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# Graphene-based field effect transistor in two-dimensional paper networks

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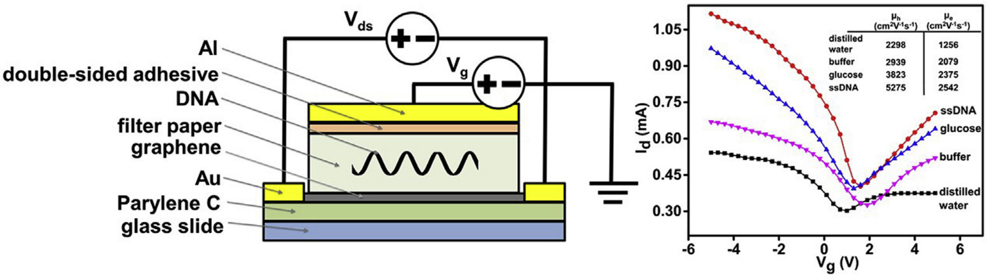
## HIGHLIGHTS

- A graphene-based field effect transistor sensor was fabricated for two-dimensional paper network formats.
- The constructed GFET on 2DPN was shown to behave similarly to solution-gated GFETs.
- Electrolyte gating effects have more prominent effect over adsorption effects on the behavior of the device.
- The GFET incorporated on 2DPN was shown to yield linear response to presence of glucose and ssDNA soaked inside the paper.

## GRAPHICAL ABSTRACT



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## ABSTRACT

We demonstrate the fabrication of a graphene-based field effect transistor (GFET) incorporated in a two-dimensional paper network format (2DPNs). Paper serves as both a gate dielectric and an easy-to-fabricate vessel for holding the solution with the target molecules in question. The choice of paper enables a simpler alternative approach to the construction of a GFET device. The fabricated device is shown to behave similarly to a solution-gated GFET device with electron and hole mobilities of ~1256 $cm^2 V^{-1} s^{-1}$ and ~2298 $cm^2 V^{-1} s^{-1}$ respectively and a Dirac point around ~1 V. When using solutions of ssDNA and glucose it was found that the added molecules induce negative electrolytic gating effects shifting the conductance minimum to the right, concurrent with increasing carrier concentrations which results to an observed increase in current response correlated to the concentration of the solution used.

## 1. Introduction

Paper is a well-known material with abundant utilities not only in writing, printing and packaging, but also in traditional laboratory techniques such as filtration and paper chromatography. Recently, paper is being rediscovered as a material for many different areas of research like electronics and biomedical applications. The upsurge in attention is due to some advantageous properties that are inherent to paper such as porosity, flexibility and capillary flow in

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addition to its ease of manipulation and low-cost. One of the developing areas of research interest for paper is two-dimensional paper networks (2DPNs) otherwise also known as paper microfluidics. 2DPNs were shown to be comparable to the conventional continuous-flow microfluidics exhibiting the same characteristic laminar flow processes. However, the paper-based microfluidic counterpart does not need any external pumps because of capillary flow and is far easier to fabricate. And in many recent research progress, a number of the capabilities of continuous-flow microfluidics have been translated into its paper format. It was demonstrated that patterns can be placed on paper to create channels for directed fluid flow which offered advantages of multiplexed analysis from a single sample introduction transcending the capabilities of sensing seen from the traditional lateral flow assays (LFAs) [1–3]. In addition, the same idea and principle was employed to carry out multi-step processes by linking multiple inlets to a shared region and precise control of flow is also shown to perform sequential steps [4,5].

