

A Review on Modified Carbon Cloth-Based Electrode for Supercapacitor

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ABSTRACT

To meet human needs on a global and financial scale, energy storage is essential. Supercapacitors (SCs) have become a focal point of attention because of their improved electrochemical capabilities, extended cycling durability, elevated specific power, and swift charge/discharge rates, outperforming conventional capacitors and batteries. Many compounds, including as metal oxides, carbon-based materials, and metal-organic frameworks (MOFs), have been researched as possible supercapacitor electrode materials. The current review article covers conductive polymers, conductive polymer composites, and carbon materials used in supercapacitors. The development process, characteristics, and uses of different electrode materials such as activated carbon, metal or metal oxide, graphene or graphene oxide, and conductive polymers based on carbon cloth electrodes for supercapacitors are also covered in the current review article.

Keywords: Supercapacitors, Electrode materials, Conductive polymer, Hybrid materials, Carbon cloth.

INTRODUCTION

The increasing need to create renewable and eco-friendly energy sources, such as solar energy, stems from the extensive use of limited fossil fuel reserves, contributing to environmental pollution and energy challenges [1, 2]. The sun's strength, however, differs from place to place on Earth; it is strongest at the equator and is weaker as one approaches the Polar Regions. It also changes with the time of day; for example, it is stronger in the afternoon and unavailable at night [3, 4]. Devices that use wind energy or other renewable energy sources that produce inconsistent power also have similar problems. This could result in an inadequate power supply, which could cause electrical appliances to malfunction and make it more difficult to provide a consistent power supply across longer distances. Storing energy from fluctuating renewable sources in electrical or electrochemical formats is crucial for efficient supply and utilization, aiming to mitigate associated issues. These considerations have sparked significant interest in technologies such as supercapacitors and batteries, which excel in storing electrical and electrochemical energy derived from renewable sources [5].

Supercapacitors (SCs) are categorized into three classifications depending on how they store energy: electrostatic double layer capacitors, pseudo-capacitors, and hybrid capacitors [6]. Electrostatic double layer

capacitors consist of an electrolyte, two carbon-based electrode materials, and a separator. These capacitors can store charges either electrostatically or through a non-Faradic process that does not involve charge transfer between the electrolyte and electrode [6, 7]. Electrostatic double layer capacitors utilize the principle of the electrochemical double layer for energy storage, while pseudo-capacitors employ electrodes made of metal oxides or conducting polymers with significant electrochemical pseudo-capacitance. Charge storage in pseudo-capacitors occurs through a Faradaic mechanism, such as oxidation-reduction reactions, which involve the transfer of charges between the electrolyte and electrode [6, 8]. Hybrid capacitors utilize electrodes with an asymmetrical configuration, where one electrode primarily exhibits electrostatic capacitance, while the other demonstrates electrochemical capacitance. These hybrid capacitors represent a fusion of performance characteristics that were previously inaccessible, combining the strengths of both pseudo-capacitors and electrostatic double layer capacitors. By integrating the desirable traits of pseudo-capacitors and electrostatic double layer capacitors, hybrid capacitors form a unified supercapacitor with enhanced capabilities [6].

Other terms for supercapacitors that are used in different situations include large-capacitors, energy-storage, electric double-layer, and farad capacitors. Exploring carbon materials derived from naturally available lignocellulose biomass as electrode materials for flexible or wearable electronic devices represents a forthcoming fashion that has been under development for many years, albeit with existing technological challenges yet to be overcome [9, 10]. Several carbon-based materials, such as graphene integrated with carbon fabric, activated carbons (AC), and carbon with nitrogen doping, have been extensively studied due to their widespread availability and cost-effectiveness [11, 12]. They do not, however, have a really strong performance. Researchers are becoming more aware of the practical uses of supercapacitors. An electrode assembly, an electrolyte solution, and a separator make up a supercapacitor. The type of electrode material used has a big influence on how well supercapacitors work [9, 13]. A wide variety of composite materials have been created, encompassing advanced porous three-dimensional structures, which include carbon-based materials [14], carbon materials doped with elements [15], metallic compounds [16], multi-metal compounds [17], and polymers [18]. An increasing number of wearable, highly flexible electrodes with strong plasticity have been sought for as civilization has advanced. Carbon cloth has garnered acclaim as an excellent electrode material, offering high performance while being cost-effective, flexible, and highly conductive [9, 19–21]. The research interest in carbon cloth (CC) has surged due to its suitability as an electrode material in flexible supercapacitors, particularly for applications in wearable and portable devices. Electrode materials for flexible supercapacitors based on CC show great promise. CC offers many benefits, including high conductivity, porosity, big surface area, low weight, high cost, and outstanding flexibility [9, 22]. It is also very simple to create an electrode material with structural advantages [9].

