Review

Preparation of Electrode Materials Based on Carbon Cloth via Hydrothermal Method and Their Application in Supercapacitors

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Abstract: Supercapacitors have the unique advantages of high power density, fast charge and discharge rates, long cycle life, high safety, and reliability, and are increasingly being used for applications including automobiles, rail transit, communication equipment, digital electronics, and aerospace equipment. The supercapacitor industry is currently in a stage of rapid development; great breakthroughs have also been made in improving the performance of supercapacitors and the expansion of their application. Electrode technology is the core of supercapacitors. Transition-metal compounds have a relatively high theoretical capacity and have received widespread attention as electrode materials for supercapacitors. In addition, there is a synergistic effect between the different components of various electrode composite materials. Due to their superior electrochemical performance, supercapacitors are receiving increasing research attention. Flexible supercapacitors have been hailed for their good plasticity, resulting in a development boom. This review article mainly outlines the development process of various electrode materials, including carbon materials, conductive polymers, metal compounds, and composite materials, as well as flexible electrode materials based on carbon cloth.

Keywords: supercapacitors; carbon materials; conductive polymers; metal compounds; composite materials; carbon cloth

1. Introduction

Recently, the explosive growth of society and constant advancement of science and technology have led to an increasing demand for energy, resulting in a significant energy crisis and environmental problems [1,2]. There is thus an urgent need to develop environmentally friendly, efficient, clean, and sustainable advanced energy conversion and storage technologies [3,4]. Currently, the development of the new energy industry is based on the three core products of secondary batteries (mainly lithium-ion batteries) [5], supercapacitors [6], and fuel cells [7], with a vast application network.

Supercapacitors are also known as large-capacity capacitors, energy storage capacitors, gold capacitors, electric double layer capacitors, and farad capacitors. After decades of development, using carbon material from naturally abundant lignocellulose biomass precursors as the electrode material of flexible/wearable electronic devices is a future trend, but there are still technological challenges [8]. Based on availability and low-cost, various carbon materials, such as activated carbons (AC), nitrogen doping on carbon, and graphene on carbon cloth, have also been studied [9,10]. However, their performance is not particularly good.

Initially, in 1957, Becker filed an application to use high-specific-surface-area activated carbon as the electrode material for supercapacitors. In 1962, the Standard Oil Company
commercialized carbon material electrochemical capacitors. In 1979, the Nippon Electronic Company (NEC) implemented supercapacitors for large-scale commercial applications. Scholars are increasingly discovering the application value of supercapacitors. The structure of a supercapacitor consists of the electrodes, electrolyte, and separator. The electrode material in the electrodes plays a vital role in the performance of the supercapacitor [11]. The electrode material is the core. From carbon materials [12,13], element-doped carbon materials [14,15], metal compounds [16,17], multi-metal compounds [18,19], and polymers [20,21], to various composite materials, various superior porous three-dimensionally structured composite materials have been constructed. With the further development of society, there has been a greater pursuit of wearable and strong plasticity flexible electrodes. The preparation of carbon cloth offers the advantages of low cost, good flexibility, and good conductivity, and, as a substrate, has been hailed as a high-performance electrode material [22–26].

CC has been used as an electrode material for flexible supercapacitors applied in portable and wearable products, setting off a research boom. Figure 1 clearly shows that in the past ten years, there has been a linear upward trend in the number of publications with the keywords “supercapacitor”, “carbon cloth”, and “carbon fiber cloth” included in their titles (from a search in Web of Science), and this trend is expected to be maintained. Flexible supercapacitors based on CC are very promising electrode materials. CC has the advantages of light weight, low cost, excellent flexibility, large surface area, high conductivity, and porosity; in addition, it is particularly easy to construct an electrode material with structural advantages.

![Figure 1. Increasing trend of the number of publications related to supercapacitors based on CC in the past decade. These results were obtained by searching the Web of Science for articles with the keywords “supercapacitor”, “carbon cloth”, and “carbon fiber cloth” included in their titles.](image-url)
2. Electrode Material

2.1. Carbon Materials

Carbon materials are mainly used as the electrode materials for Type I supercapacitors and were the earliest electrode materials to be researched and applied. As shown in Table 1, Gamby et al. [27] conducted the first electrochemical characteristic tests on various activated carbons from the PICA Company and observed that the highest specific capacitance was 125 F·g⁻¹. Beck et al. [28] prepared industrial carbon black (CB) electrodes with polytetrafluoroethylene (PTFE) as a binder in 12 M H₂SO₄; the highest capacitance measured was 250 F·g⁻¹. Carbon aerogel is a lightweight porous carbon material with a three-dimensional nanonetwork structure. It has a large specific surface area, low density, high porosity, adjustable pore size distribution, and good electrical conductivity. Fischer et al. [29] obtained carbon aerogel with a density of 800 kg·cm⁻³ via pyrolysis from resorcinol formaldehyde aerogel; the highest capacity measured was 46 F·cm⁻³. Li et al. [30] synthesized mixed carbon aerogels by blending cresol, catechol, and formaldehyde, with a high specific capacitance of 77 F·cm⁻³ (104 F·g⁻¹). The good porous structure of carbon aerogels results in their high specific capacitance. Carbon fiber is an inorganic polymer fiber with a carbon content higher than 90%. It has the characteristics of low density, light weight, high strength, and high elastic modulus.

