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# Advances in the Application of Graphene in Lithium-ion Batteries

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**Abstract:**

The search for effective energy storage options has escalated due to the rising global energy demands and environmental concerns. Lithium-ion batteries (LIBs) emerge as a key technology. However, the performance of traditional LIBs still needs to be improved. Therefore, choosing new nanomaterials for the modification of LIBs has become an effective solution. Graphene, as a nanomaterial, is an excellent candidate material for modification. This paper delves into the application of graphene in enhancing the performance of LIBs. Graphene, with its superior electrical conductivity and substantial specific surface area, offers the potential to enhance the performance of both anode and cathode materials in LIBs. This can lead to significant improvements in the overall cycle life, energy density, and power density of these batteries. This work emphasizes the application of graphene in cathode and anode materials. In cathode materials, the layered structure of graphene enables lithium-ion with high theoretical capacity. Meanwhile, graphene helps improve the electron and lithium ion mobility of anode materials. Despite the promising developments, challenges such as lithium loading capacity and coulombic efficiency still remain. The continued exploration of graphene applications is expected to drive LIBs to unprecedented performance metrics. This is in line with the global quest for sustainable and efficient energy storage technologies.

**Keywords:** LIBs; graphene; electrode materials; performance.

## 1. Introduction

With the significant growth of the global population, energy issues have become a key concern for people. The exploration and utilisation of renewable energies has become crucial for the protection of our common home. In order to achieve this goal, it is necessary to progressively utilise cheap and sustainable energy sources such as solar, wind and tidal energy. In this way, dependence on non-renewable fossil fuels will be replaced [1]. As a quintessential instance of electrochemical energy storage, battery technology plays a crucial role in the mechanisms of energy conservation and dissemination. In the realm of modern science, there is a collective push within the academic community to develop energy storage solutions that are environmentally sustainable and highly efficient. Such innovations are critical to the creation of alternative energy sources capable of replacing traditional energy sources and fossil fuels.

In this process, rechargeable lithium-ion batteries (LIBs) have been shown to be superior to other potential battery technologies due to their long life cycle, low self-discharge characteristics and high energy/power density [2-3]. However, the performance of LIBs is difficult to meet diverse demands. Therefore, selecting materials with excellent performance to modify LIBs has become the first choice for researchers. Graphene is a high-performance nanomaterial. The use of graphene for modifying LIBs has become a research hotspot in recent years.

Therefore, this research delves into the possibles uses of graphene to enhance the performance of batteries and assesses its contributions to the evolving field of LIBs technology.

## 2. LIBs and Graphene

Understanding the mechanism of LIBs is the foundation for their modification. Lithium is the elemental cornerstone of lithium-electron batteries. Of all the elements discovered to date, lithium has the lowest reduction potential and the highest battery potential. This property is the main reason why LIBs are favored in contemporary applications. Furthermore, lithium boasts the third smallest atomic radius among all monovalent ions listed in the periodic table. This endows lithium batteries with exceptional power density [4]. In view of this, LIBs, are now the primary energy storage technology in use. Its application scenarios include electric cars, mobile devices, and energy storage. However, research on high-performance battery materials is essential due to the rising demand for better safety, longer cycle lives, and better energy densities. A lithium-ion battery ($Li^+$) is separated from the battery anode by an external voltage while the battery is charging. It then moves through the electrolyte to the anode surface and becomes embedded in the lattice structure. To complete the transformation of electrical energy into chemical energy, electrons travel to the anode through the external circuit. During the discharge process, lithium ions ($Li^+$) and electrons move from the anode towards the cathode. The lithium ions travel through the electrolyte, and the electrons traverse the external circuit. During the entire charge/discharge cycle of a lithium battery, lithium ions are constantly inserted or delaminated between the cathode and anode. Consequently, the conductivity of the electrode materials exerts a substantial influence on the energy density, power density, and the ability for rapid charging of LIBs. There is a vast array of structural designs and a multitude of carbon materials. It is generally known that they are chemically and physically stable. They have been extensively utilized as active electrode materials and as conductive additives in the formulation of LIBs. Graphene is a carbon material with excellent mechanical robustness, a fast electron mobility of $2\times10^5$ $cm^2/(V\cdot s)$, a huge specific surface area of 2630 $m^2/g$, and superb thermal conductivity of 5000 $W/(m\cdot K)$ [5-7]. Because of these characteristics, graphene has a lot of promise for use in the field of LIBs.

