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Overview of Graphene as Promising Electrode Materials for Li-ion Battery

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Abstract:

The energy and storage sector of today's world is constantly facing challenges in terms of the performance, functionality of the fundamental materials. Graphene is a Carbon-based material that is extensively investigated as anode material for rechargeable secondary Lithium-ion batteries (LIBs) because of its amazing superlative properties i.e. mechanical, optical, electrical, thermal, and sensing properties. Graphene has extraordinary electron mobility (2.5×10^5 cm^2 V^{-1} s^{-1}) and a large surface area (2630 m^2g^{-1}) and these interesting properties make it a suitable material for the energy storage device. Also Nanostructure evolution of graphene, its electrochemical performance raised to a new stage. In this review, we focus on the electrochemical performance of graphene and Graphene-based nanocomposite materials in Lithium-ion Batteries and also focuses on the synthesis route of graphene which is used both industrially and commercially.

Keywords **—**Graphene, Lithium-ion Battery, Electrochemical Performance, Electrode materials.

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1. INTRODUCTION

Lithium-ion batteries (LIBs) are widely used as energy storage devices with Compared to other types of batteries due to their various types of major advantages together with high energy density, high columbic, and energy efficiency, zero memory effect, comparatively long cycle life, and edibility in modification, which make LIBs desirable for a wide range of products such as smartphones, laptop computers batteries and mobile power banks.[26] The energy and storage sector of today's world is repeatedly facing provocation in terms of performance, functionality of the fundamental materials.[38] To fulfill the demand for electrochemical energy storage, especially for portable and the lightweight electronic devices, secondary lithium-ion batteries were introduced into the global market for a long period of time. Sony for the first time commercialized lithium-ion battery (LIB) in 1991. But also sodium-ion battery is opening the possibilities of beginning its commercial journey as a swap of LIB.[27] [28]

A secondary lithium-ion battery or Li-ion battery (abbreviated as LIB) is mainly a reversible battery, 1st outlined by chemist M Stanley Whittingham at Exxon within the Seventies. Lithium-ion batteries are ordinarily used for moveable physics and electrical vehicles and are growing in quality for military and regional applications. Lithium is the lightest of metals and it can easily float on water. The electrochemical properties of lithium are exceptional and it is also a highly reactive material. This characteristic gives Lithium the potential to achieve very high energy and power densities in high-density battery utilization such as automotive and standby power. A lithium cell can yield voltage from 1.5 V to 3 V depends on the types of materials used.[29] [30]

Lithium-ion batteries are gain enormous attention in terms of their marvel electrochemical properties. The lithium-ion battery is rechargeable, lightweight, and comparatively low cost. However, researchers are always finding a possible solution for reducing LIB cost and its weight and size.[30][31]

1.1 WORKING PRINCIPLE

Lithium-ion batteries are working on an interesting concept based on the electrochemical potential associated with the metal. Electrochemical potential means the tendency to lose electrons of a metal.

Now a general electrochemical potential series is shown below

Li	Mg	Al	Zn	Fe	H	Hg	Cu	Ag	F
3.04 V	2.37 V	1.66 V	0.76 V	0.44 V	0 V	-0.24 V	-0.34 V	-1.69 V	-2.8 V
← - Potential in Voltage									

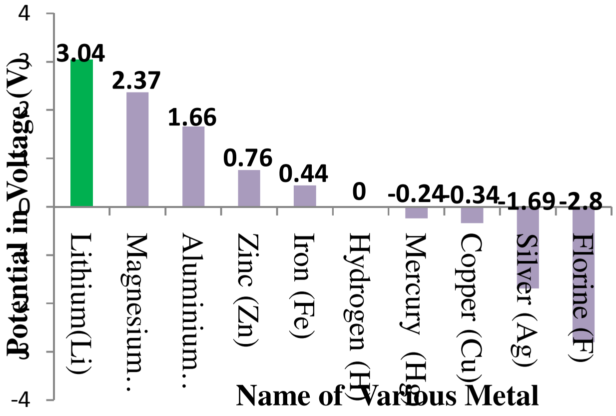

Figure 1 Electrochemical Potential Series of Various metals.

