

## RECENT ADVANCES IN APPLICATIONS OF GRAPHENE

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[Received-07/12/2012, Accepted-10/01/2013]

### ABSTRACT

Graphene, a two-dimensional, single-layered sheet of  $sp^2$  hybridized carbon atoms has attracted much attention from researchers due to its interesting mechanical, thermal, electrochemical and electronic properties. These exceptional properties have opened up new opportunities for the application of this nanomaterial in the future devices and systems. This review article aims to present an overview of the advancement of research in applications of graphene and its derivatives in different areas such as field emission, sensors, energy storage, electronics, catalysis, and biomedical field as well as a brief discussion on the challenges and perspectives for future research.

**Key word:** Graphene, field emission, sensors, energy storage, super capacitor, biomedical applications.

### INTRODUCTION

Graphene has attracted much attention from researchers due to its interesting mechanical, electrochemical and electronic properties. Graphene, a single atomic layer of  $sp^2$ -bonded carbon atoms tightly packed in a two dimensional (2D) honeycomb lattice, has evoked great interest throughout the scientific community since its discovery [1-4]. As a novel nanomaterial, graphene possesses unique electronic, optical, thermal, and mechanical properties [5-8]. Graphene and its derivatives have shown outstanding potentials in many

fields such as nanoelectronics [9], engineering nanocomposite materials [10-16], energy storage [17-21], field effect transistor (FET) [22-24], organic light emission diodes (OLED) [1], sensors [25], catalysis [26] and biomedical applications (biosensor, biodevices, drug and gene delivery, cancer therapy etc.) [25, 27, 28]. Up until now, several methods have been developed for fabrication, growth or synthesis of graphene and its derivatives. Pristine graphene is obtained from the mechanical exfoliation of graphite using adhesive tapes [5,29]. Chemical vapor deposition (CVD) has been used to grow

single and few-layer graphene sheets on metal surfaces such as Ni and Cu [30-39]. Graphene layers can also be obtained by carbon segregation on carbon-containing substrates like SiC through high temperature annealing [40-44]. Oxidation and exfoliation of graphite oxide followed by the chemical reduction has been used to prepare reduced graphene oxide (rGO) or chemically functionalized graphene (CFG) [45-47]. Featuring unique physical and chemical properties and having reliable synthetic methods for both solid and solution-phase processes, graphene and its derivatives have been incorporated into a number of functional materials to form composites [48-54] and have been used as building blocks for various kinds of applications.

**APPLICATIONS OF GRAPHENE AND ITS DERIVATIVES.**

The advancement of new-found nanomaterials provides a fascinating opportunity for development in different fields because of their structures, components and properties. In

comparison with its precursor, carbon nanotube (CNT), graphene exhibits some merits like low cost, two external surfaces, facile fabrication and modification and absence of toxic metal particles [55,56]. Thus graphene and its derivatives are expected to find applications in many fields such as nanoelectronic devices, chemical and biological sensors, energy storage and biomedical fields which have been summarized in Table-1.

**1. Electronic nanodevices**

Because of high electrical conductivity, mechanical flexibility and low cost, graphene and its derivatives have got wide spread applications in light emitting diode (LED), field effect transistor (FET), memory and photovoltaic devices.

**1. a. Field effect transistor (FET)**

Due to unique band structure, the carriers in graphene are bipolar, with electrons and holes that can be continuously tuned by a gate electrical field [5]. The observation of electric field effect in graphene was first

