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Review

Review of Flexible Supercapacitors Using Carbon Nanotube-Based Electrodes

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Abstract: Carbon nanotube (CNT)-based electrodes in flexible supercapacitors have received significant attention in recent years. Carbon nanotube fiber fabrics (CNT-FF) have emerged as promising materials due to their high surface area, excellent conductivity, and mechanical strength. Researchers have attempted to improve the energy density and rate performance of CNT-FF supercapacitor electrodes through various strategies, such as functionalization with conductive materials like MnO_2 nanoparticles and/or incorporation of graphene into them. In addition, the utilization of CNTs in combination with thin metal film electrodes has also gained widespread attention. Research has focused on enhancing electrochemical performance through functionalizing CNTs with conductive materials such as graphene and metal nanoparticles, or by controlling their morphology. This review paper will discuss the recent developments in supercapacitor technology utilizing carbon nanotube-based electrodes, including CNT fiber fabrics and CNTs on thin metal film electrodes. Various strategies employed for improving energy storage performance and the strengths and weaknesses of these strategies will be discussed. Finally, the paper will conclude with a discussion on the challenges that need to be addressed in order to realize the full potential of carbon nanotube-based electrodes in supercapacitor technology.

Keywords: carbon nanotube; supercapacitor; flexible; electrode; composite; wearable

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1. Introduction

Energy storage is a crucial aspect of modern society and is necessary to meet the growing demand for renewable and clean energy [1–8]. Supercapacitors, also known as electrochemical capacitors, are energy storage devices that have gained significant attention in recent years due to their high energy density and fast charging/discharging capability [9–15]. The development of new electrode materials has been an area of active research in supercapacitor technology, as this leads to improvements in energy and power density (Figure 1) [16–22].

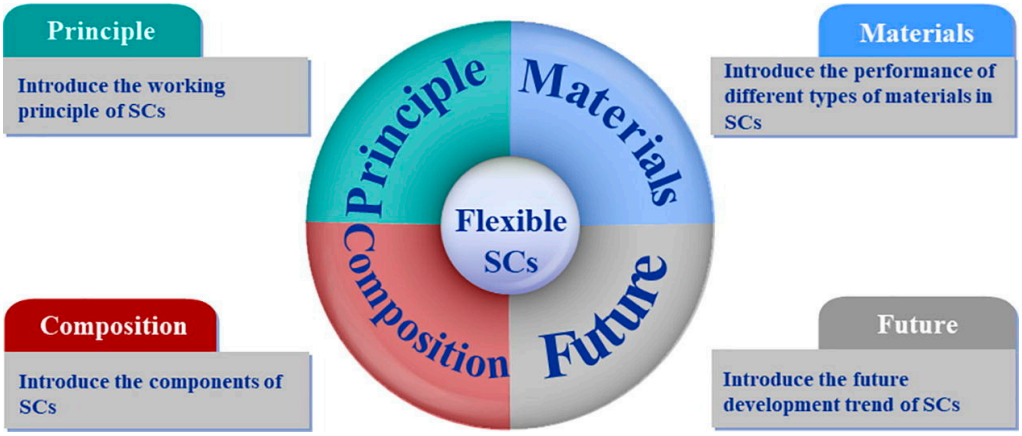


Figure 1. Main issues in flexible supercapacitors (SCs). Reproduced with permission from ref. [23]. Copyright 2015 Elsevier.

Supercapacitors are generally divided into two categories: pseudo capacitors and electronic double layer supercapacitors (EDLCs) [24,25]. Pseudo capacitors accompany redox reaction at the surface of electrode. Otherwise, having non-faradaic reaction, EDLCs accumulate charges between the electrode and electrolyte by forming a double layer, so carbon materials such as carbon nanotubes (CNTs), carbon black (CB), and graphene are beneficial for a large interfacial area due to their porosity. Among them, CNTs have high electrical conductivity, specific surface area, chemical/thermal stability, and good contact with the electrolyte [16,26–33]. Carbon materials can also be formed in various dimensions such as 1D carbon fiber, 2D carbon film, and 3D carbon composite. Therefore, CNTs with those unique properties are expected to serve as EDLCs electrode materials in various forms of supercapacitor.

Synthesis and characterization of new materials are very important topics for several applications including supercapacitors [34–38]. One such promising material is carbon nanotube fiber fabrics (CNT-FF), which have emerged as a candidate for supercapacitor electrodes due to their combinatorial merits in high surface area, excellent conductivity, and mechanical strength [10,39]. CNT-FF is a type of fabric-like structure made by weaving together carbon nanotubes, which have high surface area and electrical conductivity. These unique properties of CNTs make them an excellent option for supercapacitor electrodes as they provide a large surface for charge storage and fast charging/discharging. Additionally, the high mechanical strength of CNT-FF makes it suitable for flexible devices, reducing the risk of mechanical degradation during cycling.

The focus of the research into the use of CNT fiber fabrics as supercapacitor was on boosting energy density and rate performance. Researchers have employed various strategies such as functionalizing CNT fiber fabrics with conductive materials such as graphene and metal nanoparticles, and incorporating these materials into hybrid CNT-based fiber fabrics. These efforts have resulted in impressive energy storage performance, including specific capacitance values of up to 402 F/g and energy densities of up to 28 Wh/kg. However, it is important to note that these studies were limited in their comparison with other electrode materials and their lack of information on the long-term stability and durability of CNT-FF electrodes, which are critical factors for practical applications.

Another promising approach in supercapacitor technology is the utilization of CNTs in combination with thin metal film electrodes. The combination of high surface area, electrical conductivity, and stability of CNTs with thin metal film electrodes has gained widespread attention in recent years. Research has been conducted to optimize the energy density and rate performance of supercapacitors utilizing this combination. One strategy for enhancing performance has involved functionalizing the CNTs with conductive materials, such as graphene and metal nanoparticles, while another method has involved controlling the

morphology of the CNTs on thin metal films. These efforts have resulted in improved energy storage performance, including specific capacitance values of up to 300 F/g and improved rate performance.

This review paper will discuss the recent developments in supercapacitor technology utilizing carbon nanotube-based electrodes, including CNT fiber fabrics and CNTs on thin metal film electrodes. The various strategies employed for improving energy storage performance and the strengths and weaknesses of these strategies will be discussed. Finally, the paper will conclude with a discussion of the challenges that need to be addressed to realize the full potential of carbon nanotube-based electrodes in supercapacitor technology.

