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REVIEW ARTICLE

Emerging Materials for Next Generation Supercapacitors: Exploring the Latest Trends and Innovations

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ABSTRACT: Supercapacitors have emerged as a leading energy storage technology due to their exceptional power density, rapid charge/discharge cycles, long operational lifespan, and environmentally friendly characteristics. Unlike traditional batteries, which rely on chemical reactions to store energy, supercapacitors store energy electrostatically, allowing for much faster energy release and recharging. These attributes make them ideal for applications where quick bursts of power are needed, such as in electric vehicles (EVs), renewable energy systems, and portable electronics. However, despite their advantages, supercapacitors face limitations, particularly in terms of energy density, which is significantly lower than that of conventional batteries, and high manufacturing costs, which restrict their commercial viability. This review aims to provide a comprehensive overview of recent advancements in supercapacitor technology, focusing on the development of novel materials and innovative device architectures that aim to address these challenges. Key materials covered include carbon-based nanomaterials (graphene, carbon nanotubes), metal oxides (MnO_2, RuO_2), conductive polymers (polyaniline, polypyrrole), and emerging hybrid materials. These materials have shown great promise in enhancing the electrochemical performance, energy density, and cycling stability of supercapacitors. Additionally, advanced fabrication techniques, such as chemical vapor deposition (CVD) and template-assisted methods, are reviewed for their potential to optimize electrode structures and lower production costs. The review also explores various applications of supercapacitors across industries, including electric vehicles, grid energy storage, aerospace, defense, and consumer electronics. By summarizing the latest trends in materials, architectures, and applications, this article serves as a valuable resource for researchers, engineers, and industry professionals looking to innovate in the field of energy storage. The review concludes by discussing the potential of supercapacitors to revolutionize energy storage systems and identifies key areas where further research and development are needed to overcome existing limitations.

Keywords: Supercapacitors, Energy Storage, Graphene, Conductive Polymers, Hybrid Materials, Carbon Nanotubes, Energy Density, Electrode Design

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1. INTRODUCTION

Supercapacitors, also known as ultracapacitors or electrochemical capacitors, represent a remarkable advancement in energy storage technology. With unique characteristics that distinguish them from conventional batteries and capacitors, supercapacitors offer high power density, rapid charge and discharge rates, and extended cycle life, making them a promising solution for a range of energy storage applications [1]. Unlike batteries, which store energy through chemical reactions, supercapacitors rely on physical processes to store and release energy. This results in faster response times, higher efficiency, and the ability to deliver large amounts of power in a short period. These attributes make supercapacitors ideal for various applications, including transportation, renewable energy systems, consumer electronics, and industrial sectors [2].

The ability of supercapacitors to deliver quick bursts of energy is particularly beneficial in high-power applications, such as regenerative braking in electric vehicles and peak power shaving in renewable energy systems. In electric vehicles, supercapacitors can capture and store the energy generated during braking, which can then be used to assist in acceleration. This not only improves energy efficiency but also extends the overall range of the vehicle. Similarly, in renewable energy systems, supercapacitors can help manage the intermittent nature of energy sources such as solar and wind power by providing rapid power storage and release capabilities. This ensures that energy is available when needed, improving the overall stability and reliability of the energy grid [3, 4].

The performance of supercapacitors is heavily influenced by the materials used in their construction, particularly the electrode materials. These materials determine the energy storage capacity, power density, and cycle stability of the device. The most commonly used electrode materials in supercapacitors include carbon-based materials, metal oxides, conductive polymers, and their composites (Figure 1). Each of these materials offers unique properties that affect the specific capacitance and charge-discharge characteristics of the device [5, 6].

Carbon-based materials, such as activated carbon, carbon nanotubes, and graphene, are widely used in supercapacitors due to their high surface area, excellent electrical conductivity, and relatively low cost [7-9]. These materials provide a large surface area for charge storage, which is crucial for achieving high capacitance. Additionally, their excellent conductivity ensures efficient charge transport, improving the overall power density of the device. However, carbon-based materials have limitations in terms of energy density, which has driven researchers to explore alternative materials.

Various metal oxides materials have been investigated as potential electrode materials for supercapacitors due to their higher energy density compared to carbon-based materials [10, 11]. These materials can store more energy per unit volume or mass, making them attractive for applications that require longer energy storage durations. However, metal oxides often suffer from poor conductivity, which can limit their power density. To address this issue, researchers have developed composite materials that combine metal oxides with carbon-based materials, resulting in improved performance [14].

Conductive polymers, such as polyaniline and polypyrrole, are another class of materials that have been explored for supercapacitor applications [12-13]. These materials offer high capacitance and good conductivity, making them suitable for use as electrode materials. However, they often suffer from poor cycle stability, which can lead to a decrease in performance over time. To overcome this limitation, researchers have developed hybrid materials that combine conductive polymers with other materials, such as metal oxides or carbon-based materials, to enhance their stability and performance.

In addition to the choice of materials, the synthesis process used to fabricate the electrode materials plays a critical role in determining their structure, morphology, and electrochemical properties [3-4]. Various synthesis techniques, such as sol-gel, hydrothermal, electrospinning, electrodeposition, and chemical vapor deposition (CVD), allow for precise control over material composition, crystallinity, particle size, and porosity. These factors directly impact the surface area, pore size distribution, and interfacial properties of the materials, which in turn influence the accessibility of electrolyte ions and the electrochemical reaction kinetics.

For example, the sol-gel method allows for the fabrication of highly porous materials with a large surface area, which is beneficial for achieving high capacitance. The hydrothermal method, on the other hand, can produce materials with well-defined nanostructures, which can improve the ion transport properties of the electrode. Similarly, electrospinning can be used to fabricate nanofibers with a high surface-to-volume ratio, which can enhance the performance of the supercapacitor by increasing the amount of charge that can be stored.

The combination of suitable materials and appropriate synthesis techniques has led to significant improvements in the performance of supercapacitors. By carefully selecting the materials and tailoring the synthesis process, researchers have been able to enhance the charge storage capacity, power delivery, cycle life, and stability of supercapacitors. These advancements have expanded the range of potential applications for supercapacitors, from high-power applications such as electric vehicles to portable electronics and renewable energy systems.

In recent years, there has been substantial progress in supercapacitor research, with a particular focus on improving energy density, which has traditionally been a limitation for this technology. While supercapacitors have historically excelled in power density, their energy density—the amount of energy stored per unit volume or mass—has been lower than that of batteries. This has restricted their use in applications that require longer energy storage durations.

To address this challenge, researchers have focused on developing new electrode materials with higher energy storage capacity.

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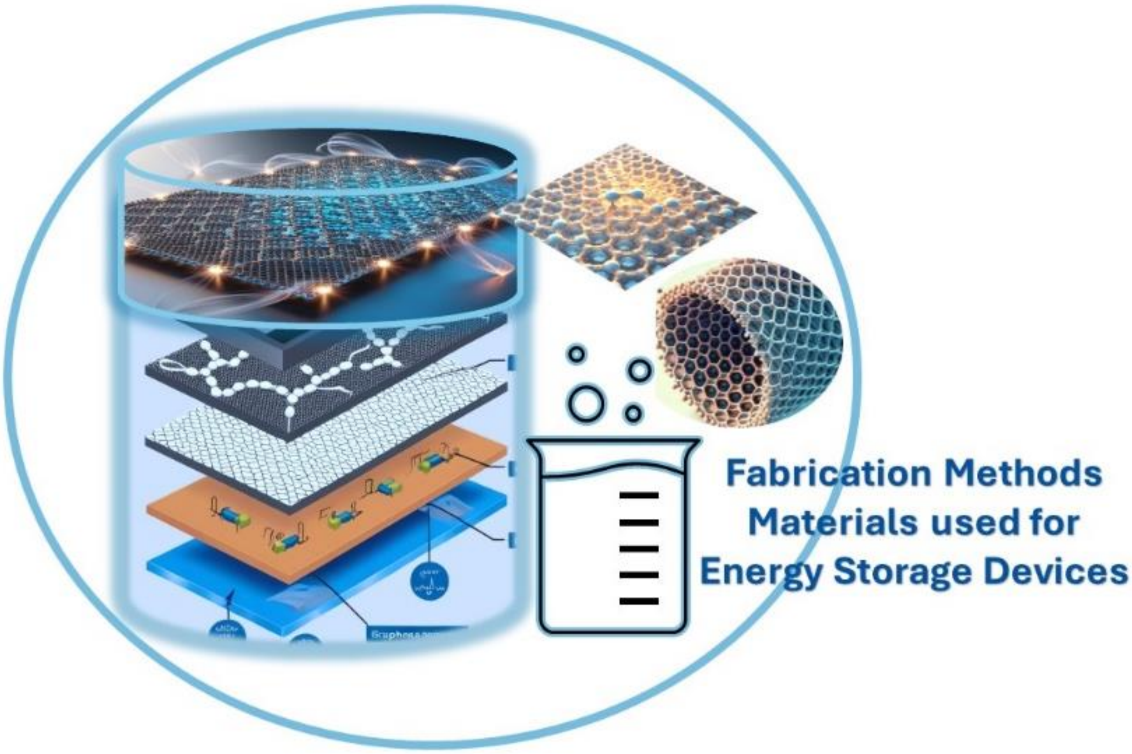

Fig. 1. Energy storage devices: Supercapacitor parameters affecting performance.

Carbon-based materials, such as graphene, have been widely explored for their potential to increase energy density due to their high surface area and excellent conductivity [7, 8]. Additionally, researchers have investigated the use of metal oxides and conducting polymers as alternative electrode materials to further improve energy storage capacity [9-13].

Novel electrode architectures have also been developed to maximize the accessible surface area, enhance ion diffusion, and improve overall electrochemical performance [17]. These architectures include hierarchical structures, nanostructured materials, and 3D frameworks, which enable increased charge storage and faster ion transport. For example, hierarchical structures combine materials with different pore sizes to optimize both ion storage and transport, resulting in higher energy density and faster charge-discharge rates.

Another area of research that has gained significant attention is the development of advanced electrolytes for supercapacitors. The choice of electrolyte plays a crucial role in determining the performance of the device, as it affects the ion transport properties and the voltage window of the supercapacitor [18-19]. Researchers have explored various electrolyte formulations, including ionic liquids, gel electrolytes, and hybrid electrolyte systems, to enhance energy density, thermal stability, and ion conductivity [20-23]. These advancements in electrolyte design have significantly improved the power delivery and overall performance of supercapacitors [24, 25].

In addition to improving the performance of standalone supercapacitors, researchers have also explored the integration of supercapacitors with other energy storage technologies, such as batteries and fuel cells, to create hybrid energy storage systems. These hybrid systems leverage the strengths of each technology, combining the high power density and fast response of supercapacitors with the higher energy density and longer duration of batteries or fuel cells. This approach offers improved energy efficiency, extended operating life, and enhanced overall performance [26].

For example, in electric vehicles, hybrid energy storage systems that combine supercapacitors and batteries can provide both high-power bursts for acceleration and longer energy storage for extended driving range. Similarly, in renewable energy systems, hybrid systems can store energy generated from intermittent sources, such as solar and wind, and provide reliable power when needed. These systems offer the potential to improve the overall efficiency and reliability of energy storage solutions, making them a compelling option for a wide range of applications.

The applications of supercapacitors continue to expand as advancements in materials and device design improve their performance. In transportation, supercapacitors are being used to enhance the efficiency of electric vehicles by providing rapid energy recovery during braking and assisting with acceleration. In renewable energy systems, supercapacitors are helping to stabilize the grid by providing

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fast-response power storage and release capabilities. They are also being used in consumer electronics, where their ability to deliver rapid bursts of power is beneficial for devices such as smartphones and laptops [27].

Moreover, supercapacitors are finding applications in aerospace and defense, where their high power density and reliability make them suitable for use in mission-critical systems. For example, supercapacitors can provide backup power for aircraft systems during takeoff and landing, ensuring uninterrupted operation in the event of a power failure. In the defense sector, supercapacitors are being used in advanced weapon systems and communication devices, where their ability to deliver high power in a short period is essential.

In this comprehensive review, we have explored the latest advancements in supercapacitor technology, focusing on electrode materials, electrolytes, device architectures, and hybrid energy storage systems. These advancements have significantly improved the energy density, power delivery, and overall performance of supercapacitors, expanding their range of potential applications. By continuing to innovate in materials design and device fabrication, researchers have the opportunity to unlock new possibilities for supercapacitors and reshape the energy storage landscape.

The future of supercapacitors is bright, with ongoing research focused on overcoming existing limitations and exploring new applications. As the demand for efficient and sustainable energy storage solutions continues to grow, supercapacitors are poised to play a critical role in meeting the energy needs of the future. This review serves as a valuable resource for researchers, engineers, and industry professionals, providing insights into the potential of supercapacitors and inspiring further advancements in this exciting field.

2. EMERGING MATERIALS FOR HIGH-PERFORMANCE SUPERCAPITORS

In recent years, the field of materials research for supercapacitors has seen substantial progress, with a growing focus on the development of novel materials and innovative strategies to enhance supercapacitor performance. Emerging trends in this area include the exploration of carbon-based materials [28-38], transition metal oxides [30-31], conducting polymers [32-34], composite materials [35-36], 2D materials, nanostructured materials, and eco-friendly sustainable alternatives [37-40]. These advancements (Figure 2) reflect the ongoing quest to improve energy storage capacity, increase power density, extend cycle life, and reduce costs. Such progress is vital for the widespread adoption of supercapacitors in applications ranging from portable electronics to electric vehicles and large-scale energy storage systems. By optimizing these materials, researchers are laying the groundwork for the future of supercapacitor technologies.