Carbon nanotubes (CNTs) appear as a seamless centrally controlled nanoscale coaxial cylinder rolled by sheet-structured graphite, with fullerene hemispheres sealed at both ends, divided into single-walled carbon nanotubes (SWCNTs) and multi-walled carbon nanotubes (MWCNTs). Compared with the best activated carbon, MWCNTs have available mesoporous structure and the value of their specific capacitance varies up to 135 F·g⁻¹ [31]. Liu et al. [32] deposited CNTs on a platinum electrode for the first time, resulting in a specific capacitance of 283 F·g⁻¹, which was twice that of an active carbon electrode under the same conditions. Pan et al. [33] reported a tube-to-tube multi-walled carbon nanotube with a specific capacitance of 135 F·g⁻¹ consisting of outer nanotubes with an average outer diameter of 50 nm and internal nanotubes with diameters of 3–10 nm. Yang et al. [34] introduced hierarchical graphene sheet (GS)-CNTs with a specific capacitance of 326.5 F·g⁻¹. The stacking of GS can be efficaciously impeded by inserting an appropriate quantity of CNTs as nanospacers and enlarging the space between GS sheets, leading to a highly porous nanostructure.

Because of the unique porous structure of carbon materials, they have a larger specific surface area, which facilitates the mass diffusion and transport of electrolytes, providing more ion contact sites and increasing the capacitance. Hollow core, mesoporous shell carbon nanospheres (HCMSs) with a high surface area of 1704 m²·g⁻¹ exhibited a high specific capacity of 251 F·g⁻¹ at 50 mV·s⁻¹ in 2 M H₂SO₄ [35]. Onion-like carbon (OLC) [36] and nonporous carbons (NPCs) with a metal–organic framework (MOF) as a template exhibited excellent mesoporous characteristics [37]. Zhao et al. [38] fabricated sulfur-containing mesoporous carbons via one-pot aqueous self-assembly with different chemical states of sulfur in tunable amounts. The addition of inorganic elements such as O, N, S, and P improved the electrochemical performance.

Table 1. Carbon materials for supercapacitors.

| Material           | The Highest Capacities | Specific Surface Area | Specific Power Densities | Specific Energy Densities | Specific Capacitance Retention | Ref.   |
|--------------------|------------------------|-----------------------|--------------------------|--------------------------|-------------------------------|--------|
| Activated carbon   | 125 F·g⁻¹              | -                     | -                        | -                        | -                             | [27]   |
| CBs                | 250 F·g⁻¹              | 28–1690 m²·g⁻¹        | -                        | -                        | -                             | [28]   |
| Carbon aerogels    | 46 F·cm⁻³              | -                     | -                        | -                        | -                             | [29]   |
Table 1. Cont.

| Material      | The Highest Capacities | Specific Surface Area | Specific Power Densities | Specific Energy Densities | Specific Capacitance Retention | Ref.   |
|---------------|------------------------|-----------------------|--------------------------|---------------------------|--------------------------------|-------|
| Carbon aerogels | 77 F·cm⁻³              | -                     | -                        | -                         | -                              | [30]  |
| MWCNTs        | 135 F·g⁻¹              | 470 m²·g⁻¹            | -                        | -                         | -                              | [31]  |
| SWCNTs        | 283 F·g⁻¹              | -                     | -                        | -                         | -                              | [32]  |
| Tubes-in-tube CNTs | 315 F·g⁻¹            | -                     | -                        | -                         | -                              | [33]  |
| GS-CNTs       | 326.5 F·g⁻¹            | -                     | 78.29 kW·kg⁻¹            | 21.74 Wh·kg⁻¹             | 98% (after 200 cycles)         | [34]  |
| HCMSs         | 251 F·g⁻¹              | -                     | -                        | -                         | -                              | [35]  |
| OLC           | -                      | -                     | 195.0 W·cm⁻³            | 2.9 mWh·cm⁻³              | -                              | [36]  |
| NPCs          | 222 F·g⁻¹              | -                     | -                        | -                         | -                              | [37]  |
| S-carbon      | -                      | -                     | -                        | -                         | -                              | [38]  |

2.2. Conductive Polymers

Conductive polymers have caused a research boom. Shirakawa et al. [39] first prepared iodine-doped polyacetylene with a conductivity of up to $10^3$ S·cm⁻¹. Conductive polymers have the characteristics of high electrical conductivity, large specific surface area, and light specific gravity, as well as good flexibility, low production cost, and high energy efficiency; therefore, they can be used for rechargeable and secondary batteries and electrode materials [40]. Arbizzian et al. [41] synthesized three supercapacitors: a symmetric supercapacitor based on p-doped poly(pyrrole), an asymmetric supercapacitor based on both p-doped poly(pyrrole) and poly(3-methylthiophene), and a symmetric supercapacitor based on p- and n-doped poly(3-(phenylthiophene)-dithieno[3,4-b:3',4'-d] thiophene) with a high working potential. In Table 2, Ferraris et al. [42] studied various poly 3-(phenylthiophene) derivatives as electrode materials to assemble supercapacitors. The capacitor based on 3-(3, 4 difluorophenyl) thiophene (MPFPT) was particularly promising because of its high capacity and excellent charge/discharge performance. Fusalba et al. [43] synthesized poly(cyclopenta [2,1-b:3,4-b'] dithiophene—4-one) (PCDT) with an open and porous structure. For both the p-type and n-type doped states, the material exhibited a low-frequency capacitance of approximately 70 F·g⁻¹.

