The Application of Carbon Nanotube Electrode in Lithium-Ion Batteries

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Abstract. Carbon nanotubes (CNTs) have emerged as promising materials for enhancing lithium-ion battery (LIB) performance due to their exceptional electrical conductivity, mechanical strength, and unique nanostructure. This review examines the application of CNTs in both anode and cathode materials, highlighting their advantages and existing challenges. In anodes, single-walled CNTs (SWCNTs) effectively mitigate silicon particle volume expansion while providing high conductivity, enabling capacities up to ~3200 mAh • g^{-1} . Multi-walled CNTs (MWCNTs) serve as robust scaffolds for silicon coatings, improving coulombic efficiency from 12% to 94% after 10 cycles. However, issues such as irreversible lithium loss, SEI formation, and high production costs persist. For cathodes, annealed MWCNTs form 3D conductive networks that enhance $LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$ (NCM) performance, achieving 173.16 mAh • g^{-1} at 0.1 C and 95.8% capacity retention after 100 cycles. Nevertheless, energy-intensive annealing and dispersion challenges hinder scalability. Key limitations include defect-related side reactions, van der Waals aggregation, and electrolyte decomposition. Future research should focus on hybrid architectures, cost-effective synthesis, and scalable manufacturing to realize CNTs' full potential in next-generation LIBs.

Keywords: CNT; Anode; LIBs.

1. Introduction

Global warming crisis rising, fossil fuel resources diminishing, and air pollution caused by car internal combustion engines are all forcing society to accept using sustainable and renewable power sources. This is why an extremely large number of alternative sources of energy have appeared: like solar, wind. At the same time, the development of electric and hybrid cars with low carbon dioxide emissions is going full speed ahead. Solar power and wind power power up and down pretty intermittently as well in the middle of the night, so having something that can do it by itself for a while is kind of important. This need makes energy storage systems more important for our modern life [1]. To address the challenges brought forth by contemporary society as well as newly rising environmental issues; it's a must to create brand-new, cheaper and friendly to environment energy transformation and saving systems. When we talk about Electro Chemical Energy Storage System, they are Lithium - Ion Battery, Fuel Cell and ECS. And because much of the system's performance depends on the characteristics of the materials they're made from, plenty of research and development has gone into developing these key materials. But, in order for us to provide the more challenging application requirements that are expected in the future, from smaller, smarter cell phones to hybrid cars, to heavy industrial unit will need better materials. This could be done by finding new materials and a better knowledge of the nano-electrochemical interface. Lithium-ion batteries have high energy density and open-cell voltage. Though the lithium-ion battery performs pretty well, its design remains many challenges. The first main challenge is improving anode material capacity and cycle-life. Graphite used to be the only anode material for lithium-ion batteries. The reason behind that is because of its high in-plane e^- conduction due to non-local π-bonds and its good structure that can be easily intercalated by Li^+ ions. However, the capacity of lithium-ion batteries containing graphite as a negative electrode cannot go on forever. Lithium ions can only fit between alternates in the graphite layer, so it needs 6 carbons per atom of lithium (LiC_6). It ends up in 372mah/g specific capacity and also the practical capacities are 280-330 mah/g, depending upon the kind of graphite being used. Carbon nanotubes (CNTs), which are allotropes of graphite, have proven valuable. Due to its unique structure and property, its lithium storage capacity is much higher than graphite [2,3]. SWCNTs and MWCNTs are said to have extremely high electrical conductivities, reaching 10^6 $S.m^{-1}$ and 10^5 $S.m^{-1}$, respectively, and very strong tensile strengths between 60 GPa. In recent years, a lot of attention has been paid to CNT-based lithium-ion battery anodes, with different results obtained depending on the treatment method used. The latest development progress in the application of carbon nanotube structures and forms in the anode of lithium-ion batteries are introduced in a general way [4].