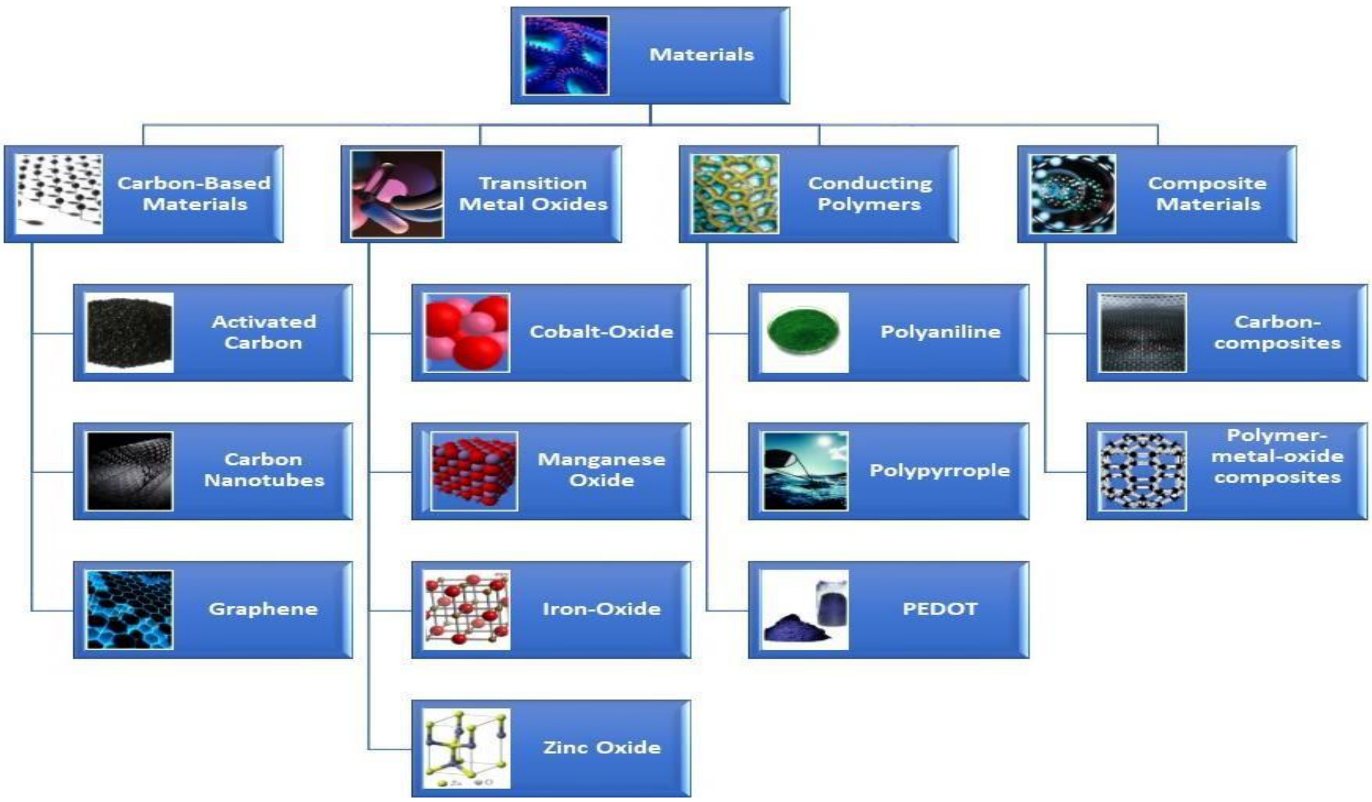

Fig. 2. Advanced materials for Supercapacitor electrode fabrication.

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2.1. Carbon-Based Materials

Carbon-based materials, such as activated carbon [41], carbon nanotubes [42], and graphene [43-44], continue to be extensively studied for supercapacitors. Researchers are investigating sophisticated synthesis processes and hierarchical architectures intending to enhance surface area and optimize pore size distribution, which will end up resulting in improved energy storage capacities. Ghosh et al. [45] focused on the importance of finding sufficient energy sources for modern scientific applications, considering the depletion of fossil fuels. The study presents a simple synthesis route for biomass-derived hard carbon and explores its potential as an electrode material for electrochemical supercapacitors. The team synthesized four distinct hard carbons from varied biomass sources and evaluated them as supercapacitor electrodes. The KOH-activated hard carbon exhibited a specific capacitance of 479.23 F/g, as determined from cycle voltammograms. The study provides a detailed analysis of the relationship between the obtained results and the material properties, emphasizing the need for characterization techniques to assess the quality and reliability of materials for electrochemical supercapacitor applications.

Activated Carbon: Activated carbon [46] (Figure 3) is a widely studied and highly sought-after material for supercapacitor applications, particularly compared to other carbon-derived materials. Its unique properties and exceptional performance characteristics set it apart from the field of energy storage. Compared to other carbon materials, such as carbon nanotubes or graphene, activated carbon exhibits a significantly higher specific surface area thanks to its extensive network of micropores and mesopores [47]. This enlarged surface area enables various active sites for charge storage, resulting in improved capacitance and energy density. Furthermore, activated carbon demonstrates excellent electrochemical stability and allows long-term cycling without significant degradation. Its abundance, low cost, and ease of synthesis make activated carbon a versatile and practical choice for supercapacitor electrode materials. Through ongoing research and development, activated carbon continues to be a prime focus in advancing the performance and scalability of supercapacitors for various energy storage applications. Various sources, including coal [48] (Figure 4), crude oil, and biomass [49], were used to create carbon components. These natural materials can serve as precursors for carbon- based materials. These materials' characteristics, however, significantly shrunk to the nanoscale. The dimensions of carbon nanostructures play a crucial role in determining the properties exhibited by carbon-based materials, such as their behavior toward light, heat, and electricity. These variances imply that they are organized around several carbon nanostructures with varied diameters. Activated carbon derived from both olive husks and banana stems holds promise for supercapacitor applications.

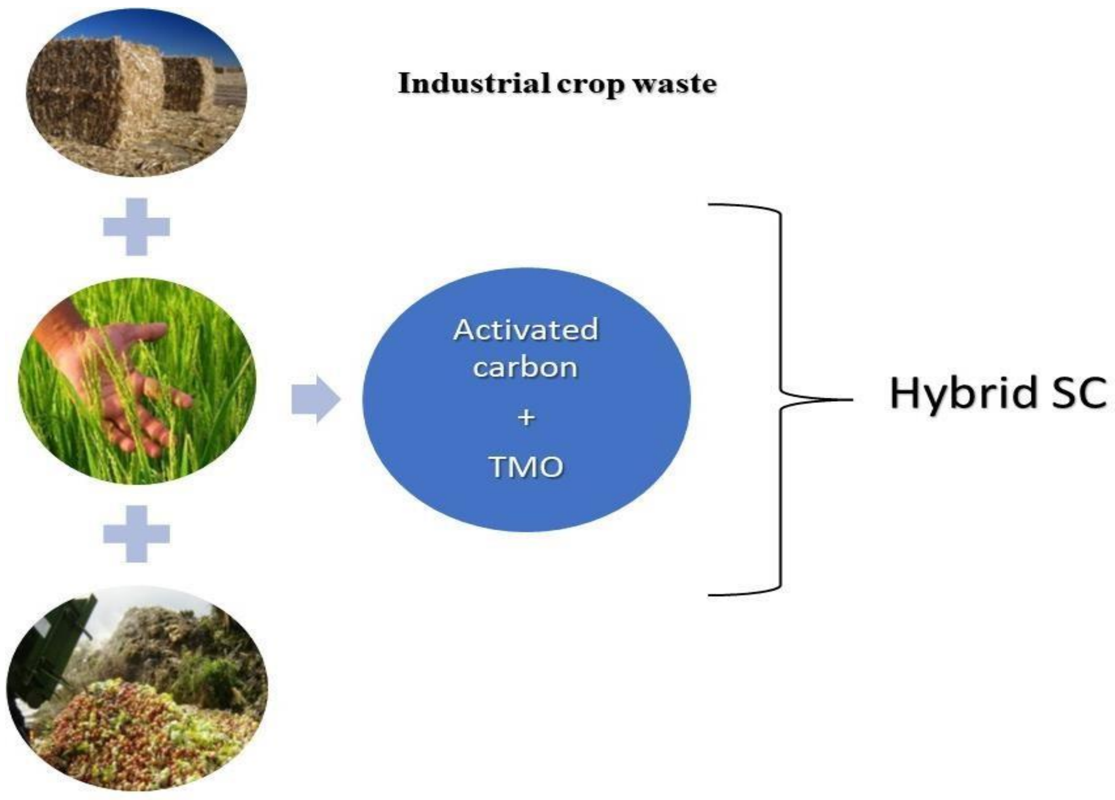

Fig. 3. The production of activated carbon from industrial agricultural waste for the synthesis of a hybrid supercapacitor.

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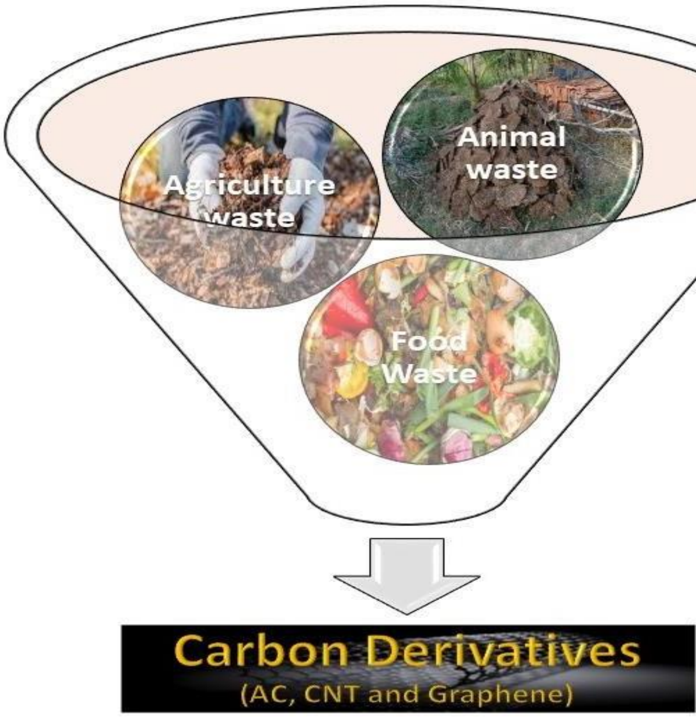

Fig. 4. Different natural sources of carbon derivatives.

Olive husk-derived activated carbon is characterized by its porous structure and high surface area, facilitating effective energy storage in supercapacitor electrodes. It offers a sustainable advantage by reusing agricultural waste.

Banana stem-derived activated carbon also demonstrates a porous structure and significant surface area, making it an attractive candidate for supercapacitor electrodes. This material offers a sustainable solution by repurposing agricultural waste. Current research is focused on optimizing its properties to enhance supercapacitor performance, with potential applications dependent on the specific attributes and availability of different carbon sources. For instance, Taer et al. [50] developed activated carbon electrodes from banana stems, achieving a specific capacitance of 170 F/g and a surface area of approximately 835.9 m^2/g. Using a conventional thermal carbonization method and cyclic voltammetry, they were able to analyze the electrochemical properties of these electrodes effectively.

Similarly, olive husk-derived activated carbon, prepared through a hydrothermal synthesis method and activated using dual agents (KOH/NaOH), has shown promising results. Nasser et al. [51] fabricated an electrode from this material, achieving a high specific capacitance of 549 F/g at a current density of 1 A/g with a surface area of 2900 m^2/g. This electrode demonstrated excellent capacitance retention of 79.5%, sustained high current densities up to 30 A/g, and exhibited remarkable cyclic stability, with a rate capability of 93.2% after 10,000 cycles at 10 A/g. Additionally, the symmetric supercapacitor (SC) device using this material delivered a specific energy of 38.8 Wh/kg and a power density of 650 W/kg at 1 A/g, making it a cost-effective and high-performance candidate for energy storage.

As concerns over pollution and dwindling fossil fuel resources intensify, research into carbon materials derived from waste has gained momentum. The carbonization of agricultural and industrial waste is increasingly seen as a key avenue for developing sustainable, high-performance carbon-based materials like biochar, graphene, graphene oxide, carbon nanotubes (CNTs), and activated carbon (AC). The field of green nanotechnology has emerged in response to these global sustainability challenges, focusing on the conversion of bio-waste into functional nanomaterials.

One green approach to producing activated carbon for supercapacitors involves minimizing the carbon footprint through eco-friendly methods. Agricultural waste materials such as coconut shells, olive husks, or bamboo are often pyrolyzed or chemically activated using sustainable energy sources like solar or biomass-derived heat. This reduces reliance on fossil fuels while repurposing waste materials. Additionally, using environmentally benign activation agents, or steam-based processes, further minimizes harmful emissions. These practices ensure that activated carbon production aligns with global sustainability goals, making it an appealing option for green supercapacitor technologies.

Devi et al. [52] explored the synthesis of activated carbon using walnut shells through a hydrothermal decomposition method, with autoclave temperatures ranging from 200 to 250°C under high-pressure conditions. The resulting activated carbon had a surface area of 408.8 m^2/g, offering abundant charge storage sites, making it highly effective for supercapacitor applications.