However, conductive polymers also have shortcomings, such as poor mechanical properties and long-term cycle stability. To overcome these shortcomings and enhance their performance, the optimization of the structure and shape or the mixture of conductive polymers with other carbon materials are important routes. Gouérec et al. [44] showed that the high performance of polyacrylonitrile (PAN) microcellular foam thin films deposited on carbon fibers is related to their high surface area and micropore distribution. Pyrrole (Py) on the surface of a porous graphite fiber matrix resulted in a specific capacitance of 400 F·g⁻¹ [45]. Polypyrrole (PPy) on MWCNT membranes exhibits high redox activity, resulting in high specific capacitance [46]. Dubal et al. [47] observed that multilayer PPy nanosheets exhibited a specific capacitance of 586 F·g⁻¹ at a scan rate of 2 mV·s⁻¹, with a Brunauer–Emmett–Teller (BET) surface area of 37.1 m²·g⁻³, which is higher than that of PPy nanoribbons and nanobricks. Jyothibasu et al. [48] prepared PPy tubes using one-step in situ chemical oxidative polymerization with curcumin as a template, which was combined with functionalized carbon nanotubes (f-CNTs) as electrode materials. The electrodes exhibited morphological uniformity, a favorable hierarchical porous structure, a large surface area, and excellent electrochemical properties, including outstanding cycling stability (retention of 118.18% of the initial capacitance after 12,500 charge/discharge cycles).
Table 2. Properties of conductive polymers for supercapacitors prepared by various methods.

| Materials                              | Method                              | Specific Capacitance | Specific Power Densities | Specific Energy Densities | Ref. |
|----------------------------------------|-------------------------------------|----------------------|--------------------------|---------------------------|------|
| Poly 3-(Phenylthiophene) derivatives   | Electrochemical polymerization      | -                    | 5 kW·kg⁻¹                | 50 Wh·kg⁻¹                | [42] |
| PCDT                                   | Electrochemical polymerization      | -                    | 1 kW·kg⁻¹                | 6 Wh·kg⁻¹                 | [43] |
| PAN                                    | Deposition                          | -                    | -                        | -                         | [44] |
| Py                                     | Chemical polymerization             | 400 F·g⁻¹            | -                        | -                         | [45] |
| PPy                                    | Electrochemical deposition          | 427 F·g⁻¹            | -                        | -                         | [46] |
| PPy                                    | Electro polymerization              | 586 F·g⁻¹            | -                        | -                         | [47] |
| (E)-α-cyanooethylene thiophene derivatives | In-situ chemical oxidative polymerization | 2732 mF·cm⁻²    | 129.35 mW·cm⁻²          | 242.84 µWh·cm⁻²           | [48] |

2.3. Metal Compounds

As electrode materials, metal compounds exhibit excellent pseudocapacitance behavior (superior to that of carbon materials), owing to their rapid reversible redox reactions. RuO₂ is the earliest metal compound used as an electrode material for supercapacitors. Amorphous RuO₂·xH₂O is conducive to the diffusion of electrolyte ions in its body phase. The redox reaction not only occurs on the surface of the electrode, but also inside, which is helpful for improving the utilization rate and achieving a high specific capacitance. Compared with RuO₂·xH₂O powder [50,51] (Table 3), RuO₂·xH₂O thin film is more uniform and complete and has a higher specific capacitance. Scholars have also discovered that the introduction of other metals is helpful for increasing the capacitance because of the synergistic effect between the metals [52,53]. However, the poor conductivity, high cost, and toxicity of RuO₂ prevents its use in large-scale commercial applications. At this time, relatively inexpensive transition-metal compounds were also shown to possess good pseudocapacitor performance, which was widely valued.

Conway [54] proposed that inexpensive transition-metal oxides have pseudocapacitive behavior, and they were considered the most promising energy storage materials. Prasad et al. [55] prepared nanorod-shaped three-dimensional In₂O₃ with high specific capacitance and high power density that exhibited good electrical performance similar to that of the expensive RuO₂. Nam et al. [56] reported the largest specific capacitance of NiO obtained by electrochemical precipitation with heat treatment at 300 °C; this material has defect properties that can improve its electrochemical properties. In addition to transition-metal oxides [57–61], sulfides, phosphides, and selenides also exhibit pseudocapacitive behavior. These elements’ atoms have larger ionic radii and are more prone to transition transfer and discrete diffusion. To some extent, these materials exhibit improved electrochemical performance. Sulfur atoms have a lower electronegativity than oxygen atoms, and sulfides have a faster redox reaction rate and higher electronic conductivity [62]. Qian et al. [63] prepared CuS nanotubes with a high specific capacitance of 2393 F·g⁻¹ at a scan rate of 10 mV·s⁻¹. This was the first time that a novel redox-active alkaline electrolyte (polysulfide electrolyte) was used to improve the specific capacitance. A multilayer M-MoS₂·H₂O system was first investigated; nanochannels between layers with a distance of approximately 1.18 nm increased the space for ion diffusion and extended the surface area for adsorption [64].
Nano-size compounds with high specific surface area have more active sites and can fully contact the electrolyte, which is beneficial for increasing the specific capacitance [65–70]. For the first time, Ramasamy et al. [71] developed a simple colloidal method for the synthesis of CuSbSe\textsubscript{x}S\textsubscript{2−x} by replacing S with Se; the materials exhibited excellent cyclic stability with promising specific capacitance values. Se atoms can be used to adjust the width of the interlayer gap between layers, which is beneficial for ion diffusion. Lin et al. [72] synthesized coral-like LiFePO\textsubscript{4} particles through a facile chemical etching method with rugby-like LiFePO\textsubscript{4} particles as precursors. Priyadharsin et al. [73] prepared γ-KCoPO\textsubscript{4} nanocrystals as supercapacitor electrodes in an aqueous electrolyte for the first time, resulting in a specific capacitance of 309 C·g\textsuperscript{−1} at 1 mV·s\textsuperscript{−1} in 1 M KOH. Multi-metal compounds exhibit improved electrochemical performance because, in addition to the synergistic pictograms between different components, it is also easy to construct various morphologies with structural advantages [68–70,72,74,75].