Among all the emerging materials, Carbon Nanotubes (CNTs) have attracted considerable attention as one - dimensional nanostructured allotropes with excellent electrical, mechanical and structural properties. In contrast to graphite, CNTs is mainly made up of a hollow cylinder with nanometer diameter and high aspect ratio. It makes it easy for lithium ions to move around and gives lithium places to stay. SWCNTs and MWCNTs have excellent electrical conductivity reaching up to 1-6 $S \cdot m^{-1}$ and 1-5 $S \cdot m^{-1}$ respectively, and strong tensile strength [5,6]. So, these things point out that CNTs might fix problems that come up with graphite and really make LIB anodes work better. Recently, efforts have been put into engineering CNT-based anode by different means of chemicals and physiochemical modifications to obtain high capacity and excellent rate capability as well as cycle stability.

This paper presents a current review of the application of carbon nanotubes structure and morphology in Li-ion battery electrodes. It mainly discusses synthesis methods, surface functionalization, and composites affecting the electrochemical properties of the CNT-based anode and cathode. The next sections will be major studies in which progress, problems, and potential solutions were shown. In addition to the existing literature, some ideas for next-generation lithium-ion batteries are suggested for the improvement of CNT-based materials.

2. Case

2.1. Carbon Nanotubes

Carbon nanotubes are literally cylindrical nanostructures made up of carbon atoms with a hexagonal structure. They possess great electrical conductivity, mechanical strength, and chemical stability are well known and make them suitable for use in a number of applications, including lithium-ion batteries. High surface area and aspect ratio of CNTs are mainly attributed to their ability to enhance electron transport and ion diffusion. These same properties enable them to improve the durability of the electrode by withstanding volume expansion during charge/discharge operations.

The main CNTs in electrodes is divided into Single-walled carbon tubes (SWCNTs) and multi-walled carbon tubes (MWCNTs). The advantages and disadvantages were introduced as follows.

2.2. Lithium-ion Anode

Graphite is in fact the most widely used anode material in commercial lithium-ion batteries due to its ability to hold lithium ions intercalated between its layers. Nowadays, Silicon as an anode material offers a much higher theoretical capacity (~10 times that of graphite). However, its large volumetric expansion (~300%) during lithium intercalation and deintercalation results in cracking and degradation, which is a commonly reported problem. Application of CNTs into anodes has been shown in research to create conductive networks that are capable of hosting volume changes, minimizing mechanical degradation and increasing cycle stability. assist to produce high-performance lithium-ion battery anodes.

2.3. Lithium-ion Cathode

The cathode is the source of lithium ions and governs energy density and voltage, typical cathode materials include Lithium cobalt oxide ($LiCoO_2$), NMC ($LiNiMnCoO_2$), and lithium iron phosphate ($LiFePO_4$), they are among the most widely used cathode materials in lithium-ion batteries. This is well established in battery research. CNTs can be utilized to enhance electrical conductivity by the development of networks that reduce cathode material resistance. This has been evidenced in experiments. CNTs also enhance structural stability, particularly in layer-type cathodes, by preventing stress and being resistant to cycle degradation.

3. Analysis & challenge

3.1. SWCNT in anode

A SWCNT/Si anode's preparation method is as follows: Silicon MPs and NPs served as the active substances. Single walled carbon nanotubes (SWCNTs) were used as a conductive additive with 0.4 wt% SWCNT dispersion in water. The ratio of the mass between the active material and the conductive agent was set as 90:10. In order to evaluate the battery capacity and cycling stability according to the ratio between micro-sized and nano-sized silicon, three different silicon mixtures were created, as can be seen in Fig. 1. The silicon particles and the dispersion with SWCNTs were uniformly mixed at 2000 rpm for 10 minutes using a planetary centrifugal mixer. And the obtained anode slurry was coated on a copper foil with thickness of about 150 μm by a doctor's blade. Then it was dried at 65℃ for 30min, then dried under vacuum at 80 ℃ overnight. [7]