According to these values, Li is the highest tendency to lose an electron. Li metal is lightweight. Similarly, exposing a huge amount of charge storing capacity. Due to this reason, Li-ion battery occupies a wide range of electronics sectors, power devices, electric vehicles automobile parts engines, laptops, etc.

LIB batteries are two major working mechanisms one is the Discharging mechanism and the last one is the charging mechanism which is illustrated by the following figure

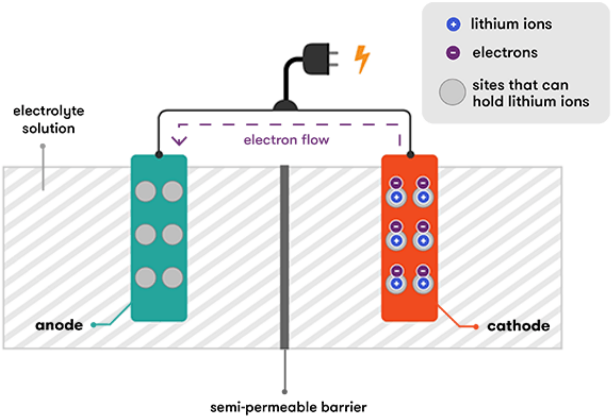

Figure 2 Lithium-ion battery charging and discharging mechanism

While discharging Li-ions are detached from the anode and migrate across the electrolyte and are inserted into the crystal structure of the host compound of the cathode. Again during charging, lithium in positive electrode material is ionized and travels from layer to layer, and added into the negative electrode. Ordinary lithium-ion batteries used graphite as an anode and lithium metal oxide compound as a cathode. The overall cell reaction is mainly divided into two categories as charging reaction and discharging reaction. The reaction is described below

During discharging

At anode: $Li_xC \rightarrow xLi^+ + xe^- + C$

At cathode: $Li_{1-x}MO_2 + xLi^+ + xe^- \rightarrow LiMO_2$

During charging

At anode: $xLi^+ + xe^- + C \rightarrow Li_xC$

At cathode: $LiMO_2 \rightarrow Li_{1-x}MO_2 + xLi^+ + xe^-$

But when the battery is in over-discharge then Li_2O is produced from $LiCoO_2$

$$Li^+ + e^- + LiCoO_2 \rightarrow Li_2O + CoO$$

Therefore in extreme overcharge CoO_2 is produced.

$$LiCoO_2 \rightarrow Li^+ + CoO_2 + e^-$$

1.2 CONSTRUCTION

The three initial structural components of a lithium-ion battery are the positive and negative electrodes and electrolytes. Ordinarily, the negative electrode of a traditional lithium-ion cell is manufactured from natural carbon. The positive electrode is a metal oxide, and the electrolyte is a lithium salt in an organic solvent. The battery consists of an anode of Lithium, diffuse as ions, into a carbon. The cathode material is made up of Lithium liberating compounds, typically the three electro-active oxide materials such as Lithium Cobalt-oxide ($LiCoO_2$), Lithium Manganese-oxide ($LiMn_2O_4$), and Lithium Nickel-oxide ($LiNiO_2$).

Li-ion cell has a four-layer structure.

- A positive electrode material is known as an anode.
- A Negative electrode material is known as Cathode.
- A separator is a fine porous polymer film.
- An electrolyte made with lithium salt in an organic solvent. The electrolytes are selected in such a way that there should be an effective transport of Li-ion to the cathode during discharge.

However, in this review, we will concentrate only on the anode materials, and in the last section of this review, we will demonstrate that Graphene is a promising anode material for secondary lithium-ion batteries due to its amazing superlative properties.

