

Advances in Graphene-Based Sensors and Devices

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Since its discovery in 2004, graphene has been the most widely used nanomaterial that has sparked tremendous interest in researchers, for highly diversified applications [1,2]. It is a two-dimensional planar sheet of sp²-bonded carbon atoms that are densely packed in a honeycomb crystal lattice. The ground-breaking research on graphene by Andre Geim and Konstantin Novoselov from the University of Manchester was awarded the Nobel Prize in Physics for 2010.

Graphene has been extensively investigated for the development of optoelectronic devices, super capacitors and various types of high performance sensors, due to its large surface-to-volume ratio, unique optical properties, excellent electrical conductivity, remarkably high carrier mobility (in excess of 20000 cm²V⁻¹s⁻¹), high carrier density (10¹³ cm⁻²), high thermal conductivity, room temperature Hall effect, ambipolar field-effect characteristics, high signal-to-noise ratio (due to low intrinsic noises), and extremely high mechanical strength (200 times greater than steel, tensile modulus of 1 TPa). The large surface area of graphene enhances the surface loading of desired biomolecules, either through passive adsorption, or by covalent crosslinking to the reactive groups of biomolecules. On the other hand, the excellent conductivity and small band gap of graphene are beneficial for the conduction of electrons between the biomolecules and the electrode surface. Graphene has about two-fold higher effective surface area and greater cost-effectiveness than carbon nanotubes. Additionally, it has greater homogenous surface that is responsible for highly uniform and efficient functionalization.

Graphene has been synthesized by several methods that include exfoliation of graphite, electric arc discharge, epitaxial growth on electrically insulating surfaces, opening of carbon nanotubes, growth from metal-carbon melts, pyrolysis of sodium ethoxide, sonication of graphite, reduction of carbon dioxide, Chemical Vapor Deposition (CVD), and reduction of graphene oxide. The mechanical exfoliation is unable to produce large graphene sheets and also has low throughput. However, CVD can produce large areas of single layer graphene for mass production. The chemical or thermal reduction of graphene oxide is another mass production method that is most commonly employed due to its higher cost-effectiveness. It has also been reported that graphene synthesized by chemical redox reaction possess many structural defects that are highly beneficial for electrochemical applications.

A wide range of chemical modification and biomolecular binding strategies [3], have been developed for the induction of specific functional groups (such as carboxyl, hydroxyl, sulfonate, acid chloride and amine) on graphene and for binding the chemically-modified graphene to the biomolecules, respectively. Similarly, several methods have also been developed for preparing graphene-nanocomposites by conjugating graphene to nanomaterials, and/or polymers.

Graphene has been widely used in biosensors and diagnostics [1,2], for the detection of a wide range of analytes such as glucose, glutamate, hydrogen peroxide, benzene, ethylbenzene, xylenes, cyclohexane, nicotinamide adenine dinucleotide, hemoglobin, cholesterol, protein biomarkers (alpha fetoprotein, carcinoembryonic antigen, prostate specific antigen, human epidermal growth factor receptor 2, epidermal growth factor receptor, immunoglobulin G (IgG) and

IgE), saccharides, and cancer cells. It has immense potential for the development of electrochemical biosensors, based on the direct electron transfer between the enzyme and the electrode surface. Moreover, the graphene-functionalized electrodes have been demonstrated to have superior analytical performance, negligible interference from biological substances and drug metabolites, and excellent anti-fouling ability [4]. Graphene-based non-enzymatic electrodes have been used for the detection of ascorbic acid, uric acid, dopamine and hydrogen peroxide. Graphene-based nano-electronic devices have also been employed for DNA sensors (for detecting single- and double-stranded DNA, nucleobases and nucleotides), gas sensors (for detecting hydrogen, carbon monoxide, ammonia, chlorine, nitrogen dioxide, nitric oxide, oxygen, ethanol, water vapours, iodine, methane, hydrogen cyanide, trimethylamine and 2,4-dinitrotoluene), pH sensor, detection of environmental contaminants (paraxon, hydroquinone, catechol, hydrazine, heavy metal ions (Ag⁺, Cd²⁺, Ca²⁺ and Hg²⁺), methyl jasmonate and nitromethane), detection of pharmaceutical compounds (Rutin, paracetamol, 4-aminophenol and aloe-emodin), detection of bacteria (*Escherichia coli*), and development of Field-Effect Transistors (FET) (for the detection of DNA hybridization, negatively-charged bacteria and IgE). Graphene oxide has potential anti-bacterial properties that are currently being investigated for food-packaging to keep the foods fresher for extended periods of time.