Altinci et al. [49] presented an innovative green

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approach using pistachio shells as a carbon precursor and sodium thiosulfate ($Na_2S_2O_3$) as an activating agent. The hydrothermal carbonization, followed by chemical activation at 800°C in the presence of KCl salt, produced a biocarbon sample (NCNaK-1) with a sponge-like morphology and a surface area of 775 m²/g. Nitrogen (1.5 wt%) and sulfur (6.5 wt%) dopants further enhanced its electrochemical performance. The NCNaK-1 electrode exhibited a specific capacitance of 166 F/g at 0.5 A/g in 1 M KOH electrolyte, with 96% retention over 5000 cycles, demonstrating outstanding cycling stability. Additionally, the supercapacitor device assembled from these electrodes achieved an energy density of 2.7 Wh/kg and a power density of 250 W/kg (Figure 5). This eco-friendly and cost-effective process highlights the versatility of biocarbon materials for use in supercapacitors, as well as other energy storage systems like lithium-ion batteries and gas storage devices.

In a related study, Taer et al. [53] examined the performance of binderless activated carbon monoliths (ACMs) derived from pre-carbonized rubber wood sawdust. These ACMs were activated under a CO_2 atmosphere at 900°C for various durations to assess their physical and electrochemical properties. Their research evaluated parameters such as specific capacitance, equivalent series resistance, energy density, power density, and surface structure, highlighting the impact of activation time on these characteristics.

In another promising development, Sandhiya et al. focused on the fabrication of an all-solid-state flexible supercapacitor (ASSFSC) using nitrogen-doped porous activated carbon derived from poultry waste (PW) [6]. The increasing demand for sustainable energy storage solutions has prompted researchers to explore the use of bio-waste materials for developing high-performance carbon materials. In this context, the authors synthesized N-doped activated carbon through chemical activation of poultry waste using potassium hydroxide (KOH). The activated carbon demonstrated a remarkable improvement in specific capacitance, energy density, and cyclic stability, making it a promising candidate for commercial energy storage devices.

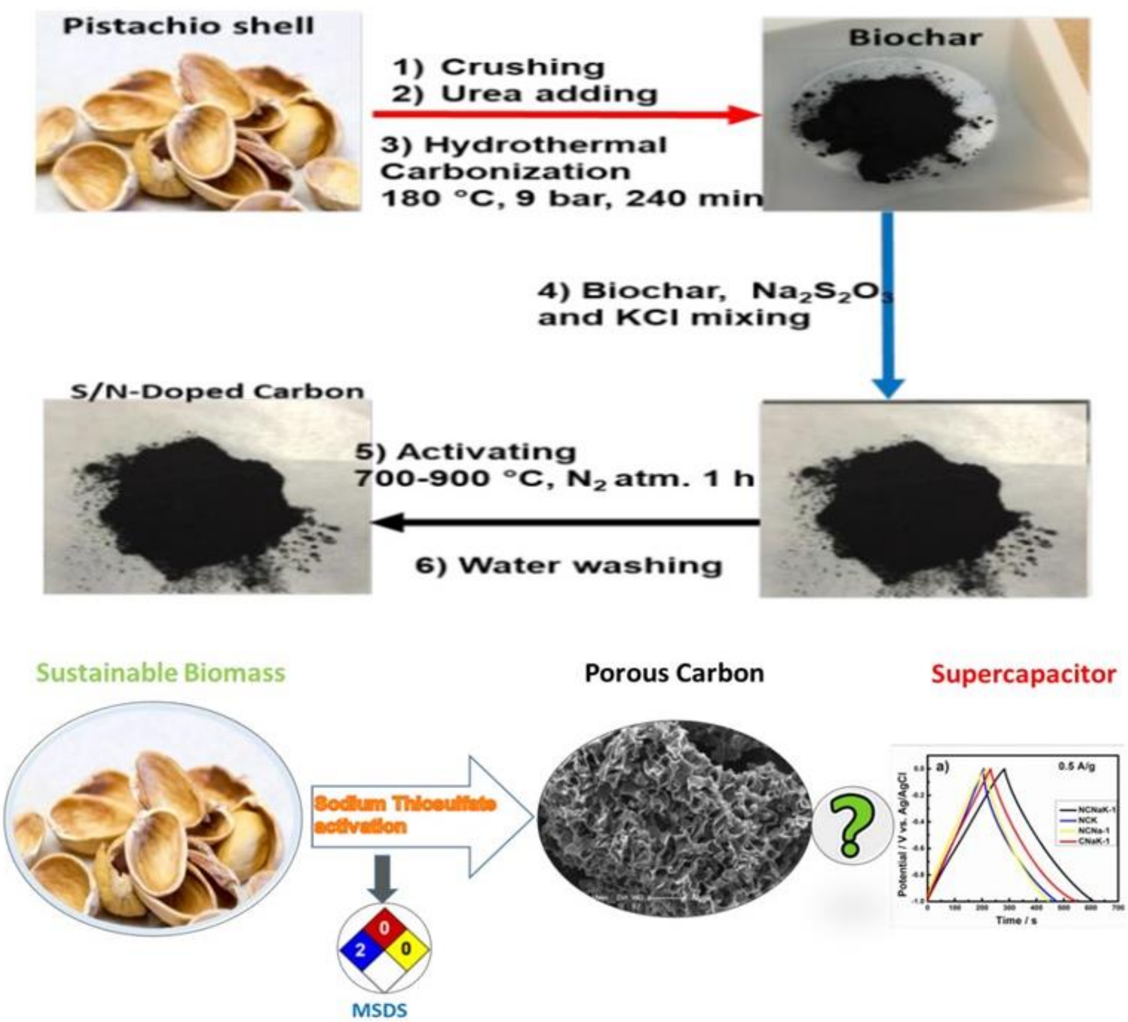

Fig. 5. Synthesis of biocarbon and its supercapacitor electrode performance. Reprinted with permission from Ref. [49] Altinci, O.C. and Demir, M., **2020.** Beyond conventional activating methods, a green approach for the synthesis of biocarbon and its supercapacitor electrode performance. *Energy & Fuels*, *34*(6), pp.7658-7665. Copyright © American Chemical Society.

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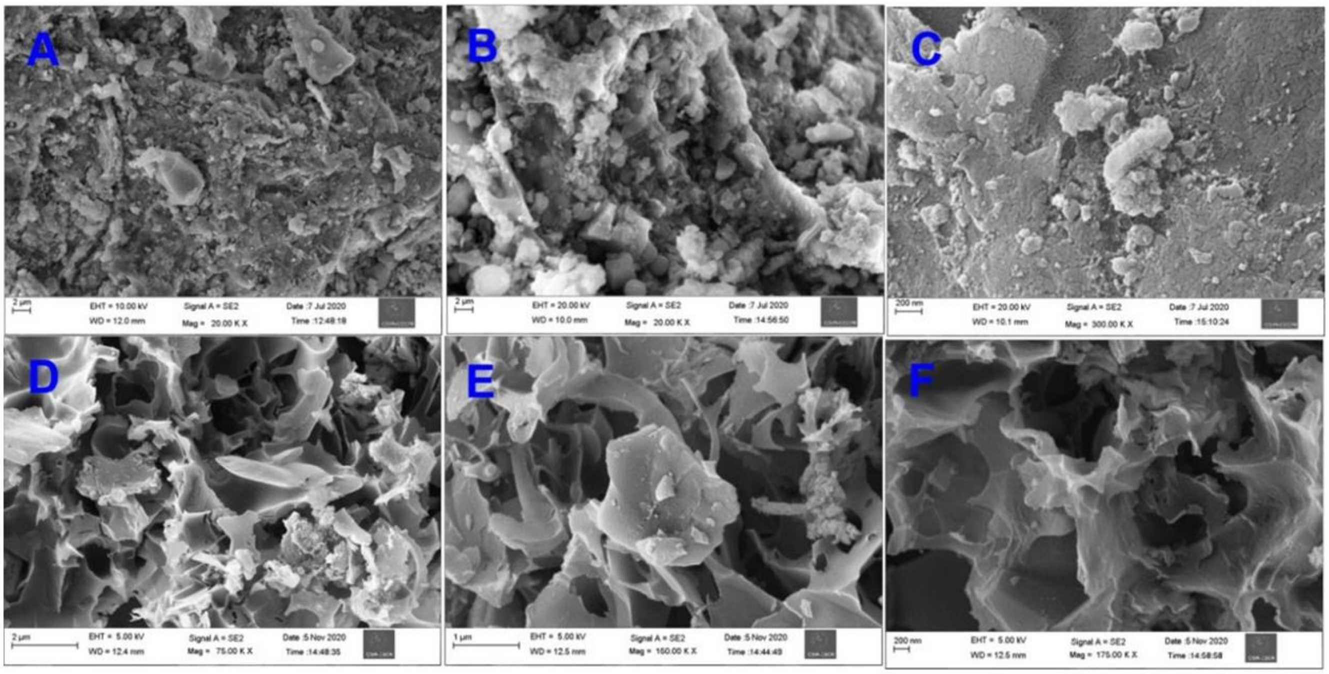

Fig. 6. FE-SEM images of (A-C) PW-BA and (D-F) PW-700 at various magnifications. Reprinted with permission from Ref. [6] Sandhiya, M., Nadira, M.P. and Sathish, M., **2021.** Fabrication of flexible supercapacitor using N-doped porous activated carbon derived from poultry waste. *Energy & Fuels*, *35*(18), pp.15094-15100. Copyright © American Chemical Society.

The activation process significantly enhanced the surface area and graphitization degree of the PW-derived carbon, which played a key role in boosting its electrochemical performance. Specifically, the maximum specific capacitance achieved by the activated carbon (PW-700) was 520 F/g, which is 7.2 times higher than that of the non-activated carbon (PW-BA). Additionally, the energy density of the symmetric supercapacitor fabricated with PW-700 rose from 16 to 23 Wh/kg, while the cycling stability increased from 83% to 99% after 50,000 charge-discharge cycles. The high degree of graphitization and the porous nature of the material were responsible for these improvements, as they provided more active sites for charge storage and enhanced conductivity (Figure 6).

The ASSFSC device fabricated using PW-700 and poly(vinyl alcohol) (PVA)/H_2SO_4 as the electrolyte and separator exhibited excellent electrochemical stability, with 90% capacitance retention over 25,000 cycles and a high Coulombic efficiency of 99%. The device also achieved an energy density of 21.5 Wh/kg at 0.5 A/g and demonstrated superior cycling performance without significant degradation, making it highly suitable for commercial applications.

Gouda et al. explored the fabrication of high-performance supercapacitor electrodes using activated carbon (AC) derived from willow catkin (WC) and modified with nickel oxide (NiO) and cobalt oxide (Co_3O_4) nanoparticles [54]. Willow catkin, an abundant natural waste material, was subjected to chemical activation with KOH and carbonization at 600°C in an inert atmosphere to produce porous AC. The AC was further modified by incorporating different weight percentages (10%, 25%, 50%, and 75%) of NiO and Co_3O_4 nanoparticles, which were synthesized using nickel sulfate and cobalt nitrate precursors. The modified nanocomposites were calcined at 300°C for 3 hours to improve their electrochemical properties. The electrochemical performance of the nanocomposite electrodes was evaluated in 3M KOH aqueous electrolyte using cyclic voltammetry (CV), galvanostatic charge-discharge (GCD), and electrochemical impedance spectroscopy (EIS). The results indicated that the incorporation of NiO and Co_3O_4 significantly improved the specific capacitance and energy density of the electrodes compared to the pristine AC. The AC electrode displayed a relatively low specific capacitance of 105 F/g at a current density of 1.0 A/g. However, the nanocomposites with 25 wt% NiO and Co_3O_4, denoted as 25NiO@Co_3O_4-AC and 25Co_3O_4@NiO-AC, demonstrated exceptional electrochemical performance. The 25NiO@Co_3O_4-AC electrode achieved a maximum specific capacitance of 800.9 F/g and an energy density of 136.6 Wh/kg, while the 25Co_3O_4@NiO-AC electrode reached 691.8 F/g with an energy density of 116.2 Wh/kg. These improvements are attributed to the homogeneous dispersion of the metal oxide nanoparticles, which enhanced ion diffusion and electrode conductivity. However, increasing the content of metal oxides beyond 25 wt% led to agglomeration, which blocked the porous structure of the AC and hindered electrolyte ion diffusion, reducing the specific capacitance. Despite this, the 25Co_3O_4@NiO-AC electrode exhibited excellent cycling stability, retaining 98.1% of its capacitance after 5000 charge-discharge cycles at a high current density of 10 A/g.

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This research demonstrates the potential of NiO and Co_3O_4-modified AC derived from willow catkin as a cost-effective, eco-friendly, and high-performance electrode material for supercapacitors. The findings provide valuable insights into developing novel hybrid materials for energy storage applications.

The extensive literature review on activated carbon-based electrode supercapacitors has provided a comprehensive understanding of their unique properties, outstanding electrochemical performance, and diverse synthesis methods. Activated carbon emerges as a highly sought-after material for supercapacitor applications, surpassing other carbon-derived materials such as graphene and carbon nanotubes. Its exceptionally high specific surface area, outstanding electrical conductivity, and electrochemical stability make it an excellent choice for energy storage systems. The versatility of activated carbon is evident in its numerous applications, including portable electronics, electric vehicles, grid stabilization, and renewable energy integration, reflecting its crucial role in the quest for sustainable energy solutions. Moreover, the literature highlights various sources of activated carbon, including agricultural waste, biomass, and natural materials, promoting the concept of green nanotechnology by recycling and repurposing these materials for energy storage. The synthesis of activated carbon from waste materials contributes to environmental sustainability while producing nanocomposite electrodes with enhanced specific capacitance for supercapacitor applications. Collectively, the findings from the literature review underscore the significance of activated carbon-based electrode supercapacitors in addressing global energy storage challenges. The ongoing investigation of synthesis processes, new materials, and green nanotechnology technologies is projected to improve the efficiency, scalability, and practical applications of these energy storage devices. As researchers strive to advance in this sector, activated carbon-based supercapacitors are set to play a critical role in reaching a sustainable and greener energy future.