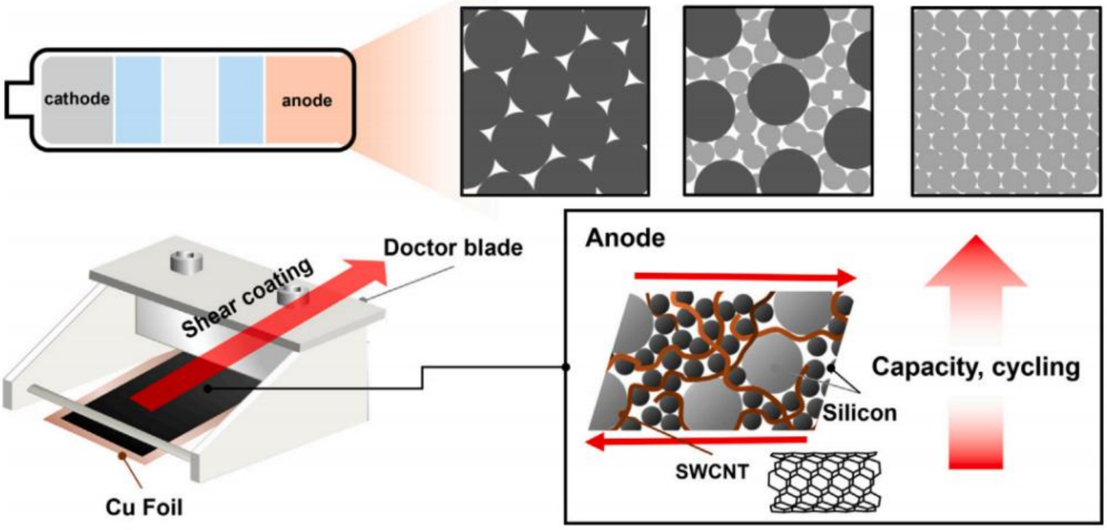

Fig. 1. Schematic illustration of silicon anode slurries: the anode electrode with increased packing density is achieved by mixing silicon microparticles and nanoparticles. [7]

When we add single walled carbon nanotubes (SWCNTs) to help electricity move better and give some physical help, they wrap like a blanket around the Si bits. This structure avoids volume expansion and maintains electrode. High capacity and improved cycling stability were achieved by means of the nano and micro-Si particles. Other than the current density of 10.0 A/g which is exceedingly high, the SWCNT-Si electrode delivers rate performance at all other current densities. The Si MPs and Si NPs mixture anodes have a clear trend of decreasing electrical conductivity when Si NPs are added. However, its conductivity can also remain at a relatively high level of about $2*10^3$ S/m. And that means that to achieve the best equilibrium between Si MPs and Si NPs, which guarantees the conductive channel formed by SWCNTs will be safe from NP agglomeration. [7]

3.2. MWCNT in anode

The MWCNT-Si composite anode was prepared as shown in Fig. 2: Multi-walled carbon nanotube (MWCNT) tissue with a thickness of 73 μm and an areal density of 2 mg cm^{-2} was used as the scaffold material. The MWCNT tissue was pre-treated under vacuum for more than 10 hours to remove adsorbed moisture. Silicon coating was then deposited onto the MWCNT tissue via magnetron sputtering (Quorum Q150T ES) using an arsenic-doped n-type silicon wafer as the sputtering target. The sputtering process was conducted under argon gas at a current of 80 mA, with durations of 5, 10, and 20 minutes, resulting in silicon weight loadings of 0.5, 1.0, and 2.0 mg cm^{-2}, respectively. Prior to each sputtering step, the silicon target was activated to remove the outer passivation layer of silicon oxide. The resulting Si-coated MWCNT (Si/MWCNT) composite sheets were cut into 0.6 cm × 0.6 cm squares for electrochemical testing [8].

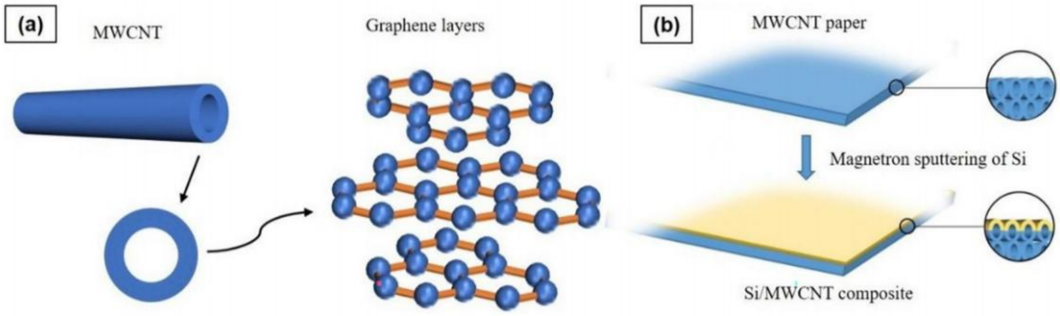

Fig. 2 Fabrication process of MWCNT-Si composite anode [8]