1.3 Graphene-Based Anode in LIB

Carbon-based materials including carbon fiber, carbon nanotube (CNT), carbon nanoribbons, graphite, graphene, etc. as battery electrode materials have advanced in this research field toward the studious stage. By comparing with all the carbonaceous electrode materials, graphene acquires the most potential to face the challenges of the power and storage sectors.[33]

The marvel material graphene is literally one atom thick sheet of carbon organized in an SP^2- bonded hexagonal framework. Further Graphene is a two-dimensional one-atom-thick planar sheet of sp^2 bonded carbon atoms, which is shown in Figure 2. In extension to its planar state graphene can be categorized into 0D/1D/2D dimensional Graphene. Consequently, graphene can be expressed as the mother of all carbon forms.[1]

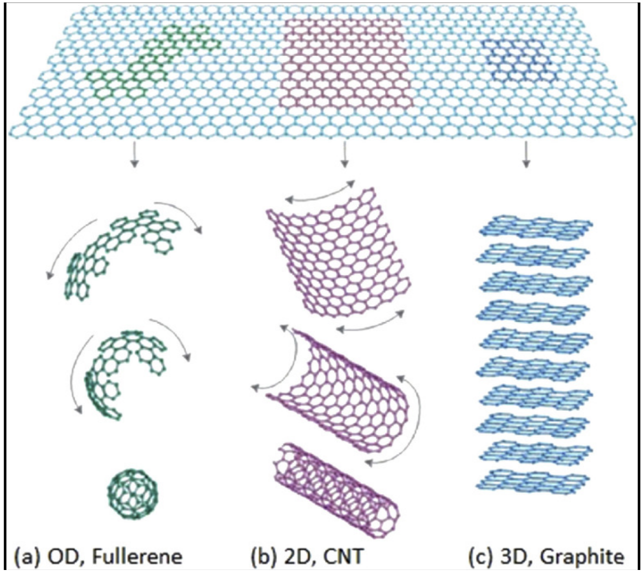

Figure 3 Mother of all Graphene forms. Graphene is a 2D building material for carbon material of all other dimensionalities. It can be wrapped up into 0D Buckyballs, rolled into 1D nanotubes, or stacked into 3D graphite.[2]

Nobel laureates Geim and Novoselov discover this marvel materials graphene and they show numerous applications of graphene depending on its wide range of properties. Graphene exposes many charming characteristics including mechanical, optical, electrical, thermal, and sensing properties. Graphene has extraordinary electron (e-) mobility (2.5×10^5 cm^2 V^{-1} s^{-1}) and a large surface area (2630 m^2g^{-1}) and these interesting properties make it a suitable material for energy storage devices. The service and performance of graphene have been checked by researchers around the world for versatile application along with its usages as an electrode (anode/cathode) and electrolyte for secondary rechargeable batteries. Despite having such interesting potential, the future of commercial-scale synthesis of graphene is now in a challenging stage. Production of a large number of high-quality graphene productions is hope or challenge for the future. In this review, our purpose is to inspect the growth of graphene-based electrode materials used in LIB.[2]

2. Graphene Synthesis Route

Most of the Graphene is synthesized from Natural Graphite but this method is unconventional and very difficult as well as costly. Many researchers and investigators produced graphene by using many routes and some of them produce Graphene on large scale and industrially. Now all the possible routes for the synthesis of Graphene are represented by the following Hierarchy chart.