Due to its high carrier mobility and low noise, it is an ideal material for FETs. The high electrical conductivity and high optical transparency of graphene, further makes it a prospective candidate for developing transparent conducting electrodes that can have tremendous applications in liquid crystal displays, touchscreens, organic light-emitting diodes, and organic photovoltaic cells. It is an ideal material for spintronics, as it has small spin-orbit interaction and absence of nuclear magnetic moments in carbon. Additionally, it has potential for significantly improving the energy storage density of the existing ultracapacitors, based on its extremely high surface area to mass ratio.

The commercially-available graphene, known as graphene nano platelets, is crystalline or flake form of graphite, that consists of many graphene sheets stacked together (Figure 1). Bilayer graphene, having two layers of graphene, has also been made and shown to have extreme potential for optoelectronic and nanoelectronic applications, due to its tunable band gap, quantum hall effect and potential for excitonic condensation. Moreover, graphene oxide papers and graphene oxide

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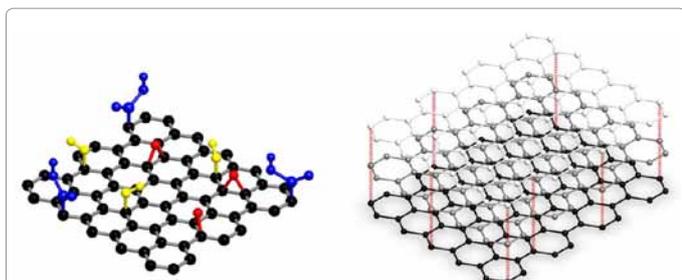


Figure 1: Single-(left) and multi-layered (right) graphene. The yellow, red and blue groups in single-layered graphene are hydroxyl, ether and carboxyl groups, respectively.

flakes in polymers have also been developed, that have peculiar chemical and enhanced photo-conducting properties. Recently, graphene-based membranes, which are impermeable to all gases and liquids, but allow water vapours to pass through, have also been prepared. They will enable the cost-effective distillation of ethanol at room temperature that will be highly beneficial for alcoholic beverage industry and biofuel production. Similarly, graphene filters have been made and shown to have analytically superior performance for desalination, in comparison to the existing techniques. Some other forms of graphene, such as graphene nanoribbons and graphene quantum dots, have also been synthesized and shown to have tremendous potential for ultrasensitive detection, as they are highly sensitive to the field effect and the chemical disruption at edges.

The evaluation of the toxicity and biocompatibility of graphene

is critical for *in vivo* applications. However, due to the absence of international guidelines for evaluating the toxicity of nanomaterials [5], researchers have obtained highly contradictory results. Graphene has been shown to be non-cytotoxic in cell culture experiments, while it affects the growth of Gram-positive and Gram-negative bacteria. Therefore, the international regulatory guidelines for assessing the toxicity of nanomaterials and strict compliance with the industrial and healthcare requirements [6], is of critical importance, in order to develop commercially-viable graphene-based sensors and devices. Moreover, there is need to improve the analytical methods for reproducible, facile and scalable preparation of graphene. However, based on their superior analytical performance, graphene-based sensors and devices will have significant applications in healthcare, environmental monitoring, and industrial settings.

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