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Nanotube-on-graphene heterostructures for three-dimensional nano/bio-interface

Sun Sang Kwon [a], Jae Hyeok Shin [a], Jonghyun Choi [b], SungWoo Nam [b], Won Il Park [a,*]

[a] Division of Materials Science and Engineering, Hanyang University, Seoul 04763, Republic of Korea
[b] Department of Mechanical Science and Engineering, University of Illinois at Urbana-Champaign, Urbana, IL, 61801, United States

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ABSTRACT

We report the synthesis, fabrication, and characterization of a nanotube-on-graphene (NT-on-Gr) field-effect sensor array for electrical detection of the biological activity of living cells. In order to form vertical nanotubes on a graphene surface, Ge/Si core-shell nanowires were vertically grown on graphene, followed by cap opening and Ge core-etching processes. Source-drain current versus water-gate potential measurements in electrolyte solutions with various pH values showed typical gate-dependent ambipolar characteristics with a decrease in pH sensitivity versus that of a flat graphene field-effect sensor. This is associated with limited solution gating of Si nanotubes that form nanoscale fluidic channels and thus interconnect the solution with the graphene field-effect sensor. The Si nanotubes also bridged interconnections between cells and the graphene field effect sensors, which were then able to record electrical spike peaks caused by cell networks.

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1. Introduction

Living biological systems generate electrical potential via nanoscale ion channels located across the cell membrane. Accordingly, the biological sensors that utilize nanoscale building blocks with one or several dimensions matching the target length-scale have potential advantages in the fundamental study of biological activities. In particular, the study of electrogenic cells and tissues has been carried out using a variety of recording techniques, such as patch-clamp electrodes, multielectrode arrays, and planar field effect transistors (FETs) [1–3]. However, these conventional techniques suffer from limitations. For instance, patch-clamp electrodes may be either invasive or even destructive for cellular measurements, and their measurement resolution is not ideal for subcellular recording. Semiconductor nanomaterials with controlled structures, such as nanowires (NWs), nanotubes (NTs), and graphene have been developed to overcome such challenges and record the properties of biological species with a higher resolution [4–7]. One or several dimensions of the nanomaterials match the length-scale of target biological species and could provide information about electrical signal propagation in biological systems at a far higher spatial resolution than previously demonstrated [8–11]. Furthermore, the microfabricated device arrays can be readily fabricated on flexible polymer substrates, which enable minimally invasive and gentle contact of device chips with non-planar biological systems [12,13]. For example, Qing et al., Gao et al. and Tian et al. demonstrated that flexible nanoelectronic devices based on laterally transferred NWs, NTs and/or graphene sheets offer the potential for conformal interrogation of biological systems and electrical recording and/or regulation of the biological activities of living cells [14,6,15]. However, these devices are in a 2-dimensional (2D) planar structure and have limitations in nanoscopic, 3-dimensional (3D) localized, and tunable detection. Furthermore, the 2D nature of these devices precludes the interrogation of intracellular events.

To overcome such challenges and to record biological process inside a cell, 3D electrode probes based on metallic nanopillars have been developed [16,17]. These 3D nanopillar probes function as point-like, minimally invasive probes capable of entering cells through natural pathways due to their nanoscale dimensions. More advanced device concepts utilize active nanoprobes based on NW FETs, in which their performance does not depend on device impedance; consequently, they can be much smaller than conventional micropipettes or microelectrodes. The nanoprobe devices are usually made of carefully designed semiconductor NWs, such as kinked NWs or branched NWs; however, the device configuration and design are relatively complex and thus limit the potential

* Corresponding author.
E-mail address: wipark@hanyang.ac.kr (W.I. Park).

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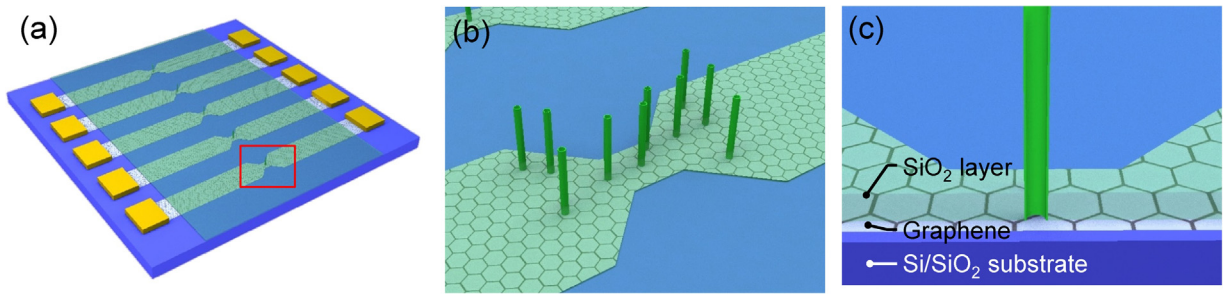

Fig. 1. Schematic 3-D view of the NT-on-Gr FET sensor array (a), magnified image highlighting the NT-on-Gr region (b), and cross sectional image of the NT-on-Gr FET sensor (c).

for multiplex and multifunctional biotic and abiotic interfaces [15,7,18].