2. Flexible Supercapacitor with Various Types of CNT-Based Electrodes

2.1. CNT-FF-Based Electrodes

Supercapacitors, also known as electrochemical capacitors, have gained significant attention as energy storage devices in recent years due to their high energy density and fast charging/discharging capability. One of the key research areas in supercapacitors is the development of new electrode materials that can improve their energy and power density. CNT-FFs have emerged as promising materials for supercapacitor electrodes due to their unique combination of merits in high surface area, excellent conductivity, and mechanical strength.

CNT-FF is a type of fabric-like structure made by weaving carbon nanotubes together. The high surface area of the CNTs makes them excellent supercapacitor electrodes due to their large surface for charge storage. Their electrical conductivity is also much higher compared with other carbon-based materials, making fast charging and discharging reactions possible. Additionally, CNT-FF have a high mechanical strength, making them suitable for flexible devices and reducing the risk of mechanical degradation during cycling. Research into using CNT-FF as supercapacitor electrodes has been largely focused on boosting the energy density and rate performance. For example, in the work by Yang et al., researchers fabricated CNT-FF using a solution-blowing method as shown in Figure 2a, which resulted in a remarkable specific surface area of 15.995 m^2/g [12]. When used as supercapacitor electrodes, the CNT-FF displayed impressive energy storage performance, including a specific areal capacitance of up to 353 mF/cm^2 and an energy density of up to 247 $\mu W/cm^2$. Strengths of this study include the high surface area of CNT-FF and the impressive energy storage performance. However, one potential weakness could be the scalability of the solution-blowing method used in creating the CNT-FF. Liang, Y., et al. knitted CNT yarns using a weaving machine followed by heating at 400 °C for 60 min under air condition [39]. After oxidation, the CNT-FF is immersed into HNO_3 and H_2SO_4 solution and magnetically stirred at 45 °C for 60 min. Then, after cleaning with deionized water, vacuum drying is performed at 60 °C. This method exhibited improved cyclic voltammetry (CV) curves and longer discharge time compared with bare, oxidized, and acidized CHT-FF. High surface area and an additional oxygen-containing functional group by oxidation and acid treatment caused the increased capacitance of 1033 mF cm^{-2} at a current density of 2 mA cm^{-2}. In addition, the activated CNT-FF had excellent cyclic capacitance retention for 10,000 cycles. This weaving process makes it easy to fabricate a large-scale electrode for flexible supercapacitor. Another work by Zhang et al. involved functionalizing CNT-FF with MnO_2 nanoparticles to form hybrid CNT-MnO_2 fiber fabrics (Figure 2b) [14]. The results showed improved electrochemical performance, with a specific capacitance reaching up to 231.3 F/g and an energy density of up to 86 nWh/cm. Strengths of this study include the significant improvement in electrochemical performance achieved through functionalization with MnO_2 nanoparticles. However, one potential weakness could be the cost and scalability of synthesizing the MnO_2 nanoparticles.

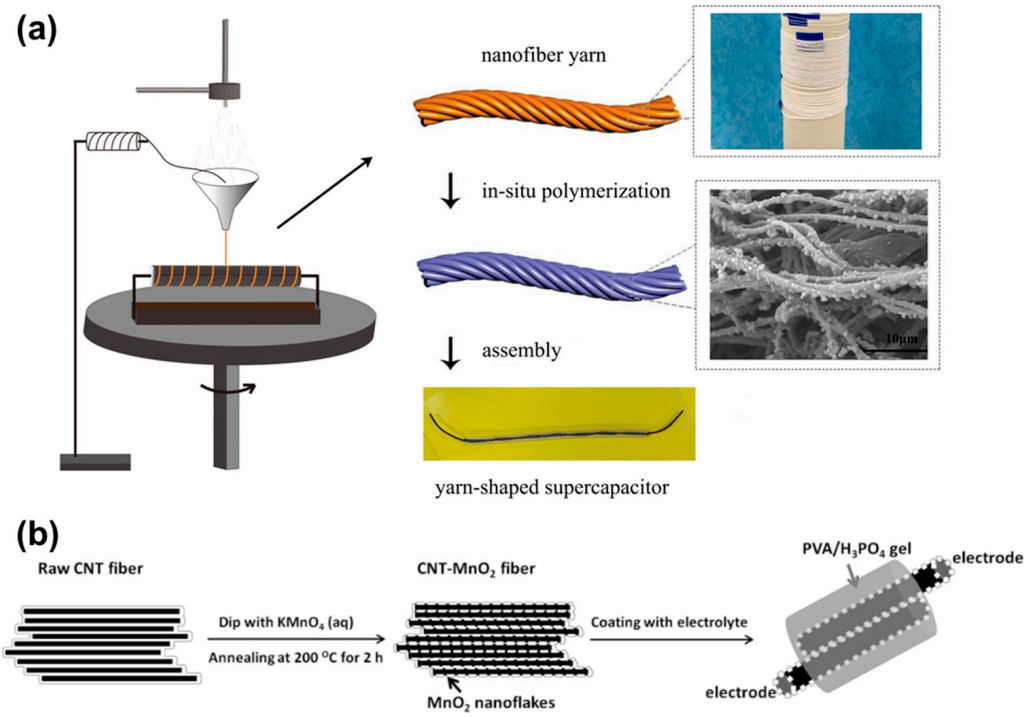


Figure 2. (**a**) Process diagram of the supercapacitor based on CNT-FF. Reproduced from ref. [12] under the Creative Commons Attribution 4.0 International (CC BY 4.0) License). (**b**) Schematic diagram of the manufacturing method of the CNT–MnO_2 supercapacitors. Reproduced from ref. [14] under the Creative Commons Attribution 4.0 International (CC BY 4.0) License).

Researchers have also sought to enhance the mechanical and electrical conductivity of CNT-FF. This has been achieved through various strategies, such as incorporation of conductive materials such as graphene and metal nanoparticles or utilization of hybrid CNT-graphene fiber fabrics. For instance, Wang et al. coated CNT-FF with graphene to create hybrid CNT-graphene fiber fabrics (Figure 3a–f) [10]. These hybrid fiber fabrics showed improved conductivity and mechanical strength, with a specific capacitance of up to 350 mF/cm^2. Strengths of this study include the improvement in electrical conductivity and mechanical strength achieved by using graphene. However, one potential weakness could be the cost and scalability of synthesizing graphene on a mass production scale. In the work by Arshad et al., CNT-FF were functionalized with NiO nanoparticles to form hybrid CNT-NiO fiber fabrics (Figure 3g) [1]. The resulting hybrid fiber fabrics demonstrated superior electrochemical performance, including a specific capacitance of up to 402 F/g and an energy density of up to 28 Wh/kg. Strengths of this study include the superior electrochemical performance achieved through functionalization with NiO nanoparticles. However, one potential weakness could be the cost and scalability of synthesizing the NiO nanoparticles.

