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Review Article

Properties of Electrode Materials and Electrolytes in Supercapacitor Technology

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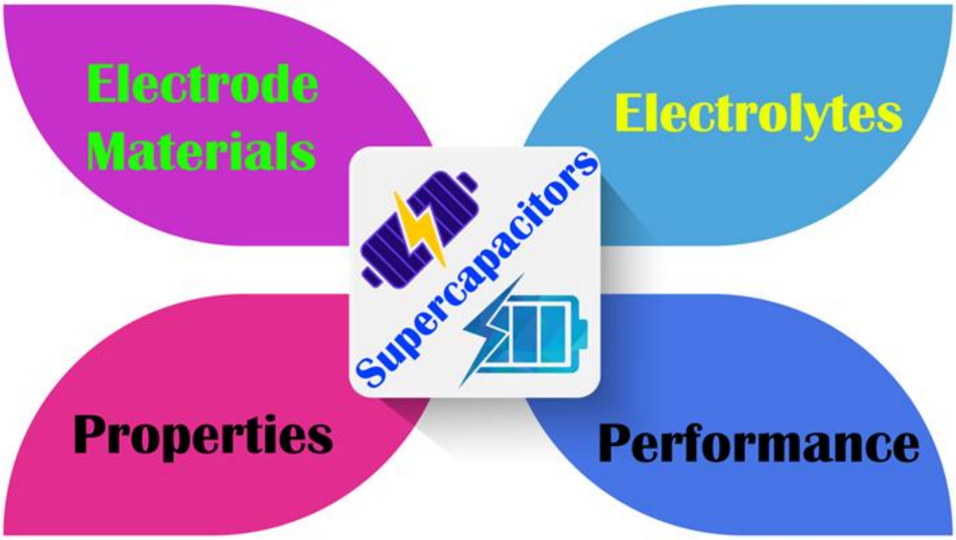
Abstract

This thorough review article offers a cutting-edge analysis of the essential characteristics and developments in electrode materials and electrolytes for supercapacitor technology. We start by going over the basics of supercapacitors and how important characterization methods like electrochemical impedance spectroscopy, galvanostatic charge-discharge, and cyclic voltammetry work. Specific capacitance, energy, and power densities, three essential characteristics that are crucial for assessing supercapacitor performance, are carefully covered in this work. We also analyze the many kinds of capacitors, including hybrid supercapacitors, electric double-layer capacitors, pseudocapacitors, and supercapacitors, and explain their working principles and material-specific characteristics. The study highlights the importance of metal oxides and hydroxides, carbon-based materials, conductive polymers, and novel and hybrid materials such as MXenes and metal-organic frameworks. The special qualities of each material class, such as large surface area, electrical conductivity, and particular redox properties, are highlighted in this section. These qualities are crucial for maximizing the performance of supercapacitors. The topic of electrode materials is discussed in detail, including their benefits and the difficulties and chances to improve energy storage, stability, and affordability. Parallel to this, the study thoroughly examines various electrolyte kinds, a sometimes overlooked yet essential part of supercapacitor technology. Discussed include ionic conductivity, operating voltage windows, safety profiles, and electrochemical stability of aqueous, organic, ionic liquid, gel, and solid-state electrolytes. This paper highlights the relationship between supercapacitor performance and electrolyte type, explaining how electrolyte selection affects total energy density, power density, and operational longevity. This review article covers supercapacitor technology in detail and with a wide scope and is an invaluable resource. It is a fundamental work for scholars and practitioners new to the area. It offers sophisticated insights that may encourage creativity and application-specific advancement in this quickly changing field. The study presents a comprehensive analysis of the present and future developments in supercapacitor materials and technology, establishing it as a vital resource in the continuous search for cutting-edge energy storage solutions.

Keywords: Supercapacitor technology, electrode materials, electrolyte innovations, energy storage efficiency, advanced capacitive mechanisms

Graphical abstract



1. Introduction

Supercapacitors, also known as ultracapacitors, represent a class of energy storage devices at the intersection of conventional capacitors and batteries. Characterized by their rapid charge and discharge capabilities, high power density, and longer cycle life, supercapacitors have carved a niche in energy storage, complementing and, in some cases, replacing batteries in various applications [1-4]. In today's technology-driven world, the role of supercapacitors is increasingly pivotal, catering to the growing demand for efficient energy storage solutions in renewable energy systems, electric vehicles, portable electronics, and power backup systems. The unique ability of supercapacitors to bridge the gap between the high energy storage capacity of batteries and the high power delivery of traditional capacitors makes them indispensable in applications where energy and power are crucial [5-9].