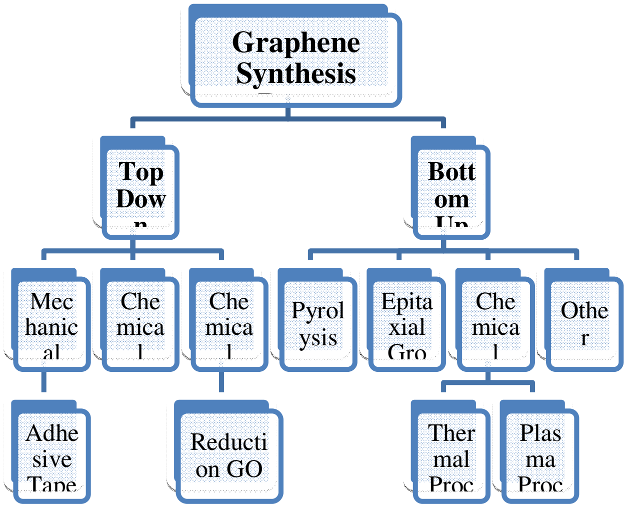

Figure 4 Routes for Graphene Synthesis.

Natural graphite is the main raw material for the extraction of graphene. For this reason, we do not need to create graphene architecture, rather need to exfoliate it from graphite. Novoselov and Geim are the first to exfoliate graphene back in 2004. They used the scotch tape method to peel a layer of graphene from a highly ordered pyrolytic graphite sample. Still, this mechanical means of graphene production yield a product of the highest quality but cannot be used commercially because of the constraint of large-scale production.[3]

However, in this review, we will describe a few specific processes for Graphene synthesis because these methods are very consistent and most available. Many researchers choose these routes for the production of a large amount of Graphene. Graphenes are mainly conventional natural graphite. Researchers choose Graphite as a raw material for the production of industrial graphene. The top-down and bottom-up approach is the main synthesis methods which are subdivided into many categories.[4]

2.1 Top-down Method

The top-down approach involves the exfoliation of graphene from a precursor material mostly graphite. The mechanical method, chemical method, electrochemical method, etc. are various top-down methods. Among them, the chemical method gained huge attention due to its advantages of moderate quality and quantity production at low cost and ease in synthesis procedures. Usually in this method, graphite is oxidized to produce graphene oxide (GO) by chemical means and then subsequently reduced into graphene. This reduction can be done by various techniques, i.e. thermal, chemical, solvothermal, photocatalytic, etc. In recent years, several modifications have been done in other top-down approaches.[5]

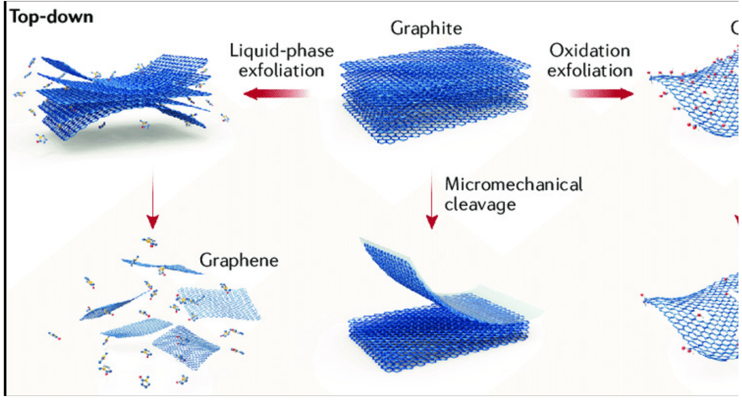

Figure 5 Top-down method for Graphene Synthesis.[5]

2.2 Bottom-up Method

Bottom-up approaches involve the growth of graphene on a precursor substrate. Chemical vapor deposition (CVD), epitaxial growth on SiC, pyrolysis, etc. are the various bottom-up approaches of graphene synthesis. Very high-quality graphene layers can be found by the Chemical vapor deposition and epitaxial growth methods, and therefore, these two methods are widely used in the laboratory for graphene research purposes.[6]