Graphene is made up of carbon atoms bonded by $sp^2$ hybridisation, and theses atoms tightly located to form a single two-dimensional honeycomb lattice. Carbon atoms are covalently connected in-plane, resulting in σ-bonds with three surrounding carbons and an out-of-plane π-bond. This $sp^2$ hybridized connection network gives graphene its unique properties. Graphene has the highest thermal conductivity in carbon material. At ambient temperatures, it exhibits a carrier mobility of approximately 15000 $cm^2/(V\cdot s)$. The velocity of its electrons can reach one-third of the speed of light, which is a feat that surpasses the performance of conventional conductors [8]. Graphene can be directly added to the conductive agent to prepare electrode slurry. Then, it is mixed with the active material, coated electrodes. By this way, graphene dispersed between the active material, constituting a continuous and efficient point-to-point contact mode conductive network. It can provide a long conductive path for e-rapidly. The high conductivity of graphene reduces the interfacial resistance, and it facilitates $Li^+$ conduction between the positive and negative electrodes.

## 3. Utilization of Graphene as Cathode Materials

There are several common cathode materials widely used in LIBs. The most commonly used include lithium iron phosphate ($LiFePO_4$). However, it has relatively low conductivity for $Li^+$ and electrons. This limitation affects the overall performance of the battery. Graphene is known for its superior electrical conductivity and expansive specific surface area. Therefore, graphene is increasingly used as a cathode material for LIBs. Composite of graphene with cathode active material can form a three-dimensional porous structure. This composite enhances the specific surface area and the number of active sites on the graphene, while it also facilitates the movement of $Li^+$. For example, graphene was introduced into lithium iron phosphate (LFP) cathode materials by Edigar et al. This improves the efficiency and cycling stability of the transport between electrons and lithium ions, while it also extends the cycle life [9]. Ali et al changed the $LiMn_2O_4$ (LMO) cathode by ultrasound-assisted electrochemically manufactured graphene, which was synthesized in the study by electrochemical exfoliation technique [10]. In this work, higher grade graphene (EG1) is used. Electrochemical experiments show that a cell with an LMO/EG1 cathode has a higher coulombic efficiency (89%) than a pure LMO cell (81%). In addition, the LMO/EG1 battery has a 94.8% retention rate of discharge capacity after 100 cycles at 0.1C, which is significantly higher than the 71.4% of the LMO battery. This achieves the improvement of LIB charging and discharging performance. On the other hand, Han et al. successfully created $C_6O_6$/Gr composites by an eco-friendly ball milling procedure and studied their potential as LIB cathodes. A large number of effectively exposed $C_6O_6$ active sites are obtained in the prepared sample. It integrates highly conductive graphene and a well-developed stable structure. These enable large penetration of the electrolyte, efficient diffusion of ions and rapid transfer of electrons [11].

Creating a stable solid electrical interaction (SEI) in the cathode material is the other significant functions of graphene. This SEI layer is important because it acts as a barrier between the electrolyte and the electrode material. By preventing the electrolyte from directly touching the electrode, the SEI layer reduces the likelihood of unwanted side reactions. These reactions can damage the battery and shorten its lifespan. Kim et al. used a polyethylene glycol (PEG) thin carbon layer encased in graphene. This coating inhibits the development and production of the SEI layer by obstructing the electrolyte-silicon particle interface. The Si/C-PEG/G composite, which consists of carbon-coated silicon nanoparticles integrated with graphene, demonstrated remarkable electrochemical performance. Notably, it achieved a coulombic efficiency of 99.5% by the 10th cycle. Furthermore, at a current density of 0.1 A/g, it exhibited an optimal capacity of approximately 1820 mA·h/g [12].