Polyaniline (PANI) stands out as a commonly utilized material in pseudo-capacitors (PCs), known for its excellent conductivity among conductive polymers. Its widespread adoption is due to easy synthesis, affordability, remarkable conductivity, distinctive doping mechanism, high theoretical specific capacitance (around 1200 F g^{-1}), impressive wave absorption capabilities, and outstanding electrochemical performance [23]. Since its first synthesis in 1886, PANI has established itself as the leading conductive polymer with promising attributes [23, 24]. An aniline monomer is oxidized by an electrochemical or chemical process to produce PANI [25]. Aniline, a prominent amine, finds extensive use in dye production, pharmaceuticals, resin manufacturing, and as a rubber vulcanization accelerator. Consequently, PANI emerges as the most cost-effective and thermally stable inherently conductive polymer [23, 26]. The utilization of PANI as a common material in pseudo-capacitors has led to significant advancements in supercapacitor research, leveraging its aforementioned benefits [23, 27]. PANI exhibits a considerable total surface charge potential during charge and discharge cycles as it transitions between different oxidation states, contributing to its high capacitance. However, extended charging/discharging periods can lead to structural degradation of the main chain in PANI, causing expansion, contraction, or degradation, ultimately affecting conductivity and stability negatively [26]. A promising approach involves the design of structured PANI nanorods and

nanoparticles to mitigate these issues [23, 28]. Compared to randomly arranged structures, well-organized PANI nanostructures demonstrate enhanced rate capability, increased specific surface area, excellent cycle stability, and superior energy storage capacity [23, 29]. Additionally, significant research efforts have focused on synergistic composite strategies involving PANI and other active materials. By addressing PANI's stability concerns, increasing metal oxide conductivity, and improving the specific capacitance of carbon materials, this strategy hopes to boost overall electrochemical performance [23]. To achieve these qualities in all composites with PANI, carbon cloth materials are one of the possible materials. PANI and CC-based electrodes together provide a high-performance energy storage device, for this reason. The conductive polymers, conductive polymer composites, and carbon materials utilized in supercapacitors are the main topics of this review. As indicated in Table 2, PANI-containing composite materials are presented in the field of supercapacitors in this article. Lastly, Table 3 illustrates the improved carbon cloth-based supercapacitor electrode that is included in this research.

