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Review

Advances in Carbon Coatings for Current Collectors in Lithium-Ion Battery Applications: Focus on Three-Dimensional Carbon Nanowalls

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Abstract: Current collectors are key components of lithium-ion batteries, providing conductive pathways and maintaining interfacial stability with the electrode materials. Conventional metal-based current collectors, such as aluminum and copper, exhibit excellent conductivity and mechanical strength. However, they have considerable limitations, including electrochemical corrosion, interfacial resistance caused by the formation of passive layers, and mechanical degradation due to repeated cycling. To overcome these challenges, various carbon-based coatings, including amorphous carbon, graphene, and carbon nanotubes, have been developed. These coatings enhance the current collector performance by improving the collector conductivity, chemical stability, and interfacial adhesion. Vertically aligned graphene-like structures known as carbon nanowalls (CNWs) have garnered attention owing to their unique architecture, resulting in high surface area, exceptional conductivity, and excellent thermal and mechanical properties. In this mini-review, the recent advancements in carbon-based coating technologies and their role in enhancing the performance of current collectors were summarized, focusing on the innovative applications of CNWs in next-generation energy storage systems.

Keywords: current collectors; lithium-ion batteries; carbon-based coatings; carbon nanowalls

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1. Introduction

Lithium (Li)-ion batteries have become an indispensable energy storage technology, supplying power to various applications such as portable electronics, electric vehicles, and energy storage systems [1–3]. As shown in Figure 1A, the performance of these batteries is affected by their key components, i.e., the cathodes, anodes, electrolytes, and current collectors [4]. Among these, the current collector is essential for providing electrical pathways for electron transport and maintaining interfacial stability with the electrode active materials [5].

Traditionally, metal-based current collectors, such as aluminum (Al, for cathodes) and copper (Cu, for anodes), are extensively used in commercial batteries because of their high electrical conductivity, lightweight, and mechanical robustness. However, the key limitations of metal-based collectors include their susceptibility to electrochemical corrosion, which can degrade the material, reduce the collector efficiency, and increase the electrode–current collector interfacial resistance owing to the formation of passive layers, such as Al_2O_3 and CuO, during operation [6,7]. These limitations are intensified in high-energy-density and high-voltage battery systems. Moreover, repeated charge–discharge cycles cause other problems, such as delamination and mechanical fatigue, compromising both the efficiency and reliability of the system [8]. Research has been focusing on advanced surface modification and coating technologies to address these problems. Several techniques, such as carbon coatings, metals, and electrically conductive polymeric layers, are promising in enhancing the corrosion resistance of metal-based current collectors, reducing the interfacial resistance, and improving the mechanical adhesion of the electrode materials on the collector and overall performance of the collector [9–12]. Metal coatings, such as Au, Ag, and Sn, provide excellent lithiophilicity, enhancing uniform lithium deposition and mechanical stability while protecting against corrosion. However, their relatively high cost and density increase the overall weight of the battery system, limiting their applicability in lightweight and cost-sensitive designs. Polymer coatings offer excellent flexibility, lightweight properties, and strong adhesion to electrode materials, making them ideal for flexible and wearable battery applications. They enhance ionic conductivity and provide mechanical reinforcement, helping to mitigate stress during cycling. However, their relatively low electrical conductivity limits their performance in high-power applications. Additionally, polymers may degrade over time in harsh electrochemical environments, requiring careful selection and modification for long-term stability [13]. Carbon materials offer several unique advantages as coatings for current collectors compared to other materials such as metals and polymers. Carbon coatings provide enhanced adhesion between the active material and the current collector due to their rough surface and expansive area, reducing interfacial resistance through high conductivity. They protect against corrosion and electrolyte decomposition, improving device cyclability and preventing dendritic growth with their robust structure. Additionally, they stabilize metallic substrates by preventing direct solvent contact via the SEI layer, ensuring selective lithium-ion transfer [9].

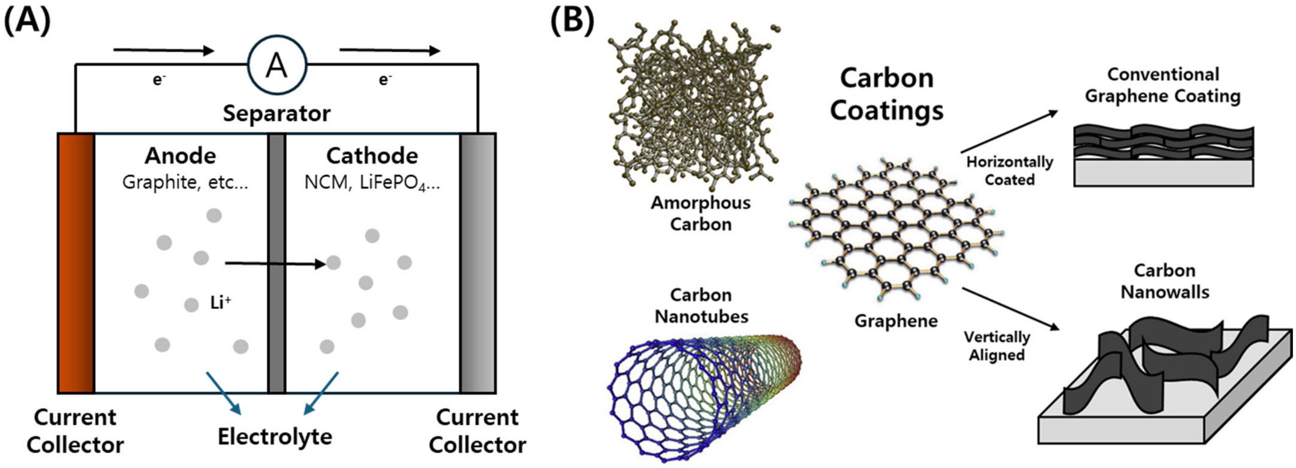

Figure 1. (**A**) Schematic of the structural configuration of a Li-ion battery during discharge. (**B**) Examples of various carbon-coating materials used for current collectors.

Existing carbon coatings, such as amorphous carbon, graphene, and carbon nanotubes (CNTs), exhibit various advantages and limitations depending on their specific applications and configurations [9]. Three-dimensional (3D) carbon nanowalls (CNWs) have recently emerged as an innovative coating material [14,15] (Figure 1B). CNWs comprise vertically aligned, two-dimensional (2D) graphene-like sheets, which give them a unique 3D structure with a high surface area, exceptional conductivity, and uniform interfacial contact [16], allowing them to stabilize the electrochemical reactions at the electrode–current collector interface, optimize the conductive pathways in the active materials, and extend the lifespan of the electrodes.

This mini-review reports the advancements in carbon coatings for improving current collectors in Li-ion batteries, focusing on 3D CNWs, and it starts with a discussion on conventional carbon coatings and progresses to explore the distinctive properties and application potential of CNWs.

2. Basic Requirements for Current Collectors in Li-Ion Batteries

Current collectors are a main component of Li-ion batteries, providing the electrical pathways necessary for the electron transfer between the active materials and the external circuit. As battery systems evolve to have higher energy densities and longer lifespans and to work under severe operating conditions, the requirements for current collectors also increase. These requirements can be broadly categorized into electrical, mechanical, chemical, and thermal requirements.

2.1. Electrical Requirements

The primary role of a current collector is to provide a low-resistance pathway for electron transport, which is critical for efficient battery operation throughout its lifespan [17] because high electrical conductivity minimizes ohmic losses. Resistance in a current collector can increase owing to surface oxidation, material degradation, or mechanical stress, leading to reduced efficiency, increased heat generation, and, ultimately, performance loss [18]. Thus, current collectors must maintain stable conductivity even under high current densities and repeated cycling.

