trimetallic nanoclusters (Ag, Au, and Pt). (b) Cross sectional diagram of the sensor. Color-code: gold-yellow, silver-light gray, and brown-platinum. (c) Stainless steel shadow mask used in experiment.

2.2. Simulation

This part aims to demonstrate the detection of various concentrations of DNA by first-principles computations and to confirm the experimental results. Quantumwise Atomistix Toolkit (ATK) and its graphical user interface Virtual Nanolab (VNL) were used to design and simulate the proposed sensor. Device current has been generated and measured with the ATK simulator where the simulator has different methodologies to generate the electronic transport properties of the built sensor. In order to, speed up the simulation process, high-performance computing (HPC) environment has been used with 7 nodes each with 36 processors. In total 252 processors were used to conduct the simulation.

Figure 2 displays the schematic of the simulated nanoscale electronic device. Figure 2(a) displays a two-dimensional schematic view of the designed GO-FET sensor and figure 2(b) illustrates the cross-sectional view of the investigated sensor. The GO-FET sensor is made of three regions source, drain and an active channel with a gate terminal underneath. The source and drain are made of gold and the channel is made of GO. The gate is made of two layers: a dielectric layer of SiO2 with relative dielectric $K = 4$ and a metallic layer. The GO channel has an approximate width of 17 Å and length of 42 Å. A pair of 17 Å gold electrodes is connected at the edges of the GO sheet. The software employed different approaches to generate the transport properties of quantum systems. Non-equilibrium Green’s function formalism.
Figure 2. (a) Schematic diagram of the GO-FET sensor from ATK. (b) Cross-sectional view of the GO-FET sensor. The designed sensor is made of pair of gold electrodes (source and drain), a GO channel and a gate below the channel. Color code: carbon-gray, hydrogen-white, gold-yellow, and oxygen-red.

Figure 3. (a) Composite nanocluster of silver, gold, and platinum of size 1 nm. (b) GO-FET decorated with 1 nm composite nanocluster of silver, gold, and platinum. The gate potential (0 V) and bias voltage between the right and left electrodes (−0.1 and +0.1 V) are fixed. Color code: carbon-gray, hydrogen-white, gold-yellow, oxygen-red, silver-light gray, and platinum-brown.

(NEGF) and density functional theory (DFT) has been used for the designed sensor simulation in ATK–VNL.

Decorating the GO channel with composite metal nanocluster affects the sensor behavior because the metallic nanoclusters change the electronic structure of the sensor. Composite nanocluster of gold, silver, and platinum displayed in figure 3(a) were used in this work because of their high affinity to DNA. The GO-FET was decorated with 1 nm nanocluster as displayed in figure 3(b). The effect of decorating the sensor with nanocluster on the sensor current and sensitivity was investigated. It was noticed that GO-FET sensor performance and sensitivity is enhanced after adding the nanocluster.

The GO-FET sensor electronic transport properties are generated by the ATK–VNL package. Poisson equation with marginal conditions is utilized where Dirichlet boundary condition is selected for the electrostatic potential in the Z-direction and Neumann boundary condition is chosen for the X and Y directions. These are the appropriate conditions for a device with a metallic gate. For correlation exchange in ATK–DFT calculator, the parameterization of Perdew–Zunger (PZ) for local density approximation (LDA) as well as Troullier–Martins norm-conserving pseudo potentials are applied. The 50 k-points are chosen in the Z direction transmission. Throughout the calculations the density mesh cut-off is 75 Hartree. The gate potential is set to 0 V, and the bias voltage between the source and drain electrodes is set to −0.1 V.
and 0.1 V. The transverse current is measured to detect various concentrations of DNA. The computational method and mathematical formalism used by the software has been explained in our previous work [80, 81]. All the required simulations have been generated through high performance computing environment (HPC).

3. Results

3.1. Experimental detection of DNA

The FET sensor based on graphite oxide was utilized to detect various concentrations of DNA. This sensor identifies various elements by measuring the variation in the electrical signals.