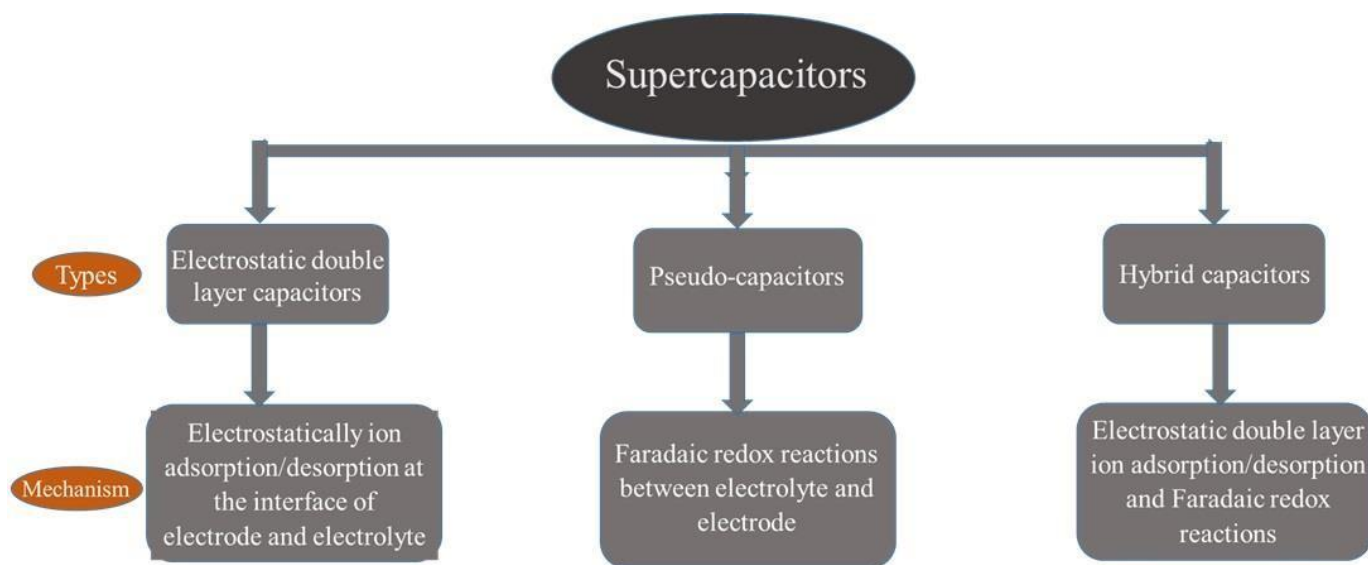


Fig. 1: Types of supercapacitors according to the energy storage mechanism [6].

ELECTRODE MATERIALS

Carbon Materials in Supercapacitor

Carbon materials were among the earliest materials investigated and utilized as electrodes, particularly in supercapacitors [9]. Beck et al., (2001) [30] pioneered the development of industrial carbon black (CB) electrodes in 12M H_2SO_4 using polytetrafluoroethylene (PTFE) as a binder, achieving the highest observed capacitance of $250 F g^{-1}$. Li et al., (2002) [31] combined cresol, catechol, and formaldehyde to create mixed carbon aerogels with a high specific capacitance of $77 F cm^{-3}$ ($104 F g^{-1}$), attributed to their highly porous structure. Additionally, Pan et al., (2007) [32] discovered that multi-walled carbon nanotubes with an average outer diameter of 50 nm and an internal diameter ranging from 3 to 10 nm displayed a specific capacitance of $135 F g^{-1}$ due to their unique structural characteristics.

The most attractive aspect of CC, as opposed to other carbon materials, is its remarkable mechanical strength combined with its increased flexibility and mechanical integrity. Moreover, CC can be incorporated

into microdevices of various forms and sizes in addition to being used directly as a flexible, freestanding electrode for supercapacitors. Yu et al., (2015) synthesized a high-rate fiber-shaped supercapacitor utilizing activated carbon, which has the potential to be sewn into a glove or woven into a dragonfly knot [33, 34].

Conductive Polymers

Research in the field of conductive polymers has experienced significant growth. Arbizzian et al., (1996) [35] developed three types of supercapacitors: a symmetric supercapacitor utilizing p-doped poly(pyrrole), an asymmetric supercapacitor based on poly(3-methylthiophene), and p- and n-doped poly(dithieno [3,4-b:3',4'-d] thiophene) with a high working potential. Fusalba et al., (1999) [36] developed poly(cyclopenta [2,1-b;3,4-b'] dithiophen-4-one) (PCDT) with an open and porous structure, exhibiting a low-frequency capacitance of approximately 70 F g^{-1} for both the p-type and n-type doped states. However, despite their advantages, conductive polymers face challenges such as limited cycle stability and mechanical strength over extended periods. Strategies like optimizing their structure, and form, or integrating them with other carbon materials are crucial for overcoming these drawbacks and enhancing their performance [9]. Because of their wide surface area and micropore distribution, polyacrylonitrile (PAN) microcellular foam thin films work exceptionally well on carbon fibers, as demonstrated by Gouerec et al., (2001) [37]. Using functionalized carbon nanotubes (f-CNTs) as electrode materials, Jyothibasud et al., (2020) [38] fabricated polypyrrole (PPy) tubes through one-step in situ chemical oxidative polymerization combined with curcumin as a template.