In this study, we demonstrate a practically simple 3D nanosensor platform based on a combination of a graphene FET and a SiNT as an artificial ion channel (Fig. 1). This nanotube-on-graphene heterostructure (NT-on-Gr HS) device exploits hollow NT tips that can connect the graphene FET sensor to an external object such as a cell, with the following advantages over conventional electrophysiology techniques. First, the diameters of the SiNTs can be decreased to tens of nanometers, significantly smaller than those of the glass pipettes in conventional patch-clamp technique. Such feature leads to a high spatial resolution, minimal invasiveness and bio-compatibility when interfacing with living biological species. Second, an ultra-high sensitivity is attainable even at the extremely small probe size via the active field-effect sensing mechanism of a single atomic layer graphene transistor, which is an improvement over the passive current measurements of the conventional method. Third, the mechanically flexible NT-on-Gr HS is able to achieve a conformal, flexible interface with biological systems. In particular, by introducing an array of nanoprobes, our approach has the potential to not only enhance the information resolution, but also improve the multiplexed recording of large scale biological information in the future.

2. Experimental

2.1. Graphene ribbon

A single-layer graphene film was grown on copper foil (25 μm thick) by chemical vapor deposition (CVD), using 35 sccm hydrogen (H_2) and 15 sccm methane (CH_4) as a carrier gas and carbon source, respectively. The graphene film on the Cu foils was coated with a ~200 nm poly(methyl methacrylate) (PMMA) layer to minimize the mechanical fracture of the graphene during the transfer. The sample was moved to a 0.05 M ammonium persulfate solution, in which the underlying copper foil was gradually etched from the back of the floating sample. After complete etching of the copper foil, the free-standing PMMA/graphene layer was transferred to an $18 \times 18\ mm^2$ piece of Si substrate coated with a thermally oxidized 300-nm-thick SiO_2 layer. The substrate was then immersed in an acetone solution for 2 min to remove the PMMA layer. The transferred graphene film was patterned to have a ribbon shape with a 5 μm width by photolithography and oxygen (O_2) plasma etching. Subsequently, the sample was immersed in an acetone solution again to remove the remnant residue, followed by annealing at 300 °C [19,20].

2.2. Nanotube-on-graphene

The NT-on-Gr HS was fabricated by CVD growth of vertically-aligned Ge/Si core-shell NWs on the graphene ribbon surface and subsequent Ge core etching. First, the surface of the graphene ribbon was decorated with 50-nm-diameter Au colloidal particles (Sigma Aldrich). Then, the sample was loaded into a CVD reactor to grow GeNWs via an Au-nanocluster-catalyzed vapor-liquid-solid (VLS) mechanism. Subsequently, a Si shell layer was coated on the core GeNWs. For the growth of vertically-aligned GeNWs on the graphene surface, the Au nanoparticle-coated graphene sample was annealed under vacuum at 425 °C for 20 min and cooled to 320 °C, followed by introduction of a 50 sccm H_2 and 5 sccm germane (GeH_4) reactant source at a reactor pressure of 15 Torr. After Ge NW growth for 20 min, the CVD reactor was cooled to room temperature and re-heated to 455 °C to deposit a conformal Si layer on the GeNWs. After coating a 20-nm-thick Si layer, the sample was moved to an e-beam evaporation chamber, in which a 50-nm-thick SiO_2 layer was deposited on the chip surface as a supporting and passivation layer. For selective etching of the core Ge wires, a PMMA layer was spin-coated with a thickness less than the height (length) of the Ge/Si NWs. Subsequently, the GeNW tops were opened by removing the PMMA, SiO_2, and Si layers with O_2 plasma treatment (50 W for 5 s) and wet etching in a buffered hydrofluoric acid (5% in a deionized water) and potassium hydroxide (KOH) solution (10 wt.% in a deionized water). Finally, the GeNW cores were etched in a hydrogen peroxide (H_2O_2) solution, followed by immersion in an acetone solution to remove the PMMA layer (Fig. 2a).

2.3. pH response measurement

For pH response measurement, both ends of the graphene ribbon were coated with silver electrodes to act as source and drain contacts in a graphene FET sensor (Fig. 3a). A PDMS pool was carefully placed onto the sensor chip such that its open window ($4 \times 12\ mm^2$) overlapped with the central regions of the graphene ribbon while avoiding direct exposure of the metal electrodes to the electrolyte. As a reference gate electrode, a silver/silver chloride (Ag/AgCl) wire was also inserted into the PDMS pool, and the sensor responses were monitored using a semiconductor parameter analyzer (HP4145). The pH buffer solutions were prepared in 10 mM PBS (pH 7.4), and a specific amount of hydrogen chloride (HCl) or potassium hydroxide (KOH) was added to adjust the pH value between 6.55–8.25.

2.4. PDMS-cell sheet

To obtain a PDMS-cell sheet, thin PDMS sheets ($3 \times 6\ mm^2$) were coated with poly-L-lysine solution and placed in a cell culture dish. Cultured HEK 293 cells were detached with trypsin solution (Sigma Aldrich) and subcultured on the prepared PDMS sheet. The PDMS-cell sheet was incubated in minimum essential medium Eagle (MEM, Gibco) supplemented with 10% fetal bovine serum (FBS, Gibco) and was maintained at 37 °C and 5% CO_2 atmosphere for 5 days [21].

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