Because advanced batteries are designed for rapid charging, current collectors must support high current flows while minimizing resistive heating and electrical losses. This is essential for maintaining optimal battery performance under demanding conditions [4,7]. Moreover, current must be uniformly distributed across the electrode surface to prevent localized hot spots [19], which can accelerate material degradation and reduce the battery lifespan. Therefore, appropriate material selection and geometry optimization for the current collectors are essential steps in the battery design. Maintaining optimal electrical performance in the current collector enhances the power delivery and contributes to the overall safety and reliability of battery systems.

2.2. Mechanical Requirements

Mechanical integrity is also a critical requirement for current collectors, particularly in applications involving high-capacity or high-stress electrodes. During the charge–discharge cycles, the volume of the electrodes often changes, generating mechanical stresses that can propagate to the current collector [20,21]. The current collector must exhibit appropriate mechanical properties to resist cracking, deformation, or delamination under these stresses [22,23]. For Li-ion batteries, high-purity Al and copper Cu foils are commonly used as current collectors due to their excellent electrical conductivity and compatibility with battery chemistries [24,25]. The mechanical strength of these foils is closely linked to their purity. Typically, high-purity Al exhibits tensile strengths ranging from approximately 91 MPa to 120 MPa [26], while Cu foils offer higher tensile strengths ranging from 245 MPa to 295 MPa [24]. However, to further enhance the mechanical robustness of current collectors, some studies have explored the use of alternative materials such as nickel (Ni) and titanium (Ti) [4,27]. These materials provide significantly higher tensile strength compared to Al and Cu, making them promising candidates for high-stress or high-capacity battery applications where structural integrity is critical.

Flexibility is important for current collectors, especially in flexible or wearable batteries. A flexible collector can accommodate the strain and stress caused by changes in the electrode volume without losing its structural integrity [28]. Moreover, strong current collector–electrode adhesion is crucial to prevent detachment, which can increase the interfacial resistance and reduce the battery capacity [23].

2.3. Chemical Requirements

Chemical stability is essential for current collectors to ensure a long lifespan, especially when used with reactive electrolytes and high-voltage electrodes. Conventional metal current collectors, such as Cu and Al, are susceptible to electrochemical corrosion and oxidation. For instance, Cu can dissolve under certain anodic conditions [29], and Al forms an insulating oxide layer at high voltages [30,31]. Electrochemical reactions, such as passive layer formation or corrosion, can increase the resistance within current collectors and degrade their conductivity, resulting in capacity loss or safety risks [32]. For advanced battery systems, chemical stability is particularly important because high-voltage and high-energy-density electrodes intensify chemical degradation. Therefore, a chemically stable current collector must be corrosion-resistant and maintain its conductivity under different operating conditions.

Innovative materials and coatings that increase the chemical resistance of conventional current collector materials are being explored to address these challenges. Utilizing alternatives, such as corrosion-resistant alloys [33] or advanced coatings on traditional metals [34–36], can substantially extend the lifespan and improve the reliability of current collectors. The electrode–current collector interface should be optimized to minimize the detrimental side reactions to enhance the overall efficiency and safety of the battery system.

2.4. Thermal Requirements

The thermal stability of current collectors is becoming increasingly important because Li-ion batteries are used in applications that require high power and fast charging, such as electric vehicles and energy storage systems [9,37]. During rapid charge–discharge cycles, current collectors are subjected to considerable thermal stress owing to localized heating and resistive losses. This localized heating can increase the temperature, leading to the thermal expansion of the current collector. This expansion may disrupt the interface with the electrode, increasing the resistance, reducing the efficiency of the battery, or even causing thermal runaway [38]. To prevent these issues, current collectors must effectively manage heat generation and dissipation. Thus, the current collector should have low thermal expansion coefficients and high thermal conductivity to maintain a stable interface and prevent material failure at elevated temperatures [39,40].

Another solution for improving the thermal performance of current collectors is using coatings to enhance heat dissipation and thermal management by inducing rapid heat transfer. These advanced coatings are often made from carbon-based materials such as amorphous carbon, graphene, and CNTs [9]. These carbon coatings are particularly effective because of their high thermal conductivity and ability to uniformly distribute heat across the collector surface [41], which helps maintain stable temperatures, reducing the risk of thermal runaway and minimizing material degradation during charge–discharge cycles.

The basic requirements for current collectors are summarized in Table 1.

Table 1. Summary of Basic Requirements for Current Collectors in Li-ion Batteries.

Requirement	Details
Electrical Requirements	High conductivity to minimize ohmic losses and maintain efficient charge transfer under repeated cycling.
Mechanical Requirements	Must resist cracking, deformation, and delamination during cycling; flexibility is essential for wearable applications.
Chemical Requirements	Resistant to corrosion and oxidation; maintain conductivity in reactive environments.
Thermal Requirements	Manage heat dissipation effectively to prevent thermal runaway and maintain structural integrity under high-power operations.

3. Overview of Carbon Coating Technologies

Each type of conventional carbon material, i.e., amorphous carbon, graphene, and CNTs (Figure 2), offers unique properties and advantages. This section explores the application methods of these coatings to current collectors, the coating effects on battery performance, and the challenges associated with each coating type.

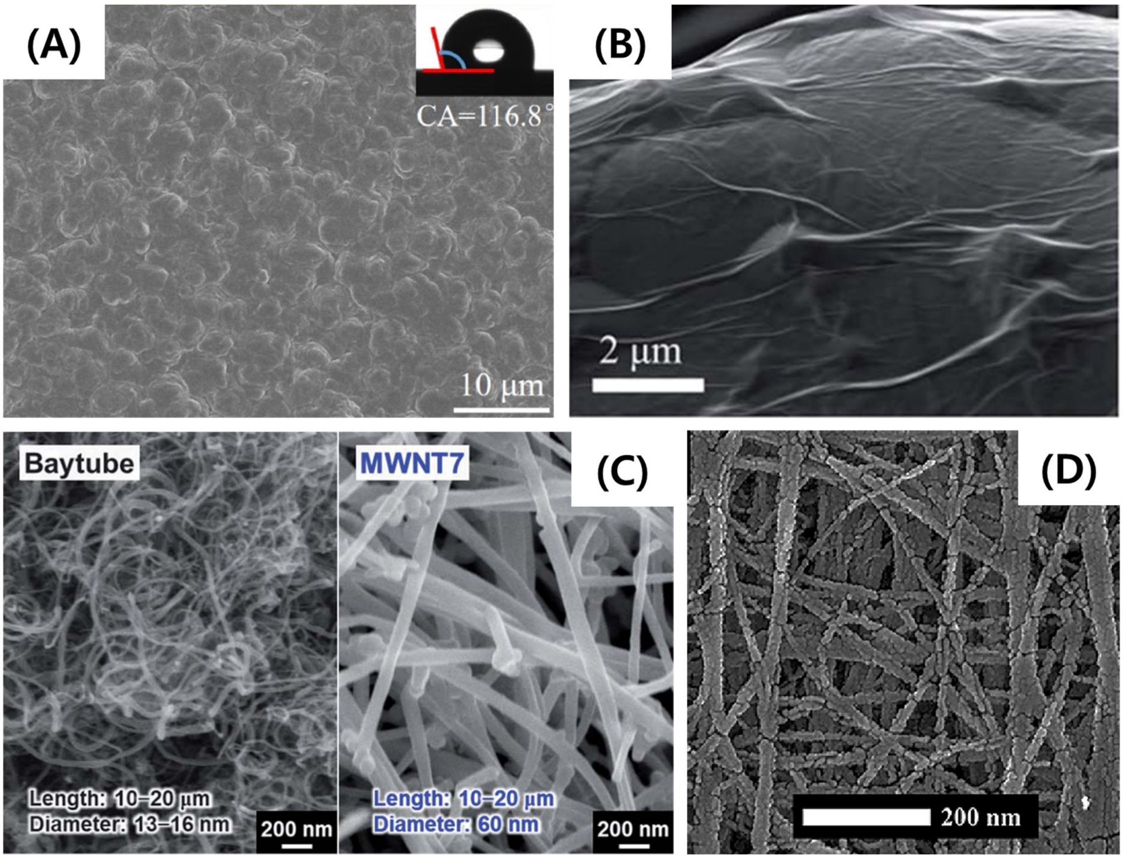

Figure 2. Scanning electron microscopy images of various carbon-based coatings and coated current collectors: (**A**) amorphous carbon-coated Cu, (**B**) graphene-coated Cu, (**C**) CNT-coated Al current collectors, and (**D**) Al-coated CNT current collectors.