Table 1. The capacitance performance and capacitance retention rate of pure conductive polymers at 0.3 A g^{-1} reported at [39]

Conducting Polymer	Capacitance (F g^{-1})	Capacitance Retention Rate (%)
PANI	642.6	64.0
PPy	586.2	56.4
PTh	697.1	51.6
PEDOT	718.2	54.3

Conductive Polymer Composite for Supercapacitor Application

Conductive polymer composites, with a distinctive combination of electrical conductivity, processing simplicity, and flexibility, have generated a lot of interest for their possible use in supercapacitors. Supercapacitors, sometimes referred to as electrochemical capacitors or ultracapacitors, are energy-storage devices that fill the gap between batteries and conventional capacitors with their high power density and quick charge/discharge times. For a number of reasons, conductive polymer composites are important in the field of supercapacitor applications. The conductive polymers such as polyaniline (PANI), polypyrrole (PPy), and polythiophene (PTh) are commonly employed due to their favorable electrochemical properties and electrical conductivity characteristics [40, 41]. The scientist will gain from choosing suitable conductive polymers to produce high-performance supercapacitor applications. Composite structures that enhance both electrical conductivity and capacitance of the composite material are typically created by blending conductive polymers with other substances such as metal oxides or carbon-based materials (like carbon nanotubes, graphene, and carbon black). This combination results in improved electron transport speed and enhanced charge storage capacity compared to pure conductive polymers, leading to better electrochemical

performance [41, 42]. Xu et al., (2020) [39] synthesized a ternary composite supercapacitor using zinc sulfide/reduced graphene oxide (ZnS/RGO) via a hydrothermal technique, then incorporating various conductive polymers (PANI, PPy, PTh, and PEDOT) through in situ polymerization. They subsequently assessed the capacitor's reliability and capacitance performance. The results showed a remarkable enhancement, with values reaching 160% at 1 A g^{-1} and 1045.3 F g^{-1} after 1000 cycles, along with power densities of 18.0 k W kg^{-1} and $349.7 \text{ W h kg}^{-1}$. Among these, the PANI ZnS/RGO ternary electrode composite exhibited the most superior cycle stability and capacitance performance, as concluded from the findings [41].

CHARACTERISTICS AND USES OF VARIOUS COMPOSITE MATERIALS FOR ADVANCED SUPERCAPACITORS

Activated Carbon/Polyaniline Hybrids

Activated carbon (AC) is mainly non-crystalline and contains a minimum of 80% carbon content, making it a carbon-based material. Amorphous carbon's intrinsically low conductivity, but it will inevitably impede its advancement in the realm of energy storage. Researchers typically employ high-temperature local graphitization and synthetic graphite coatings to increase amorphous carbon's conductivity to increase the applicability of ACs [23, 43]. Due to its huge specific surface area, AC is generally well-suited for the adsorption of solid particles from gases and liquids [44]. Prior research has indicated that the coupling of PANI and AC in composite materials results in a favorable combination of increased PANI pseudo-capacitance and considerable carbon porosity [23, 45–46]. An inevitable challenge in developing conductive polymer materials lies in the interface resistance within nanostructures [47]. To get over these drawbacks, numerous researchers have cleverly mixed PANI molecules with AC materials, making them the most promising options for supercapacitors. Lyu et al., (2018) [48] synthesized carbon spheres by utilizing yeast as a structural template and carbon source. The research utilized in situ polymerization to produce hybrids of yeast-derived N-doped carbon microspheres (YCs) combined with PANI. The resulting YCs' electrical conductivity is excellent. In a three-electrode setup, the YC/PANI composite demonstrated a significant specific capacitance of 500 F g^{-1} at a current density of 1 A g^{-1} .

Table 2. The electrochemical behavior of different supercapacitors made using PANI composite materials.