**TABLE 1 : REPRESENTS THE APPLICATIONS OF GRAPHENE IN DIFFERENT FIELDS**

	APPLICATIONS		REFERENCES
GRAPHENE	ELECTRONIC NANODEVICES	FIELD EFFECT TRANSISTORS	22, 23, 29, 57, 58, 59, 60, 61, 62, 63, 64, 65, 66, 67, 68, 69, 70, 71, 72, 73, 74, 75, 76, 77 & 78.
		TRANSPARENT CONDUCTIVE FILMS	31, 56, 79, 80, 81, 82, 83 & 84.
	ENERGY STORAGE DEVICES	Li- ION BATTERIES	87, 88, 89 & 90.
		ULTRA CAPACITORS	18, 47, 91, 92, 93, 94, 95, 96, 97, 98, 99, 100, 101, 102, 103 & 104.
		FUEL CELL AND SOLAR CELLS	79, 105, 106, 107, 108, 109, 110, 111, 112, 113, 114, 115 & 116.
	SENSORS	ELECTROCHEMICAL SENSORS	118, 127, 128, 129, 130 & 131.
		BIOSENSORS	25, 132, 133, 134, 135, 136, 137, 138, 139, 140, 141, 142, 143, 144, 145, 146, 147, 148, 149, 150, 151 & 152.
	BIOMEDICAL ENGINEERING	GENE DELIVERY	153, 154, 155, 156, 157, 158, 159 & 160.
		DRUG DELIVERY	27, 28, 161, 162, 163, 164 & 165.
		TISSUE ENGINEERING	11, 142, 166, 167, 168, 169 & 170.
		CANCER THERAPY	27, 28, 162, 171, 172, 173, 174, 175 & 176.

reported by Novoselov et al. in 2004. According to this report, graphene based FETs demonstrated ambipolar characteristics with electron and hole concentration of  $10^{13}$  sq.cm with mobilities upto 10,000 sq.cm per volt. sec. at room temperature. Graphene FET devices with a single back gate have been investigated by several other workers [29, 57, 58].

For the application as transistor, graphene should be in the form of quasi one dimensional (1D) structure with narrow width and atomically smooth edges termed as graphene nanoribbons(GNRs). These GNRs exhibits band gap useful for FET application with excellent switching speed and high carrier mobility at room temperature. Thus the quasi 1D GNRs become semiconductors with finite energy band gap [22, 59-65].

Although band gap have been demonstrated in GNRs, these were quite different from those of graphene in terms of carrier mobility and fabrication [66]. Several workers have demonstrated various methods to fabricate GNR including chemical and lithographic methods [22, 23, 64].

Several theoretical studies have been reported to predict the performance of GNR FETs as function of their edge roughness [67], chirality [68], chemical doping [69], carrier scattering [70] and contact [71]. Various models have also been developed to predict the performance of GNR FETs [72-77]. Thus they can be valuable tools for designing efficient FETs.

Lu et al. fabricated a high mobility flexible graphene field-effect transistor with self-healing gate dielectrics for a wide range of applications in flexible electronics [24]. Szafranek and his co-workers have demonstrated current saturation and voltage gain in bilayer graphene field effect transistor [78].

### 1. b. Transparent conductive films

With high electrical conductivity, high carrier mobility and moderately high optical transmittance in the visible range of spectrum,

graphene material show promise for transparent conductive films (TCFs) and is expected to be one of the mostly sought material for future optoelectronic devices [31, 79, 80]. Graphene TCFs have been used as electrodes for dye-sensitized solar cells, liquid crystal devices (LCDs) and organic light emitting diodes (OLEDs) [81, 82].

The high hole transport mobility, large surface area and inertness against oxygen makes graphene a promising candidate for photovoltaic applications. Graphene has been used as a novel acceptor for bulk heterojunction polymer photovoltaic cells, showing remarkably reduced photoluminescence and efficient energy transfer [56].

The high mobility and excellent mechanical properties of transparent graphene films makes it a suitable candidate for microelectronic applications. Kim et al. evaluated the fold ability of graphene films, transferred to a polyethylene terephthalate (PET) substrate coated with a thin PDMS layer by measuring resistance as a function of bending radii [31].

Liu et al. fabricated flexible graphene film on PET substrate from large size GO by thermal annealing and the green production of rGO films had potential application in flexible electronics [83]. Kang and his co-workers developed graphene films grown on metal substrate by chemical vapor deposition method and safely transferred onto desired substrate for its application for display and solar cells [84].