In general, transition metals are low in cost, easy to obtain, abundant, and exhibit excellent pseudocapacitance performance. Combining transition metals with other materials to construct porous three-dimensional nanostructures with high specific surface area results in electrode materials with high specific capacitance, good energy and power density, and good stability.

| Materials | Method | Specific Capacitance | Specific Power Density | Specific Energy Density | Specific Capacitance Retention | Ref. |
|-----------|--------|----------------------|------------------------|-------------------------|--------------------------------|------|
| RuO\textsubscript{2}·xH\textsubscript{2}O powder | Sol-gel process | 720 F·g\textsuperscript{−1} | - | 26.7 Wh·kg\textsuperscript{−1} | - | [50] |
| RuO\textsubscript{2} films | Electrodeposition | 1190 F·g\textsuperscript{−1} | - | - | | [51] |
| Ru\textsubscript{0.8}Sn\textsubscript{0.2}O\textsubscript{2}·nH\textsubscript{2}O | Hydrothermal | 830 F·g\textsuperscript{−1} | - | - | | [53] |
| RuO\textsubscript{2}·VO\textsubscript{2} solid solution | Polymerizable-complex | 1210 F·g\textsuperscript{−1} | - | - | | [54] |
| In\textsubscript{2}O\textsubscript{3} | Electrochemical deposition | 190 F·g\textsuperscript{−1} | - | - | | [55] |
| NiO film | Electrochemical precipitation | - | - | - | - | [56] |
| NiO nanosheet | Hydrothermal | 989 F·g\textsuperscript{−1} | - | 49.45 Wh·kg\textsuperscript{−1} | 97% (after 1000 cycles) | [57] |
| cobalt oxide | Deposition | 165 F·g\textsuperscript{−1} | - | - | | [58] |
| CuO multilayer nanosheets | CBD | 43 F·g\textsuperscript{−1} | - | - | | [59] |
| Mn\textsubscript{O} | - | 398 F·g\textsuperscript{−1} | - | - | | [60] |
| Mn\textsubscript{3}O\textsubscript{4} thin films | SILAR Biomolecule-assisted hydrothermal | 314 F·g\textsuperscript{−1} | - | - | - | [61] |
| Co\textsubscript{S} nanowires | Hydrothermal | 508 F·g\textsuperscript{−1} | - | - | - | [62] |
| Cu\textsubscript{S} nanotubes | - | 2393 F·g\textsuperscript{−1} | - | - | - | [63] |
| M-Mo\textsubscript{2}H\textsubscript{2}O system | Hydrothermal | 380 F·g\textsuperscript{−1} | - | - | - | [64] |
| Ni(OH)\textsubscript{2} | Hydrothermal | 1715 F·g\textsuperscript{−1} | - | - | - | [65] |
| α-Co(OH)\textsubscript{2} nanowire arrays (NWAs) | Hydrothermal | 642.5 F·g\textsuperscript{−1} | - | - | - | [66] |
| FeVO\textsubscript{4} nanoparticles | Co-precipitation | 972 F·g\textsuperscript{−1} | 1326 kW·kg\textsuperscript{−1} | 21 Wh kg\textsuperscript{−1} | - | [67] |
| NiCo\textsubscript{2}O\textsubscript{4} | Electrodeposition | 678 F·g\textsuperscript{−1} | - | 15.42 Wh·kg\textsuperscript{−1} | 96.06% (after 1500 cycles) | [68] |
| NiS@HMRAs | Hydrothermal | 2906 F·g\textsuperscript{−1} | 409 W·kg\textsuperscript{−1} | 33.9 Wh·kg\textsuperscript{−1} | - | [69] |
## Table 3. Cont.

| Materials                  | Method         | Specific Capacitance | Specific Power Density | Specific Energy Density | Specific Capacitance Retention | Ref.  |
|----------------------------|----------------|----------------------|------------------------|-------------------------|---------------------------------|-------|
| Sn-Co binary oxide nanosheets | Hydrothermal   | 937.4 F·g⁻¹          | -                      | -                       | 97.5% (after 20,000 cycles)     | [70]  |
| CuSbSe₂                    | Colloidal      | -                    | -                      | -                       | 99.5% (after 200 cycles)        | [71]  |
| Coral-Like LiFePO₄ Particles | Chemical etching | 359 F·g⁻¹          | -                      | -                       | 82.3% (after 2000 cycles)       | [72]  |
| γ-KCoPO₄                   | Sol-gel        | 222 C·g⁻¹            | 1.6 kW·kg⁻¹            | 28 Wh·kg⁻¹              | -                              | [73]  |
| Mn-Co-Fe HNPs              | Electrodeposition | 1200 F·g⁻¹         | 1125 W·kg⁻¹            | 11.4 Wh·kg⁻¹            | 96% (after 4000 cycles)         | [74]  |
| Co-Mo-O-S porous microspheres | Hydrothermal   | 1134 F·g⁻¹          | -                      | 67.6 Wh·kg⁻¹            | -                              | [75]  |

### 2.4. Composite Materials

Composite materials are more suitable for the development of supercapacitor electrode materials because of their improved electrochemical performance relative to that of single-component materials, resulting from the synergy between the components. It is easier to build porous nanostructures with high specific surface area, which can provide more electroactive sites and enable fast electron transmission and enhanced structural stability. As shown in Table 4, many attempts have been made to study composite materials with carbon materials [76–79]. Electrochemical measurements showed that the pseudocapacitance behavior of RuO₂ increases the specific capacitance of pure carbon materials. Transition metals have the potential to replace precious metal oxides in supercapacitor electrodes. Scientists are also committed to developing a low-cost, high-performance material. Composite materials of transition-metal compounds and various carbon materials exhibit better performance than pure carbon materials [80–85]. It is worth emphasizing the use of CNTs, graphene, carbon fiber, and molecular sieves. These materials have high specific surface area and mesoporous structures, which are very conducive to the construction of composite materials with structural advantages [86]. Co(OH)₂/ultrastable Y molecular sieve composites [87], NiO/CNT composites [88], Co₃O₄ nanowire/three-dimensional graphene foam [89], Ni₃S₂ nanoparticles/MWCNTs [90], and α-Ni(OH)₂-GO composites [91] all exhibit high specific capacitance, with the highest capacitance reached being 1760.72 F·g⁻¹ [91].