3.1.1. Raman spectrum

Raman spectroscopy is used to investigate electronic and structural features of materials. The D-band provides information about defects while the G-band provides information about saturated carbon structure. Figure 5 was generated by applying 532 nm laser radiation and 20% of the ND filter. Figure 5 shows that the D-band was observed around 1300–1400 cm$^{-1}$ which shows the structural defects, while the G-band was observed around 1550–1630 cm$^{-1}$ which indicates the stacked structures. The figure also shows the Raman spectra of graphite oxide sample displayed in figure 4(a) and the Raman spectra of graphite oxide decorated with composite nanoclusters of gold, silver, and platinum displayed in figure 4(b). Metallic nanoclusters enhance Raman spectrum [82, 83]. Thus, figure 5 shows that the Raman spectra of GO with composite nanoclusters has higher counts than the Raman spectra of GO sample. This difference is due to the intrinsic characteristics of metallic nanoclusters leading to different ionization energies [83].

3.1.2. Ultraviolet–visible spectroscopy

Ultraviolet–visible (UV–vis) spectroscopy was used to identify the graphite oxide peak. Figure 6 displays the UV–vis spectra of graphite oxide. The graphite oxide has an absorption peak at 230 nm which is attributed to the $\pi - \pi^*$ plasmon peak.

3.1.3. Size distribution of nanoclusters

The sensor is utilized to detect various concentrations of DNA. These sensors identify various elements by measuring the variation in the electrical signals. The nanocluster size distribution is displayed in figure 7 for a sputtering discharge power of 10.8 W, inert gas flow rate of 40 sccm, and aggregation length of 70 mm. Figure 7 shows the size distribution for the trimetallic nanocluster of silver, gold, and platinum as measured using the QMF where the average diameter size is 3.64 ± 0.18 nm. The nanoclusters deposition rate was 0.1 Å s$^{-1}$ and it was left for 4 min resulting in nanoclusters with a thickness of 2.4 nm.

3.1.4. Nanoclusters characterization

Commercial glass slides are placed on the sample holder with the sensor while the nanoclusters are being deposited to confirm the existence and composition of the nanoclusters. Figure 8 shows the EDS spectrum which confirms the existence of the composite nanoclusters of gold, silver, and platinum. The figure displays the amount of mass percentage for each of the different nanoclusters where the mass percentage for silver was 0.54%, while the mass percentage for gold was 1.02%, and the mass percentage for platinum was 0.21%. The different atomic percentage within that nanoclusters than that within the target; can be assigned to the different sputtering yield of the different elements. The existence of other elements such as carbon, silicon, and oxygen is because the composite nanoclusters were sputtered on glass substrates for the EDS analysis. The various atoms with the nanoclusters (Ag, Au, and Pt) have high affinity to DNA which enhances the sensor performance [84].

3.1.5. FET characteristics

The sensor electrical characteristics are measured at room temperature. Figure 9(a) shows the drain current ($I_D$) versus drain–source voltage ($V_{ds}$) characteristics for the sensor without nanoclusters and with nanoclusters. The $V_{ds}$ varies from $-0.6$ V to $0.6$ V at room temperature. The figure displays a linear dependence between $I_D$ and $V_{ds}$ where $I_D$ increases with increasing $V_{ds}$. The $I_D–V_{ds}$ characteristics reveal n-type transistor behavior where applying a positive gate voltage leads to an increment of the conducting electrons within the transistor channel which improves the channel conductivity and increases the drain current. The addition of metallic nanoclusters resulted in higher drain current at the same applied voltage because trimetallic nanoclusters of gold, silver and platinum exhibit higher conductivity as compared with graphite oxide.
3.1.6. Sensor testing