The journey of supercapacitors began in the early 20th century, with the initial concept dating back to the General Electric experiments in 1957. However, it wasn't until the 1970s and 1980s that significant strides were made in the development of supercapacitors, with the introduction of carbon-based electrodes and the exploration of various electrolytes [10]. The evolution of supercapacitor technology has been marked by continuous advancements in materials and design, leading to enhanced performance and broader applications. The supercapacitor technology has grown exponentially from the initial use of activated carbon (AC) and aqueous electrolytes to the recent developments in nanomaterials, conductive polymers, and ionic liquids. This evolution has been driven by the growing energy demands of modern technology and the need for sustainable and efficient energy storage solutions [11-23]. At its core, a supercapacitor consists of two electrodes, an electrolyte, and a separator. The electrodes, typically made of highly porous materials, are where the energy is stored, either through electrostatic charge accumulation (in EDLC) or through fast surface redox reactions (in pseudocapacitors). The electrolyte provides the medium for ion transport between the electrodes, while the separator prevents physical contact between the electrodes and allows for the passage of ions [24-27].

The working principle of supercapacitors hinges on two primary mechanisms: electrostatic double-layer capacitance and pseudocapacitance [28-30]. In electrostatic double-layer capacitance, ions from the electrolyte adsorb onto the surface of the electrodes, forming a double layer and storing charge non-Faradaically. This mechanism is predominant in EDLCs, where the energy storage is purely physical and reversible [31]. In contrast, pseudocapacitance involves Faradaic (redox) processes at or near the electrode surface, contributing to higher capacitance and energy density. Pseudocapacitors leverage materials that undergo these Faradaic reactions, such as metal oxides and conductive polymers [32-34]. The efficiency and performance of supercapacitors are largely dictated by the properties of the electrode materials and the electrolytes. The electrode materials, with their high surface area, conductivity, and electrochemical stability, determine the charge storage capacity and power delivery of the supercapacitor. On the other hand, the type of electrolyte influences the operational voltage window, ionic conductivity, and overall stability of the device. The exploration and optimization of these components have been central to the advancements in supercapacitor technology [35-37].

In recent years, significant research has been focused on developing advanced electrode materials and electrolytes to enhance the performance of supercapacitors. Carbon-based materials, such as AC, carbon nanotubes, and graphene, have been extensively studied for their high surface areas and excellent electrical conductivities [38-41]. Conductive polymers and metal oxides have also garnered attention for their pseudocapacitive properties, offering higher energy densities [42-46]. Emerging materials like MOFs and MXenes are being explored for their unique structures and electrochemical properties [39, 47-49]. MOFs and MXenes stand out in energy storage research due to their unique structural characteristics and superior electrochemical properties, including high porosity, customizable surface functionalities, and exceptional electrical conductivity. These attributes enable them to deliver enhanced performance in electrochemical supercapacitors and batteries, promising significant advancements in energy storage technologies [48, 50]. Similarly, the development of electrolytes has seen a shift from traditional aqueous and organic electrolytes to more advanced options like ionic liquids and gel or solid-state electrolytes [51-54]. These new electrolytes offer extended operational voltage windows, improved safety profiles, and the potential for flexible supercapacitor designs.

This review paper aims to provide a comprehensive overview of the current state-of-the-art in electrode materials and electrolytes for supercapacitors. By delving into the properties, advantages, and challenges associated with various materials and electrolytes, this paper sheds light on the key factors influencing the performance and applicability of supercapacitors. The insights offered in this review are intended to guide future research and development efforts in supercapacitor technology, paving the way for more efficient, reliable, and sustainable energy storage solutions. As the world increasingly turns towards renewable energy sources and seeks efficient energy storage systems, the role of supercapacitors, with their rapid charging capabilities and high power densities, becomes ever more critical. The advancements in electrode materials and electrolytes discussed herein are at the forefront of this technological evolution, marking a significant stride in the journey towards advanced energy storage systems.