## 4. Utilization of Graphene as Anode Materials

Multilayer graphene shows notable benefits for the embedding of $Li^+$ in the energy storage domain. The layered architecture of the material facilitates the storage of $Li^+$ on either side of its layers, leading to the formation of $Li_2C_6$ compounds. This configuration endows the material with a theoretical specific capacity that can reach as high as 744 mA·h/g [13]. That is twice as high as that of conventional graphite materials. In addition, monolayer graphene achieves effective adsorption of $Li^+$ through the randomly arranged nanopore structure on its surface as well as between the layers. The nanoscale characteristics of graphene are pivotal in determining its electrochemical efficacy when utilized as an anode in LIBs. A novel silicon-doped graphene material (SiG) was studied by Liu et al. The graphene lattice was successfully doped with silicon impurity atoms by microwave radiation method. The application of this material in LIBs showed a higher specific capacity than pristine graphene, reaching twice the specific capacity. Electrochemical testing at multiple current densities has demonstrated that SiG electrodes exhibit superior capacity and rate performance compared to pure graphene electrodes. With 400 cycles at a substantial current density, the SiG-450 electrode maintains a specific power rating of 145 mA·h/g, demonstrating its exceptional cycling performance. In addition, the cyclic voltammetry (CV) curves of the SiG-450 electrode at different sweep speeds showed a similar behaviour of pristine graphene. After 400 cycles, two small oxidation peaks appeared, which were attributed to the effect of silicon heteroatoms. During the full-cell evaluation, the SiG/$LiFePO_4$ cell maintained 86% of its initial capacity after 200 cycles, highlighting the viability of SiG as an anode material for real-world applications [14]. Li et al evenly embedded silicon nanoparticles (Si NPs) and graphene oxide (GO) in a bitumen matrix using a straightforward solvent dispersion technique. The composites were prepared by a carbonization process. GO formed a multi-interfacial structure inside the composites, which reduced the exposed surface and improved the electrical conductivity. Empirical data show that Si/G/P composites exhibit commendable electrochemical behavior when used as anodes. Their cycling stability reached 93.6% capacity retention after 1000 cycles at a current density of 2 A/g. Additionally, these electrodes display a remarkable reversible capacity, maintaining a capacity of 820.8 mA·h/g at a current density of 50 mA/g. In a full battery setup with a $LiFePO_4$ cathode, the system retains 95% of its capacity after undergoing 100 cycles at a current density of 85 mA/g [15].

Despite the great lithium storage capacity of graphene anodes in LIBs, they face many difficulties in practical applications. Low coulombic efficiency, short cycle life, high operating voltage, low lithium loading capacity, and electrochemical degradation under high current density conditions are just a few of the specific problems. In order to overcome these problems, current research directions have focused on creating hybrid anode materials based on graphene doped with metal oxides or other dopants. Besides, There is also potential for improving performance through structural design. Through morphological engineering, graphene electrodes can be designed into porous, spherical, strip-shaped structures to enhance their surface area and electrochemical activity.

## 5. Conclusion

Incorporating graphene into LIBs is anticipated to enhance the capabilities of energy storage systems. It has superb mechanical strength, huge specific surface area and excellent electrical conductivity. These unique properties help to extend the life of LIBs and improve their efficiency. This study has scrutinized various applications of graphene aimed at augmenting the performance of both anode and cathode materials.

The graphene with layered structure is promising for lithium-ion intercalation in the field of anode materials. Its theoretical capacities exceed those of traditional materials. Novel approaches like silicon-doped graphene have demonstrated a notable increase in particular capacity and speed performance. For cathode materials, the incorporation of graphene has been shown to improve electron and lithium-ion transport. Using graphene to create three-dimensional porous structures has shown to be a particularly successful tactic, since it increases the active surface area and promotes ion transport.

Despite these encouraging advances, the practical application of graphene-based anodes still faces obstacles, such as lithium loading capacity, operating voltage and coulombic efficiency. The goal of the current research is to better improve the electrochemical performance of lithium batteries by developing graphene-based hybrid materials containing metal oxides. These challenges will be incrementally overcome to promote the application of this technology. These innovations are anticipated to embody high-performance metrics, eco-friendly characteristics, and economic prudence, thereby addressing the demands of sustainable energy solutions in a global context.

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