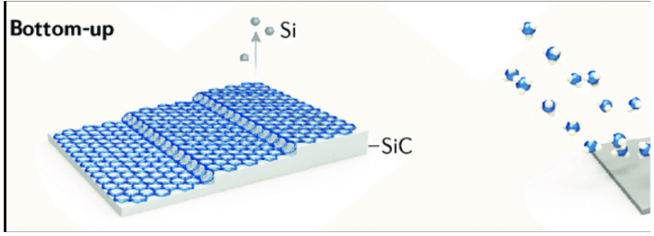

Figure 6 Bottom-up method for Graphene synthesis.[6]

2.3 *Summarizations of Few Top-down and Bottom-up Synthesis methods.*

Top-Down method						
Name of Methods	Dimensions		Essential Chemicals	Advantages	Disadvantage’s	Ref
	Thickness	Lateral				
Micromechanical exfoliation	Slight layers	μm up to cm	Highly ordered pyrolytic graphite (HOPG), unique crystal graphite,	Broad size and unmodified graphene sheets, 2% absorption rate.	Low yields, High Costly process.	[7,8]
Direct sonication of graphite	Single and Double layers	μm up to sub-μm	Graphite	Unmodified graphene, Low costly process.	A low quantity of graphene yields.	[9]
Electrochemical exfoliation	Single and slight layers	500-700 nm	Polymer composites, Graphene oxide (GO), $KMnO_4$, and $NaNO_3$ in H_2S	Single-step production and exfoliation yields high Electrical conductive graphene.	High ionic liquid cost.	[10,11]

			O_4/H_3PO_4 as oxidizing agents.			
Bottom-Up method						
Methods	Dimensions		Essential Chemicals	Advantages	Disadvantage's	Ref
	Thickness	Lateral				
Chemical vapor Deposition (CVD)	Slight layers	Very large in cm	High-quality graphite, Transition metal including Ni, Cu, Pd	The larger size and high-quality graphene produced.	Small scale production, Not used commercially.	[12]
Epitaxial growth on Silicon Carbide (SiC)	Slight layers	Up to cm size	Silicon carbide (SiC) surface	Yields graphene show a large surface area.	Very small scale production.	[13]
Unzipping of carbon nanotubes (CN)	Multiple or double layers	Less μm	Carbon nanotube (CNT)	Easily size controlled by selection of CNT	Expensive process	[14]

Table 1 Top-down and Bottom-up methods summary

3. Graphene as Anode in LIB

Investigator says that Graphene acts as promising electrode material in LIB as a result of its some awesome appealing properties like high surface to volume ratio, ultra-thin thickness, electrical conductivity, structural flexibility, etc. Similarly, these materials are very much needed for use as an electrode in LIB. Liu et al. work on graphene and they demonstrate how to expand the capacity of graphene.[15] They successfully integrated two Li atoms on the twin sides of graphene single layer maintaining stoichiometry of Li_2C_6 having a specific capacity of 540 $mAHg^{-1}$better than Li-intercalated graphite.[15] [33]

Bhardwaj et al. showed that the capacity increments of graphene in the year 2010. By using carbon nanoribbons they successfully expand the Li-ion storage capacity. They extracted GNRs by unzipping pristine multi-walled carbon nanotubes. The authors showed oxidized GNSs outperformed in terms of energy density than all other materials tested (GNSs and MWCNTs). Oxidized GNRs had the first charge capacity of ~1400 $mAhg^{-1}$ with low columbic efficiency for the first cycle (~53%) and reversible capacity in the range of 800 $mAhg^{-1}$ [16][33-37]. Now lithium-ion insertions and De-insertions of properties of various Graphene families are shown in figure 7.