Carbon Nanotubes: Among various materials investigated for supercapacitor electrodes, carbon nanotubes (CNTs) have emerged as a compelling choice, offering exceptional properties that enhance the overall performance of these energy storage systems. Zhu et al. [55] discuss the growing interest in utilizing flexible supercapacitors (SCs) as power supply units for portable and wearable electronics. Carbon nanotubes (CNTs) are highlighted as promising materials for flexible SC electrodes due to their exceptional mechanical properties, high electrical conductivity, large surface area, and versatility. The review focuses on various flexible CNT assemblies, including 1D fibers, 2D films, and 3D aerogels and sponges, detailing their design strategies and fabrication techniques. The article summarizes recent advancements and cutting- edge applications of these structures as electrodes in flexible SCs, categorizing them based on device configurations such as sandwiched, interdigital in-plane, and cable type. The flexible CNT-based electrodes demonstrate advantages in terms of bendability, stretchability, compressibility, and extended cycle lifetimes. The review also addresses existing challenges and outlines future research opportunities in this evolving field. Gupta et al. [56] presented that Carbon nanotubes (CNTs) are cylindrical nanostructures of tubular organized carbon atoms. Based on their structure, CNTs can have many configurations, such as single-walled carbon nanotubes (SWCNTs) or multi-walled carbon nanotubes (MWCNTs), depending on how many layers they contain (Figure 7).

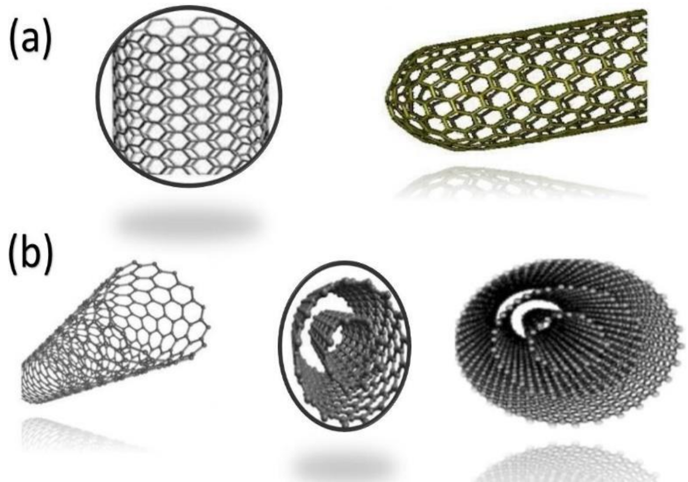

Fig. 7. Types of CNT on (a) basis of ends: Open (left) and closed (right), (b) basis of number of walls: SWCNT (left), DWCNT (middle), MWCNT (right).

Carbon nanotubes (CNTs) are extremely important in fabricating supercapacitors due to their outstanding characteristics. Their nanotubular shape encompasses a large surface area, facilitating more charge storage in the form of ions at the electrode-electrolyte interface, resulting in enhanced capacitance. CNTs exhibit excellent electrical conductivity, enabling efficient electron transport within the electrode material, reducing internal resistance, and improving charge-discharge rates for rapid energy delivery. Moreover, their remarkable mechanical strength assures structural integrity throughout charge-discharge cycles and enhances efficiency and extended device lifetimes.

CNTs offer the advantage of tunability, allowing researchers to customize their properties for specific supercapacitor applications by altering their diameter, length, and functionalization. Also, their flexibility enables integration into various electrode architectures, making them suitable for flexible and wearable energy storage systems. By forming composites or hybrids with other materials, CNTs can further enhance capacitance and stability. Another important feature is their environmental friendliness since they are composed of carbon, which is abundant and eco-friendly when relative to several other electrode materials. The main objective herein is to provide an in-depth analysis of the

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recent developments and utilization of carbon nanotubes as supercapacitor electrodes. We will delve into the synthesis techniques employed to produce CNTs with desired characteristics, focusing on their impact on supercapacitor performance.

A method to enhance the performance of supercapacitors using binderless composite electrode monoliths (BCMs), made from self- adhesive carbon grains (SACG) derived from fibers obtained from oil palm empty fruit bunches was proposed by Farma et al. [57]. Three variations were used to prepare monoliths: mixed with KOH (5% by weight), mixed with CNT and KOH together (each 5% by weight), and untreated for reference. They carbonized them at 800°C in a nitrogen environment. Later these monoliths were activated by CO_2 gas at 800°C for 1 hour. They found that the use of KOH treatment led to BCMs with higher specific capacitances and enhanced the capacitor's ability to store electrical charge. Further addition of CNTs resulted in BCMs with smaller internal resistance. This implies that CNTs contributed to improving the charge and discharge efficiency of the supercapacitor. Supercapacitor cells that utilized these improved BCMs as electrodes exhibited enhanced specific energy and specific power. This suggests that the modifications made to the BCMs positively impacted the overall energy storage and delivery capabilities of the supercapacitor. Deraman et al. [58] extracted self-adhesive carbon grains (SACG) fibers from oil palm empty fruit bunches and prepared binderless composite electrode monoliths using CNTs. Green monoliths (GMs) were prepared from three different types of precursors: SACG (for reference), SACG treated with 0.4 Molar H_2SO_4, and a mixture of SACG and 5% CNTs (by weight), treated with 0.4 Molar H_2SO_4. The GMs were subjected to carbonization at 600°C in a nitrogen (N_2) gas environment. Subsequently, the carbonized GMs were activated by CO_2 gas at 800°C for 1 hour to produce activated carbon monoliths (ACMs).

Zhang et al. demonstrated the preparation of a novel porous 3D honeycomb-structured carbon material derived from seaweed powder, aimed at enhancing the performance of supercapacitors (SCs) [5]. The carbon material, labeled CS-2, was synthesized through a simple gas-phase cycle reaction and exhibited key features necessary for efficient energy storage: a high specific surface area (SSA) of 1206.97 m^2 g^{-1}, good surface wettability, and excellent electrochemical properties. The 3D honeycomb structure of CS-2 facilitated effective ion diffusion, improving conductivity and charge-transfer abilities, which are crucial for SC electrode materials. Characterization techniques, including cyclic voltammetry (CV) and galvanostatic charge-discharge (GCD) tests, confirmed the superior performance of the CS-2 electrode. The specific capacitance reached 448.3 F g^{-1} at a current density of 0.1 A g^{-1}, and the CS-2 electrode demonstrated excellent cycle stability, maintaining 91.39% of its capacitance after 3000 cycles (Figure 8).

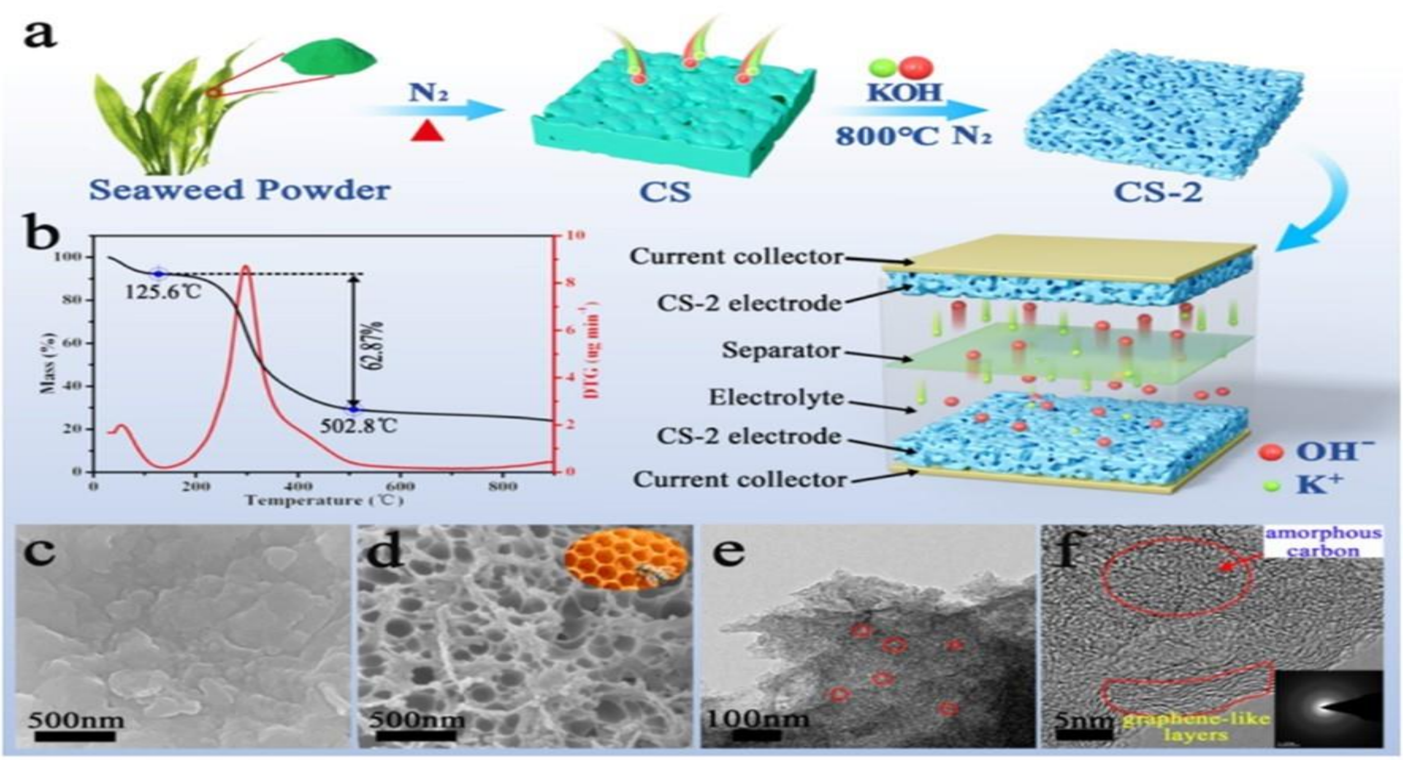

Fig. 8. Assembly strategy and micromorphology of the CS-2 electrode: (a) Schematic of the CS-2 electrode assembly; (b) TG and DTG curves of CS; SEM images of (c) CS and (d) CS-2; (e) TEM image of CS-2; (f) HRTEM image of CS-2 with SAED pattern inset. Reprinted with permission from Ref [5] Zhang, Z., Yang, W., Wu, Y., Yan, G., Li, L., Qing, Y. and Lu, X., 2021. Porous 3D honeycomb structure biomass carbon as a supercapacitor electrode material to achieve efficient energy storage. *Industrial & Engineering Chemistry Research*, *60*(30), pp.11079-11085. Copyright © American Chemical Society.

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The material's low equivalent series resistance (0.57 Ω) further indicated strong conductivity, while the contact angle measurements confirmed its outstanding wettability, with CS-2 showing the best surface wetting properties compared to other carbon-based electrodes in the series. To evaluate practical applicability, a symmetrical supercapacitor (SSC) was assembled using CS-2 electrodes and a 6 M KOH electrolyte. The SSC exhibited excellent energy storage performance, achieving a maximum energy density of 3.125 Wh kg^{-1} and a power density of 5 kW kg^{-1} at a high current density of 10 A g^{-1}. These results surpassed the performance of several previously reported carbon-based SSCs, further validating the potential of seaweed-derived biomass carbon for energy storage applications. This study demonstrates that the sustainable and low-cost carbon material derived from seaweed powder, with its high SSA, excellent porosity, and good conductivity, is an efficient candidate for use in supercapacitor electrodes. The research highlights the practical application of biomass-derived carbon in energy storage devices, paving the way for further exploration of similar materials in the development of next-generation SCs. The findings suggest that CS-2 can play a significant role in achieving high energy and power densities, making it a promising material for future SC technologies.

Hu et al. [59] reported the successful development of a flexible solid-state supercapacitor with impressive energy density and performance characteristics. By combining cotton paper-carbon nanotube electrodes with a solid-state polymer-based electrolyte, the researchers created a high-energy-density solution comparable to commercial supercapacitors that rely on liquid electrolytes. This innovative approach highlights the potential for solid-state supercapacitors to meet energy storage demands with improved safety and flexibility compared to traditional liquid-electrolyte systems. De Marco et al. [60] introduced a novel method for creating covalent, carbon-bonded organogels by cross-linking anions of single-walled carbon nanotubes (SWCNTs) using a dielectrophile. These organogels, produced via freeze-drying, form cryogels with remarkable properties, including low density (2.3 mg cm^{-3}), high surface area (766 m^2 g^{-1}), and impressive conductivity (9.4 S m^{-1}), making them strong candidates for supercapacitor electrodes. The cross-linking process is carefully controlled by adjusting the counterion concentration, influencing the unbundling of SWCNTs, grafting ratio, and overall material properties. This method is unique in that it preserves the individualization of the SWCNTs without causing damage, unlike other approaches. The strategy is based on a charge-driven dissolution mechanism, leading to the formation of a robust carbon-to-carbon bonded network within the gel. Additionally, the unreacted iodide offers opportunities for further material modification. The optimized cryogel, with a sodium concentration of 15 mM, achieves superior unbundling and grafting, resulting in an enhanced surface area and uniform pore distribution at smaller sizes. These cryogels demonstrate excellent electrical conductivity, low density, and chemical stability, making them promising for applications in energy conversion technologies and environmental solutions. Preliminary electrochemical tests using these cryogel electrodes in electrochemical double-layer capacitors (EDLCs) show favorable performance and durability over extended charge-discharge cycles (up to 105 cycles). The study also suggests that further refinement in the composition of nanotubide solutions, cross-linkers, and drying protocols could optimize the morphology of the cryogels for various applications.