### 2. Energy Storage Devices:

Due to its high theoretical surface area of  $2630 \text{ m}^2.\text{g}^{-1}$  and ability to facilitate electrons or hole transfer along its two-dimensional surface, graphene has been a promising material for electrode. There have been several reports on graphene based electrodes for both rechargeable lithium ion batteries (RLBs) and electrochemical double layer capacitors (EDLCs). Graphite, the most commonly used anode material in RLBs has been replaced by graphene for its superior

electrical conductivity, high surface area and chemical tolerance [22, 80, 85, 86].

### 2. a. Lithium Ion Battery

Lithium ion battery has been a key component of hand-held devices due to its renewable and clean nature. To meet the increasing demand for lithium ion batteries with higher energy density and durability, new electrode materials with higher capacity and stability have been developed. Paek et al. has prepared graphene nanosheets decorated with SnO<sub>2</sub> nanoparticles. The SnO<sub>2</sub>-Graphene exhibits reversible capacity of 810mAh/g and its cycling performance is drastically enhanced in comparison to that of bare SnO<sub>2</sub> nanoparticles [87]. Wang et al. have demonstrated self-assembled TiO<sub>2</sub>-graphene hybrid nanostructure to enhance high rate performance of electrochemical active material [88].

Xie et al. synthesized a SnSb nanocrystal/graphene hybrid nanostructure by a facile one step solvothermal route which can be used as a potential high capacity anode material for lithium ion battery [89]. Xiao and his co-workers demonstrated a novel air electrode consisting of an unusual hierarchical arrangement of functionalized graphene sheets (with no catalyst) which delivered an exceptionally high capacity of 15000mAh/g (highest value ever reported) [90].

### 2. b Ultra capacitor.

EDLCs are non-faradic ultra capacitor which store charges in electric double layers formed at the interface between a high surface area electrode and an electrolyte [91]. Activated carbon with high specific surface area is extensively used as electrode material in EDLCs [92]. Chemically modified graphene (CMG) has been a potential material for the use as an electrode in ultracapacitors [18, 93].

Graphene materials made by thermally expanding graphene oxide (GO) at high temperature [94] or alternatively at relatively low temperature (example 200<sup>0</sup>C) under vacuum

(less than 1 Pa) [95] has been used as ultra capacitor electrodes. There are several reports regarding graphene-based ultracapacitors using metal oxides/ graphene [96-98], CNTs [99] and polymer/graphene composite [47, 100-102] as electrodes.

Tang et al. prepared graphite oxide by modified Hummers method and the graphene thus obtained exhibited an enhanced storage capacity as an electrode material in supercapacitors [103]. Kim and his co-workers fabricated CNT-graphene nanostructure via atmospheric pressure chemical vapor deposition (APCVD) for supercapacitor applications [104]. Chen et al. fabricated a composite of graphene oxide supported by needle-like MnO<sub>2</sub> nanocrystals through a simple soft chemical route in water-isopropyl alcohol system which has potential application in supercapacitor [98].

### 2. c. Fuel cell and solar cell

Graphene materials have also been used in fuel cells and solar cells. Graphene has been identified as a catalyst support for oxygen reduction and methanol oxidation in case of a fuel cell configuration [105-109]. Conductive graphene scaffolds for platinum nanoparticles facilitates efficient collections and transfer of electrons to electrode surface.

Graphene-based materials have been used as both window electrode and counter electrode in dye sensitized solar cells [79, 110]. Graphene doped conducting polymers such as poly (3,4-ethylenedi-oxythiophene) poly (styrene sulphonate) (PEDOT: PSS) and poly (3-hexylthiophene) (P3HT) have shown better power consumption efficiency (4.5%) than cells with PEDOT:PSS as counter electrode (2.3%) [111].