3.1. Amorphous Carbon

3.1.1. Typical Coating Methods for Amorphous Carbon

Amorphous carbon coatings on current collectors are applied using various methods tailored to enhance the performance and stability of Li-ion batteries and related systems. A common coating technique is chemical vapor deposition (CVD), where methane (CH_4) is thermally cracked at high temperatures to deposit a conformal carbon layer on Al current collectors [42–44]. Magnetron sputtering enables the deposition of uniform and adherent carbon films on current collectors, providing a stable base layer for subsequent processes [45]. Carbonization methods involve treating porous aluminum foils with precursors like polyacrylonitrile (PAN), forming a protective carbon layer that enhances the structural integrity of the current collector [46]. These techniques demonstrate the versatility of amorphous carbon coatings in accommodating various current collector substrates and deposition methods.

3.1.2. Performance Enhancement with Amorphous Carbon Coating

Carbon coatings deposited using CVD can considerably improve the performance of current collectors across various battery chemistries. Wu et al. used a high-temperature

CVD process to coat Al current collectors for $LiFePO_4$ cathodes, achieving a 3–7× increase in power capability compared to uncoated Al. This considerably enhanced the cycling stability and capacity retention at high rates [43]. Wu et al. also used CVD to deposit conformal carbon coatings on both Al and Cu current collectors for $Li_4Ti_5O_{12}$ anodes. The coatings removed the native oxide layers, reduced the interfacial resistance, and improved the surface hydrophobicity, enhancing the rate performance up to 20C with minimal capacity degradation [44].

Carbon coatings deposited using sputtering techniques can substantially improve the current collector performance across various studies. Zhu et al. used magnetron sputtering for depositing amorphous carbon films on Cu current collectors for $Li_4Ti_5O_{12}$ anodes (Figure 2A), considerably reducing the contact resistance, enhancing the interface stability, and improving the charge collection efficiency. The modified electrodes exhibited a specific capacity of 162.51 mAh g^{-1} at a rate of 1C and maintained a capacity of 96.68% after 400 cycles. Moreover, the uniform and adherent carbon layer suppressed the interfacial degradation during cycling, ensuring consistent long-term performance [45].

Studies on polyacrylonitrile (PAN)-based carbon coatings have confirmed their effectiveness in enhancing current collector performance, particularly for high-rate and high-stability applications. Tong et al. developed a PAN-based carbonization method and used it to deposit PAN on a 3D porous Al foil and subsequently carbonize it at high temperatures to create a conductive carbon layer. The resulting carbon-coated porous Al demonstrated excellent rate performance, achieving capacities of 104 and 85 mAh/g at 2C and 20C, respectively. Moreover, it exhibited a high energy density of 204 Wh/kg at a power density of 3084 W/kg. The carbon layer reduced the effect of the volume changes during cycling, suppressing the surface reactions and maintaining the structural integrity of the collector. These effects enhanced the cycling stability and resulted in 93 mAh/g capacity retention after 1000 cycles [46].

The ability of hollow carbon nanosphere coatings to considerably enhance the performance of current collectors has also been confirmed. Zheng et al. proposed an innovative approach involving coating polystyrene spheres with carbon and thermally removing the template to create hollow carbon nanospheres. This unique structure facilitated uniform Li deposition on Cu current collectors, effectively suppressing dendrite formation. The battery systems containing these coatings exhibited excellent electrochemical performance, maintaining a Coulombic efficiency higher than 99% for 150 cycles. This structural stability ensured long-term and reliable charge–discharge performance [47].

3.2. Graphene Coatings

3.2.1. Typical Coating Methods for Graphene

Coating graphene on current collectors can effectively enhance the performance, stability, and safety of Li-ion batteries. The exceptional properties of graphene, including high electrical conductivity, mechanical strength, and chemical stability, can overcome numerous challenges faced by conventional current collectors [9], such as high interfacial resistance, poor adhesion with active materials, and susceptibility to corrosion over long-term cycling. Advanced techniques, including CVD, and innovative integration methods have been used to optimize the deposition of graphene coatings on conventional substrates such as Al and Cu.

3.2.2. Performance Enhancement with Graphene Coating

Li et al. applied a CVD-grown graphene-like structure to Al foil for high-rate Li-ion batteries. The coating reduced the interfacial resistance and improved the adhesion, enabling superior rate capabilities and stable cycling, even under demanding conditions [48].

Kim and Choi applied high-temperature CVD to deposit single-layer graphene on the Cu current collectors. This graphene coating considerably reduced the contact resistance and enhanced the adhesion with the graphite anodes. The graphene-modified collectors improved the cycle stability and mitigated the capacity fade associated with delamination and poor interfacial contact, which can be attributed to the superior electrical conductivity of graphene, facilitating efficient charge transfer between the active material and the collector and resulting in better rate performance [49]. Kim et al. used roll-to-roll CVD to apply a few-layer graphene coating on Al current collectors. This multilayer graphene structure enhanced the corrosion resistance, reduced the internal resistance, and enhanced the power capability. When paired with $LiCoO_2$ cathodes, the graphene-coated Al exhibited improved rate performance and stable cycling at high voltages. In addition, the coating created a strong barrier against electrolyte-induced degradation, ensuring long-term electrochemical stability [50]. Jiang et al. applied graphene coatings on Cu current collectors using low-pressure CVD. The graphene-coated collectors reduced the charge transfer resistance and enhanced the rate performance, maintaining stable cycling at high current densities, mitigating capacity degradation, and improving the adhesion between the current collector and electrode materials [51].

Hybridization approaches, which involve combining graphene with other materials, such as nanoparticles or doping elements, have been proposed to further enhance the properties of current collectors. Li et al. deposited a graphene coating with Au nanoparticles on Cu current collectors using CVD and thermal evaporation, respectively. This approach improved the conductivity and reduced the polarization of the current collector, achieving a specific capacity of 172.2 mAh/g at 4 A/g. The graphene layer coupled with Au nanoparticles effectively suppressed the electrolyte decomposition at the interface, enhancing cycling efficiency and stability [52]. Zhang et al. deposited nitrogen-doped graphene (N-graphene) coatings, with a nitrogen content of 1.31%, on 3D porous Cu current collectors using CVD. The N-graphene layer induced homogeneous Li-ion deposition, effectively suppressing dendritic growth and reducing hotspots during cycling. Using the coated collectors resulted in high Coulombic efficiency (>99%) of the battery system over extended cycling and enabled high areal capacities of up to 4 mAh/cm^2. Moreover, the graphene coating enhanced the mechanical stability, mitigating the structural degradation during repeated lithiation–delithiation processes. This study highlighted the potential of N-graphene in Li-metal batteries for improving safety, efficiency, and long-term performance [53].

In addition, novel approaches employing graphene in innovative structures have been developed. Yang et al. explored the use of graphene-coated Cu foam as a current collector (Figure 2B). This 3D porous structure induced uniform Li deposition, effectively suppressing dendrite formation, and the overall configuration achieved a Coulombic efficiency of 98.6% over 250 cycles at a current density of 0.5 mA/cm^2, as shown in Figure 3A,B. The graphene coating also provided excellent mechanical stability, mitigating the effects of the volume changes during cycling and ensuring reliable long-term performance [54].