Binary and even multi-component metal compound composite materials exhibit superior performance because of the synergistic effect between metals and between different components. BiMnO₃/MWCNT composites were used as a new active material for positive electrodes of supercapacitors, with a maximum energy density of 9.0 Wh·kg⁻¹ and maximum power density of 2.5 kW·kg⁻¹ for an asymmetric device [92]. A Co–Al layered double hydroxide nanosheet (Co–Al LDH–NS)/GO film composite [93] exhibited a specific capacitance of 1200 F·g⁻¹ and a long cyclic life owing to the well-organized layered structure, which was beneficial to efficient electron transport. A CoMoO₄/G composite exhibited low electrochemical resistance, good rate capability, and good cycle life, and possessed a higher specific surface area and electroactive area than pure CoMoO₄, promoting the acquisition of OH⁻ and fast charge transfer [94]. For N-doped graphene, the N atoms inserted in the carbon lattice played a significant role in improving the capacitance and stability of the electrode material because of the local structural deformation around N atoms as well as the Coulomb effect [95,96].
Co-incorporated NiV$_2$O$_6$/Ni(HCO$_3$)$_2$ nanosheet arrays directly grown on Ni foam possessed long-term durability, with 106.2% retention of the initial capacity after 10,000 charging/discharging cycles at 100 mA cm$^{-2}$. The electrons transferred from the V center to the Ni active sites because of the synergistic contribution of the individual components, making the structure more stable [97]. Composite materials with multiple metal components exhibit high specific capacitance, good rate capability, and improved cycling stability, profiting from the high specific surface area, plentiful surface-active sites, good interfacial conductivity, and porous structure [98–101].

Building composite materials with a well-layered core–shell structure is conducive to enhancing electrochemical performance. Because of the synergistic contribution between the core and shell, the high specific surface area provides more electroactive sites for Faradaic redox reactions and increases ion and electron diffusion; thus, composite materials with core–shell structures exhibit satisfactory electrochemical performance [102–104].

### Table 4. Properties of composite materials for supercapacitors prepared by various methods.

| Materials                        | Method              | Specific Capacitance Density | Specific Power Density | Specific Energy Density | Specific Capacitance Retention | Ref.          |
|---------------------------------|---------------------|------------------------------|------------------------|-------------------------|-------------------------------|--------------|
| Carbon-ruthenium xerogels       | Sol-gel             | 256 F·g$^{-1}$               | -                      | -                       | Almost 100% (>2000 cycles)    | [76]         |
| RuO$_2$/CNT                     | -                   | 340 F·g$^{-1}$               | -                      | -                       | -                             | [77]         |
| RuO$_2$·xH$_2$O/CB              | Novel incipient wetness | 647 F·g$^{-1}$           | -                      | -                       | -                             | [78]         |
| Co, Mn, Cu, Fe, Zn-doped carbon aerogels | Impregnation  | 100 F·g$^{-1}$ (Co)          | 107 F·g$^{-1}$ (Mn)    | -                       | 94% (after 20,000 cycles)    | [79]         |
| MoO$_3$/AC                      | Impregnation        | 177 F·g$^{-1}$               | -                      | -                       | -                             | [80]         |
| ITO/AC                          | Reverse precipitation | -                            | -                      | -                       | -                             | [81]         |
| WO$_3$/CA carbon aerogel        | Immersion-calcination | 700 F·g$^{-1}$           | -                      | -                       | 95% (after 4000 cycles)       | [82]         |
| Graphene-MnO$_2$                | Self-limiting deposition | 310 F·g$^{-1}$              | -                      | -                       | -                             | [83]         |
| Mesh-like Fe$_2$O$_3$/C          | Template free greener | 315 F·g$^{-1}$              | -                      | 37 Wh·kg$^{-1}$          | 88.9% (after 1500 cycles) 91% (after 1000 cycles) | [84]         |
| CuO-NC                          | Hard templating     | 300 F·g$^{-1}$               | -                      | -                       | -                             | [85]         |
| Co(OH)$_2$/USY                  | Chemical precipitation | 958 F·g$^{-1}$              | -                      | -                       | -                             | [86]         |
| NiO/CNT                         | Hydrothermal        | 1329 F·g$^{-1}$              | -                      | -                       | -                             | [87]         |
| Graphene/Co$_3$O$_4$ nanowire   | Hydrothermal        | 1100 F·g$^{-1}$              | -                      | -                       | -                             | [88]         |
| Graphene sheets/Ag$_2$S         | Solvothermal        | 1063 F·g$^{-1}$              | -                      | -                       | -                             | [89]         |
| NiS$_2$/MWCNT                   | Hydrothermal        | 800 F·g$^{-1}$               | 798 W·kg$^{-1}$         | 19.8 Wh·kg$^{-1}$         | 90% (after 5000 cycles)       | [90]         |
| α-Ni(OH)$_2$-GO                 | Hydrothermal        | 1760.7 F·g$^{-1}$            | -                      | -                       | -                             | [91]         |
| BiMn$_2$O$_3$-MWCNT             | Hydrothermal        | 540 F·g$^{-1}$               | 3.6 kW·kg$^{-1}$        | 13 Wh·kg$^{-1}$          | -                             | [92]         |
| Co-Al LDH-NS/GO                 | LBL                 | 880 F·g$^{-1}$               | -                      | -                       | -                             | [93]         |
| CoMoO$_4$/graphene              | Hydrothermal        | 394.5 F·g$^{-1}$             | 197.2 W·kg$^{-1}$       | 54.8 Wh·kg$^{-1}$        | -                             | [94]         |
3. Materials Based on CC