The studies reported in these articles indicate that CNT-FF have the potential to be used as electrodes in high-performance supercapacitor devices, including flexible devices, asymmetric supercapacitors, aqueous supercapacitors, and devices integrated with photovoltaic cells. However, it is important to note that the studies were limited in their comparison with other electrode materials and their lack of information on the long-term stability and durability of CNT-FF electrodes, which are critical factors to their practical applications.

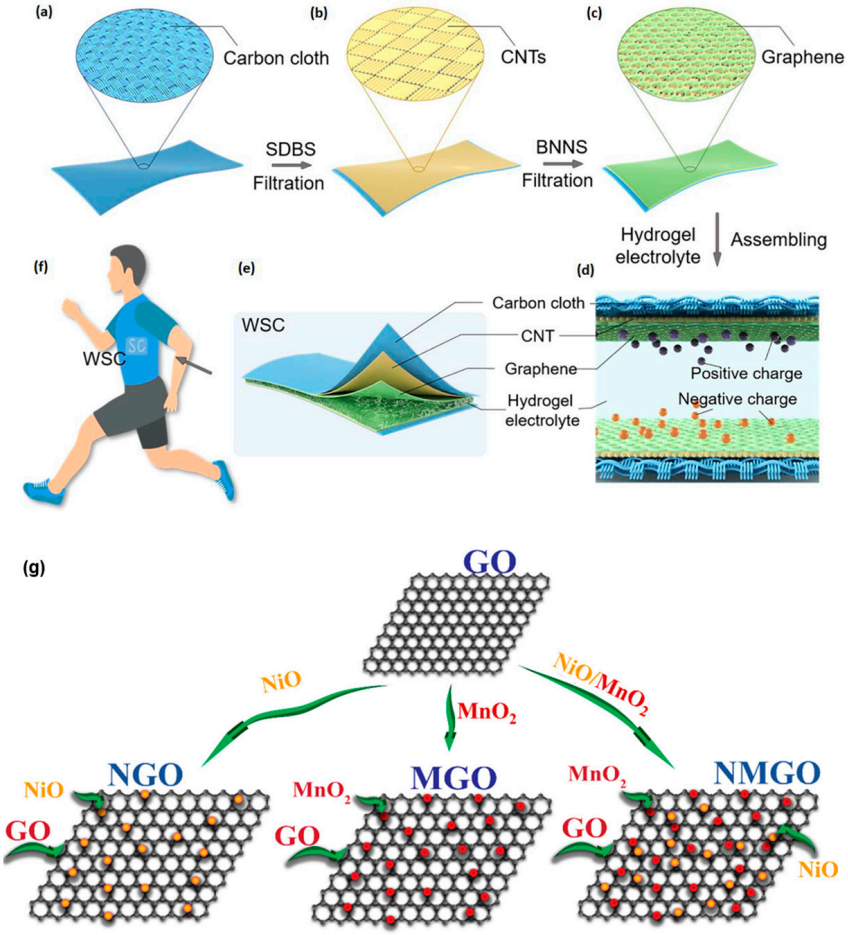


Figure 3. A schematic showing the fabrication process and structure of the wearable supercapacitor (WSC). (**a**) The carbon cloth was used as a current collector. (**b**) The CNTs were loaded on the carbon cloth through filtration, where sodium dodecylbenzene sulfonate (SDBS) was used as a dispersant. (**c**) The graphene was mounted on the CNTs layer through filtration, where boron nitride nanosheets (BNNS) were used to prevent graphene from collapsing. (**d**) The as-fabricated WSC was assembled with hydrogel used as a solid electrolyte. (**e**) The schematic structure of the WSC. (**f**) The WSCs could be integrated into clothing to power wearable devices. Reproduced from ref. [10] under the Creative Commons Attribution 4.0 International (CC BY 4.0) License). (**g**) Schematic illustration for the formation of composites. Reproduced from ref. [1] under the Creative Commons Attribution 4.0 International (CC BY 4.0) License.

2.2. Carbon Nanotube on Thin Metal Film Electrodes

The utilization of CNTs in combination with thin metal film electrodes has gained widespread attention in recent years due to the high surface area, electrical conductivity, and stability of the CNTs. Research has been conducted to optimize the energy density and rate performance of supercapacitors utilizing CNTs on thin metal film electrodes.

One strategy for enhancing performance has involved functionalizing the CNTs with conductive materials such as graphene and metal nanoparticles. Shen et al. improved electrical conductivity by functionalizing CNTs on a Ti thin film with graphene (Figure 4a), resulting in a specific capacitance of 535.7 F/g and an energy density up to 26.1 Wh/Kg [40]. Meanwhile, another study by Shahrokhian et al. utilized Ni nanoparticles on CNTs on Cu thin films (Figure 4b,c), which resulted in a specific capacitance of 12.2 F/cm^2 and improved rate performance [41]. Another method for improving the performance of CNTs on thin metal film electrodes is by controlling their morphology. Cheng et al. used a two-step growth process to synthesize CNTs on Ni thin films (Figure 4d,e), leading to a significant increase in surface area and a specific capacitance of 1200 F/g [2].