2. Basics of supercapacitor technology

Supercapacitors, also known as ultracapacitors, are a type of energy storage device that bridges the gap between conventional capacitors and batteries. They store energy through a process that involves the electrostatic accumulation of charges, offering high power density and rapid charging and discharging capabilities compared to batteries [48, 55].

2.1. Key characterization techniques

2.1.1. Cyclic voltammetry

Cyclic voltammetry (CV) is a widely used electrochemical technique for analyzing the electrochemical properties of materials, especially in the study of supercapacitors. In CV, the potential of the working electrode is varied linearly with time in a cyclic manner, and the resulting current is measured. This process generates a plot of current versus voltage, known as a voltammogram, which provides valuable information about the electrochemical behavior of the electrode material [56-58]. CV helps in evaluating the electrochemical properties of electrode materials used in supercapacitors. It provides insights into the capacitive behavior, charge storage mechanism, and reversibility of the electrode reactions. CV can distinguish between capacitive (non-Faradaic) and pseudocapacitive (Faradaic) charge storage mechanisms. In a purely capacitive process, the current response is rectangular, while pseudocapacitance shows redox peaks. CV reveals the kinetics of the electrochemical reactions occurring at the electrode/electrolyte interface, including reaction rates and potential-dependent capacitance changes. In EDLCs, the CV curve is typically rectangular, indicating ideal capacitive behavior with negligible Faradaic reactions. Meanwhile, pseudocapacitors exhibit distinct redox peaks in the CV curve, indicating Faradaic processes contributing to the capacitance [59-61].

The potential window in CV for supercapacitors is critical and is selected based on the electrochemical stability of the electrode material and the electrolyte. An ideal potential range ensures maximum capacitance without causing decomposition of the electrolyte or degradation of the electrode material. For aqueous electrolytes, the potential range is usually limited to 1.0-1.23 V due to water electrolysis limits [44, 62, 63]. For organic or ionic liquid electrolytes, a wider potential window (up to 2.5 - 2.7V) can be used. The scan rate in CV affects the shape of the voltammogram. A higher scan rate leads to a broader peak and higher current responses, indicating kinetic limitations. The choice of the electrochemical window in CV tests is crucial to avoid irreversible reactions that could damage the electrode or electrolyte [64-67]. CV can also provide information on leakage current and the electrochemical stability of the supercapacitor under repeated charge-discharge cycles. The symmetry and area under the CV curve are indicative of the efficiency and the specific capacitance of the supercapacitor, respectively. CV is a fundamental tool in supercapacitor research, offering insights into the electrochemical characteristics, charge storage mechanisms, and performance capabilities of supercapacitors. Its ability to provide detailed information about the electrode materials and their interaction with electrolytes makes it indispensable in the design and development of efficient and high-performance supercapacitors.

2.1.2. Galvanostatic charge-discharge

Galvanostatic charge-discharge (GCD) is a fundamental electrochemical technique used to evaluate the performance of supercapacitors. In GCD testing, a constant current is applied to charge and then discharge the supercapacitor, while the voltage across the supercapacitor is recorded over time. This results in a charge-discharge curve, essential for determining key performance parameters of supercapacitors, such as specific capacitance, energy density, and power density [68-71]. GCD is directly used to calculate the specific capacitance of the supercapacitor, which is a measure of its energy storage capacity. From the charge-discharge curves, energy density (how much energy can be stored) and power density (how quickly energy can be delivered) are calculated, providing a comprehensive understanding of the supercapacitor's performance. Repeated GCD cycles are used to assess the cyclic stability and lifetime of the supercapacitor, crucial for practical applications [68].

During the charge phase, the voltage increases linearly with time until a set voltage is reached. During discharge, the voltage decreases linearly. The shape of the curves provides insight into the capacitive behavior and efficiency of the supercapacitor. An initial sudden drop in voltage, known as the IR drop, is observed at the beginning of the discharge phase. It's indicative of the internal resistance of the supercapacitor. The linearity and symmetry of the charge and discharge curves are indicative of the ideal capacitive behavior and low resistance of the supercapacitor. Varying the current density in GCD tests helps to understand the rate capability and performance of the supercapacitor under different loads. Performing GCD at different temperatures can provide information about the thermal stability and performance of the supercapacitor in various environmental conditions [72]. Long-term GCD cycling tests are crucial to evaluate the durability and stability of the supercapacitor over extended periods. GCD is a vital technique in the characterization of supercapacitors, offering comprehensive insights into their charge-discharge behavior, energy storage capacity, and overall performance. Through GCD, researchers and engineers can optimize supercapacitor design for a variety of applications, ensuring maximum efficiency and longevity [73, 74].