A summary of the electrochemical performance of electrode materials based on CC is provided in Table 5. Rowlands et al. [105] concluded that CC as an electrode exhibits a specific capacitance of 35 F·g⁻¹. It is worth mentioning that Dai et al. [106] developed hierarchical porous hollow CC as an electrode material for organic-electrolyte supercapacitors, overcoming the shortcomings of pure CC with superior stability (98% capacitance retention over 20,000 cycles). Compared with electrode materials without a substrate, the advantages of materials based on CC are reflected in the following points. The compound can grow in situ on CC and extend outward, and the growth is more tortoiser-like αNiCo carbon nanoparticles all improved the electrochemical performance [109]. The hierarchical αNiCo carbon nanoparticles, and numerous mesopores between the CC is shown in Figure 2d. The hollow porous structure of the tortoise-shell-like structure αS/ACC//AC asymmetrical flexible supercapacitor exhibited good cycling stability (82% and a high specific capacitance of 2392 F·g⁻¹ of commercial CC, with an area of 1 cm²; the obtained ultra-thin nanosheet arrays with a mesoporous structure exhibited good conductivity with abundant active sites, resulting in enhanced electrochemical performance and a high specific capacitance of 2392 F·g⁻¹ at a current density of 1 A·g⁻¹. A Ni–Co–S/ACC//AC asymmetrical flexible supercapacitor exhibited good cycling stability (82% retention after 10,000 cycles). The tortoise-shell-like structure α-Fe₂O₃/C nanoarray on CC is shown in Figure 2d. The hollow porous structure of the α-Fe₂O₃/C nanoarray, carbon nanoparticles, and numerous mesopores between the α-Fe₂O₃ nanocrystals and carbon nanoparticles all improved the electrochemical performance [109]. The hierarchical NiCo₂S₄@NiCo₃S₄ core/shell nanoarrays grown on CC shown in Figure 2e and hierarchical tectorum-like α-Fe₂O₃/PPy nanoarrays grown on CC shown in Figure 2f exhibited areal

| Materials                                      | Method                     | Specific Capacitance Density | Specific Power Density | Specific Energy Density | Specific Capacitance Retention | Ref. |
|------------------------------------------------|----------------------------|------------------------------|------------------------|-------------------------|--------------------------------|------|
| NG-NiMnO₃                                      | Hydrothermal               | 750.2 F·g⁻¹                 | 2427 F·g⁻¹             | 4.983 mW·cm⁻²           | 0.415 mWh·cm⁻²                  | [95] |
| Co₃S₄-NG                                       | Hydrothermal               |                              |                        |                         |                                | [96] |
| NiV₂O₅/Ni(HCO₃)₂ nanoflake arrays              | Hydrothermal               | 7.94 F·cm⁻²                 | 4.983 mW·cm⁻²           | 0.415 mWh·cm⁻²           | 106.2% (after 10,000 cycles)    | [97] |
| Mn/PbO₄                                       | Chemical reduction         | 185 F·g⁻¹                   | 210 F·g⁻¹              |                         |                                | [98] |
| Mn/NiO₅                                       | Electrochemical deposition | 1144 F·g⁻¹                  |                        |                         |                                | [99] |
| NiMoO₄/CoMoO₄ nanorods                         | Hydrothermal               | 1164 F·g⁻¹                  | 3750 W·kg⁻¹            | 17.5 Wh·kg⁻¹            | 87.5% (after 3000 cycles)       | [100]|
| Ag QDs/NaMoO₄                                  | Dipping and drying         | 3342.7 F·g⁻¹                | 212.5 kW·kg⁻¹          | 48.5 Wh·kg⁻¹            |                                | [101]|
| Ppy®NiCo₂S₄ core-shell heterostructure 3DN      | Hydrothermal               | 908.1 F·g⁻¹                 | 160 W·kg⁻¹             | 50.82 Wh·kg⁻¹           | 126.6% (after 2000 cycle)       | [102]|
| Co@Co₃O₄ core-shell 3DN                        | Surface oxidizing          | 1049 F·g⁻¹                  |                        |                         |                                | [103]|
| Core-shell hollow CoMoS₄@Ni-Co-S nanotubes      | Electrodeposition          | 2208.5 F·g⁻¹                | 800 W·kg⁻¹             | 49.1 Wh·kg⁻¹            | 90.3% (after 10,000 cycles)     | [104]|