Chang et al. demonstrated the development of a high-performance flexible all-solid-state supercapacitor using a composite electrode made from two-dimensional molybdenum disulfide (MoS_2), single-walled carbon nanotubes (SWCNT), and cellulose nanofiber (CNF) aerogel [7]. The innovative design leverages the unique properties of these materials to enhance both the electrochemical performance and flexibility of the supercapacitor. SWCNTs inhibit the agglomeration of MoS_2 nanosheets, improving conductivity, while CNFs enhance dispersion uniformity, contributing to a high specific surface area of 328.86 m^2/g and excellent mechanical flexibility. The MoS_2–SWCNT/CNF composite electrode exhibits impressive electrochemical characteristics. The cyclic voltammetry (CV) curve retains a rectangular shape even at a high scan rate of 2000 mV/s, and the galvanostatic charge-discharge (GCD) curve shows a symmetrical triangular shape, indicating high capacitance and stability. The device achieves a specific capacity of 605.32 mF/cm^2 at a scanning rate of 2 mV/s and 30.34 F/g at 0.01 A/g, demonstrating its strong energy storage capabilities. Additionally, the supercapacitor delivers an area-specific energy of 35.61 mWh/cm^2 with an area-specific power of 0.03 mW/cm^2 (Figure 9).

The nanocomposite electrode's 3D fibrous porous network structure, with a pore volume of 0.83 cm^3/g and small pore size (~10 nm), further supports its outstanding performance. This structure, combined with the intercalation of SWCNTs, prevents MoS_2 nanosheets from stacking, improving conductivity and ensuring high surface area availability for charge storage. In terms of durability, the supercapacitor displays excellent cycling stability, retaining 91.01% of its specific capacity after 10,000 charge-discharge cycles. Its flexibility is maintained across various bending angles, making it suitable for portable and wearable electronic devices. Furthermore, the device features an extended voltage window of 1.5 V, enhancing its overall energy storage potential. This study provides significant insights into the use of two-dimensional materials like MoS_2 and SWCNTs in flexible energy storage systems. By combining these materials with CNF aerogel, Chang et al. achieved a robust, flexible supercapacitor with superior electrochemical performance. This work presents a promising approach for developing flexible devices, enhancing the applicability of supercapacitors in future portable electronics. Their high performance, durability, and flexibility position them as ideal energy storage solutions, effectively addressing the increasing demand for efficient energy sources in portable and flexible applications.

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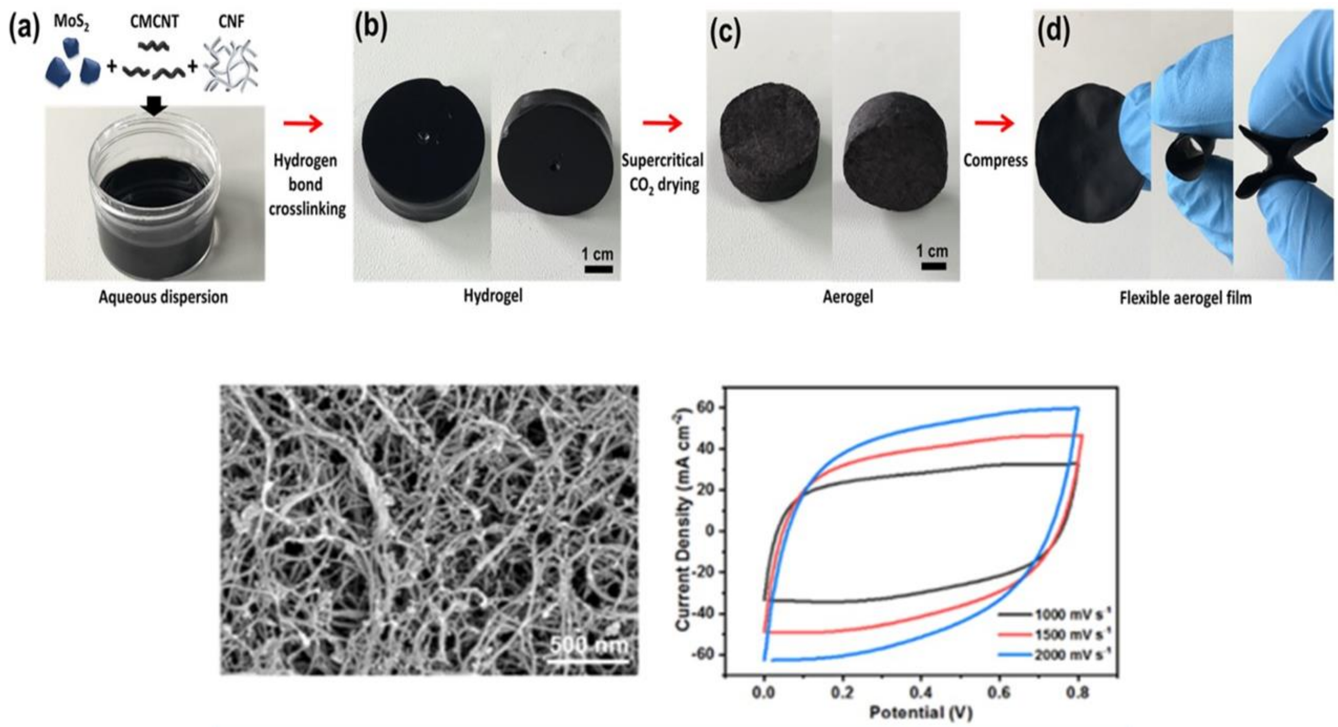

Fig. 9. Flexible and freestanding MoS_2 nanosheet/carbon nanotube/cellulose nanofibril (CNF) hybrid aerogel film for high-performance all-solid-state supercapacitors. Reprinted with permission from Ref [7] Chang, H., Zhang, L., Lyu, S. and Wang, S., 2022. Flexible and freestanding MoS2 nanosheet/carbon nanotube/cellulose nanofibril hybrid aerogel film for high-performance all-solid-state supercapacitors. *ACS omega*, *7*(16), pp.14390-14399. Copyright © American Chemical Society.

Majeed et al. [61] introduced an innovative solution in energy storage technology by developing a flexible single-wall carbon nanotube (SWCNT) film decorated with carbon-encapsulated nickel (CENi) nanoparticles. The research employs a floating catalyst chemical vapor deposition (FCCVD) technique to fabricate a flexible SWCNT film laden with a substantial quantity of CENi nanoparticles. Following air oxidation, the process culminates in the creation of carbon-encapsulated nickel oxide nanoparticle-decorated SWCNT (CENiO/SWCNT) films. This novel CENiO/SWCNT material is notable for its binder-free flexible electrode configuration, which displays exceptional durability, maintaining its specific capacitance even after 500 bending cycles at an impressive angle of 130 degrees. The performance metrics of the CENiO/SWCNT electrode are equally impressive, with a specific capacitance reaching 1422 F g^{-1}. Furthermore, the material exhibits outstanding cyclic stability, retaining 92% of its capacitance after undergoing 5000 cycles. These remarkable achievements are largely attributed to the unique combination of properties provided by the SWCNT scaffold, which offers excellent electrical conductivity and flexibility, along with the high-capacity nickel oxide nanoparticles integrated into the hybrid electrode. The structure of the electrode features small NiOx nanoparticles uniformly dispersed across the SWCNT network, achieved through the FCCVD process and subsequent air oxidation. The carbon shell that encases the NiOx particles serves multiple functions, primarily preventing particle aggregation and establishing strong connections with the highly conductive SWCNTs through π-π interactions. Consequently, the resulting CENiO/SWCNT electrodes exhibit not only high capacity and cyclic stability but also superior rate performance and flexibility, making them highly suitable for application in flexible energy storage devices.

Another significant study [62] reported the successful fabrication of electrode material with exceptional longevity and performance by integrating carbon nanotubes (CNTs) of varying lengths onto hierarchically three-dimensional carbon foam (CF), which is derived from mesophase pitch. Through a chemical vapor deposition (CVD) process, the resulting CF/CNT composites are systematically characterized using a variety of analytical techniques. The CF/CNT-50 composite showcases impressive cycling stability, retaining 96.5% of its capacitance even after an astounding 10,000 cycles. This material boasts a significant mass capacitance of 227.5 F g^{-1}, accompanied by enhanced energy density (28 Wh kg^{-1}) and power density (3700 W kg^{-1}) at a current density of 2 A g^{-1}. The composite further demonstrates commendable charge/discharge rates, collectively highlighting its exceptional charge storage properties. Such remarkable performance positions the hybrid CF/CNT materials as strong contenders for high-performance supercapacitor electrode materials. The development and utilization of

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CNT-based electrodes for binderless supercapacitors signify a significant advancement in energy storage technology. The unique characteristics of CNTs, which include a high surface area, excellent electrical conductivity, mechanical strength, and chemical stability, make them particularly suitable as electrode materials in supercapacitors. The elimination of binders not only enhances the energy and power density of these devices by reducing resistance at the electrode-electrolyte interface but also simplifies the overall manufacturing process. This reduction leads to a decrease in the overall weight and volume of the supercapacitor, minimizing the risk of delamination or degradation over time. Furthermore, CNT-based electrodes offer several advantages over conventional electrode materials, including improved energy storage performance, faster charge/discharge rates, and extended cycle life. Their adaptability to various form factors, including flexible and wearable devices, demonstrates their versatility and potential for a wide range of applications. However, challenges remain in achieving scalable and cost-effective manufacturing processes for CNT-based electrodes, as well as addressing potential environmental impacts and the long-term stability of these materials. Ongoing research is essential to optimize electrode architecture, enhance the uniformity of electrode-electrolyte interactions, and explore novel synthesis and assembly methods. In the pursuit of more efficient and sustainable energy storage solutions, CNT-based electrodes for binderless supercapacitors represent a significant advancement in the field. With continuous innovation and collaborative efforts among the scientific and engineering communities, these electrodes possess the potential to revolutionize energy storage technology. This could lead to the development of more efficient, compact, and environmentally friendly power sources suitable for a variety of applications, ranging from consumer electronics to renewable energy systems. The future of energy storage may very well hinge on the further integration of CNT-based technologies, paving the way for breakthroughs that could significantly impact our approach to energy consumption and sustainability.

Shah et al. demonstrated the synthesis and characterization of NiO-CNT and NiO-Fe-CNT composites derived from waste high-density polyethylene (HDPE) plastic, with a focus on enhancing supercapacitance through tuning the quality of carbon nanotubes (CNTs) [8]. Using transmission electron microscopy (TEM) and Raman spectroscopy, the formation of multiwalled CNTs (MWCNTs) is confirmed, revealing a high graphitization level with an ID/IG ratio of 0.77. The specific surface area (SSA) of the MWCNTs in the NiO-Fe-CNT composite is determined to be 87.8 m^2/g, in contrast to 25 m^2/g for the NiO-CNT composite. The performance evaluation demonstrates that NiO-Fe-CNT exhibits superior specific capacitance and energy density values of 1360 F g^{-1} and 1180 W h kg^{-1}, respectively, compared to NiO-CNT's 1250 F g^{-1} and 1000 W h kg^{-1}. These enhanced performances are attributed to the higher-quality MWCNTs present in the NiO-Fe-CNT composite, which also contributes to a more significant electric double-layer capacitor (EDLC) behavior (59%) compared to the NiO-CNT's 38%, indicating a hybrid supercapacitor nature. Moreover, NiO-Fe-CNT maintains a high capacitive retention rate of 96% after 1000 charge-discharge cycles, underscoring its stability and longevity (Figure 10).