Wang et al. demonstrated the influence of polymer/fullerene-graphene structure on organic polymer solar devices [112]. Hsu and his co-workers reported a layer-by-layer molecular doping process on graphene for forming sandwiched graphene/tetracyanoquinodimethane

(TCNQ)/graphene stacked films for polymer solar cells [113]. Yan et al. synthesized CdS/CdSe quantum dots (QDs) co-sensitized graphene nanocomposite for potential photovoltaic applications [114]. Liu and his co-workers deposited graphene electrochemically on carbon cloth to fabricate an anode for microbial fuel cell [115]. Yong et al. fabricated a macroporous and monolithic anode based on polyaniline hybridized three-dimensional graphene for high-performance microbial fuel cells [116].

### 3. Sensors

Due to its conductance changing as a function of extent of surface adsorption, large specific surface area and low Johnson noise [1, 57, 117], recent experimental [118-123] and theoretical [124-126] research has demonstrated monolayer graphene as a promising candidate to detect a variety of molecules, such as gases [118, 119, 121, 122] to biomolecules [120, 123]. The charge transfer between the adsorbed molecule and graphene is proposed to be responsible for the chemical response.

#### 3. a. Electrochemical Sensors

Schedin et al in the year 2007 demonstrated the sensing property of graphene towards  $\text{NO}_2$ ,  $\text{NH}_3$ ,  $\text{H}_2\text{O}$  and  $\text{CO}$  by exposing graphene to the analyte of interest by vacuum annealing at  $150^\circ\text{C}$  or by illumination to UV for short time [118]. In another work Fowler et al. reported that in addition to  $\text{NO}_2$  and  $\text{NH}_3$ , dinitrotoluene was also detected by graphene sensor [127]. Sundram et al. chemically modified graphene surface by electrodeposition of Pd nanoparticles. The electrodeposition of Pd on graphene improved the response of graphene sensors to  $\text{H}_2$  detection [128].

Hu et al. prepared graphene nanosheets-gold nanocomposite by microwave radiation which has got enhanced electrochemical response and can be used in highly sensitive electrochemical sensor [129]. Ghosh et al. in their study have mentioned the  $\text{NO}_2$  and humidity sensing

characteristics of few-layer graphene [130]. Deng and his co-workers fabricated reduced graphene oxide (rGO)-conjugated  $\text{Cu}_2\text{O}$  nanowire mesocrystals for high performance  $\text{NO}_2$  gas sensor [131].

#### 3. b. Biosensors

A promising application of graphene in sensing technology is fluorescent detection due to its excellent capability in fluorescent resonance energy transfer [FRET] FRET refers to the transfer of energy from a donor fluorophore to an acceptor fluorophore, and it serves as one of the advanced tools for measuring nanoscale distance and change. Graphene was recently confirmed through theoretical and experimental studies to be a super quencher for organic dyes and quantum dots (QDs) [25, 132].

Lu et al. reported the first graphene-based biosensor consisting of a dye-labeled ssDNA probe that could be bound and quenched by graphene oxide (GO), resulting from FRET effect between the dye and GO [25]. Jang et al developed a novel GO-related assay to investigate the helicase-mediated duplex DNA unwinding activity [133].

Molecular beacons (MBs) are elaborately designed DNA hairpin structures that are dual-labeled by a fluorophore and a quencher at two ends. MBs provide more sequence specificity than linear probes due to their inherent structural constraint, thus they have been widely used in genetic screening biosensors and biochips, detection of single nucleotide polymorphism (SNP) and mRNA monitoring in living cells [134, 135]. It has also been reported that GO-quenched MB can detect DNA with higher sensitivity and single-base mismatch selectivity than conventional MB [136-138].

Dong et al. fabricated a FRET biosensor based on change in binding affinity of GO to MB-QDs upon recognizing target DNA [139]. Balapanuru et al. prepared a GO-dye charge transfer complex by a simple ion-exchange strategies which can be used as optical sensor for dsDNA

[140]. Wang et al. have designed an aptamer-carboxy fluorescein (FAM)/ graphene oxide nanosheet (GO-ns) nanocomplex to study the cellular target monitoring, which can be efficiently used for DNA and protein analysis and intercellular tracking etc. [141].