These studies confirmed the considerable potential of graphene coatings for current collectors in Li-ion batteries.

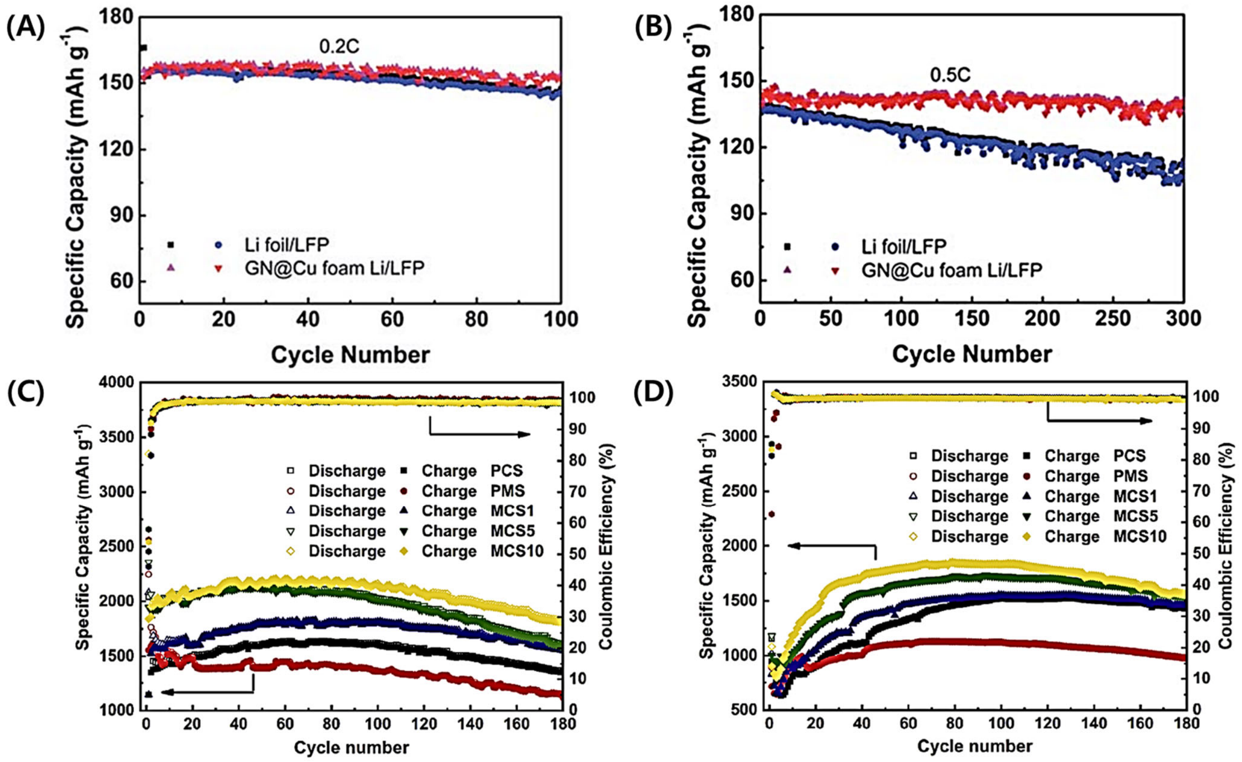

Figure 3. Cycling performance of graphene-coated Cu at (**A**) 0.2C and (**B**) 0.5C; MWCNT-coated Cu at (**C**) 1 A/g and (**D**) 3.5 A/g.

3.3. Carbon Nanotube Coatings

3.3.1. Typical Coating Methods for Carbon Nanotube

CNT coatings of current collectors have been extensively studied because of their potential to improve the performance, stability, and safety of Li-ion batteries. Owing to the exceptional electrical conductivity, high surface area, and mechanical flexibility of CNTs, they can mitigate several problems of conventional current collectors, such as high interfacial resistance, poor adhesion, and structural degradation [9]. These coatings enhance electron transport and mechanical stability and suppress dendritic Li deposition in Li metal batteries. Several deposition methods, including Langmuir–Schaefer deposition [55], spin coating [56], spray coating [57], electroplating [58], and CVD, have been explored to optimize CNT coatings for different applications. In addition, some studies have focused on using CNT films as standalone current collectors, eliminating the need for traditional metal substrates.

3.3.2. Performance Enhancement with Carbon Nanotube Coating

The Langmuir–Schaefer technique allows a uniform deposition of multiwalled CNTs (MWCNTs) onto metal substrates, ensuring excellent contact between the CNT layer and the collector surface. Rytel et al. applied this method to coat MWCNTs on Al current collectors. The resulting coatings considerably reduced the charge transfer resistance and enhanced the high-rate performance for $LiMn_2O_4$ cathodes, resulting in higher energy storage efficiency and cycling stability, which confirmed the effectiveness of this method for Li-ion battery applications [55]. The spin coating enables the formation of thin, uniform CNT layers when combined with other materials, such as metal nanowires or polymers. Zhao et al. combined MWCNTs with Cu nanowires to create a composite coating on a Cu foil, resulting in a high specific capacity (1845 mAh/g) for the silicon anodes at 3.5 A/g, with an 85.1% capacity retention after 180 cycles and improving the mechanical stability, which mitigated the effects of volume expansion during cycling [56]. Electroplating combines CNTs with metal matrices to enhance the mechanical and electrical properties of current collectors. Shimizu et al. used electroplating to coat MWCNTs onto Cu substrates, forming Cu-CNT composite films (Figure 2C). These coatings provided strong adhesion between the current collector and active material as well as efficient electron conduction pathways, achieving a reversible capacity of 1100 mAh/g for silicon anodes with 64% capacity retention after 100 cycles, as shown in Figure 3C,D [58].

In addition, CNT coatings have been used to increase the utility of new, nonmetallic substrates as current collectors, enhancing their properties to suit advanced battery designs. Spray coating offers a scalable and cost-effective method for depositing CNTs on flexible substrates. Ventrapragada et al. spray-coated CNTs on cellulose-based paper, forming lightweight and flexible current collectors. These paper-CNT electrodes, when paired with $LiFePO_4$ cathodes, achieved an energy density of 460 Wh/kg at a power density of 250 W/kg and maintained a gravimetric capacity of 150 mAh/g after 450 cycles, demonstrating superior gravimetric and volumetric performance compared to Al-based collectors [57].

Moreover, CNT films have been used as freestanding current collectors. Zhang et al. synthesized a 3D CNT sponge using CVD and coated it with an Al_2O_3 layer using atomic layer deposition, which provided a strong framework for Li deposition. The coated sponge suppressed dendritic growth and improved the coulombic efficiency (92.4% after 80 cycles at 1.0 mA/cm^2), which proves its high cycling stability [59]. Wang et al. developed cross-stacked, super-aligned CNT films modified with a thin metal layer using electron-beam deposition (Figure 2D). These CNT films exhibited low sheet resistance and improved charge transport characteristics. Standalone CNT current collectors enhanced the energy density and cycling stability of $LiFePO_4$ cathodes and $Li_4Ti_5O_{12}$ anodes [60].

These studies confirm that CNT coatings considerably enhance the performance of Li-ion batteries by reducing interfacial resistance, improving electron transport, and suppressing dendrite formation.

The details of conventional carbon coatings are summarized in Table 2.

Table 2. Summary of the amorphous carbon, graphene, and CNT-coated current collectors.