2.1.3. Electrochemical impedance spectroscopy

Electrochemical impedance spectroscopy (EIS) is a powerful and sensitive technique used to characterize the electrical and electrochemical properties of supercapacitors. In EIS, a small amplitude AC voltage is applied to the supercapacitor, and the resulting AC current is measured. By varying the frequency of the applied voltage (usually from mHz to MHz), EIS provides detailed information about the different resistive and capacitive elements within the supercapacitor. EIS helps in quantifying the internal resistance or impedance of the supercapacitor, which is crucial for high-power applications. EIS provides insights into the kinetics of charge transfer and ion diffusion within the electrodes and electrolyte. The data from EIS can be used to model the supercapacitor using an equivalent electrical circuit, helping to understand and optimize its performance. A key output of EIS is the Nyquist plot, which graphs the imaginary part of the impedance against the real part at different frequencies. Key features of a Nyquist plot for supercapacitors include high-frequency region, mid-frequency region, and low-frequency region. The intercept of the Nyquist plot with the real axis at high frequencies represents the equivalent series resistance (ESR) of the supercapacitor. ESR includes contributions from the resistance of the electrode material, the electrolyte, and the contact resistances. The semicircular portion in the mid-frequency range is associated with the charge transfer resistance (R_{CT}) at the electrode/electrolyte interface. The vertical line (or Warburg impedance) in the low-frequency range is indicative of the ion diffusion in the porous structure of the electrode.

In EIS, the frequency of the applied AC voltage is varied, and the impedance response is measured. This sweep covers a wide range of frequencies to capture various electrochemical processes. The impedance data is typically interpreted using an equivalent circuit model, which simplifies the complex electrochemical processes into electrical components like resistors, capacitors, and Warburg elements. Ideal capacitive behavior is indicated by a vertical line in the Nyquist plot at low frequencies, signifying that the impedance is primarily capacitive. EIS can be used to determine the relaxation time of the supercapacitor, which is important for understanding its charge-discharge rate capabilities. EIS measurements at different temperatures can reveal the effects of temperature on the impedance and performance of the supercapacitor. Similarly, the choice of electrolyte can be evaluated based on its impact on the impedance characteristics. The supercapacitor's performance at different frequencies can provide insights into its suitability for various applications, particularly where response time is critical. EIS is an indispensable technique in the field of supercapacitors, providing a comprehensive understanding of their internal dynamics, including resistance, capacitance, and charge transfer processes. The detailed insights gained from EIS, particularly through the analysis of Nyquist plots, are crucial for the design, optimization, and application of high-performance supercapacitors.

2.2. Fundamental parameters

2.2.1. Specific capacitance

Capacitance is a measure of a supercapacitor's ability to store charge. It's defined as the amount of charge stored per unit voltage and is expressed in farads (F). Whereas specific capacitance is a key parameter in supercapacitors, representing the capacitance per unit mass of the electrode material and is expressed in farads per gram (F/g). It is a measure of the energy storage capability of the supercapacitor and is crucial for evaluating and comparing the performance of different electrode materials. High specific capacitance indicates a greater ability of the electrode material to store electrical energy. It reflects the efficiency of the electrode material in utilizing its mass for charge storage. Knowing the specific capacitance helps in designing supercapacitors with desired energy and power densities for specific applications [75]. The specific capacitance (C_{sp}) from CV curves can be calculated using the equation (1).

$$C_{sp} = \frac{\int I \times \Delta V}{m \times \Delta V \times \nu} \qquad (1)$$

Where, m is the mass (g) of the active material in the electrode(s), ν is the scan rate (V/sec), ΔV is the operating potential window (OPW, V), and∫I×dV is the integrated area under CV curve (W). The specific capacitance (C_{sp}) from GCD profile is calculated from the discharge curve using equation (2).