The sensor was tested for different concentrations of DNA. Figure 9(b) shows the variation in the current ($V_{ds} = 0.2$ V and $V_g = 0$ V) when the graphite oxide channel sensor is exposed to 2 $\mu$l of DNA of 25.7 nM concentration. The sensor current started at $I_{max} = 3.76 \times 10^{-4}$ A and when the DNA solution is dropped
on the sensor channel at time $t = 34$ s, the current $I_d$ drop is observed and reached $I_{\text{min}} = 3.55 \times 10^{-4}$ A. The variation in the current $I_d$ was used to evaluate the sensor sensitivity since the values of $I_{\text{max}}$ and $I_{\text{min}}$ are device dependent. The change in the current was calculated as $\Delta I = I_{\text{max}} - I_{\text{min}}$. Figure 9(c) shows the current variation for the sensor decorated with the trimetallic nanoclusters. The sensor current started at $I_{\text{max}} = 4.07 \times 10^{-4}$ A and when the DNA solution is dropped on the sensor channel at time $t = 34$ s, the current $I_d$ drop is observed and reached $I_{\text{min}} = 3.76 \times 10^{-4}$ A. It is noticed that the sensor starting current with nanoclusters is higher than the sensor starting current without nanoclusters and $\Delta I$ for the sensor with nanoclusters is $310 \, \mu$A while $\Delta I$ for the sensor without nanoclusters is $210 \, \mu$A. This indicates that sensor sensitivity is improved after adding the nanoclusters.

The drop of the drain current of graphene oxide sensor after placing DNA droplet is due to the DNA negatively charged triphosphate group which induce excess hole carriers [85–87]. Several studies reported that graphite oxide or graphene adsorb the DNA molecules through $\pi-\pi$ interaction [27, 88–91]. The adsorption of the DNA ions (negative charge) by the channel of graphite oxide or graphite oxide with nanoclusters reduces the current. The more significant reduction of drain current for the sensor decorated

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Figure 7. Size distribution of composite trimetallic (Au, Ag, and Pt) nanoclusters measured using QMF where the average diameter size is $3.64 \pm 0.18$ nm.

Figure 8. Shows the EDS spectrum of composite nanoclusters of silver, gold, and platinum with mass percentage of 0.54% for silver, 1.02% for gold, and 0.21% for platinum. The existence of other elements such as calcium (Ca), sodium (Na), silicon (Si), and oxygen (O) is due to the glass substrate.
Figure 9. (a) $I_d-V_{ds}$ characteristics profile for both GO sensors: with and without trimetallic nanoclusters. (b) Variation in the drain current due to a 2 μl drop of 25.7 nM of DNA for sensor without nanoclusters, and (c) sensor with alloy nanoclusters. (d) Variations in the electrical drain current due to different concentrations of DNA.
Figure 10. (a) DNA of guanine, cytosine, and thymine. (b) GO-FET with DNA placed on the graphene oxide channel. Color code: carbon-gray, hydrogen-white, gold-yellow, phosphor-orange, and oxygen-red.

with nanoclusters confirms the more adsorption of DNA [91]. The hole concentration increases compared to the electrons where the holes trap the electrons leading to an increment of the sensor electrical resistance which results in decreasing the $I_d$ current. These results are in agreement with previous reports [85–87, 92]. The difference in $I_d$ is more when the sensor is decorated with the trimetallic nanoclusters since these metals have high affinity to DNA [84, 91].

The variation of the electrical current is measured for both sensors due to placing various concentration of DNA solution. The real time DNA detection reached a detection limit of 1.28 nM. Figure 9(d) shows the variation in the current due to different concentrations of DNA which ranges from 1.28 nM to 77.04 nM. The figure depicts that the $\Delta I$ increases due to higher concentrations of DNA in both sensors. Also, the figure indicates that GO-nanoclusters sensor results in higher sensitivity for the various concentrations of DNA. This is due to the existence of composite trimetallic nanoclusters which have high affinity to DNA, which causes more adsorption and interaction for the DNA with the sensor. Moreover, all three metals gold, silver, and platinum are good conductors which increase the current signal. The physical and chemical properties of the trimetallic composite nanoclusters enhance the absorption and the interaction of the materials on the surface of the sensor such as DNA. These observations are in good agreement with the simulation results.

3.2. Simulation of DNA detection

The performance of the GO-FET bare sensor and the one decorated with composite trimetallic nanoclusters is investigated. Figure 10(a) shows the DNA in figure 10(b) placed on top of the sensor to study the sensors performance. The addition of composite metallic nanocluster results in higher drain current at the same voltage. The sensor bias voltage at 0.200 V and the current is 7.289 μA, while after adding the nanoclusters the sensor current is 7.795 μA. The sensor performance is examined due to different concentrations of DNA. The current variation $\Delta I$ is used to evaluate the sensor sensitivity. Figure 11 shows the current variation (at $V_{ds} = 0.200$ V and $V_g = 0$ V) after placing different concentrations of DNA on each sensor channel. Figure 11 shows that $\Delta I$ is higher for the sensor after decoration with nanocluster which proofs that the sensor with nanocluster has higher sensitivity than the bare sensor. Moreover, the figure depicts that the higher concentration of DNA results in an increment in the current variation.