Kodali et al. used the nonperturbative chemical modification of graphene for protein micropatterning that are relevant to glucose sensors and cell sensors [142]. Wang et al. have discussed the facile one-step microwave assisted route towards Ni nanosphere/ reduced graphene oxide (rGO) hybrid for non-enzymatic glucose sensing [143].

Many other studies indicated the use of graphene and its derivatives for the detection of various biomolecules such as amino acids, oligonucleotides, dopamine, adenosine triphosphate (ATP) thrombin etc. [141, 144-148].

Yang et al. developed a label-free amperometric immunoassay for thrombomodulin using graphene/silver-silver oxide nanoparticles as a immobilization matrix [149]. Alwarappan et al. reported the direct electrochemistry of glucose oxidase at a gold electrode modified with graphene nanosheets. The result confirmed that graphene is capable of holding the enzyme GOD in a favorable position and retain its original structure and functionality that are essential for biosensing [150]. Feng et al. prepared graphite oxide (GO) and graphene by chemical method and applied to modify electrodes in electrochemical detection of hydroquinone and ascorbic acid [151]. Lian et al. developed a high sensitive uric acid sensor by using graphene doped chitosan as functional matrix and uric acid as template molecule and electrode position technique was used to form a controllable graphene-chitosan-uric acid composited film on glassy carbon electrode whose uric acid was removed via electrochemical induce elution [152].

#### **4. Bio-medical applications**

The application of the principles of biology to nanotechnology provides a valuable route for further miniaturization and improvement of performance of artificial devices. The synergetic future of nano graphene and biotechnology holds great promise for its applications in the fields like gene and drug delivery, Tissue engineering and cancer therapy.

##### **4. a. Gene delivery:**

Gene therapy is a powerful tool for the treatment of various diseases, both inborn and acquired by producing bioactive agents or stopping abnormal functions of the cells such as genetic disorder or uncontrollable proliferation of cells. One of the key issues in this area is to develop nonviral gene delivery vectors or carriers with high efficiency of gene transfection, in which cationic polymers are usually involved [153-155].

Liu et al. and Chen et al observed that polyethylenimine (PEI) generally used during transfection when grafted to other system decreases the transfection efficiency, but PEI modified graphene oxide proved to be a promising candidate for efficient gene delivery [156-158]. Kim et al. developed a GO based efficient hybrid gene delivery carrier through the installation of low molecular weight branched polyethylenimine (BPEI) a cationic polymer, which has been widely used as an efficient non-viral gene delivery vector [159]. Bao et al. reported the synthesis of a chitosan based functionalized GO (GO-CS) sheets and its applications in gene delivery [160].

##### **4. b. Drug Delivery**

In recent years, there has been a surge of interest in developing graphene for drug loading and delivery because of the strong interactions existing between hydrophobic drugs and aromatic regions of the graphene sheets. Liu et al. functionalized nano graphene oxide (NGO) a novel graphitic material with branched polyethyleneglycol (PEG) to obtain a NGO-PEG conjugate and use them for attaching

hydrophobic aromatic molecules like camptothecin (CPT) analogue, SN38 noncovalently via  $\pi$ - $\pi$  stacking [27, 28]. A controlled loading of two anti-cancer drugs, DOX and camptothecin (CPT), onto folic acid conjugated NGO (FA-NGO) via  $\pi$ - $\pi$  stacking and hydrophobic interaction was studied by Zhang's group [161].

In a study conducted by Sundar and Prajapati have mentioned carbon nano tube (CNT) and graphene as an excellent therapeutic agent for biomedical application. These nano particle surface functionalized with specific biomolecule based drug delivery has driven a new direction for modulating the pharmacokinetics, pharmacodynamics, biorecognition and for increasing the efficacy of targeted drugs. These new strategies would minimize the drug degradation and increase the drug availability [162].

Wen and his co-workers pointed out that incorporation of PEG shell possesses a significant diffusion barrier that adversely affects the release of loaded drug and therefore employed a redox responsive PEG detachment mechanism [163]. Tahara et al. exploited the availability of large quantities of single-walled carbon nanohorns (SWNHs) cytotoxicity and the immunological responses induced by the abundant uptake level in RAW 264.7 murine macrophages, that resulted in apoptosis and cell death [164]. Rana et al. reported the delivery of drug ibuprofen by using a chitosan grafted GO and controlled its release by adjustment of pH values [165].