For comparison, pristine MWCNT tissue without silicon coating was also evaluated as an anode material. The composite anode demonstrated improved electrochemical performance, including higher coulombic efficiency and gravimetric capacity, compared to the uncoated MWCNT tissue. The detailed characterization and electrochemical measurements confirmed the uniform distribution of silicon on the MWCNT surface, with minimal penetration into the inner layers, as evidenced by SEM-EDS and Raman spectroscopy analyses

The Si-coated MWCNT (Si/MWCNT) composite anode exhibited markedly enhanced electrochemical properties compared to pristine MWCNT tissue. The silicon coating, deposited via magnetron sputtering, improved the initial coulombic efficiency (CE) from 12% (pristine MWCNT) to 23%, with CE further increasing to 94% by the 10th cycle. The gravimetric capacity also rose from 109 mAh g^{-1} (uncoated MWCNT) to 290 mAh g^{-1}, attributed to silicon's high theoretical capacity and the conductive MWCNT scaffold. Cross-sectional analysis confirmed a conformal Si coating (~427 nm thick) on the MWCNT surface, which mitigated irreversible lithium loss by reducing SEI formation and graphene layer peeling during cycling. The hybrid anode also showed superior rate performance and capacity retention (39% after 100 cycles) due to the MWCNT network's mechanical robustness, which accommodated silicon's volume changes. [8]

Despite these improvements, the Si/MWCNT anode still faces critical challenges. The irreversible capacity loss in the first cycle remained high (77%), primarily due to SEI formation on the MWCNT sidewalls and strong Li-ion adsorption within the nanotube cores. The sputtered Si coating, while effective, did not fully eliminate "dead lithium" accumulation (0.54 mg after 100 cycles). Additionally, higher Si loadings (>1 mg cm^{-2}) induced pulverization from volume expansion, degrading cycling stability. The study notes that further optimizations—such as pre-lithiation, doping, or artificial SEI layers—are needed to address these issues. Scalability of the magnetron sputtering process and the cost of high-purity MWCNT tissues also pose practical hurdles for commercialization.

3.3. CNT in cathode

The MWCNT-based cathode samples were prepared as follows [9]: Multi-walled carbon nanotubes (MWCNTs) were synthesized via chemical vapor deposition (CVD) using iron nitrate and aluminum nitrate as catalysts, with propane as the carbon source. The as-prepared MWCNTs were then annealed at temperatures ranging from 2000 to 2800 °C under an argon atmosphere to remove metal impurities and reduce structural defects. The annealed MWCNTs were dispersed in N-methyl-pyrrolidone (NMP) with polyvinylpyrrolidone (PVP) as a dispersant (solid content ratio: 95:4:1) and ground using a sand mill at 1600 rpm for 30–40 min to form a homogeneous conductive slurry.For cathode fabrication, $LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$ (NCM) active material, polyvinylidene fluoride (PVDF) binder, and the MWCNT slurry were mixed at a mass ratio of 96:2:2 in NMP. The mixture was homogenized at 2000 rpm for 20 min, coated onto aluminum foil (thickness: 0.019 mm), and dried at 120 °C for 24 h. The cathodes were assembled into CR2032 coin cells with lithium metal anodes in an argon-filled glovebox. Samples were labeled as NCM-CNT-(annealing temperature), e.g., NCM-CNT-2200 °C, with unannealed MWCNTs and Super P (SP)-based cathodes serving as controls. The annealed MWCNT and cathode were shown in Fig. 3.