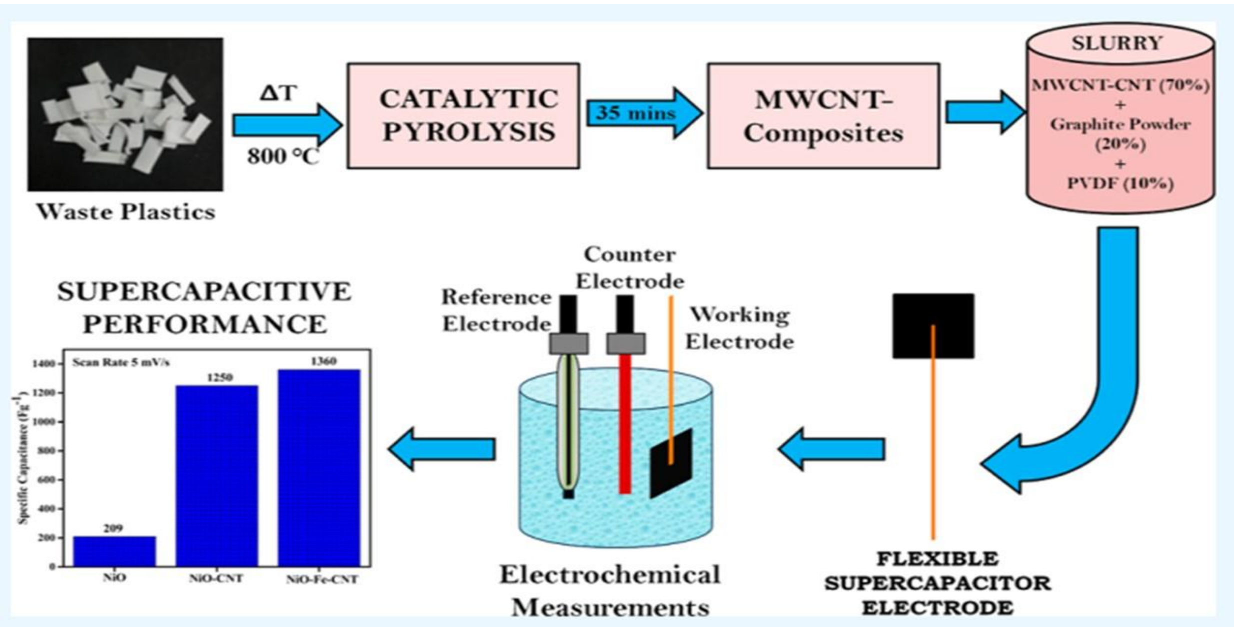

Fig. 10. Supercapacitor Performance of NiO, NiO-MWCNT, and NiO–Fe-MWCNT Composites. Reprinted with permission from Ref [8] Shah, A., Senapati, S., Murthy, H.A., Singh, L.R. and Mahato, M., 2023. Supercapacitor performance of NiO, NiO-MWCNT, and NiO–Fe-MWCNT composites. *ACS omega*, *8*(37), pp.33380-33391. Copyright © American Chemical Society.

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Table 1 presents a comparison of the key advantages and disadvantages of various carbon-based materials used in supercapacitor electrodes. The materials are evaluated based on their electrochemical performance, including specific capacitance, conductivity, surface area, cost-effectiveness, and environmental sustainability. This overview highlights the strengths of carbon-based materials, such as high surface area and excellent stability, while also addressing limitations like lower energy density and pore structure challenges.

Table 1. Advantages and Disadvantages of Carbon-Based Materials in Supercapacitor Electrode Applications.

Carbon-based Materials	Advantages	Disadvantages	Ref.
Activated Carbon	A large surface area, porous structure, high amount of charge storage through adsorption, high specific capacitance, Low cost, easily available.	Lower energy density, lower power density, slower charge/discharge rates, experience performance degradation over time due to cycling.	[63-64]
Carbon Nanotubes	A high surface area, increased charge storage, outstanding electrical conductivity, high power density, fast charge/discharge rates, mechanically robust, enhancing the durability, lightweight.	Expensive and challenging, mass production can be difficult, may raise concerns about toxicity, especially if they become airborne during manufacturing or use.	[65]
Graphene	Extremely high electrical conductivity, high power density, fast charge/discharge rates, large surface area, significant charge storage, lightweight, thinness but is remarkably strong, chemically stable, which extends the lifespan.	Large-scale production is challenging and costly, lack the well-defined pore structure, no band gap limits its energy density, environmental impact of graphene production and disposal needs further study.	[66]

2.2. Transition Metal Oxides

Transition metal oxide-based electrodes are used in supercapacitors to capitalize on their pseudocapacitive behavior, which enables higher specific capacitance, fast charge/discharge rates, enhanced energy density, and long cycle life. As research and development in this field continue, TMO-based supercapacitors hold the potential to bridge the gap between traditional supercapacitors and batteries, offering versatile energy storage solutions for various applications.

Transition metal oxides, such as ruthenium oxide (RuO_2), manganese dioxide (MnO_2), and cobalt oxide (Co_3O_4), have gained attention due to their high theoretical specific capacitance. Researchers have been working to improve the electrical conductivity and stability to use them for supercapacitors. These materials possess high specific capacitance, excellent electrical conductivity, and good redox activity, enabling efficient charge storage and rapid charge-discharge cycles. They exhibit pseudocapacitive behavior, which combines aspects of both double-layer capacitance and faradaic redox reactions. It enables the storage of energy not only through the surface adsorption of ions but also through reversible redox reactions within the bulk material. The pseudo-capacitance mechanism in transition metal oxide (TMO) electrodes stems from reversible faradaic reactions occurring on their surfaces.

The electrolyte ions adsorbed onto the TMO electrode during charging, resulting in redox reactions that promote charge storage via ion insertion and reversible redox processes. This results in a higher capacitance than electric double-layer capacitors (EDLCs). Recent advancements [67] in the field have focused on designing nanostructured architectures and creating composites with expansive specific surface areas utilizing transition metal oxides and nitrides. This class of materials includes compounds such as ruthenium oxides [68], nickel oxides [69], manganese oxides [70], vanadium oxides [71], cobalt oxides [72], titanium nitrides, vanadium nitrides, molybdenum nitrides, and niobium nitrides. The exploration of these advanced nanostructured designs and composite formulations has profound implications for further research into pseudo-capacitor electrode materials. By harnessing the unique properties of these transition metal compounds researchers can unlock higher energy and power densities, ushering in a new era of efficient and versatile energy storage solutions. A recent study [73] introduces a compelling approach by utilizing nickel nanodendrites (NDs) as a sturdy substrate for hydrous RuO_2, forming a unique electrode architecture for symmetric supercapacitors. This strategy capitalizes on the self-assembled synthesis of Ni NDs directly onto nickel foam, yielding a nanostructure with diameters ranging from 30 to 100 nm. The resulting composite

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electrode configuration exhibits exceptional electrochemical performance, presenting a high specific capacitance of 678.57 F g^{-1} and an energy density of 60.32 Wh kg^{-1} when operated at 1.6 V. The absence of carbon additives or resistive binders in the pristine metal nanostructure allows the system to retain remarkable energy density (19.73 Wh kg^{-1}) even under large current densities of 100 A g^{-1}, accompanied by a power density of 40 kW kg^{-1}. Additionally, the extended lifespan of the system, showing minimal performance degradation over 10,000 cycles, reinforces its durability. Notably, the template-less synthesis of Ni NDs using eco-friendly chemicals and low-temperature processes provides a practical advantage, while the ease of incorporating RuO_2 nanoparticles onto the foam presents potential for scalable mass production.

TMOs exhibit notably higher specific capacitance than conventional carbon-based EDLC materials. This heightened capacitance results from additional contributions via pseudocapacitive reactions, translating to increased energy storage capacity. TMOs enable swift charge and discharge rates due to internal redox reactions. This trait proves crucial in applications necessitating rapid power bursts, like regenerative braking and peak power demand management. TMO-based supercapacitors achieve elevated energy densities compared to EDLCs by harnessing both double-layer capacitance and pseudo-capacitance. Though not reaching battery energy densities, TMO-based supercapacitors offer a balanced compromise between supercapacitor power density and battery energy density. Some researchers [74] have synthesized nanocomposite materials consisting of neodymium and manganese oxide (Nd_2O_3/Mn_3O_4-0, Nd_2O_3/Mn_3O_4-1, $Nd_2O_3/Mn_3O_4^{-2}$) as well as pure nanoparticles of Nd_2O_3 using a hydrothermal process. Through various analyses, they found that the nanocomposites exhibit a combination of hexagonal and tetragonal crystal structures from Nd_2O_3 and Mn_3O_4 respectively. Microscopic examination revealed interconnected Nd_2O_3 nanoparticles and irregular nanograins of Mn_3O_4. The presence of constituent elements in the materials was confirmed through spectroscopy. Nitrogen adsorption/desorption analysis indicated that the materials possess mesoporosity, characterized by a type IV isotherm with an H_3 hysteresis loop and pore sizes within the range of 30–45 Å. Electrochemical testing demonstrated that the Nd_2O_3/Mn O $^{-1}$ electrode, with a mass ratio of 63:37, displayed a high specific capacitance of 205.29 F g^{-1} at a scan rate of 5 mVs^{-1}, outperforming other electrodes. This enhanced capacitance is attributed to the cooperative effects of redox-active Nd_2O_3 and Mn_3O_4 metal oxides. The Nd_2O_3/MnO^{-1} electrode also exhibited favorable cycling stability with 67% retention over extended cycles at 50 mV s^{-1} and a high Coulombic efficiency of 99.64% at 5A g^{-1}. These findings suggest that the $Nd_2O_3/Mn_3O_4^{-1}$ electrode holds promise as a valuable material for supercapacitor applications.

TMO materials typically exhibit enduring cycling stability due to the reversible nature of their redox reactions [75-78]. This characteristic leads to extensive cycle lifespans with minimal capacity degradation over multiple charge/discharge cycles. Wang et al. [76], focused on improving the performance of energy storage devices, specifically anodes, by creating a novel electrode structure using a combination of materials. The researchers combined manganese oxide (MnO) with a composite material made of carbon nanotubes (CNT) and graphene. This combination was used to synthesize a three-dimensional (3D) electrode structure. The primary goal of this research was to enhance the redox reactions within the anode of an energy storage device, likely a lithium-ion battery. Redox reactions are crucial for the device's energy storage and release capabilities. The 3D electrode structure created through this synthesis process possesses a large surface. This feature is important because it allows for better interaction between the electrode and the electrolyte, which can improve the electrochemical activity of the anode. The large surface area and the 3D architecture also enable fast charging and discharging of the energy storage device. This is significant for applications where rapid energy transfer is essential. The 3D architecture of the electrode provides space to accommodate volume expansion, which can occur during the charging and discharging cycles of the device. This feature helps maintain the structural stability of the electrode under stress. The combination of MnO with the CNT/graphene composite results in a synergistic effect, meaning the components work together to enhance the overall electrochemical performance of the electrode. The electrode developed in this research exhibits a high specific capacity of 526.7 milliampere-hours per gram (mA h g^{-1}) when discharged at a rate of 2.0 amperes per gram (A g^{-1}). This indicates that it can store a significant amount of energy. The electrode maintains 98% capacity retention over 1400 charging and discharging cycles. This demonstrates its durability and long-term performance stability. The author suggests that this research provides a promising pathway for the practical application of fast-charging and durable lithium-ion batteries. It also implies that the findings could be valuable for designing similar structures using other transition metal oxides.

Wei et al. demonstrated the successful design and development of a high-performance solid-state supercapacitor utilizing a 3D hierarchical nanosheet array-like composite of nickel cobaltite/reduced graphene oxide/nickel foam ($NiCo_2O_4$/rGO/NF) [11]. This innovative composite was synthesized using an aqueous coprecipitation-hydrothermal strategy facilitated by citric acid (CA). The resulting $NiCo_2O_4$/rGO/NF composite features thin $NiCo_2O_4$ nanosheets (approximately 113.6 nm × 11.2 nm), composed of $NiCo_2O_4$ nanoparticles (around 10.9 nm) that are vertically staggered on the surface of a reduced graphene oxide-modified nickel foam (NF) skeleton. This unique structural design promotes a high surface area, abundant mesoporosity, and enhanced exposure of active sites, which are essential for optimizing supercapacitor performance. The $NiCo_2O_4$/rGO/NF composite acts as a binder-free integrated electrode, achieving an exceptional specific capacitance of 2863.4 F g^{-1} (1503.3 C g^{-1}) at a current density of 1 A g^{-1}. It

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also exhibits remarkable rate performance, with a specific capacitance of 2335.2 F g^{-1} at 20 A g^{-1}, and maintains 91.7% capacitance retention after 5000 cycles, indicating excellent stability. Furthermore, the study constructs a solid-state asymmetric supercapacitor using the $NiCo_2O_4$/rGO/NF composite as the positive electrode and commercial activated carbon as the negative electrode. This configuration achieves a high energy density of 69.2 Wh kg^{-1} at a power density of 800 W kg^{-1}, retaining 48.9 Wh kg^{-1} even at a peak power density of 20004 W kg^{-1}, alongside a commendable cycling stability of 87.2% after 10,000 cycles (Figure 11). The performance enhancements are attributed to several factors, including the strong synergy among the mesoporous $NiCo_2O_4$ nanosheets, the rGO modification layer, and the 3D nickel foam skeleton. This combination significantly accelerates electron conduction kinetics and promotes efficient electron transfer. The thin nanosheet array structure provides sufficient open spaces to accommodate stresses during the redox process while offering increased active sites for ion and electron diffusion. Moreover, the integrated electrode design reduces internal resistance by eliminating the need for complex powder loading processes. This work highlight the potential of the $NiCo_2O_4$/rGO/NF composite as a practical solid-state energy storage device. Their findings demonstrate the effectiveness of the CA-assisted coprecipitation-hydrothermal strategy in achieving superior structural stability and performance in energy storage applications, showcasing the rational design of nanosheet array structures to advance supercapacitor technology.

Table 2 provides a comprehensive comparison of the advantages and disadvantages of transition metal oxides as supercapacitor electrodes [77, 78]. Transition metal oxides exhibit high specific capacitance, undergo pseudocapacitive reactions, and offer excellent energy density and long-term stability, making them ideal for high-performance energy storage. They also demonstrate chemical stability, which allows them to function in harsh environments. However, challenges such as poor electrical conductivity, mechanical instability, electrode degradation, and complex fabrication processes limit their potential. Additionally, capacity fading and limited operating voltage ranges are concerns, and the use of non-environmentally friendly elements adds to their drawbacks.