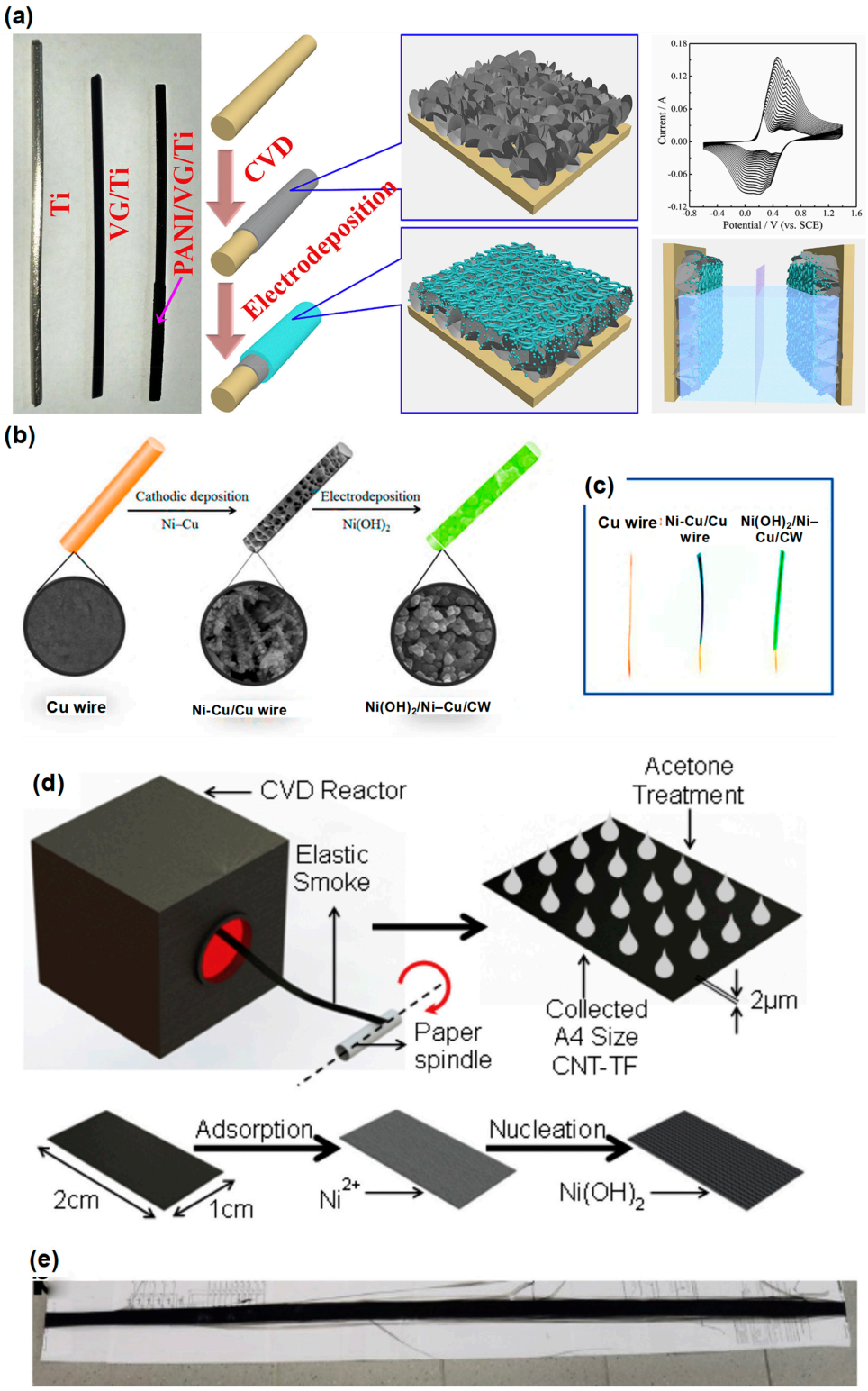


Figure 4. (**a**) Schematics of the synthesis procedure for the PANI/Gr/Ti wire electrode and symmetrical supercapacitor. Reproduced with permission from ref. [40]. Copyright 2018 Elsevier. (**b**) Schematic illustration of the preparation of $Ni(OH)_2$/Ni–Cu/CW. (**c**) Typical digital images of the Cu-wire, Ni–Cu/CW and $Ni(OH)_2$/Ni–Cu/CW. Reproduced with permission from ref. [41]. Copyright 2018 American Chemical Society. (**d**) Schematic illustration of the continuous fabrication of CNT films and the synthesis of the $Ni(OH)_2$/CNTs composite and (**e**) digital photo of the obtained CNT films with length ~1.5 m. Reproduced with permission from ref. [2]. Copyright 2015 Elsevier.

Despite the impressive results, there are some challenges that need to be addressed. For example, the volumetric energy density is relatively low in comparison with other types of electrodes, such as activated carbon. The synthetic process of conductive materials and morphologies of CNTs can be difficult to scale up. CNTs on thin metal film electrodes show immense potential for supercapacitor applications, with numerous studies demonstrating improved energy density and rate performance. However, further work is necessary to overcome the limitations and fully harness the potential of this material.

2.3. Hybrid Electrodes with Carbon Nanotube and MXene

Recently, the use of hybrid electrodes composed of CNTs and MXene materials has gained much attention for supercapacitor applications. Both CNTs and MXenes possess significant advantages, including high surface area, good electrical conductivity, and durability, making them ideal components for hybrid electrodes.

Studies have indicated that incorporating CNTs and MXenes into hybrid electrodes can lead to improved supercapacitor performance. For example, Liang, W. et al. fabricated MXene (especially $Ti_3C_2T_x$)-MWCNT electrodes and polypyrrole (PPy)-coated MWCNT electrodes as anode and cathode, respectively [4]. Ni foam substrate was immersed in the isopropanol solution containing $Ti_3C_2T_x$ and MWCNT at various ratios. Ni foam is effective due to its lightweight characteristics, porous structure, and high load mass of active material. PPy-coated MWCNT was prepared by impregnation of Ni foam with PPy-MWCNT composite in ethanol. The asymmetric supercapacitor which comprises of a $Ti_3C_2T_x$-MWCNT cathode and PPy-coated MWCNT anode showed a specific capacitance of 0.94 F cm^{-2} at scan rate of 2 mV s^{-1}. The capacitance is higher than that of pure $Ti_3C_2T_x$ by optimizing the ratio of MWCNT with increased voltage range. The asymmetric supercapacitor is also electrochemically stable at a voltage window of 0-1.6 V. Another work by Liang et al. reported that a hybrid electrode combining CNTs, Fe_3O_4, and $Ti_3C_2T_x$ MXene (Figure 5a–d) exhibited a specific capacitance of 5.52 F/cm^2 [5]. Yang et al. utilized a hybrid electrode made from CNTs and Ti_3C_2 MXene (Figure 5e–j), achieving a specific capacitance of 134 F/g [13].