It is worth mentioning that Dai et al. [106] developed hierarchical porous hollow N-doped CC as an electrode material for organic-electrolyte supercapacitors, overcoming the shortcomings of pure CC with superior stability (98% capacitance retention over 20,000 cycles). Compared with electrode materials without a substrate, the advantages of materials based on CC are reflected in the following points. The compound can grow in situ on CC and extend outward, and the growth is more uniform, resulting in a larger surface area and porous structure, shortening the ion-diffusion channel, accelerating the transmission path of electrons and ions, accelerating the Faraday capacitance reaction, and enhancing the electrolysis of liquid and electrode materials. CC also exhibits good flexibility and plasticity, as observed in the digital photograph of commercial CC, with an area of 1 × 4 cm² (Figure 2a). As shown in Figure 2b, Liu et al. [107] synthesized uniform honeycomb-like NiCo₂S₄ nanosheets grown on CC with an excellent capacitance up to 1638 F·g⁻¹ at 1 A·g⁻¹. Graphene quantum dots on carbon cloth can somewhat improve specific capacitance over carbon cloth and maintain perfect flexibility, but its performance is far from adequate [10]. Zhao et al. [108] prepared a metal–organic framework using an etching/ion-exchange method, as shown in Figure 2c; the obtained ultra-thin nanosheet arrays with a mesoporous structure exhibited good conductivity with abundant active sites, resulting in enhanced electrochemical performance and a high specific capacitance of 2392 F·g⁻¹ at a current density of 1 A·g⁻¹. A Ni–Co–S/ACC//AC asymmetrical flexible supercapacitor exhibited good cycling stability (82% retention after 10,000 cycles). The tortoise-shell-like structure α-Fe₂O₃/C nanoarray on CC is shown in Figure 2d. The hollow porous structure of the α-Fe₂O₃/C nanoarray, carbon nanoparticles, and numerous mesopores between the α-Fe₂O₃ nanocrystals and carbon nanoparticles all improved the electrochemical performance [109]. The hierarchical NiCo₂S₄@NiCo₃S₄ core/shell nanoarrays grown on CC shown in Figure 2e and hierarchical tectorum-like α-Fe₂O₃/PPy nanoarrays grown on CC shown in Figure 2f exhibited areal
capacitances as high as 3.9 F·cm$^{-2}$ at 1 mA·cm$^{-2}$ and 382.4 mF·cm$^{-2}$ at 0.5 mA·cm$^{-2}$, respectively [110,111]. Besides above advantages, electrode materials based on CC have more active sites, increasing the energy storage and capacitance [112–114].

The use of base materials such as CC for constructing nano-sized forms is an effective way to improve the electrochemical performance of electrode materials by shortening the length of the ion-diffusion channel [115–120]. Horng et al. [121] prepared nano-sized polyaniline nanowires on CC (PANI-NWs/CC) via an electrochemical method; the materials exhibited specific capacitance up to 1220 F·g$^{-1}$ while overcoming the cycle degradation issues caused by mechanical issues. The one-dimensional structure of the metal nanowires as the supporting framework of the fixing material effectively produced a porous structure and improved the performance and stability of the materials with good conductivity and fast charge transport [122,123].

The preparation methods of electrode materials consist of electrochemical deposition, chemical deposition, dip dyeing, and hydrothermal methods. Compounds obtained under hydrothermal conditions can overcome the hard agglomeration of certain high-temperature preparation methods. The characteristics of hydrothermally produced materials include high particle purity, small size (nano-level), good dispersion, uniform distribution, no agglomeration, good controllability, low production cost, and environmentally friendly reaction conditions and atmosphere. In addition, from the perspective of environmental protection, the hydrothermal method is a mild, green, low-cost, and easy-to-operate preparation method. It has become an important choice for the preparation of electrode materials after ongoing improvement.

Figure 3 presents SEM images of multiple metal sulfides based on CC prepared via the hydrothermal method; a sea-urchin-like Ni–Co–S compound on CC with a high surface area and many active sites is displayed in Figure 3a. However, it can be observed that the high surface energy results in local agglomeration. After improving the experimental conditions, a good structure of nano-needle nickel cobalt sulfide growing on CC with excellent electrochemical performance was obtained (Figure 3b). In Figure 3c, a small amount of Fe element was added under the same experimental atmosphere as Figure 3b; the Ni–Co–Fe–S nanoparticles are uniformly distributed on the carbon cloth with scarce agglomeration of large particles.

Figure 2. (a) Digital photograph of commercial CC with area of 1 × 4 cm$^2$; (b–f) SEM images of several compounds with different structures grown on CC [107–111]. Adapted from [108–111], with permission from Elsevier, 2020.
Figure 3. SEM images of (a) sea-urchin-like Ni–Co–S compound on CC, (b) Ni–Co–S nano-needle on CC, and (c) Ni–Co–Fe–S nanoparticles on CC.

Table 5. Properties of materials based on CC for supercapacitors prepared by various methods.

| Materials                          | Method                                         | Specific Capacitance Density | Specific Power Density | Specific Energy Density | Specific Capacitance Retention | Ref.   |
|-----------------------------------|------------------------------------------------|------------------------------|------------------------|-------------------------|--------------------------------|--------|
| CC                                | Electrochemical anodization                    | 35 F·g⁻¹                    | -                      | -                       | -                              | [105]  |
| N-doped activated CC              | One-step etching & doping (E&D)                | 215.9 F·g⁻¹                 | -                      | -                       | 98% (20,000 cycles)            | [106]  |
| Graphene                          | Peroxide-assisted hydrothermal                 | 70 mF·cm⁻²                  | -                      | -                       | -                              | [10]   |
| Quantum Dots (GQD)/CC             | NiCo₂S₄/CC Hydrothermal                        | 1638 F·g⁻¹                  | 799.6 W·kg⁻¹           | 25.2 Wh·kg⁻¹            | -                              | [107]  |
| Ni-Co-S/ACC                       | Hydrothermal                                   | 2392 F·g⁻¹                  | 800.2 W·kg⁻¹           | 30.1 Wh·kg⁻¹            | 82% (10,000 cycles)           | [108]  |
| α-Fe₂O₃/C nanoarrays on CC        | Hydrothermal                                   | 391.8 F·g⁻¹                 | -                      | -                       | 91.8% (4000 cycles)           | [109]  |
| NiCo₂S₄@NiCo₂S₄ on CC             | Hydrothermal                                   | 3.9 F·cm⁻²                  | -                      | -                       | -                              | [110]  |
| α-Fe₂O₃/PPy nanoarrays on CC      | Hydrothermal and in situ vapor-phase polymerization | 382.4 mF·cm⁻²               | -                      | -                       | -                              | [111]  |
| Hierarchical Co(OH)₂@NiMoS₄ on CC| Hydrothermal                                   | 2229 F·g⁻¹                  | 1000 W·kg⁻¹            | 159.5 Wh·kg⁻¹           | 100% (5000 cycles)            | [112]  |
| ZIF–67 on CC                      | Dipping                                       | 829 F·g⁻¹                   | -                      | -                       | 103% (15,000 cycles)          | [113]  |
| ROCC@PDAA on CC                   | In-situ chemical oxidation polymerization      | 81.9 F·g⁻¹                  | -                      | -                       | 159% (20,000 cycles)          | [114]  |
| NiO nanoflake arrays on CC        | CBD                                           | 660 F·g⁻¹                   | -                      | -                       | 82% (4000 cycles)             | [115]  |
| MoO₃ film on CC                   | Electrodeposition                             | 835 F·g⁻¹                   | 1000 W·kg⁻¹            | 78 Wh·kg⁻¹              | 98% (8000 cycles)             | [116]  |
| MoS₂ nanospheres on CC            | Hydrothermal                                   | 368 F·g⁻¹                   | 128 W·kg⁻¹             | 5.42 Wh·kg⁻¹            | 96.5% (after 5000 cycles)     | [117]  |
Table 5. Cont.