2.3. Conducting Polymers

Conducting polymers, such as polyaniline (PANI), polypyrrole (PPy), and poly(3,4-ethylenedioxythiophene) (PEDOT), offer attractive properties for supercapacitors, including high specific capacitance and good flexibility. Ongoing research aims to improve their cycle stability and explore novel synthesis techniques to achieve better control over their morphology and properties. These materials exhibit high electrical conductivity, excellent redox activity, and tunable charge storage capacity, enabling efficient charge transfer and rapid energy storage [79]. These electrodes have several advantages over traditional materials like carbon-based electrodes. Their ability to undergo reversible redox reactions allows for high specific capacitance and excellent energy storage capabilities. Additionally, conducting polymers offers flexibility and mechanical stability, making them suitable for flexible and wearable energy storage devices. However, challenges such as limited cycling stability and volume changes during charge-discharge cycles have been areas of research focus. Researchers are exploring various strategies, such as nanocomposite formation and nano structuring, to improve the stability and performance of conducting polymer-based electrodes [80].

Polyaniline (PANI): Polyaniline (PANI) is commonly used as an electrode material in supercapacitors due to its unique combination of properties that make it well-suited for energy storage applications. PANI is intrinsically conductive, which means it allows the efficient movement of electrons within its structure (Figure 12). This property is essential for rapid charge and discharge processes, making it ideal for applications where high-power density is required. In supercapacitors, rapid charge and discharge are critical for providing bursts of energy quickly, such as in regenerative braking systems or emergency power backup. Unlike traditional double-layer capacitors that rely on the physical separation of charge at the electrode-electrolyte interface, PANI-based supercapacitors exhibit pseudo capacitance.

This means that PANI undergoes reversible faradaic redox reactions at the electrode surface during charge and discharge. These redox reactions result in additional charge storage capacity, significantly increasing the specific capacitance of the material. This is crucial for enhancing the energy storage capability of supercapacitors. PANI can switch between different oxidation states during the charging and discharging processes. This redox activity contributes to the pseudo capacitance effect and allows for the storage of electrical energy through chemical reactions, in addition to the electrostatic charge storage in traditional capacitors. PANI exhibits good chemical stability in a variety of electrolytes, especially in aqueous solutions. This stability is essential for maintaining the long-term performance and cycle life of supercapacitors. It ensures that PANI-based electrodes do not degrade or corrode over time, providing reliable energy storage solutions.

In a comparative study, researchers [81] investigated the impact of various metal oxides (MO) on the properties of a polymeric matrix. They synthesized three different nanocomposites: PANI@Al_2O_3, PANI@TiC, and PANI@TiO_2, employing in situ polymerization with ammonium persulfate as an oxidant. The team conducted a comprehensive characterization of the materials using multiple analytical techniques. Additionally, they evaluated the materials' conductive properties through the four-point probe method. The analytical results consistently confirmed the presence of MO in the final products, as evidenced by various analyses. Spectroscopic investigations revealed intriguing interactions between the MOs and PANI. Remarkably, the study found that incorporating MO into the polymeric matrix led to enhanced thermal stability.

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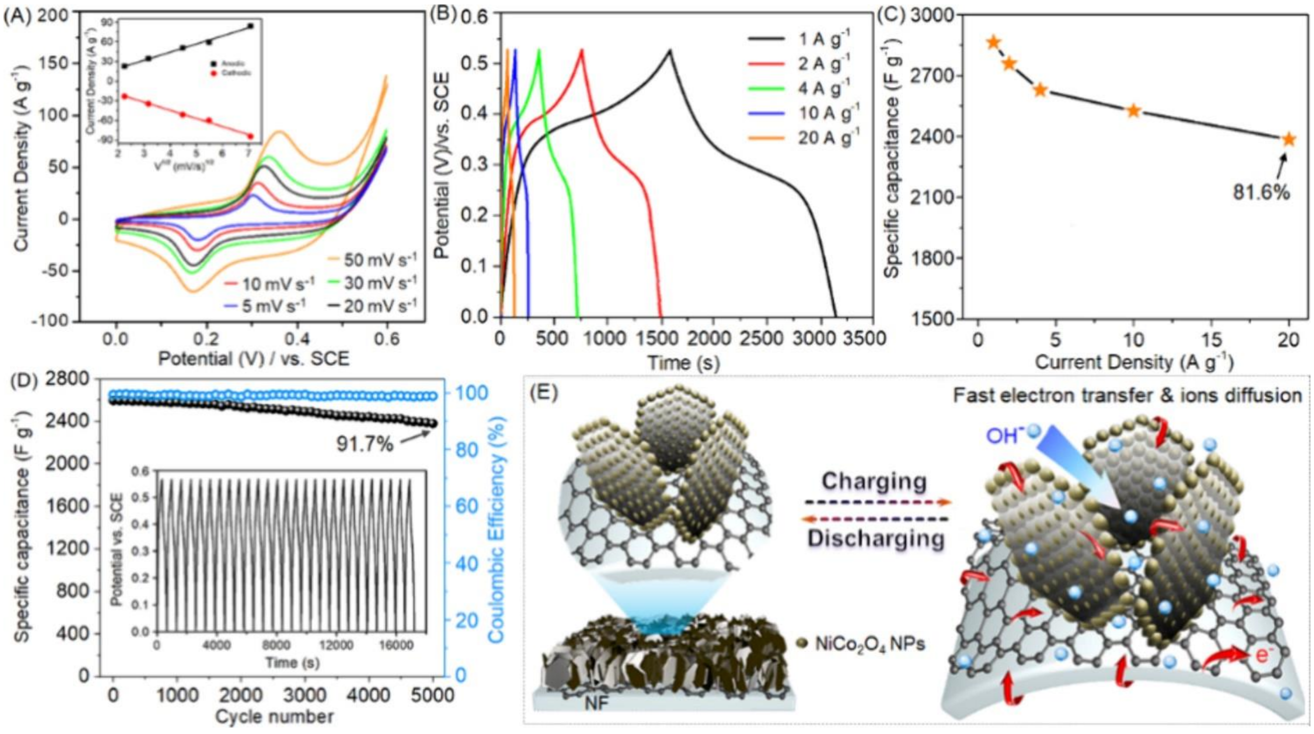

Fig. 11. Rational design of nanosheet array-like $NiCo_2O_4$ derived from layered double hydroxides, in situ grown on reduced graphene oxide-coated nickel foam, for enhanced supercapacitor applications. Reprinted with permission from Ref. [11], Wei, Z., Wang, Q., Qu, M. and Zhang, H., 2024. Rational Design of Nanosheet Array-Like Layered-Double-Hydroxide-Derived $NiCo_2O_4$ In Situ Grown on Reduced-Graphene-Oxide-Coated Nickel Foam for High-Performance Solid-State Supercapacitors. *ACS Applied Materials & Interfaces*, *16*(15), pp.18734-18744. Copyright © American Chemical Society.

Table 2. Advantages and disadvantages of Transition Metal-Oxides as supercapacitor electrodes.

	Advantages	**Disadvantages**	**Ref.**
Transition Metal Oxides	High specific capacitance, undergo pseudocapacitive reactions, higher energy density, excellent cycle life and long-term stability, chemically stable and can withstand harsh environments, enhancing their durability, higher energy density	Poor electrical conductivity result in high internal resistance, mechanical instability and electrode degradation over time, may suffer from capacity fading over extended cycling due to structural changes or electrode/electrolyte interactions, controlled morphologies and compositions can be challenging, require complex fabrication processes to achieve high performance, limited operating voltage ranges, elements used are not environment friendly.	[77, 78]

Furthermore, the research unveiled specific effects of different MOs on the properties of the nanocomposites. When TiO_2 was incorporated into the PANI matrix, it significantly improved the optical bandgap of the resulting nanocomposite. However, this improvement came at the cost of decreased electrical conductivity compared to other conducting materials. The team conducted electrochemical tests to explore the potential practical applications of these hybrid nanocomposites, including cyclic voltammetry (CV) and galvanostatic charge/discharge (GCD). The outcomes

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suggested that the PANI@TiO_2 nanocomposite exhibited promising characteristics as an electrode material, particularly for high-performance supercapacitor applications.

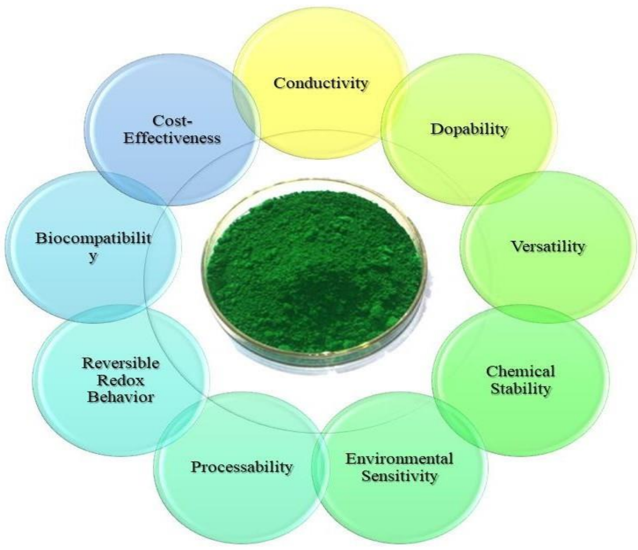

Fig. 12. Various properties of Polyaniline.

Polypyrrole (PPy): Polypyrrole (PPy) is another conducting polymer that is used as an electrode material in supercapacitors for several reasons (Figure 13). PPy is an inherently conductive polymer, allowing for efficient electron transport within its structure. This high electrical conductivity is crucial for enabling rapid charge and discharge processes in supercapacitors, leading to high power density. Like polyaniline (PANI), exhibits pseudo capacitance. This means that during the charge and discharge cycles, PPy undergoes reversible faradaic redox reactions at the electrode-electrolyte interface. These redox reactions contribute to additional charge storage capacity beyond the simple electrostatic double-layer capacitance. PPy's pseudocapacitive behavior enhances the overall energy storage capability of the supercapacitor. PPy is known for its chemical stability in various electrolytes, including both aqueous and organic solutions. This stability ensures the long-term reliability and durability of supercapacitors containing PPy electrodes, as they resist degradation and corrosion over time.

PPy can be synthesized and modified with relative ease, allowing researchers to fine-tune its properties for specific supercapacitor applications. By adjusting factors like polymer morphology, dopant type, and synthesis conditions, the performance of PPy electrodes can be optimized to meet particular energy storage requirements. The cost of producing PPy electrodes is generally reasonable, making it economically viable for large-scale production of supercapacitors. This cost-efficiency is advantageous for applications where cost considerations are essential. PPy is considered environmentally friendly because it can be synthesized using non-toxic and environmentally safe chemicals. This aligns with the growing emphasis on sustainable and eco-friendly materials in various industries, including energy storage. PPy can be used in both aqueous and organic electrolytes, providing flexibility in designing supercapacitors for various operating conditions and applications. Polypyrrole (PPy) is utilized as electrode material in supercapacitors due to its combination of high electrical conductivity, pseudocapacitive behavior, chemical stability, ease of modification, cost-efficiency, environmental friendliness, and versatility. These properties make PPy a valuable choice for enhancing the energy storage capacity and performance of supercapacitors in a wide range of applications, from portable electronics to renewable energy storage and electric vehicles.

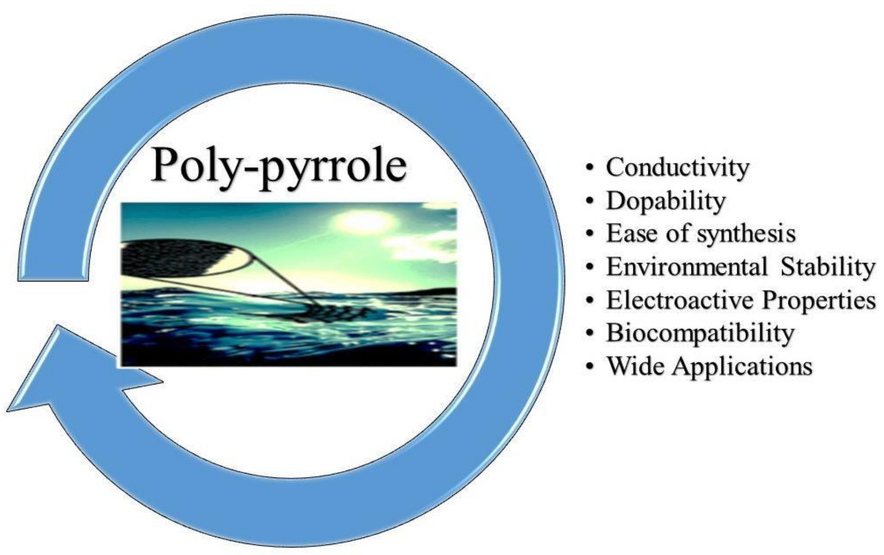

Fig. 13. Various properties of Polypyrrole.