A way to enhance the performance of hybrid electrodes even further is through the functionalization of CNTs and MXenes. Researchers have tried to modify CNTs with metal nanoparticles and MXenes with conductive materials. For instance, Sharma et al. uses functionalized CNTs and 1T metallic vanadium disulfide (VS_2) hybridized with MXene as an electrode for supercapacitor (Figure 6A). The VS_2-MX-CNT-50 // MWCNT-based asymmetric supercapacitor showed excellent electrochemical performances in supercapacitor applications, showing a specific capacitance of 505 F/g and an energy density of 7303 Wh/Kg (Figure 6B–G) [42].

Despite these promising findings, challenges persist in the development of hybrid electrodes. One major challenge is maintaining a stable and consistent mixture of CNTs and MXenes, as this can greatly impact the performance of the hybrid electrode. Moreover, scaling up the production of MXenes remains a technical challenge. Hybrid electrodes made from CNTs and MXenes show great promise for supercapacitor applications. Further research is necessary to overcome the challenges in the synthesis and functionalization of these materials to fully unlock their potential in supercapacitor technology.

2.4. Self-Assembled CNTs with $NiCoO_2$ Electrode

Self-assembled CNTs with $NiCoO_2$ electrodes have been the focus of several studies to improve the performance of supercapacitors. The combination of CNTs and $NiCoO_2$ provides a high-performing material system with enhanced energy storage and charge–discharge capability.

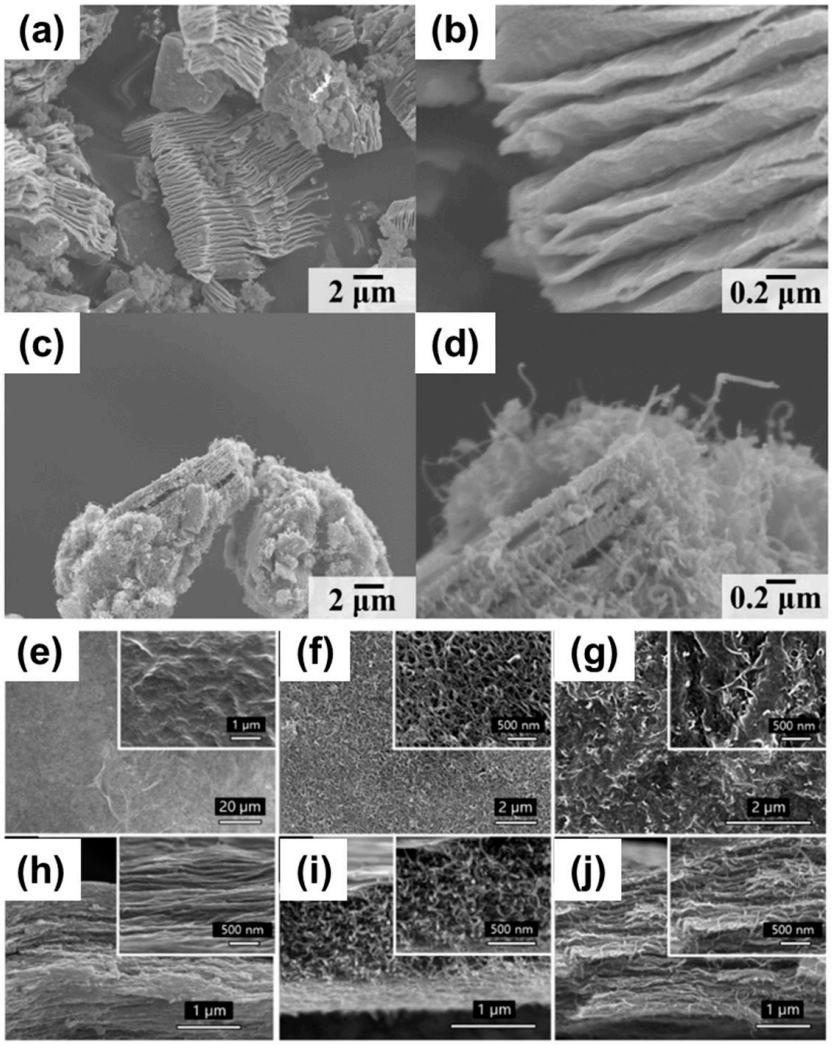


Figure 5. (**a**–**d**) SEM images at different magnifications of (**a**,**b**) as-received $Ti_3C_2T_x$ and (**c**,**d**) $Ti_3C_2T_X$-Fe_3O_4-CNT. Reproduced from ref. [5] under the Creative Commons Attribution 4.0 International (CC BY 4.0) License). (**e**–**j**) The top view SEM images of (**e**) Ti_3C_2, (**f**) CNTs, and (**g**) Ti_3C_2/CNTs film. Cross section SEM images of (**h**) Ti_3C_2, (**i**) CNTs, and (**j**) Ti_3C_2/CNTs film deposited on the graphite paper. Reproduced with permission from ref. [13]. Copyright 2018 Elsevier.

In the work by Hu, C., et al., CNT was grown on the Ni foam (NF) by immersing in CNTs solution [3]. Then, it was immersed in a solution that contains precursor of $NiCoO_2$ in deionized water and ethanol. After hydrothermal treatment and calcination under N_2 atmosphere at 300 °C, the obtained $NiCoO_2$/CNT@NF acts as an electrode (Figure 7a). The electrode has a certain structure where CNT covered with $NiCoO_2$ nanosheets is intertwined on the surface of NF due to the two-step grown approach of CNTs and $NiCoO_2$. At a current density of 5 mA cm^{-2}, it showed a specific capacitance of 929 F g^{-1} (3350 mF cm^{-2}), and it is almost 3~8 times higher than that of $NiCoO_2$@NF and CNT@NF. Furthermore, it exhibited longer discharging time, maintaining 90.1% of initial areal capacitance until 5000 cycles. The electrochemical property was enhanced by synergetic effect of CNTs and $NiCoO_2$. This is because the oxygen groups on the surface of CNTs make strong bonds with other materials and the organized nanosheet structure mentioned above. Via a simple dip-coating process, this superior electrochemical performance can be obtained by a 3D network structure. Wang et al. also reported that the self-assembled CNTs with $NiCoO_2$ electrodes exhibited high specific capacitance and excellent cycle stability [9]. They found that the specific capacitance of the CNTs/$NiCoO_2$ system (Figure 7b–e) was as high as 1587 F/g, with good cycling performance sustaining 92% performance even after

5000 cycles. The results of this study indicate that the self-assembled CNTs/$NiCoO_2$ system is a promising candidate for high-performance supercapacitors. Zhou et al. also explored the potential of self-assembled CNTs/$NiCoO_2$ system for supercapacitors [15]. The study modified CNTs' surface with metal oxides such as $NiCoO_2$ (Figure 7f), which showed a high specific capacitance of 1360 F/g, and an excellent rate capability, showing only a 1.4% loss of capacitance after 3000 cycles. The authors concluded that the CNTs/$NiCoO_2$ system had a great potential for use as high-performance supercapacitor electrodes.