Material	Electrode system	Specific capacitance	Discharge specific capacitance	Cycle stability	Cycles	Current density	Charge-transfer resistance	Capacitance retention	Ref.
ZnS/RGO/PANI	3	1045.3 F g^{-1}	1662 F g^{-1}	160%	1000	1 A g^{-1}	0.21Ω	63.9%	39
GO/PANI	2	425 F g^{-1}	–	83%	500	0.2 A g^{-1}	–	83%	49
PANI/ Zn^{2+}	3	738 F g^{-1}	–	65.4%	500	5 mA cm^{-2}	0.3Ω	–	50
rGO/ Fe_3O_4 /PANI	3	283.4 F g^{-1}	–	78%	1000	1 A g^{-1}	–	–	51
SnS_2 /NRGO/PANI	3	1021.7 F g^{-1}	–	82%	1000	1 A g^{-1}	–	71.4%	52
O- MoS_2 /PANI/rGO-160 HNSs	3	752.0 F g^{-1}	–	–	50,000	1 A g^{-1}	–	80.4 %	53
PANI-S-a/GO-10%	–	535 F g^{-1}	–	–	10,000	0.5 A g^{-1}	–	93.6%	54

Graphene or Graphene Oxide/Polyaniline Hybrids

Graphene is the thinnest known two-dimensional material with a unique honeycomb lattice structure composed of carbon atoms in the sp^2 hybrid orbital [55]. Due to its distinctive crystal arrangement, graphene exhibits remarkable characteristics like high electron mobility ($\sim 2.5 \times 10^5 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$), a high Young's modulus ($\sim 1 \text{ TPa}$), excellent physicochemical stability, and exceptional tensile strength ($\sim 42 \text{ N m}^{-1}$). These characteristics make it highly promising for a wide range of applications including biomaterials, energy storage, sensing, semiconductors, and medicine, positioning it as a revolutionary material [56].

To create a hierarchical nanostructure, Ye et al., (2017) [57] developed an ordered PANI nanowire array on a graphene sheet derived from a graphite substrate. The incorporation of graphene helps maintain excellent cycle stability by mitigating PANI's expansion and contraction during charge-discharge cycles. The resulting ordered PANI nanowire/graphene sheet exhibited impressive performance with a notable capacitance retention rate of 80.4% after 10,000 galvanostatic charge-discharge (GCD) cycles and a high capacitance of 3.57 F cm^{-2} (607 F g^{-1} at 1 A g^{-1}). In another study, Hong et al., (2017) [58] successfully developed three-dimensional reduced graphene oxide (rGO) using a diffusion-driven layer-by-layer technique. This three-dimensional rGO was employed as a scaffold for in situ polymerization to create the rGO/PANI composite. In a three-electrode system, the rGO/PANI composite exhibited a remarkable specific capacitance of 438.8 F g^{-1} at 0.5 A g^{-1} .

Hybrid Materials Utilizing Carbon Cloth for Supercapacitors

Carbon cloths consist of carbon fibers ranging from 5 to 10 μm in diameter [59, 60]. These materials possess excellent conductivity, flexibility, mechanical strength, hydrophobicity, cost-effectiveness, and environmental friendliness [61]. They have great promise in biological applications due to their high level of biocompatibility. Their 3D structures and enhanced ion mobility make carbon cloths desirable materials for electrode applications in batteries [59], supercapacitors [62], and solar cells [63]. Notably, carbon cloths serve effectively as supercapacitor substrates because of their high conductivity, large surface area, and facilitation of ion diffusion. These properties contribute to lower charge transfer resistance and increased specific capacitance of the electrodes. When compared to the popular substrate materials for supercapacitors, such as Ni foam and Fe nanostructures, these appear quite promising [59, 64-66]. Activated carbon cloths (ACC) were made by Degaldo et al., (2019) [67] using jute fibers and natural Henequen as precursors. Three different activation procedures, including chemical activation with ZnCl_2 and physical activation with steam and CO_2 , were studied throughout the preparation process on henequen and jute fibers. It was discovered that the distribution and pore structure of the resultant activated carbon cloths (ACCs) were influenced by the activation procedure [59]. Rowlands et al., (1999) [68] reported a specific capacitance of 35 F g^{-1} for carbon cloth (CC). Overcoming the limitations of pure CC, Dai et al., (2020) [69] developed hierarchical porous hollow N-doped CC as an electrode material for organic-electrolyte supercapacitors, which resulted in excellent stability (98% capacitance retention over 20,000 cycles). Liu et al., (2019) [70] successfully synthesized a consistent honeycomb-like NiCo_2S_4 nanosheet with outstanding capacitance of up to 1638 F g^{-1} at 1 A g^{-1} on CC. This process involves reducing the length of ion diffusion channels within base materials like CC, leading to nano-sized forms that significantly enhance the electrochemical performance of electrode materials [9, 71]. Horng et al., (2010) [72] utilized an electrochemical method to synthesize nano-sized polyaniline nanowires on carbon cloth (PANI-NWs/CC), achieving a specific capacitance of up to 1220 F g^{-1} . This approach also addressed mechanical issues leading to cycle degradation. A technique for producing a sheath-core PANI nanowire array on the surface of aligned CNF/CF yarn was reported by Mao et al., (2018) [73]. They employed electrospinning to produce the precursor of CF yarn@CNF using a solution containing 90% DMF and 10% PAN by weight. The precursor was then carbonized for two hours at 800°C in an N_2 environment to get CF yarn@CNF. This material is highly suitable for flexible electrodes. The resulting solid supercapacitor exhibited high power