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Poly(3,4-ethylenedioxythiophene) (PEDOT): Poly(3,4-ethylenedioxythiophene), commonly referred to as PEDOT, is a conducting polymer that is used as an electrode material in supercapacitors for several compelling reasons (Figure 14). PEDOT is an intrinsically conductive polymer, which means it can efficiently transport electrons within its structure. This property is crucial for facilitating rapid charge and discharge processes in supercapacitors, resulting in high power density. Like polypyrrole (PPy) and polyaniline (PANI), PEDOT exhibits pseudo capacitance. During the charge and discharge cycles, PEDOT undergoes reversible redox reactions at the electrode-electrolyte interface. These redox reactions contribute to additional charge storage capacity, enhancing the overall energy storage capability of the supercapacitor. PEDOT is known for its chemical stability in various electrolytes, including aqueous and organic solutions. This stability ensures the long-term reliability and durability of supercapacitors containing PEDOT electrodes, as it resists degradation and corrosion over time. PEDOT can be synthesized with a high surface area, which provides more active sites for charge storage and contributes to a higher specific capacitance. This property is essential for increasing the overall energy storage capacity of the supercapacitor. PEDOT can be incorporated into flexible and lightweight electrode materials, making it suitable for applications that require conformal or flexible energy storage solutions. This flexibility allows for the integration of supercapacitors into various form factors, including wearable electronics and flexible devices. PEDOT can be synthesized and processed using relatively simple methods. Additionally, its properties can be tailored through chemical doping and structural modifications, allowing for the optimization of its characteristics for specific supercapacitor applications. PEDOT is considered environmentally friendly because it can be synthesized using non-toxic and environmentally safe chemicals. This aligns with the growing emphasis on sustainable and eco-friendly materials in various industries, including energy storage. PEDOT can be used in both aqueous and organic electrolytes, providing flexibility in designing supercapacitors for different operating conditions and applications.

Rajesh et al. [82] presented a novel approach for the fabrication of supercapacitor electrodes by directly depositing chloride ions doped poly(3,4-ethylenedioxythiophene) (Cl-PEDOT) porous nanostructures on graphite substrates through electrochemical polymerization using ferric chloride ($FeCl_3$) as the supporting electrolyte. This method is advantageous due to its simplicity and cost-effectiveness, producing electrodes with a hierarchical porous structure that enhances charge storage and ion diffusion. The optimized deposition and cleansing process resulted in well-defined Cl-PEDOT nanofoam on graphite electrodes, which exhibited exceptional electrochemical properties. The electrodes achieved a maximum specific capacitance of 480 F g^{-1} at a current density of 2 A g^{-1} and demonstrated remarkable stability, maintaining ~95% capacitance after 10,000 charge/discharge cycles in 1M H_2SO_4. When assembled into a symmetrical supercapacitor, the electrodes showed a specific capacitance of 189 F g^{-1}, with ~86% retention over 10,000 cycles. Additionally, the device displayed high specific energy (6.19 W h kg^{-1}) and specific power (50.12 kW kg^{-1}), along with excellent rate capability at higher current densities. This work underscores the potential of Cl-PEDOT porous nanostructures for advanced supercapacitor applications. PEDOT is a valuable choice as electrode material in supercapacitors due to its combination of high electrical conductivity, pseudocapacitive behavior, chemical stability, large surface area, flexibility, ease of processing and modification, environmental friendliness, and compatibility with various electrolytes.

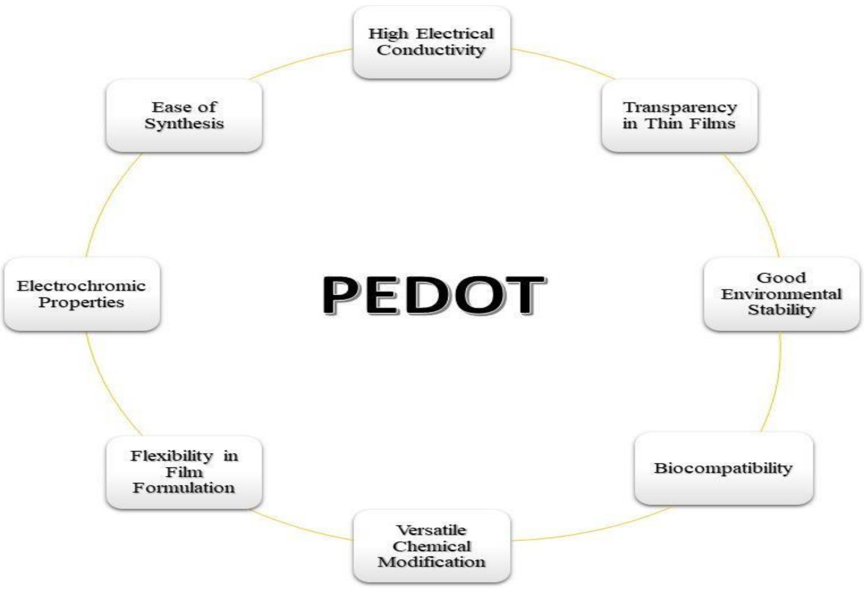

Fig. 14. Various properties of PEDOT.

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These properties make PEDOT a versatile material for enhancing the energy storage capacity and performance of supercapacitors, particularly in applications ranging from portable electronics to renewable energy storage and electric vehicles.

Polythiophene (PTh): Polythiophene (PTh) is a conducting polymer that has been explored for use as an electrode material in supercapacitors, primarily due to its electrical conductivity, environmental stability, and ease of synthesis. Here are some reasons why PTh is considered for supercapacitor electrodes (Figure 15). PTh exhibits good electrical conductivity, which is essential for efficient charge and discharge processes in supercapacitors. This property enables the rapid movement of electrons within the electrode, contributing to high power density.

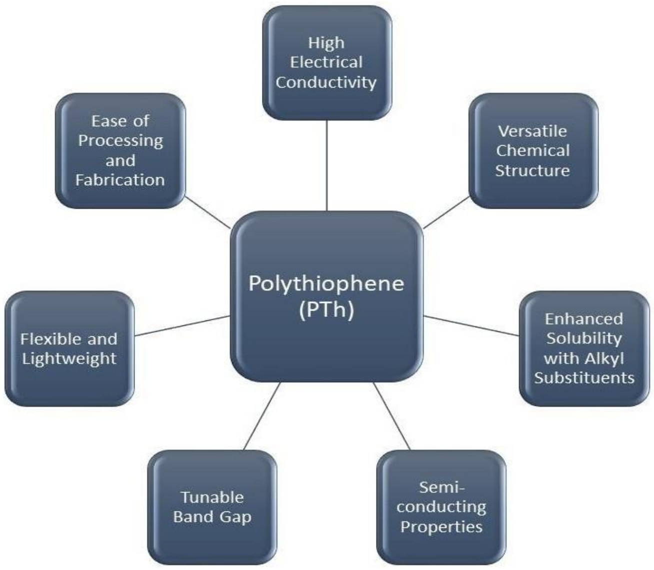

Fig. 15. Various properties of Polythiophene (PTh).

Like other conducting polymers like polypyrrole (PPy) and polyaniline (PANI), PTh can exhibit pseudo capacitance. PTh is known for its environmental stability, particularly in the context of aqueous electrolytes. It can resist degradation and corrosion over time, ensuring the long-term reliability and durability of supercapacitors containing PTh electrodes. PTh can be synthesized using relatively straightforward methods, making it accessible for research and development.

Moreover, its properties can be tailored through chemical doping and structural modifications, allowing for the optimization of its characteristics for specific supercapacitor applications. PTh can be used in both aqueous and organic electrolytes, offering versatility in designing supercapacitors for various operating conditions and applications. The cost of producing PTh electrodes is generally reasonable, making it economically viable for large-scale production of supercapacitors. This cost-effectiveness is advantageous for applications where budget considerations are essential. PTh can be deposited on flexible substrates, which is advantageous for creating flexible and lightweight supercapacitor devices. This flexibility allows for the integration of supercapacitors into wearable electronics and flexible devices. PTh synthesis can be carried out using non- toxic and environmentally safe chemicals, aligning with the growing emphasis on sustainable and eco-friendly materials in various industries. While PTh possesses several desirable properties for supercapacitor electrodes, it is essential to consider the specific requirements and constraints of the intended application. Researchers continue to investigate and develop various conducting polymers, including PTh, to enhance the performance of supercapacitors and tailor them to meet the demands of emerging technologies and energy storage solutions

Table 3 highlights the advantages and disadvantages of various conducting polymers used as supercapacitor electrodes. Polyaniline (PANI) exhibits high conductivity and pseudocapacitive behavior, but its performance is highly dependent on doping and may suffer from air stability and mechanical fragility issues. Polypyrrole (Ppy) offers relatively high specific capacitance and long cycling stability but faces challenges with doping and has lower capacitance as a cathode material. PEDOT is valued for its conductivity, flexibility, and stability, but it is costly to produce and sensitive to moisture. Polythiophene (PTh) is easy to synthesize and environmentally stable but has limitations in voltage range, mechanical strength, and long-term stability.

4. CONCLUSION

In this review, we have provided an extensive exploration of supercapacitor technology, with a specific focus on recent advancements in materials, fabrication techniques, and applications. Supercapacitors have garnered significant attention as a promising alternative to traditional batteries due to their remarkable power density, rapid charge-discharge capability, and long cycle life. However, their relatively low energy density and high production costs remain major obstacles to widespread adoption in large-scale energy storage systems. The review has highlighted the critical role that materials research plays in improving supercapacitor performance. Carbon-based materials, such as graphene and carbon nanotubes, have been shown to significantly enhance conductivity and surface area, offering excellent prospects for increased energy and power densities. Metal oxides, such as manganese dioxide and ruthenium oxide, as well as conductive polymers like polyaniline and polypyrrole, have demonstrated high electrochemical stability and capacitance. Hybrid materials, which combine the advantages of these diverse material classes, have emerged as one of the most promising avenues for further performance improvements.

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Table 3. Advantages and disadvantages of conducting polymer as a supercapacitor electrode.

Conducting polymer	Advantages	Disadvantages	Ref.
PANI	Highly conductive, pseudocapacitive behavior, Ease of synthesis, flexibility	Highly dependent on doping, and it may have limitations related to air stability, solubility, and mechanical fragility.	[83]
Ppy	Has relatively high specific capacitance, long cycling stability, and compatibility with neutral electrolytes. Materials offer advantages such as flexibility, straightforward fabrication.	Doping/de-doping difficulty, relatively low specific capacitance per unit g (only for cathode materials)	[84]
PEDOT	Conductivity, employed in creating bendable and stretchable electronic devices, stability, optical transparency.	Relatively high production cost, sensitivity to moisture, limited solubility, and potential environmental concerns associated with its manufacturing processes.	[85]
PTh	Flexible, simple to synthesize, desirable cycle stability, and environmental stability.	Limited voltage window, low mechanical strength, and potential long-term stability issues.	[86]

These hybrids are able to address some of the limitations associated with individual materials, including issues of cycle stability and energy density. In addition to material advancements, the review has explored novel fabrication techniques, such as solution-based methods, chemical vapor deposition (CVD), and template-assisted processes, which enable the creation of optimized electrode architectures. These techniques are critical for achieving the fine structural control necessary for maximizing the performance of supercapacitors. Moreover, the scalability of these fabrication methods is essential for reducing costs and ensuring that supercapacitors can be produced in large quantities for commercial applications. Supercapacitors are already being utilized in a wide range of applications, including electric vehicles, renewable energy systems, portable electronics, aerospace, and defense. Their ability to deliver high power quickly and their long operational life make them well-suited for industries requiring efficient, reliable energy storage solutions. However, challenges remain in increasing their energy density to levels comparable with batteries and reducing manufacturing costs to make them more commercially viable. In conclusion, the future of supercapacitors depends heavily on ongoing research and development in materials science, device engineering, and system integration. With continued advancements, supercapacitors have the potential to significantly impact the global energy storage landscape, offering a sustainable and efficient solution to modern energy demands.

5. FUTURE PERSPECTIVES

As the demand for advanced energy storage technologies continues to grow, supercapacitors hold immense potential to meet the evolving needs of various industries, from electric vehicles to renewable energy systems and consumer electronics. However, to fully realize this potential, significant advancements are required in both the fundamental design of supercapacitors and their practical deployment. The future of supercapacitor technology will be shaped by innovation across multiple dimensions, including materials development, fabrication methods, system integration, and environmental sustainability. Future research must address existing challenges such as increasing energy density, lowering production costs, and improving the environmental footprint of supercapacitor production. By focusing on these key areas, supercapacitors could move from niche applications to become a mainstream solution in global energy storage. In this section, we outline several critical directions that the field must take to push the boundaries of supercapacitor performance and pave the way for broader adoption in next-generation technologies.

Development of High-Energy-Density Materials: While significant progress has been made in improving power density, enhancing energy density remains a critical focus area. Future research should explore novel materials, such as metal-organic frameworks (MOFs), 2D materials beyond graphene, and composite structures that combine high surface area with efficient charge storage.

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Cost-Effective, Scalable Fabrication Techniques: Addressing the scalability and cost issues of advanced supercapacitor materials will be essential for commercial applications. Techniques like 3D printing, roll-to-roll manufacturing, and green synthesis methods should be developed to produce high-performance supercapacitors at lower costs.

Integration with Renewable Energy Systems: Future research should explore how supercapacitors can be seamlessly integrated with renewable energy systems, such as solar and wind, to buffer energy supply and enhance grid stability. Investigating hybrid systems that combine batteries and supercapacitors for optimal energy management could be a key area for innovation.

Advanced Electrolytes and Solid-State Supercapacitors: The development of solid-state electrolytes or ionic liquids with a wider electrochemical window could significantly boost both energy density and safety. Research should focus on creating stable, non-toxic, and high-conductivity electrolytes suitable for next-generation supercapacitors.

Environmental Sustainability: Efforts should be made to investigate the environmental impact of supercapacitor production and disposal. Biodegradable materials and recycling strategies for end-of-life supercapacitors should be prioritized to align with global sustainability goals.

Wearable and Flexible Supercapacitors: The future of portable electronics will likely include wearable devices that require flexible, lightweight, and durable energy storage solutions. Future research could focus on developing flexible supercapacitors with novel substrates and electrolytes that can maintain performance under mechanical stress.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests.

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