#### 4. c. Tissue Engineering

Dikin et al. demonstrated that GO sheets dispersed in water can assembled into a well ordered structure under a directional flow, yielding ultra strong GO or rGO paper [166]. This graphene paper was used for culturing mouse fibroblast cell line (L929) and the results confirmed graphene as a good candidate for

adhesion and proliferation of L929 cells [11, 167].

Ryoo et al studied the behavior of NIH-3T3 fibroblast cells on graphene/ CNTs and suggested high biocompatibility of these nanomaterials especially as surface coating materials for implants without inducing notable deleterious effects while enhancing some of the cellular functions [168]. Lim and his co-workers studied the fabrication and characterization of graphene hydrogel and its suitability in tissue engineering applications [169]. Many other workers have exploited the properties of graphene for its use in the field of tissue engineering [142, 170].

#### 4. d. Cancer Therapy

Due to its unique conjugated structure, large surface area and relatively low costs, graphene has open a new horizon in the field of pharmacological applications in-vitro and in-vivo [27, 28]. Graphene and its derivatives have been used for several biomedical applications including anti-cancer therapy [162, 171].

Yang et al. have studied in-vivo tumor uptake and efficient photothermal therapy by intravenous administration of PEGylated nano graphene sheets (NGS) in several xenograft mouse model [172]. Shen and his co-workers used the multifunctional nanocomposite based on graphene oxide (GO) for in-vitro hepatocarcinoma diagnosis and treatment [173]. Zhou et al used GO as a photosensitive drug delivery system to explore the anti-cancer activity in-vitro in PDT [174]. Tian et al used a photosensitizer molecule chlorine e6 (Ce6) loaded on polyethylene glycol (PEG) functionalized graphene oxide (GO) for photodynamic therapy (PDT) [175]. Many other workers have used graphene and its derivatives in cancer therapy [27, 176].

#### Conclusion:

Graphene is a cheap and multifunctional material with unique physical and chemical properties. Better understanding of physics and

chemistry at the surface of graphene and interaction of chemicals and biomolecules at the interface of graphene will play an important role in applying graphene as nanoscaffold in catalysis, chemical/biosensing, imaging and drug delivery. In addition graphene is an excellent electrode material for electroanalysis and electrocatalysis, and there is still much room for the scientific research and application development of graphene-based theory, materials, and devices. As well as the GS nanocomposites could be promisingly applied in many fields such as nanoelectronics, ultracapacitors, sensors, nanocomposites, batteries and gas storage. However, in spite of the considerable advances, substantial fundamental research is still necessary to provide a basic understanding of these materials to enable full exploitation of their nanoengineering potential.

Graphene-based nanomaterials these days have led to an explosive growth of the research works on their biomedical applications can be observed from the literature in the past few years, especially in the areas of biosensors, bioelectronics and cancer therapy. Owing to the progress in graphene chemistry, fabrication of water-soluble, well-defined graphene or its derivatives with high quenching capability becomes feasible, and will benefit the development of novel FRET sensors. graphene-based sensing platform demonstrated high sensitivity and low detection threshold owing to its large specific surface area and fast electron transfer kinetics. For cancer therapy, graphene-based drug delivery has combined with other techniques, including photothermal therapy and gene delivery, to improve the overall therapeutic efficacy.

Although a lot of effort has already been put together to utilize each and every property of graphene for the development and welfare of mankind still there is much to be done. For example, taking electrochemical sensing into

consideration, there is an urgent need in this area to fabricate reliable, reproducible, and low-cost sensors with high detection sensitivity using well-defined graphene. There is still much to be done for the scientific research and technological development of graphene-related theory, materials, and applications. We would like to conclude by stating that opportunities and challenges coexist with regard to the applications of graphene-based nanomaterials.

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