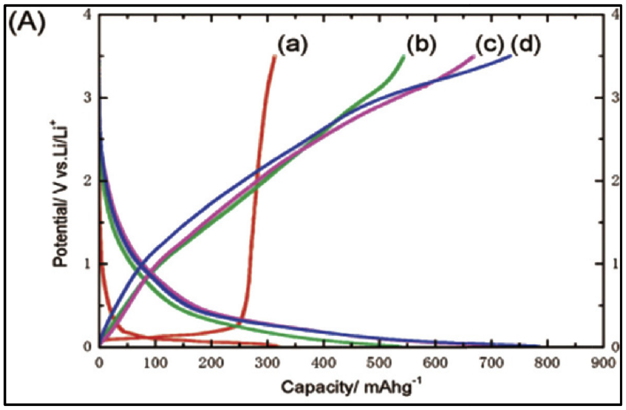

Figure 7 Charge and discharge properties of (a) Graphite (b) graphene nanosheet (c) GNS incorporated with carbon nanotube[17]

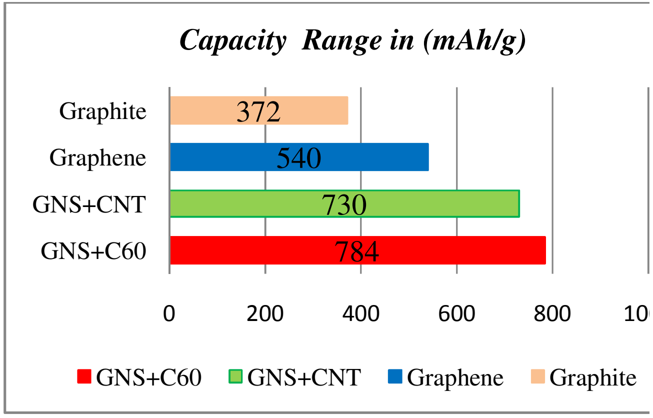

Figure 8 Comparison of Capacity of Various Carbon-based electrode materials [18].

Summarizations of LIB Anode materials involving Graphene

Materials	**Synthesis Route**	**Capacity and Cycle Performance**	**Ref**
SnO_2/graphene	Hydrothermal	The preliminary discharge capacity of 1588 mAh/g After 40 cycle its remain 730 mAh/g	[19]
Si/graphene	Hydrazine reduction/ chemical reductions	The preliminary discharge capacity of 2753 mAh/g After 50 cycle its remain 590 mAh/g	[20]
CuO/graphene	N-methyl-2-pyrrolidone solvent	Preliminary discharge capacity 640mAh/g Later 50 cycle its remain 582 mAh/g	[21]
Fe_3O_4/graphene	Graphene oxide (GO) reduction	The initial discharge capacity of 1426 mAh/g After 50 cycle its remain 1160 mAh/g	[23]

Table 2 Summary of LIB anode materials containing Graphene which is incorporated with some other non-carbon metals.

4. Conclusion

Compared to old-fashioned carbon conductor materials, Graphene, because of its low initial coulombic potency, high charge and discharge profile, and different shortcomings, cannot directly alter pristine carbon as associate degree anode material on an extensive scale. After all, Graphene will exhibit high electrical physical phenomenon, smart mechanical strength, wonderful flexibility, nice chemical stability, and high specific expanse. Once used as conductor material, Graphene will effectively cut back the scale of the active material, forestall agglomeration of nanoparticles, and improve electrons and ions transmission capability, additionally enhancing the electrode's mechanical stability. Firstly, graphene's flexibility makes it a perfect material to buffer the metal electrode's volume growth and contraction throughout the charge-discharge method. Together with, Graphene exhibit higher lithium metal particle storage capability and advanced cycle life to replace the traditional carbon. Furthermore, Graphene retains its cycle life when several cycles than traditional graphite type anode materials. Additionally, the investigator has taken numerous approaches to produce graphene on a massive scale. When graphene is incorporated with other alloy-type materials such as silicon, tin, copper oxide its electrochemical performance is enhanced greatly.

To bring this graphene-based anode into a commercial scenario as an upgraded version anode for the future-generation rechargeable battery, researches in this area has reached a potential state. Hopefully, it is not much when the effective application of graphene-based, as well as graphene-based composite materials as an anode in rechargeable batteries, will be treated as a breakthrough in battery performances.

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