Coating Materials	Methods	Substrates	Notes	Ref.
Amorphous Carbon	CVD	Al	Enhanced cycling stability Reduced interface resistance	[43]
Amorphous Carbon	High-temperature CVD	Al and Cu	Reduced interface resistance Increased capacity at a high charge–discharge rate	[44]
Amorphous Carbon	Magnetron Sputtering	Cu	Improved capacity retention	[45]
Amorphous Carbon	Pyrolysis of resorcinol formaldehyde	Cu mesh	Reduced nucleation overpotential Enhanced Coulombic efficiency Prolonged cycle life	[61]
Amorphous Carbon	Carbonization of PAN	Porous Al	Enhanced mechanical stability Improved high-rate performance	[46]
Hollow carbon nanosphere	Template synthesis	Cu	Dendrite-free Li metal anodes Improved Coulombic efficiency	[47]

Table 2. *Cont.*

Coating Materials	Methods	Substrates	Notes	Ref.
Graphene	CVD	Al	Improved corrosion resistance Enhanced performance at high voltages	[48]
Graphene	High-temperature CVD	Cu	Improved adhesion with graphite Reduced contact resistance Enhanced cycle stability	[49]
Graphene	CVD	Al	Enhanced corrosion resistance Improved rate and cycle performance	[50]
Graphene	Low-pressure PVD	Cu	Reduced internal resistance Improved high-rate performance Cu oxidation prevention	[51]
Graphene/ Au NP	CVD/ Au deposition	Cu	Increased electron transport Enhanced Li$^+$ storage properties	[52]
Graphene-like Graphite	Reduction in GO	Al mesh	Suppressed cross-talk reactions Efficient anion intercalation Stable cycling performance.	[62]
N-doped Graphene	CVD	Porous Cu	Uniform Li deposition Improved cycling stability	[53]
Graphene	Reduction in GO	Cu foam	Dendrite-free Li deposition Improved Coulombic efficiency	[54]
Graphene/ Si	Reduction in GO	Cu mesh	High specific capacity Improved cycling stability Reduced charge transfer resistance	[63]
MWCNT	Langmuir-Schaefer Method	Al	Reduced charge-transfer resistance Improved power performance of the cathodes	[55]
MWCNT	Spin Coating	Cu Nanowire	Enhanced long-term cycling High specific capacity	[56]
MWCNT/Cu	Electroplating	Cu	Increased cycling stability and charge capacity Enhanced conductivity	[58]
MWCNT	CVD	Cu mesh	Enhanced specific capacity Improved cycling stability	[64]
MWCNT	Spray Coating	Paper	Higher capacity compared 17% areal capacity improvement 450 cycles stability	[57]

4. Emerging Carbon Coating Method: Carbon Nanowalls

CNWs are vertically aligned, graphene-like structures that create a unique 3D architecture on a substrate [15]. In contrast to traditional carbon coatings, which are typically planar or randomly oriented, CNWs consist of thin 2D graphene sheets that grow perpendicularly from the substrate surface. This vertical arrangement provides a high surface area and porosity, considerably enhancing the suitability of CNWs for applications requiring improved electron transport, material adhesion, and thermal dissipation. The edges of the graphene sheets in the CNWs are highly active, providing additional sites for chemical reactions, which allow their use in various electrochemical applications, including Li-ion batteries [15].

4.1. Formation Mechanism of Carbon Nanowalls

CNWs are prepared mainly using plasma-enhanced CVD (PECVD), and their formation mechanism involves several stages in which carbon precursors, such as methane (CH_4) or acetylene (C_2H_2), dissociate into reactive radicals under plasma conditions [16].

A schematic of the CNW formation mechanism is illustrated in Figure 4A. First, carbon radicals are adsorbed on the substrate surface, forming an amorphous carbon buffer layer, which then develops defects and dangling bonds due to ion bombardment. These defects and dangling bonds act as nucleation sites, around which migrating carbon species aggregate, forming nano-islands that lead to the growth of small graphene nanosheets. These nanosheets initially grow in a random orientation. However, their edges then become preferential sites for further deposition of reactive carbon species [16,65].

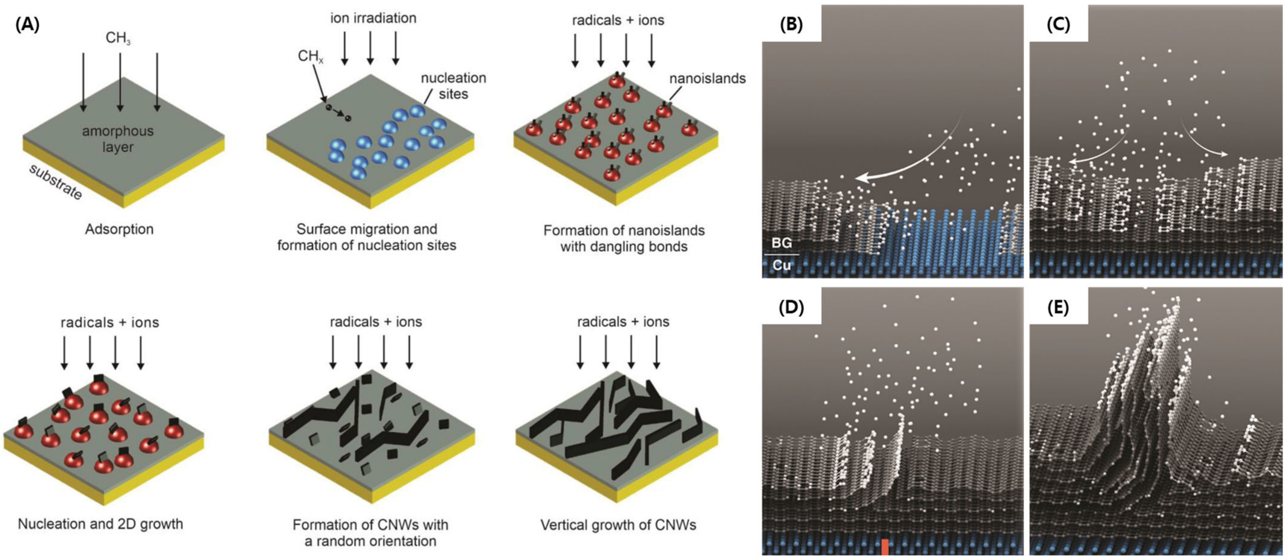

Figure 4. (**A**) Schematic representation of the carbon nanowall (CNW) formation mechanism, illustrating key stages such as adsorption, nucleation, and vertical growth. Growth evolution of CNW from the Cu substrate interface (**B**) initial formation of basal graphene (BG0), (**C**) coalescence of the BGs, (**D**) vertical bending of one BG domain, and (**E**) formation of vertical CNWs. White arrows in (**B**,**C**) indicate the flow of hydrocarbon plasma.

Vishal et al. utilized electron energy loss spectroscopy to investigate the growth mechanism of the CNW, as shown in Figure 4B–E. The process begins with the formation of basal graphene (BG) layers, which are horizontally aligned on the substrate. These layers develop defects and bend at coalescence points, which serve as nucleation sites for vertical growth. As the process continues, charged carbon species from the plasma interact with the edges of the basal planes, leading to vertical growth characterized by a mixture of graphitic and turbostratic carbon structures [66].

Ion bombardment under plasma conditions is essential because it determines the vertical alignment of the graphene sheets by creating stress and activating the edge sites. This alignment is further affected by the substrate properties such as surface roughness and electrical conductivity. Moreover, high-energy ions control the migration and diffusion of carbon species, ensuring the vertical stacking of the layers and the characteristic nanowall structure. Temperature and plasma power considerably affect the quality and morphology of CNWs because their crystallinity and uniformity are enhanced with the increase in temperatures and optimization of the ion flux [65,67].

Tailoring the deposition parameters, such as the gas flow rates and bias voltages, enables control over the CNW thickness, height, and interwall spacing. Using some techniques, such as high-voltage nanosecond pulses, during PECVD introduces additional control by modulating the ion flux, which leads to precise adjustments in the nucleation density and growth rate, allowing the synthesis of sparse or dense CNW arrays depending on the application requirements [65].