Figure 12 shows a comparison between sensors with monometallic nanoclusters of Au, Ag, and Pt and trimetallic nanocluster (Ag, Au, and Pt) due to different concentrations of DNA. The figure illustrates that the sensor decorated with trimetallic nanoclusters has higher drain current change than the other sensors. Thus, trimetallic nanoclusters of Au, Ag, and Pt were used in experiment. Noble trimetallic nanoclusters are being used because of their promising features such as high sensitivity and selectivity and their multi-functional influence due to the existence of three metals within the nanocluster.
In simulation, the variation in the electrical drain current of the sensor decorated with monometallic nanoclusters of Au, Ag, and Pt and trimetallic nanoclusters of their combination due to different concentrations of DNA was studied. Simulation results in this work indicates that the adsorption of DNA is the highest when the FET channel was functionalized with trimetallic nanoclusters of gold, silver, and platinum. While using monometallic nanoclusters of gold, silver, and platinum resulted in lower sensitivity. Thus, trimetallic nanoclusters of silver, gold, and platinum were used in experiment.

Figures 9(b) and (c) display two trials only while figure 9(d) summarizes $\Delta I$ for all the trials which are fourteen. Moreover, the simulation results include four trials as shown in figure 11 and fourteen trials as shown in figure 12. Figures 9(b)–(d), 11 and 12 show that the sensor with nanoclusters has higher sensitivity. Moreover, figures 9(d), 11 and 12 show that the variation in current is increased due to the increment of DNA concentration. Our experimental results are in good agreement with our simulation results where the sensor with nanoclusters has higher sensitivity (variation in current) and the increment in DNA concentration resulted in higher variation in current $\Delta I$. These observations are in good agreement.
with previous research work [91]. Two sensors were utilized; one with nanoclusters and one without. After each measurement the silicon wafer with the gold electrodes was cleaned with deionized water and the graphite oxide channel was placed to continue the other trials. The FET sensor electrical characteristics were checked before each trial. Figure 9(d) shows how the increment of DNA concentration resulted in higher variation in current for both sensors; the sensor with nanoclusters and the sensor without nanoclusters. Moreover figures 11 and 12 shows how the higher DNA concentration in simulation resulted in higher variation current. The simulation was used as a proof of concept to support the experimental results. The software simulation of the sensor shows the same behavior as in experiment where the increment of DNA concentration results in higher variation in current and adding the nanoclusters improves the sensor performance. Moreover, it is worth noting that QuantumATK supply computational details for nanoscale sensors which can be used as a proof of concept to support the experiment which is based on bigger size sensors. The DNA placed on the sensor is displayed in figure 10(a). The low concentration refers to one sample of DNA displayed in figure 10(a) and high refers to double the amount in figure 10(a). The software does not support concentration measurements of DNA. Other studies are using FET with \( V_g = 0 \) V and measure the variation in current due to different concentrations of DNA (figure 2(b) in Yin et al article) [91]. Our observations are in good agreement with Yin et al where the FET devices show a decrease of \( I_{ds} \) (at \( V_g = 0 \) V) with increase of the DNA concentration [91]. Moreover, the decrease in current is more significant for the channel decorated with platinum nanoparticles [91]. The decrease in current is due to the DNA adsorption. Several studies have also shown that DNA molecules can be adsorbed on the graphite oxide or graphene surface through the \( \pi-\pi \) interaction [27, 88–90]. The sensor electrical characteristics were checked before each trial. The fabrication method utilized the shadow mask displayed in the figure 1(c) to fabricate the sensor with the same electrode dimensions and channel thickness to get identical resistances. The geometry of the sensor used in experiment and the sensor built using ATK is displayed in figures 1(c) and 2(a).