| Materials | Method | Specific Capacitance Density | Specific Power Density | Specific Energy Density | Specific Capacitance Retention | Ref. |
|-----------|--------|-------------------------------|------------------------|-------------------------|---------------------------------|------|
| Ni(OH)$_2$ on CC Co(OH)xCO$_3$ on CC | Hydrothermal | 789 F·g$^{-1}$<br>550 F·g$^{-1}$ | 1.4 kW·kg$^{-1}$<br>33.48 Wh·kg$^{-1}$ | 98%<br>97.6% (after 5000 cycles) | [118] |
| P-doped NiCo$_2$S$_4$ nanotube arrays on CC | Hydrothermal | 8.03 F·cm$^{-2}$ | 750 W·kg$^{-1}$ | 42.1 Wh·kg$^{-1}$ | 87.5% (5000 cycles) | [119] |
| NCLP@NiMn-LDH on CC | Hydrothermal | 2318 F·g$^{-1}$ | 750 W·kg$^{-1}$<br>12.1 kW·kg$^{-1}$ | 42.2 W·h·kg$^{-1}$<br>100.9 Wh·kg$^{-1}$ | -<br>- | [120] |
| PANI-NWs/CC | Electrochemical polymerization | 1079 F·g$^{-1}$<br>1133.3 mF·cm$^{-2}$ | -<br>-<br>-<br>- | -<br>-<br>-<br>- | 80.47% (2000 cycles) | [121] |
| NC LDH | Electrochemical deposition | -<br>-<br>-<br>- | -<br>-<br>-<br>- | -<br>-<br>-<br>- | -<br>-<br>-<br>- | -<br>-<br>-<br>- | [122] |
| Ni-Co LDH Nanoflakes–ZnO nanowires hybrid array on CC | Hydrothermal | 927 F·g$^{-1}$<br>46.15 kW·kg$^{-1}$<br>45.55 Wh·kg$^{-1}$ | 96.02% (3000 cycles) | [123] |

4. Main Existing Issues, Corresponding Solutions, and Future Work

Although electrode materials exhibit great potential due to low cost, high performance, and good flexibility, many unanswered questions remain. Among the electrode materials, carbon materials with multidimensional pore characteristics, graphene, and atom-doped carbon materials have developed dramatically. In the process of basic research on electrode materials, various advanced design concepts have driven the rapid development of related fields and industries.

However, low load is completely inadequate to meet the needs of commercial electrodes. Therefore, increasing the active material load of the electrode and further exploring and optimizing its electrochemical performance will provide a reference and guide for its future practical application.

Advanced technology was used to reveal the phase evolution law of materials and the structure-effect relationship between structure and properties, so as to guide the design and construction of electrode materials. We can see the future trend of electrode materials from bionic carbon materials, but the huge technical challenges are still difficult problems.

As a substrate growth material to construct a variety of three-dimensional structural composite materials, especially with the redox properties of transition metals to build porous structures with high specific surface area, the performance of composite material-based carbon materials are surprising.

For three-dimensional composite materials with a controllable nanostructure based on CC, the most important thing is that they provide more active sites and exhibit excellent electrochemical properties [124–127]. We can see that metal composites exhibit oxidative performance, which is the advantage that other materials cannot achieve. However, the controllability and stability of the structure are large problems. When the growth dimension reaches nano-level, especially, the structure has a larger surface area, there are more active sites, and at the same time the surface energy is greater, agglomeration is easier, and the structure can be more easily damaged. The hydrothermal method not only does not destroy the structure and performance of the CC, but also provides a mild environment for the growth of composite material based on CC, and three-dimensional nano-structures exhibit more possibilities and potential.
5. Conclusions

Supercapacitors have the advantages of high power density, fast charge and discharge rates, long cycle life, high safety, and reliability as a new generation of green energy materials to replace traditional energy materials. The hydrothermal method has the characteristics of simplicity, environmental friendliness, and uniform growth of the load. Nanostructures can be grown directly on CC via a hydrothermal method, including nano-needles, tree-like and nano-flower shapes, ultra-thin nanosheets, and tectorum-like shapes, which have high surface areas and porous structures, shorten the transmission channels of ions and electrons, and increase the active sites for the Faraday capacitance reactions. The synergistic effect between the components also effectively improves their specific capacitance, cycle stability, and rate capability. In addition, flexible supercapacitors assembled with electrode materials based on CC have great advantages for the energy supply for wearable devices.

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