4.2. Fundamental Properties of CNWs

As shown in Figure 5A,B, CNWs are vertically oriented graphene-like structures with high aspect ratios, nanoscale thickness (5–30 nm), and variable heights ranging from hundreds of nanometers to several micrometers depending on the deposition conditions [68]. They exhibit a high specific surface area owing to their interconnected structure, making them ideal for energy storage and catalysis applications [65,67]. Vishal et al. confirmed through TEM analysis that CNWs possess a structure highly similar to graphene (Figure 5C). Specifically, the CNWs are composed of walls structured with graphitic carbon (Figure 5D) and turbostratic carbon (Figure 5E) layers formed on basal graphene. The graphitic carbon regions are primarily defect-free, while the turbostratic carbon regions exhibit a higher degree of defects and planar mismatches [66].

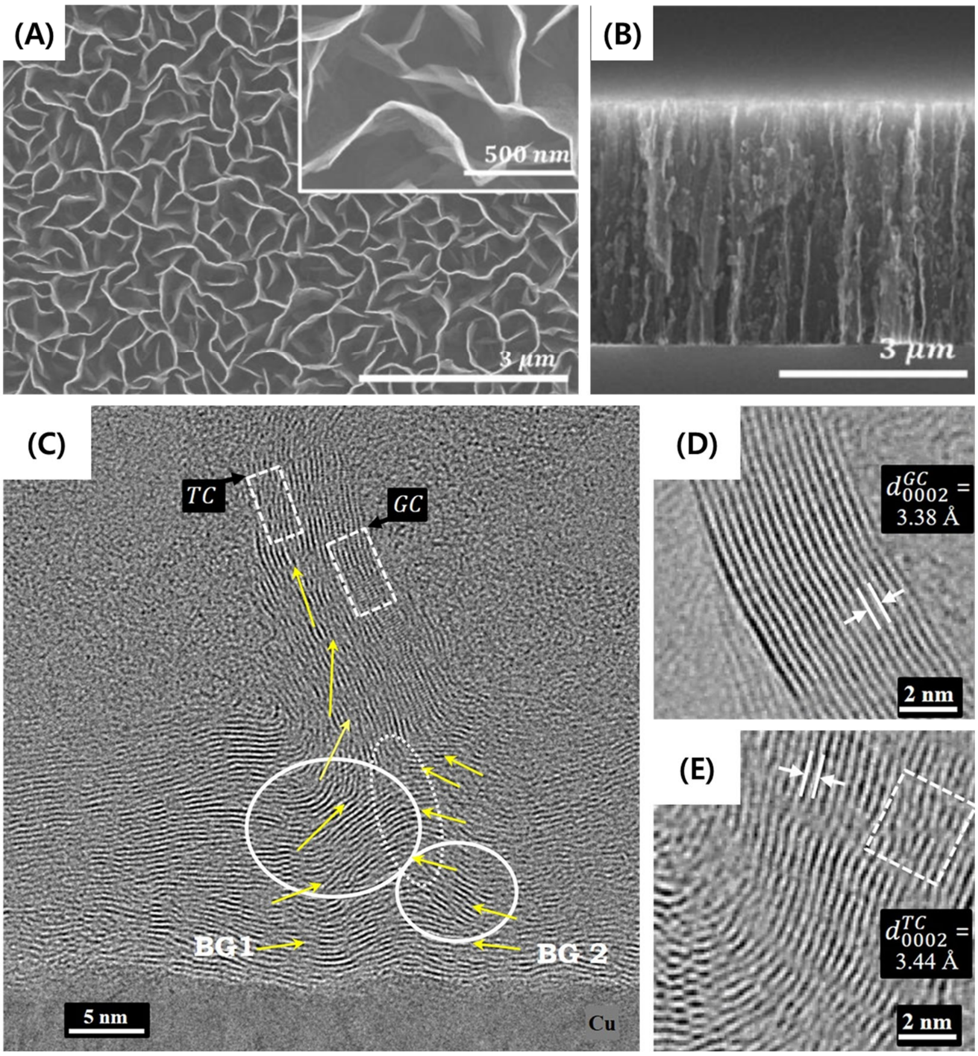

Figure 5. Scanning electron microscopy images of the CNW layer; (**A**) top and (**B**) cross-sectional view. High-resolution transmission electron microscopy (HRTEM) images of (**C**) the CNW. High-magnification HRTEM image showing (**D**) graphitic carbon and (**E**) turbostratic carbon in the vertical section of the CNW. Yellow arrows in (**C**) indicate the defects in the CNW layer.

The electrical conductivity of CNWs is determined based on their graphitization and defect density. According to the Raman spectroscopy results, CNWs grown for longer durations exhibited a higher I_G/I_D ratio, indicating greater graphitization and resulting in improved conductivity. Their sheet resistance decreases with the increase in the growth times, with values as low as 600 $\Omega/\square$ compared to 2000 $\Omega/\square$ for shorter growth times [65,67,69]. These properties enhance the electrical conductivity of CNWs in electrochemical systems [69,70]. CNWs exhibit high thermal stability with an intrinsic thermal conductivity of approximately 300 W/mK, which is comparable to that of other carbon-based nanomaterials such as CNTs, making CNWs suitable for high-temperature applications [65,69]. Moreover, CNWs maintain their structural integrity under stress owing to their strong vertical alignment and interwall connectivity. These properties also enhance mechanical flexibility [67,69].

4.3. Enhancing Performance of Current Collectors Using CNWs

CNWs have emerged as a transformative coating material for advanced current collectors in various energy storage systems, including Li-ion, Li metal, and Li–sulfur batteries. Their unique, vertically aligned, graphene-like structures provide them with high surface area as well as excellent electrical conductivity and thermal properties, enhancing the performance and durability of current collectors. Research across various battery chemistries highlights the contributions of CNWs to the reduction in interfacial resistance, improvement of cycling stability, and mitigation of critical problems such as dendrite formation and thermal management.

Liu et al. used PECVD to deposit CNW coatings on Cu current collectors for Li-ion batteries. The CNW-coated collectors considerably improved the interfacial contact with the graphite anodes, reducing the interfacial resistance from 155.9 Ω (commercial carbon-coated Cu) to 14.1 Ω, substantially enhancing the rate performance and resulting in a discharge capacity of 190 mAh/g at 3C compared to 160 mAh/g for commercial carbon-coated Cu and 90 mAh/g for bare Cu. The CNW structure considerably stabilized the cycling performance, retaining capacities higher than 190 mAh/g after 100 cycles (Figure 6A,B) [14].

Ichikawa et al. used nanosecond-pulsed PECVD to synthesize sparse, vertically aligned CNWs on silicon substrates. This tailored density and alignment of the CNWs induced uniform ion distribution during cycling. Moreover, the vertically oriented structure promoted efficient electron mobility, considerably improving capacity retention and cycling stability under high-rate conditions. These advancements demonstrated the potential of CNWs in applications requiring high energy densities, such as electric vehicles and portable electronics [65]. Wang et al. demonstrated that hierarchical graphene (hG) coatings grown on Al current collectors considerably improved their interfacial properties. The hG structure, consisting of a planar graphene basal layer and vertically aligned branches, increased the electrochemically active surface area from 2.5 cm^2/cm^2 (bare Al) to approximately 32.2 cm^2/cm^2, which reduced the interfacial resistance by over two orders of magnitude compared to bare Al, reducing its contribution to the total electrode resistance. Moreover, the adhesion strength between the electrode material and the current collector reached 20.50 N/m owing to the increase in the van der Waals interactions between the electrode and the current collector. Electrochemical tests demonstrated the superior performance of the battery systems comprising hG-coated Al current collectors. In half-cell tests using $LiFePO_4$ electrodes, the hG-coated collectors exhibited specific discharge capacities of 43 mAh/g at 20C, whereas bare Al failed at 4C. Full-cell tests using graphite anodes indicated that the system with hG-coated Al current collectors exhibited a power density of approximately 5300 W/kg and an energy density of approximately