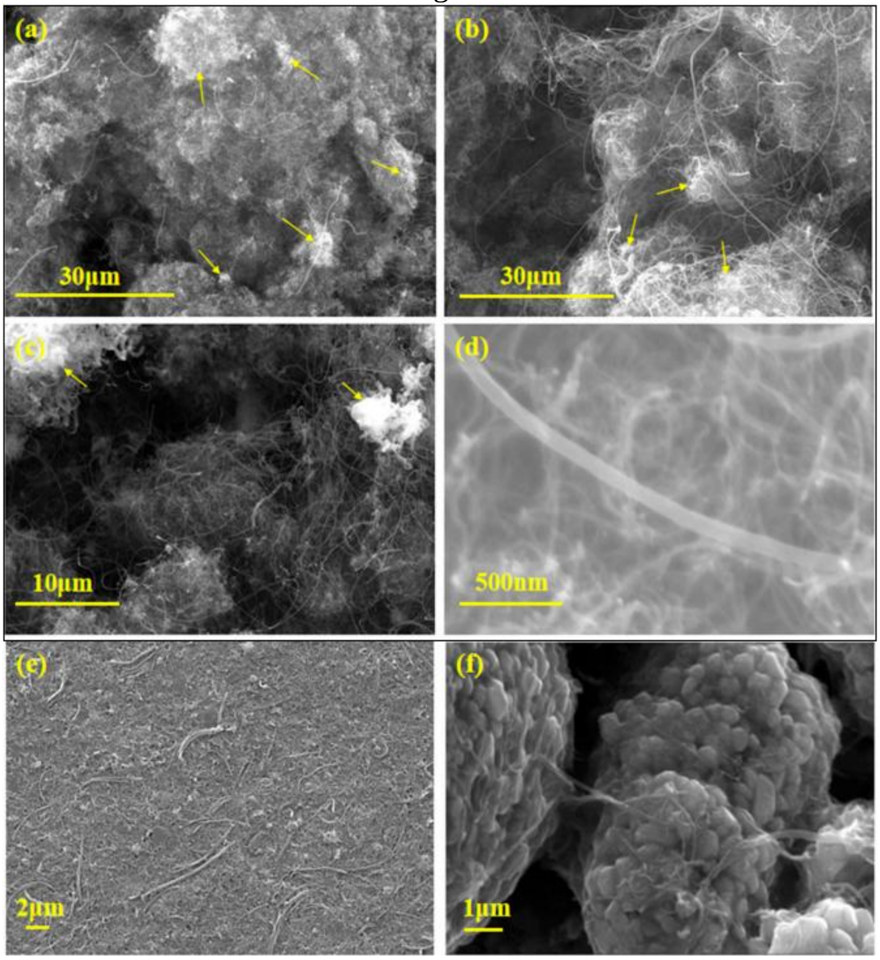

Figure 3. Characterization of MWCNTs, SEM images of (a) undiluted MWCNTs; (b) MWCNTs annealed at 2000°C; (c) MWCNTs annealed at 2400 °C; (d) MWCNTs annealed at 2800 °C; (e) MWCNTs after standing and dispersion; (f) annealed MWCNTs slurry cathode sheet [9]

When incorporated into the $LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$ (NCM) cathode, high-temperature-annealed multi-walled carbon nanotubes (MWCNTs) significantly enhance electrical conductivity and form a three-dimensional conductive network, improving electron transport and lithium-ion diffusion. The MWCNT-based cathode (NCM-CNT-2200 °C) exhibited the highest initial discharge capacity of 173.16 $mAh \cdot g^{-1}$ at 0.1 C and maintained 95.8% capacity retention after 100 cycles at 0.5 C, along with a superior rate performance (121.75 $mAh \cdot g^{-1}$ at 5 C). This improvement is attributed to the effective removal of metal impurities and defect reduction in MWCNTs through annealing, as well as the uniform dispersion achieved via sand-mill processing, which ensures optimal contact between active particles and conductive pathways. [8]

Despite these advantages, challenges remain. The high-temperature annealing process, while effective in purifying MWCNTs, is energy-intensive and may not be scalable for industrial production. Additionally, excessive annealing (e.g., 2800 °C) can over-reduce defects, potentially diminishing lithium-ion diffusion pathways and leading to lower electrochemical performance. Furthermore, the strong van der Waals forces between MWCNTs still pose dispersion challenges, requiring mechanical or chemical treatments that may damage their structure. Future research should focus on optimizing annealing conditions and developing cost-effective, scalable dispersion methods to further enhance the performance and practicality of MWCNT-based conductive agents in lithium-ion batteries.