4. Discussion

The developed sensor experimental performance is confirmed by simulation where both simulation and experimental results reveal a promising sensor for real time DNA detection. The results generated from simulation and experiment indicates that exposing the sensor channel to various concentrations of DNA decreases the current which is explained due to adsorption difference of DNA molecules by the bare graphite oxide channel and the graphite oxide channel with trimetallic nanoclusters. The addition of DNA to the channel increases the holes concentration which traps the electrons and increases the resistance leading to current decrement [85]. The current decrease due to DNA adsorption results are in good agreement with previous work that confirmed increases the holes concentration upon adsorption of DNA [88, 91, 92]. Also, the current change increases due to higher concentrations of DNA which indicates the higher adsorption due to higher concentration. Our work results agree with previous work where the current decreases due to the increment of the DNA concentration [88, 91]. Moreover, both simulation and experiment indicate higher sensitivity due to the presence of trimetallic nanoclusters on the sensor channel as compared with the bare channel. The higher sensitivity is due to the high affinity to DNA of the trimetallic nanoclusters as explained by pervious research work [60, 84, 87]. The various metal ions show diverse DNA binding affinities. Platinum ions have high affinity to DNA [84]. The adsorption intensity of DNA increases due to the addition of gold or silver ions. The increment in the absorptivity of DNA is due to the binding of gold or silver ion to the DNA phosphate group and leaving the pyrimidine and purine bases of DNA exposed [84]. The DNA binding constant to gold is higher than that to platinum and silver [84].

Noble metal nanoclusters deposited on FET channel are widely utilized to improve the DNA detection sensitivity [93–96]. These nanoclusters modifies the surface to volume ratio of the sensor resulting in higher sensitivity [38]. Functionalizing the graphite oxide channel of the FET sensor with trimetallic nanoclusters enhances the adsorption of DNA which results in higher variation in current. Therefore, higher sensitivity. In simulation, the variation in the electrical drain current of the sensor decorated with monometallic nanoclusters of Au, Ag, and Pt and trimetallic nanoclusters of their combination due to different concentrations of DNA was studied. Simulation results in this work indicates that the adsorption of DNA is the highest when the FET channel was functionalized with trimetallic nanoclusters of gold, silver, and platinum. While using monometallic nanoclusters of gold, silver, and platinum resulted in lower sensitivity. The trimetallic nanoclusters have higher catalytic activity compared to monometallic nanoclusters of Ag, Au, and Pt. Trimetallic composites of Ag/Au/Pt have been used in biosensing applications and resulted in impressive outcomes [60]. These impressive outcomes are due to the multi-functional effect of the three metals within the nanocluster. Moreover, earlier work showed that using trimetallic nanoclusters such as
Cu/Au/Pt have higher catalytic activity [60]. Trimetallic nanoclusters enable the researchers to get the required properties by monitoring their surface and composition [52–57].

The DNA adsorption to the GO-FET clearly resulted in n-doping effect. Hence, a higher decrease in current is noticed with the addition of the trimetallic nanoclusters due to the higher surface to volume ratio of the channel, the strong catalytic activity of the trimetallic nanoclusters and the higher adsorption of DNA. However, the more significant decrease in current for the channel decorated with trimetallic nanoclusters compared to the GO channel is due to the more adsorption of DNA to the channel.

5. Conclusion

In this article, a sensor for DNA detection was designed, fabricated, and studied for the purpose of detecting various concentrations of DNA. The sensor was fabricated based on FET structure on Si/SiO₂ wafer. The channel material was either bare graphite oxide or graphite oxide decorated with trimetallic nanoclusters. The two source and drain electrodes were made by thermal evaporation of gold (Au). Composite nanoclusters of silver, gold, and platinum were deposited on the graphite oxide channel by sputtering and inert gas condensation technique inside an ultra-high compatible system. The sensor was also simulated to confirm the experimental results using DFT along with NEGF calculations. The results indicate that the sensor decorated with nanoclusters exhibit better performance than the bare sensor due to high affinity of the trimetallic nanoclusters to DNA. The simulation results are in good agreement with the experimental results. The sensor fabrication method is simple, effective, and shows great possibility for mass-producing of transistor-based sensors for the purpose of DNA detection at low cost.

Conflict of interest

The authors declare no competing financial interest.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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