220 Wh/kg, outperforming the system comprising bare Al (3700 W/kg and 210 Wh/kg, respectively) [71].

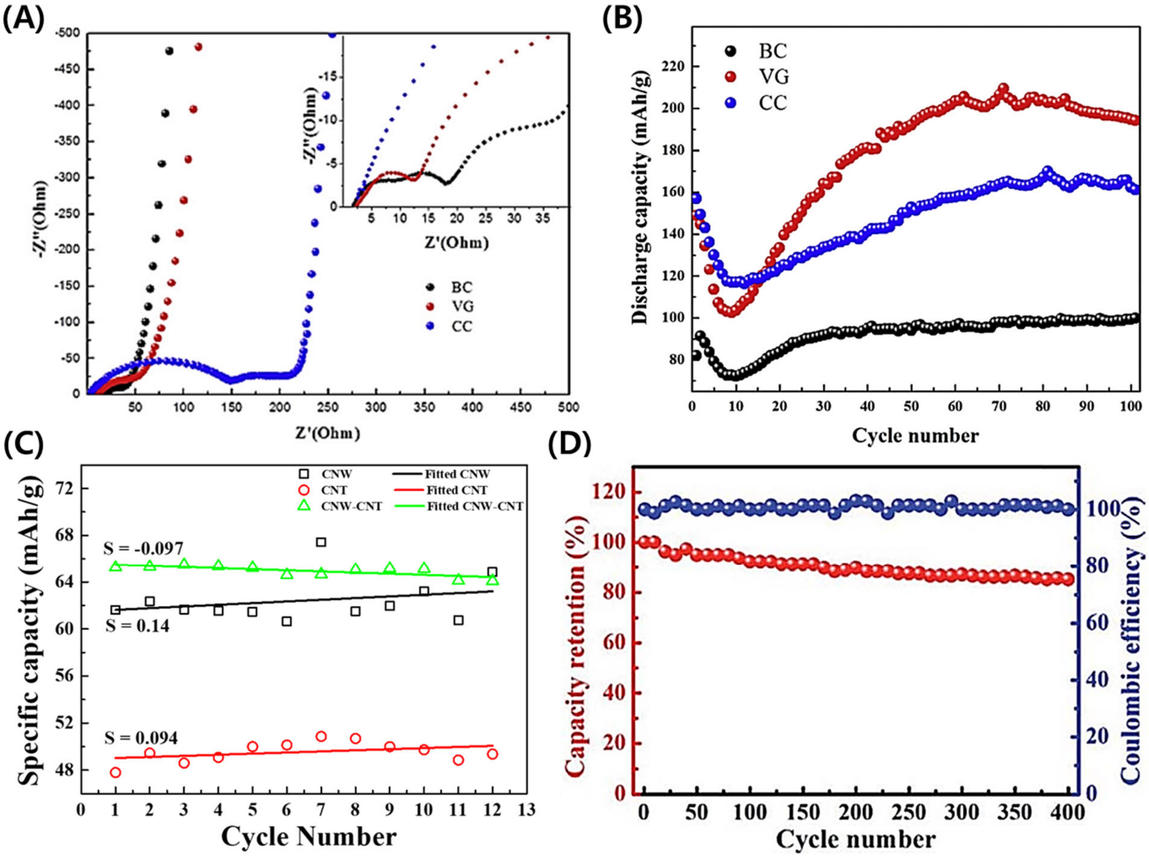

Figure 6. (**A**) Electrochemical impedance spectroscopy and (**B**) discharge capacity of CNW-coated Cu. (**C**) Specific capacity of the CNW-CNT-Cu and (**D**) capacity retention and Coulombic efficiency of CNW-coated V_2O_5 nanosheets.

In addition to using them alone, CNWs have been successfully integrated with other materials to further enhance their performance. Kim et al. introduced a novel composite structure combining CNWs and carbon nanofibers (CNFs) to enhance the performance of Li-ion battery anodes. CNWs were synthesized using methane and hydrogen via microwave PECVD, and CNFs were fabricated via electrospinning followed by thermal treatment. The CNF/CNW composite exhibited a high surface area, low resistance from CNWs, and high flexibility and durability from CNFs. Raman analysis revealed reduced defect structures (low I_D/I_G ratio) and enhanced graphitization in the composite compared to CNWs or CNFs alone. The composite anode exhibited superior electrochemical properties. The cyclic voltammetry results indicated that the oxidation–reduction reactions were improved, and the charge-transfer resistance was calculated using electrochemical impedance spectroscopy to be 93 Ω, which is considerably lower than that (155 Ω) of CNWs alone. The coulombic efficiency was also improved, reaching 91% during cycling. The CNF/CNW structure maintained stable cycling over 1000 cycles, confirming its enhanced durability and structural integrity at high current densities. Electrochemical performance tests indicated that the CNF/CNW composite achieved a high current density (2 A/g) with stable capacity retention, outperforming traditional carbon-based materials. The integration of CNFs with CNWs mitigated the problem of structural collapse in vertically aligned CNWs due to repeated cycling, ensuring long-term stability [72]. Lee et al. used CNWs and CNTs as the coating materials for the anode. CNWs were synthesized on Cu current collectors using PECVD, and CNTs were deposited on CNWs using a water dispersion method. Electrochemical tests demonstrated the higher specific capacity of the CNW/CNT composite (64.94 mAh/g) compared to those of CNWs (62.4 mAh/g) or CNTs (49.54 mAh/g) alone, which can be attributed to the synergistic effects of the two carbon structures (Figure 5C) [68]. He et al. designed a fiber-shaped Zn-ion battery comprising a hierarchical cathode composed of ultrathin V_2O_5 nanosheets grown on a 3D nitrogen-doped porous CNW framework. The CNW framework, synthesized using metal–organic frameworks supported on CNT fibers, offered high porosity and conductivity, inducing efficient ion diffusion and electron transport. V_2O_5 nanosheets considerably increased the active material loading, resulting in an exceptional rate performance and a volumetric capacity of 457.5 mAh cm^{-3} at 0.3 A cm^{-3}, of which 47.5% was retained at 30 A cm^{-3}. The strong bonding between the CNW framework and V_2O_5 minimized the contribution of inactive materials by eliminating the negative effects of “dead mass” typically caused by binders and conductive additives. This optimization facilitated efficient electron transport and enhanced ion diffusion, resulting in a capacity retention of 85.3% over 400 cycles and a nearly 100% coulombic efficiency (Figure 5D). This system outperformed numerous aqueous energy storage devices, achieving a volumetric energy density of 40.8 mWh cm^{-3} and a power density of 5.6 W cm^{-3}, which shows the system’s potential for durable, high-performance wearable energy storage devices [73].

CNWs exhibit exceptional thermal conductivity, which is an essential property in high-power energy storage applications. Mishra et al. (2016) investigated the thermal properties of CNW, indicating their potential in thermal management applications for nanoelectronics and energy devices. The CNW synthesized using electron cyclotron resonance-plasma enhanced chemical vapor deposition (ECR-PECVD) exhibited a thermal conductivity of approximately 250 W/mK at room temperature. Although this value is lower than that of single-layer graphene (4800–5300 W/m·K), it is considerably higher than that of graphene nanofoams (~11 W/m·K), rendering VGNs an efficient heat dissipation material. A monotonic redshift in the G-band was observed in the temperature-dependent Raman spectra of VGNs, with a temperature coefficient of -1.47×10^{-2} cm^{-1}/K, demonstrating the sensitivity of the VGNs to thermal changes and efficient phonon interactions. The interconnected and uniform structure of VGNs induces effective heat conduction, which prevents localized hotspots in high-power applications. Furthermore, VGNs demonstrated remarkable mechanical resilience under extreme conditions, maintaining their structural integrity under pressures of up to 40 GPa. The combination of thermal conductivity, structural stability, and sensitivity to temperature variations makes VGNs a versatile material for several advanced technologies, such as Li-ion batteries and nanoelectronics, in which efficient heat dissipation and durability are critical to maintaining device performance and safety [74].