density (0.52 mW cm^{-2}), energy density ($21.4 \mu\text{Wh cm}^{-2}$), and specific capacitance (234 mF cm^{-2} at 0.1 mA cm^{-2}).

Table 3. Characteristics of supercapacitor materials derived from carbon cloth (CC) using different preparation techniques.

Materials	Method	Specific Capacitance	Specific Power Density	Specific Energy Density	Specific Capacitance Retention	Cycles	Ref.
Porous graphene film by EPD/CC	Electrophoretic deposition process	27 F g^{-1} at 5 mV s^{-1}	670.0 W kg^{-1}	1.64 W h kg^{-1}	> 95%	1000	74
Graphene film by EPD/CC	Electrophoretic deposition process	9 F g^{-1} at 5 mV s^{-1}	252.2 W kg^{-1}	0.67 W h kg^{-1}	> 95%	1000	74
NG-PAA/PANI composite coating on CC	Hydrothermal reaction, <i>in-situ</i> oxidative polymerization and casting solution method	521 F g^{-1} at 0.5 A g^{-1}	1.1 kW/kg	5.8 Wh kg^{-1}	83.2%	after 2000	75
T-Fe ₂ O ₃ /PPy NAs on CC	Easy self-sacrificing template and in situ vapor-phase polymerization method	382.4 mF cm^{-2} at 0.5 mA cm^{-2}	165.6 mW cm^{-3}	0.22 mWh cm^{-3}	97.2%	after 5000	76
Hierarchical hybrid FeCo ₂ O ₄ @polypyrrole core/shell nanowires on CC	Hydrothermal reaction and oxidative polymerization process	2269 F g^{-1} at 1 A g^{-1}	–	68.8 Wh kg^{-1}	90.2%	after 5000	77
N-doped activated CC	Single-step doping and etching	215.9 F g^{-1}	–	–	–	–	69
Hierarchical Co(OH) ₂ @NiMoS ₄ on CC	Hydrothermal	2229 F g^{-1}	1000 W Kg^{-1}	159.5 Wh kg^{-1}	100%	5000	78
ACC140_Fe	Deposition method	116.8 F g^{-1}	–	–	–	–	79
Hierarchical CFC/CoFe ₂ O ₄	Mild hydrothermal	226.2 F g^{-1} at 1 A cm^{-3}	$1151.2 \text{ mW cm}^{-3}$	75.4 mWh cm^{-3}	95.8%	after 2000	80

CONCLUSIONS

This review thoroughly validates the capability of hybrid materials based on carbon cloth, demonstrating

significant performance as supercapacitors. Adding various supporting elements, such as conductive polymers, metal, metal oxide, etc., helps to improve the qualities of carbon cloth-based hybrid composites. The specific power density, specific capacitance retention, and specific capacitance of carbon cloth electrodes modified by various composite particles are all improved. This study describes how the hydrothermally synthesized hierarchical $\text{Co}(\text{OH})_2@ \text{NiMoS}_4$ on the CC electrode exhibits a good specific capacitance and 100% specific capacitance retention.

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