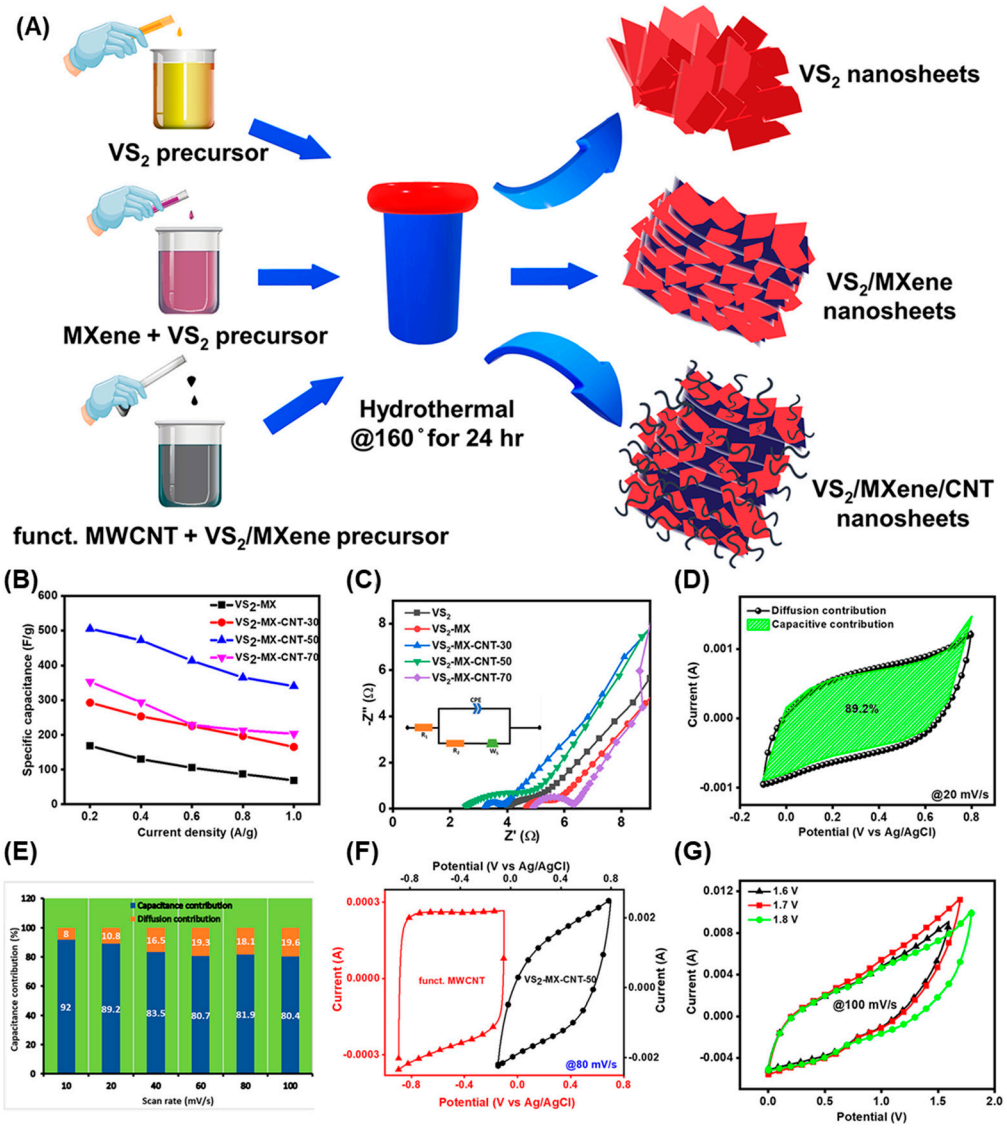


Figure 6. (**A**) General scheme of hydrothermal synthesis procedure employed to prepare VS_2, VS_2-MX, and VS_2-MX-CNT. (**B**) Comparison plot showing the specific capacitance of VS_2-MX, VS_2-MX-CNT-30, VS_2-MX-CNT-50, and VS_2-MX-CNT-70 samples at a current density from 0.2 A/g to 1 A/g. (**C**) Nyquist plot showing the EIS fit graphs in the circuit (see Inset). (**D**) Contribution of capacitive and diffusion-controlled areas of VS_2-MX-CNT-50 at a scan rate of 20 mV/s. (**E**) Contribution of capacitive and diffusion-controlled reactions at scan rates ranging from 10 to 100 mV/s for VS_2-MX-CNT-50 ternary hybrid electrode. (**F**) Comparison of MXene and VS_2-MX-CNT-50 CV profiles at a scan rate of 80 mV/s to estimate the potential window. (**G**) The stability of potential window of VS_2-MX-CNT-50 // MWCNT asymmetric devices from 1.6 to 1.8 V at 100 mV/s, as shown by CV curves. Reproduced with permission from ref. [42]. Copyright 2022 Wiley.