In most 2DPNs the applications are geared toward sensing biomolecules such as DNA [6], proteins [7] or certain metabolites and infectious diseases [8] mainly through qualitative colorimetric detection [9] or quantitative electrochemical detection [10]. In this report, we demonstrate the incorporation of a Field Effect Transistor (FET) type sensor onto a paper platform with the use of graphene as a channel. Graphene, a two-dimensional sheet of $sp^2$ hybridized carbon, has also gathered research attention in sensing applications owing to its good electronic properties particularly its ultra-high mobility and ambipolar field effect in addition to the very large surface area of its two-dimensional structure. With advances in large-scale synthesis through chemical vapor deposition (CVD) techniques, a great number of reports have utilized graphene as a sensing platform in particular as an FET. The exceptional electronic properties of graphene has great potential for sensing and paves a way for better precision and accuracy which are important in sensing tools especially in applications like medical diagnostics [11]. At present there are still challenges towards the achievement of precise and accurate measurements even with the significant improvements in many sensing systems. For example, many sensing tools rely on the measurement of hydrogen peroxide produced from the reaction of an analyte with its appropriate oxidase enzyme. It is an indirect measurement and is subject to many interferences from other electroactive species [12]. Graphene-based field effect transistors (GFETs) offer the advantage over other sensing approaches such as optical and electrochemical detection in that it is feasible to perform label-free detection with good sensitivity [13]. In fact, GFETs have been employed to detect DNA and its hybridization and other biomolecules by monitoring conductivity changes [14–16]. In addition, GFET has been reported to be incorporated as a sensor in continuous-flow microfluidics [17]. GFETs by itself has recently been shown to have versatile qualities by being incorporated in various formats especially in flexible substrates owing to the graphene material being also flexible [18]. In this regard, our report will help transcend the capabilities of 2DPNs and further translate processes carried out in continuous-flow microfluidics into its paper format by incorporating paper as a dielectric material for a GFET. There is a considerable amount of research aimed at simplifying sensing platforms without compromising performance in sensitivity and accuracy. Graphene has enabled the further development of paper sensing platforms for highly sensitive and rapid detection of analytes [19].

## 2. Experimental details

### 2.1. Preparation of graphene film by chemical vapor deposition (CVD)

The graphene sheets used in this study were synthesized via chemical vapor deposition (CVD) [20,21]. More specifically, a piece of copper foil was shortly etched for 30 s in a $FeCl_3$ solution with mild sonication followed by blow-drying using a nitrogen gun. The foil was then placed inside the CVD furnace where it was treated with Ar gas flowing at 30 sccm for 20 min and then annealed by adding $H_2$ gas flowing at 16 sccm to a temperature of 1050 °C. Graphene was grown by introducing $CH_4$ gas into the furnace as a carbon source at 16 sccm for 20 min while maintaining the flow of Ar and $H_2$.

### 2.2. Fabrication of graphene-based field effect transistor (GFET)

The prepared graphene was subsequently transferred onto a glass slide precoated with 1 μm Parylene C. The transfer process involves spin-coating a layer of PMMA solution on the side of the copper foil with graphene and then baking the PMMA to dry at 120 °C. Keeping the PMMA side face up, the underside copper was etched by floating the foil in a $FeCl_3$ solution for 5 h. After the copper is completely etched, graphene attached to PMMA was carefully scooped out using a glass slide and floated back on a dilute HCl solution and then DI water each for 1 h. Finally, the graphene is scooped into the desired Parylene-coated glass slide after which the PMMA layer was removed by bathing the slide in acetone for 5 min at 60 °C.

The construction of the FET device was done by forming 50 nm gold contacts on two ends of the graphene sheet via thin film sputtering. A Whatman Grade No. 1 filter paper was manually cut into small pieces, which was then stuck with a sheet of aluminum foil using a double-sided adhesive. The paper was soaked with the solution in question (e.g. glucose solution and ssDNA solution) and then carefully positioned on top of the graphene sheet. A schematic illustration and an image of the actual device are shown in Fig. 1a. The ssDNA used as a model analyte was an oligonucleotide for HIV-1 detection with a sequence 5′-AGTAGTGTGTGCCCGTCTGTTGTGTGACTCTGGTAACTAGAGATCCCTCAGAC-3′ [6].

### 2.3. Characterization

Raman spectroscopy of graphene was done using an InVia Renishaw Raman Microscope with a 514 nm excitation Argon laser. AFM images were obtained using a Veeco Multimode Scanning Probe Microscope. Device electrical characterization was carried out using a Signatone S-1160B Manual Probe Station.

## 3. Results and discussion

Our approach to incorporating a GFET device onto a 2DPN is to use a solution-soaked paper as a gate dielectric material for the device. Fig. 1a illustrates the configuration of the GFET as explained in the experimental section. Graphene is constructed as a channel between two contacts, and its current can be modulated by application of an electric field through a gate electrode on top. Paper with absorbed solutions serves a dielectric material in between the channel and gate where charged species from the solution can contribute to the field effect. Parylene C was coated on the surface of the glass slide to provide a hydrophobic surface for better graphene adhesion. Gold serves as good ohmic contacts for electronic characterization and is resistant to oxidation. For the gate electrode, sputtering gold on top of the filter paper was found to not be an