4. Summary and Suggestion

SWCNTs as anodes have advantages like ultrahigh electrical conductivity (10^6 $S \cdot m^{-1}$) and exceptional capacity when combined with silicon (~3200 $mAh \cdot g^{-1}$), along with their unique ability to wrap around Si particles, alleviating volume expansion. On the other hand, SWCNT anodes have disadvantages such as high production costs, safety concerns related to nanoscale handling, and excessive solid electrolyte interphase (SEI) formation during cycling. SWCNTs have significant potential, but improvements such as cost-effective synthesis methods (e.g., biomass-derived carbon sources) and surface passivation to minimize SEI could enhance their viability for commercial batteries.

MWCNTs as anodes offer a more economical alternative with excellent mechanical stability, serving as durable scaffolds for silicon coatings. Their hierarchical structure accommodates volume changes better than conventional materials. However, challenges like irreversible lithium loss within nanotube cores and limited silicon loading capacity (<1 mg cm^{-2}) restrict their performance. To address these issues, pre-lithiation techniques and hybrid composites (e.g., MWCNT-graphene hybrids) could be explored to improve initial coulombic efficiency and cycling stability.

CNTs as cathodes significantly enhance conductivity and structural integrity, particularly in high-voltage materials like NCM ($LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$). Their 3D networks facilitate electron transport and lithium-ion diffusion, enabling superior rate performance. Yet, challenges persist, including energy-intensive high-temperature annealing and dispersion difficulties due to van der Waals forces. Future research should focus on low-temperature synthesis methods (e.g., plasma-assisted CVD) and advanced dispersion techniques (e.g., sand-mill processing with optimized surfactants) to improve scalability.

Those advantages were comclude in Table.1. To fully unlock the potential of CNTs in lithium-ion batteries, interdisciplinary efforts are needed. Innovations in hybrid architectures (e.g., CNT-graphene composites), green synthesis routes, and advanced manufacturing (e.g., roll-to-roll electrode printing) could bridge the gap between lab-scale success and industrial adoption. Standardized testing protocols for CNT-based electrodes will further accelerate commercialization, paving the way for next-generation batteries in electric vehicles and grid storage.

Table 1. Comparation of various CNT-added electrodes

Type	advantages	disadvantages
SWCNT in anode	-high electrical conductivity -high capacity -improved cycling stability	- Irreversible Li loss in cores -Energy-intensive annealing
MWCNT in anode	- Lower cost than SWCNTs - Robust scaffold for Si coatings - Better mechanical stability	- Irreversible Li loss in cores -Energy-intensive annealing
CNT in cathode	-Improves NCM cathode stability - Reduces binder dependency	-High-temperature annealing required -Dispersion challenges (van der Waals forces)

5. Conclusion

This review systematically examines the application of carbon nanotubes (CNTs) in lithium-ion battery (LIB) electrodes, focusing on their roles in enhancing anode (e.g., silicon-based) and cathode (e.g., NCM-based) performance. The study demonstrates that CNTs, with their exceptional electrical conductivity, mechanical strength, and unique nanostructure, significantly improve battery energy density, cycling stability, and rate capability.

For anodes, single-walled CNTs (SWCNTs) form conductive networks around silicon particles, mitigating volume expansion while enabling high capacity (e.g., ~3200 $mAh \cdot g^{-1}$ for Si composites).

Multi-walled CNTs (MWCNTs) serve as cost-effective scaffolds for silicon coatings, improving coulombic efficiency (e.g., from 12% to 94% after 10 cycles) and cycle life. However, challenges such as irreversible lithium loss, excessive solid electrolyte interphase (SEI) formation, and high production costs remain unresolved.

In cathodes, high-temperature-annealed MWCNTs create 3D conductive pathways, boosting the discharge capacity (173.16 $mAh\cdot g^{-1}$ at 0.1 C) and retention (95.8% after 100 cycles) of NCM materials. Yet, energy-intensive annealing and dispersion difficulties due to van der Waals forces limit scalability.

In summary, CNTs offer transformative potential for LIBs, but their commercialization requires advances in defect control, cost-effective synthesis, and scalable electrode fabrication. Future work should prioritize hybrid architectures (e.g., CNT-graphene composites) and green manufacturing to bridge lab-scale innovations with industrial applications.

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