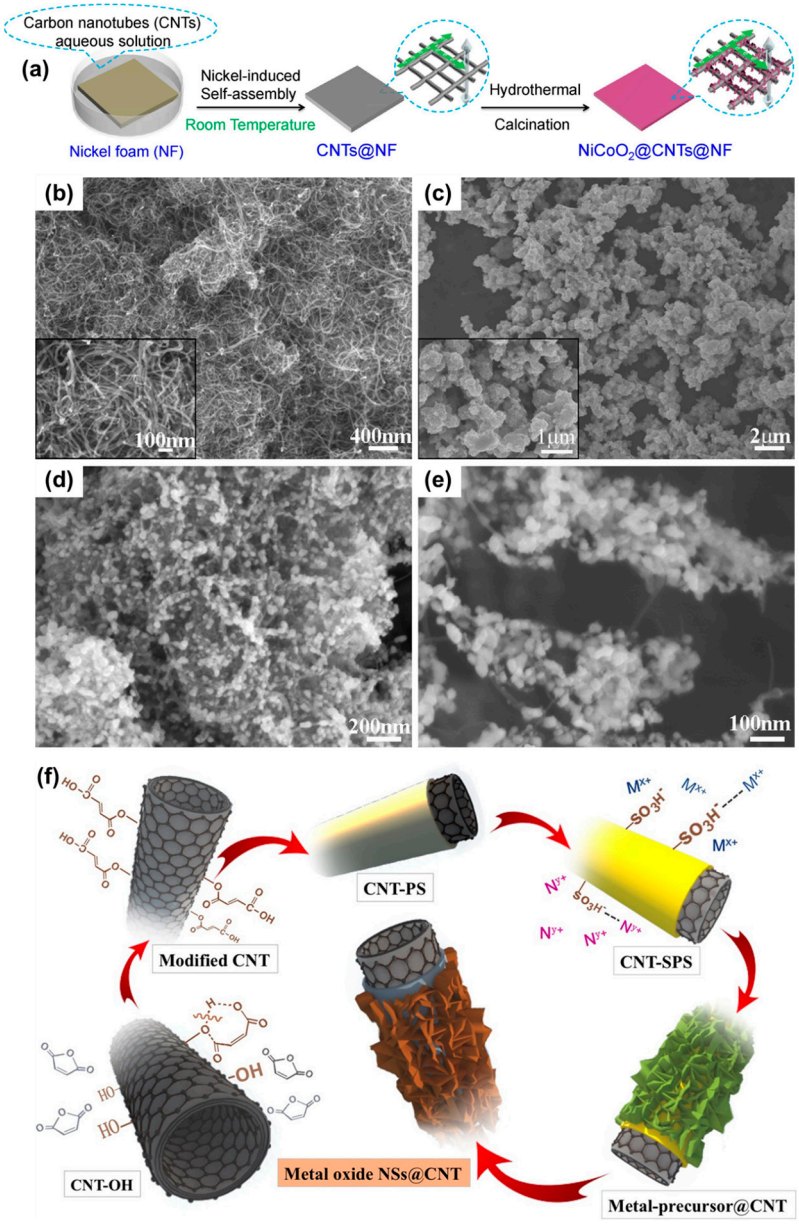


Figure 7. (**a**) Schematic illustration of the fabrication process of the $NiCoO_2$@CNTs@NF integrated electrode. Reproduced with permission from ref. [3]. Copyright 2021 Elsevier. (**b**–**e**) SEM images of (**a**) pure CNT, (**b**) pure $NiCoO_2$, and (**c**,**d**) $NiCoO_2$@CNT. Reproduced with permission from ref. [9]. Copyright 2022 Elsevier. (**f**) Schematic illustration of the synthetic procedure for TMO@CNT hybrid materials through pre-coating CNT with sulfonated polystyrene. Reproduced from ref. [15] under the Creative Commons Attribution 4.0 International (CC BY 4.0) License.

However, the practical implementation of the self-assembled CNTs/$NiCoO_2$ system is still facing several challenges. One major challenge is the consistency and stability of the CNTs self-assembling process, as this greatly impacts the performance of the supercapacitor. Additionally, the cost-effective and large-scale production of the CNTs/$NiCoO_2$ system remains a technical challenge for commercialization. Self-assembled CNTs with $NiCoO_2$ electrodes have shown great potential as high-performance supercapacitor electrodes. Despite the challenges, the results of the studies indicate that the self-assembled CNTs/$NiCoO_2$ system is a promising candidate for future energy storage systems. Further research is needed to address the challenges and optimize the electrochemical performance of self-assembled CNTs/$NiCoO_2$ supercapacitors.

2.5. One-Dimensional Fiber-Based Supercapacitor

A 1D fiber-based supercapacitor is beneficial in terms of volumetric electrochemical performance. It is important for wearable devices that there is no useless area for high volumetric capacitance. Thus, Ma et al. fabricated partially unzipped carbon nanotube (PUCNT)/reduction graphene oxide (rGO) fiber (Figure 8A–G) [43]. They oxidized CNT with $KMnO_4$, and the oxidized CNT (PUOCNT) was well dispersed in solution with grafted hydrophilic groups. The solution containing PUOCNT and GO grew on the fiber by wet-spinning, and finally the PUCNT/rGO fiber was fabricated by chemical reduction. It exhibited larger CV curves at a scan rate of 10 mV s^{-1} and longer discharging time at a current density of 100 mA cm^{-3}, which means improved electrochemical performance. The volumetric capacitance of the PUCNT/rGO fiber supercapacitor is 62.1 F cm^{-3} and the capacitance remains over 90% for 20,000 cycles. It is a result of preventing the rGO sheet from being stacked by intercalation of CNT fibers, minimizing the dead volume because the stacking of the graphene layers during fabrication of graphene-based electrodes decreases the surface area ion diffusion rate, which causes performance degeneration. A similar approach using intercalation of CNT between the rGO layers is studied by Heo et al. (Figure 8H–Q) [44]. They made a yarn-type supercapacitor by using SWNTs-coated traditional Korean paper, which obtained a high specific capacitance of 366 F/g and energy density of 114 Wh/Kg as well as excellent cyclic stability, showing no degradation of capacitance after 10,000 cycles due to the same complementary effect.