Table 3 summarizes various CNW coatings for current collector applications.

Table 3. Summary of the CNW-coated current collectors.

Coating Materials	Methods	Substrates	Notes	Ref.
CNW	PECVD	Cu	High-rate capability Enhanced charge–discharge performance	[14]
CNW	PECVD	Si	Improved wall-to-wall distance control	[65]
CNW	PECVD	Al	Optimized electron pathway Reduced interfacial resistance Enhanced power performance	[71]

Table 3. *Cont.*

Coating Materials	Methods	Substrates	Notes	Ref.
CNW	PECVD	Ni foam	Reduced charge overpotential Improved electron transfer	[75]
CNW	Microwave plasma CVD (MWCVD)	Stainless Steel	Enhanced anode-specific capacity Better electrolyte integration	[76]
CNW/CNFs	PECVD	Al	Enhanced charge–discharge Enhanced cycling stability	[72]
V_2O_5/CNW	High-voltage PECVD	Carbon Nanofiber	Enhanced structural stability Reduced interfacial resistance	[73]
CNW/Cu	PECVD	Cu	Enhanced electronic conductivity Reduced impedance	[77]
CNW/Si	PECVD	Cu	High specific capacity Mitigated volume expansion Prolonged cycle life.	[77]
CNW/Si	PECVD	Ti	High specific capacity Improved cycling stability	[78]
CNW/Si	PECVD	Ni foam	Enhanced discharge capacity Improved capacity retention Stable cycling performance	[79]
CNW/CNT	PECVD/ Drop casting	Cu Foil	Enhanced electrochemical performance	[68]

4.4. Challenges and Future Directions

The commercialization of CNWs is limited by several challenges, such as cost and scalability. The PECVD process, which is an essential step in the CNW synthesis, requires high energy input and substrate temperatures of 600–800 °C [16]. Such high-temperature processes not only limit the choice of materials but also lead to increased processing costs. To address this issue, alternative methods, such as lower-temperature PECVD processes, have been explored. Kulczyk-Malecka et al. synthesized vertically aligned graphene (VG) using microwave-assisted PECVD at low temperature (<300 °C) without external substrate heating, enabling VG growth on temperature-sensitive substrates such as polymers. They obtained highly graphitized, vertically aligned structures with thicknesses up to 596 nm under optimized conditions, i.e., CH_4:H_2 ratio = 1:8, microwave power = 2.5 kW, and controlled plasma distances. Raman and scanning electron microscopy analyses confirmed reduced defects and improved crystallinity. Moreover, the electron transfer kinetics were enhanced, and larger electroactive areas were observed, making this process suitable for flexible electronics and energy storage applications [80].

Another key challenge is the scalability of CNW synthesis. CNW synthesis has been demonstrated at the laboratory scale, and the transition to industrial-scale manufacturing remains complex. Scaling the PECVD processes for large-area production is difficult because several dimensions of CNWS, such as the wall height and spacing, must be precisely controlled to achieve the required morphology [65]. Wang et al. demonstrated the feasibility of roll-to-roll PECVD systems for hierarchical graphene coatings. However, it is still under development for CNWs. The ability to produce high-quality, large-area CNWs without sacrificing morphological precision is critical for commercial viability [81]. Another method to overcome this challenge is to increase the deposition rates. High deposition rates benefit scale-up by reducing production time and costs and enabling faster market responsiveness, making large-scale manufacturing more efficient and economical. Ji et al. developed a helicon wave plasma chemical vapor deposition (HWP-CVD) method that achieved an exceptionally high growth rate (0.26 μm/min), which was considerably faster than that of conventional PECVD processes, enabling a rapid synthesis of vertically aligned graphene nanosheets. Linear graphene growth with time was observed, allowing precise control over the morphology while maintaining high-quality graphene layers [82]. Therefore, interdisciplinary collaboration is required to address the limitations of CNW synthesis, scalability, and application-specific customizations through novel deposition methods, material integration, and sustainability practices to achieve their full potential in next-generation energy storage systems.

CNWs are not only pivotal for enhancing battery performance but also hold significant potential for multifunctional applications, particularly in electromagnetic interference (EMI) shielding. This capability is especially crucial in advanced fields like electric vehicles (EVs), where ensuring the seamless operation of electronic systems is paramount. In EVs, EMI shielding plays a vital role in maintaining the reliability and safety of sensitive electronics. During battery charging and discharging cycles, a wide spectrum of electromagnetic waves is generated, ranging from low frequencies associated with power conversion to high frequencies linked to electronic systems. Without effective shielding, these waves can interfere with vehicle electronics, degrade the performance of battery management systems (BMS), and compromise overall safety. While traditional metallic casings, such as those made from aluminum, provide effective EMI shielding, the shift towards lightweight alternatives like carbon-based composites necessitates equivalent or superior shielding solutions [83]. A notable study by Wang et al. highlighted the effectiveness of vertical graphene nanowalls (VGNs), a specific structural form of CNWs, in EMI shielding applications. VGNs were synthesized via PECVD on Cu and Ni foam substrates. The unique hierarchical and porous structure of the VGNs provided enhanced internal reflection and absorption of electromagnetic waves, resulting in significantly improved shielding performance. Specifically, VGN-coated Cu foam demonstrated shielding effectiveness (SE) of 81.7 dB in the X-band frequency range (8.2–12.4 GHz), representing a substantial 25.9% improvement over bare Cu foam. Similarly, VGN-coated Ni foam achieved an SE of 74.8 dB, underscoring the versatility of the material across different substrates. These results demonstrate the ability of VGNs to offer superior EMI shielding performance, attributed to their high surface area, conductivity, and structural efficiency [84]. By leveraging these properties, CNWs emerge as a lightweight and efficient solution for EMI shielding, holding promise for next-generation applications in EVs and other advanced technologies.

5. Conclusions

Carbon-based coatings, including amorphous carbon, graphene, and CNTs, have emerged to enhance current collectors in Li-ion batteries by addressing problems such as corrosion, interfacial resistance, and mechanical instability. Among these, CNWs are particularly notable due to their unique vertically aligned structures, offering high surface area, electrical conductivity, and interfacial stability. CNWs are composed of thin, vertically aligned graphene-like sheets that create a three-dimensional architecture. This structure not only enhances electron transport but also facilitates uniform ion distribution, making CNWs particularly effective in reducing interfacial resistance and mitigating issues like dendrite formation in Li-metal batteries. CNWs have demonstrated exceptional resistance, capacity retention, and cycling stability, especially in Li-ion and Li-metal batteries. Their ability to stabilize the electrode-current collector interface is attributed to their large surface area and high conductivity, which optimize charge transfer and mechanical adhesion. Furthermore, the high thermal stability and excellent heat dissipation properties of CNWs enable their use in high-power applications, where thermal management is critical to battery safety and performance.

However, several challenges remain in CNW synthesis, including industrial scalability. Novel methods, such as low-temperature PECVD and roll-to-roll production methods, were introduced to achieve cost-effective and scalable CNW synthesis, enabling their use with heat-sensitive substrates and in large-scale manufacturing. Moreover, CNWs hold significant potential for multifunctional applications. They demonstrate exceptional EMI shielding properties, attributed to their high surface area, electrical conductivity, and vertically aligned architecture, underscoring their suitability for lightweight and high-performance EMI shielding solutions.

Collectively, these characteristics suggest that CNWs hold immense potential as valuable materials in the field of current collectors for Li-ion battery applications.

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