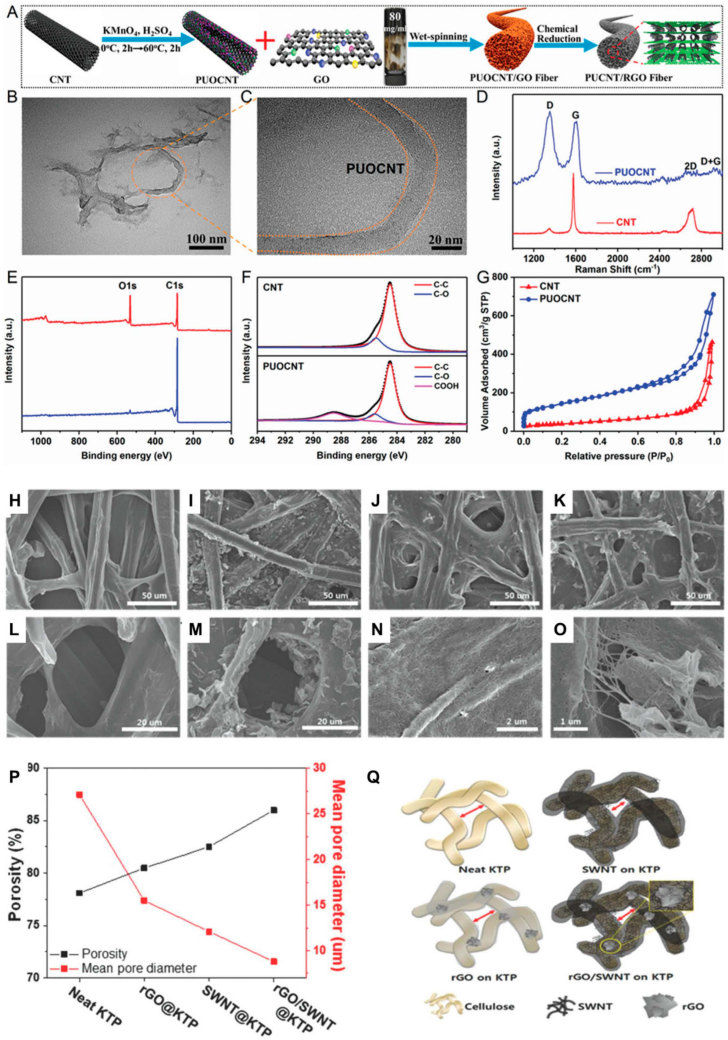


Figure 8. (**A**) Schematic illustration of PUOCNT synthesis and of PUCNT/RGO fiber fabrication. (**B**,**C**) TEM images of PUOCNT. (**D**) Raman spectra, (**E**) XPS survey spectrum, (**F**) XPS C1s spectrum,

and (**G**) nitrogen adsorption and desorption isotherms of CNT and PUOCNT. Morphologies and textural properties of conductive KTP sheets depending on coating carbon solutions. Reproduced with permission from ref. [43]. Copyright 2021 Wiley. SEM images of (**H**,**L**) neat KTP, (**I**,**M**) rGO on KTP, (**J**,**N**) SWNT on KTP, and (**K**,**O**) rGO/SWNT on KTP with different magnifications. (**P**) Porosity and mean pore diameter changes as function of carbon materials. (**Q**) Schematic illustration showing the structures of conductive KTP sheets as function of carbon materials. Reproduced with permission from ref. [44]. Copyright 2018 Wiley.

3. Conclusions and Perspective

The summary of the various types of CNTs and their electrochemical performance are given in Table 1.

Table 1. A summary of the various types of CNTs and their electrochemical performances.

Materials	Electrochemical Performance	[Ref]
CNTFF	Areal capacitance of up to 353 mF/cm^2 and an energy density of up to 247 μW/cm^2	[39]
CNT-MnO_2 fiber fabrics	A specific capacitance reaching up to 231.3 F/g and an energy density of up to 86 nWh/cm	[14]
CNT-graphene fiber fabrics	A specific capacitance of up to 350 mF/cm^2	[10]
CNT-NiO fiber fabrics	A specific capacitance of up to 402 F/g and an energy density of up to 28 Wh/kg	[1]
Functionalizing CNTs on a Ti thin film with graphene	A specific capacitance of 535.7 F/g and an energy density up to 26.1 Wh/Kg	[40]
CNTs on Ni thin films	A specific capacitance of 1200 F/g	[2]
MXene ($Ti_3C_2T_x$)-MWCNT electrodes	A specific capacitance of 0.94 F cm^{-2} at scan rate of 2 mV s^{-1}	[4]
A hybrid electrode combining CNTs, Fe_3O_4, and $Ti_3C_2T_x$ MXene	A specific capacitance of 5.52 F/cm^2	[5]
CNTs and Ti_3C_2 MXene	A specific capacitance of 134 F/g	[13]
VS_2-MX-CNT-50//MWCNT	A specific capacitance of 505 F/g and an energy density of 7303 Wh/Kg	[42]
$NiCoO_2$/CNT@NF	A specific capacitance of 929 F g^{-1}	[3]
CNTs/$NiCoO_2$ system (3D)	A specific capacitance of 1587 F/g	[9]
Self-assembled CNTs/$NiCoO_2$ system	A high specific capacitance of 1360 F/g	[15]
SWNTs-coated Korean traditional paper	A specific capacitance of 366 F/g and energy density of 114 Wh/Kg	[44]

CNT-based materials have emerged as promising materials for supercapacitor electrodes due to their high surface area, electrical conductivity, and mechanical strength. CNT-based materials can improve structural property which leads to enhanced electrochemical performance in the forms of 1D fiber, 2D film, and 3D composite. The larger surface area by porous structure of CNTs results in high areal capacitance, and intercalation of CNTs makes volumetric capacitance higher while their low density increases specific capacitance. Furthermore, there are huge potentials when combining with other materials expected to have synergetic effects because this creates various possibilities to combine with other active materials, sometimes with pre-treatment. Therefore, the scope of research regarding the use of CNTs as a supercapacitor material is expected to grow faster and broader in the future, while demands for flexible and wearable energy devices increase.

There have been numerous studies aimed at improving the value of the specific capacitance, energy density, and rate performance of supercapacitors utilizing these materials. One strategy for enhancing performance has involved functionalizing the CNTs with conductive materials such as graphene and metal nanoparticles, while another method has

involved controlling the morphology of the CNTs on thin metal films. CNT-FFs have shown impressive energy storage performance in many studies, including a specific capacitance of up to 402 F/g and an energy density of up to 28 Wh/kg. Similarly, CNTs on thin metal film electrodes have also demonstrated high specific capacitance and improved rate performance. Hybrid electrodes combining CNTs with Mxene or $NiCoO_2$ also showed excellent electrochemical performance. However, there are some challenges that need to be addressed, including the scalability of the synthesis processes and the long-term stability of the electrodes. In conclusion, CNT-based electrodes have the potential to be used as electrodes in high-performance supercapacitor devices. However, more research is needed to address the limitations and fully realize their potential for practical applications. In addition, the thickness of working electrodes is very important for supercapacitor applications. Therefore, more studies addressing the effect of thickness on the detailed electrochemical performance of supercapacitors are required. The production yield of CNTs can also be a critical factor for their use in supercapacitor electrodes. Several factors such as the synthetic method or the purity of the raw materials determine the production yield of CNTs. To improve the production yield of CNTs, alternative carbon sources such as biomass have been studied because they are more sustainable and cost-effective raw materials for CNT synthesis. However, further research will be required regarding the industrial use of CNTs in supercapacitor applications.

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