$$C_{sp} = \frac{I \times \Delta t}{m \times \Delta V} \qquad (2)$$

Where, I is the discharge current (A), Δt is the discharge time (sec), m is the mass of the active material in the electrode(s) (g), and ΔV is the OPW during discharge (V). The intrinsic properties of the electrode material, such as porosity, electrical conductivity, and surface chemistry, significantly affect specific capacitance. The type of electrolyte and its ionic conductivity play a crucial role in charge storage and thus impact specific capacitance. The design and architecture of the electrode, including its thickness, surface area, and pore size distribution, influence the accessibility of electrolyte ions to the electrode surface, affecting capacitance. Conditions like scan rate in CV and current density in GCD affect the measured specific capacitance. Understanding and maximizing specific capacitance is vital for the development of high-performance supercapacitors. This involves synthesizing advanced electrode materials with high surface areas, optimizing electrode structures, and selecting suitable electrolytes. The specific capacitance is a crucial benchmark for researchers and engineers in the field to develop more efficient energy storage devices [76]. Specific capacitance is a fundamental metric in supercapacitor technology, integral to assessing and enhancing their energy storage capabilities. Accurate measurement and understanding of specific capacitance, derived from CV and GCD techniques, are essential for advancing supercapacitor technology and its applications in various fields.

2.2.2. Energy density

Energy density is a crucial parameter for supercapacitors, indicating how much energy a supercapacitor can store per unit mass or volume. It is a direct measure of the efficiency and effectiveness of a supercapacitor as an energy storage device. It's expressed in watt-hours per kilogram (Wh/kg) or watt-hours per liter (Wh/L). Higher energy density means more energy storage in a given volume or mass, making the supercapacitor more efficient for practical applications. Energy density (E) is typically calculated from the discharge curve of GCD profile obtained in a two-electrode system using equation (3).

$$E = \frac{1}{2} \times C_{sp} \times \Delta V^2 \times \frac{1000}{3600} \qquad (3)$$

Where, C_{sp} is the specific capacitance obtained from GCD (F/g or F/cm^3), ΔV is the OPW during discharge (V), while 1000/3600 is a conversion factor. For accurately measuring energy density, it is recommended to use the GCD profile of a two-electrode system. This setup mimics the actual working conditions of a supercapacitor, providing a realistic assessment of both energy density and overall device performance. The two-electrode system takes into account the balance and interactions between the positrode and negatrode, which is crucial for practical applications of supercapacitors. There are certain limitations of CV curves and three-electrode systems for energy density measurement. CV measures capacitance under a varying voltage rate, which does not accurately represent the constant current conditions under which supercapacitors typically operate. This can lead to an overestimation or underestimation of the energy storage capacity. The three-electrode system (with a working electrode, counter electrode, and reference electrode) is primarily used for fundamental studies and material characterization. In this setup, only the working electrode material is evaluated, and the results do not reflect the interactions between two identical or different electrodes as in practical supercapacitor cells. The three-electrode system does not accurately represent the real-world performance of a supercapacitor, which typically operates with two electrodes. The energy density calculated from a three-electrode setup might not be representative of the actual energy density in a practical two-electrode supercapacitor. In a practical supercapacitor, both electrodes often interact and contribute to the overall capacitance and energy storage. A three-electrode system does not capture this interplay between electrodes. The energy density of supercapacitors is best evaluated using GCD curves in a two-electrode system. This approach ensures that the measurements reflect the true energy storage capacity of the supercapacitor under conditions that are similar to its real-world application, making it a more reliable metric for assessing and comparing supercapacitor performance.

2.2.3. Power density

Power density is a critical parameter for supercapacitors, defining how quickly the stored energy can be delivered or discharged and is expressed in watts per kilogram (W/kg) or watts per liter (W/L). It is a key metric that differentiates supercapacitors from other energy storage devices, like batteries, which are typically characterized by higher energy densities but lower power densities. A high power density means the supercapacitor can release its stored energy rapidly, making it suitable for applications requiring quick bursts of power. Power density is crucial for understanding how efficient a supercapacitor is in delivering its stored energy under different operational conditions. Power density (P) is commonly calculated using the discharge GCD curve obtained in a two-electrode system using equation (4).

$$P = \frac{E \times 3600}{\Delta t} \qquad (4)$$

Where, E is the energy density (Wh/kg or Wh/L) and Δt is the discharge time (sec). This formula ensures that the power density is calculated under realistic operational conditions, reflecting the actual performance of the supercapacitor. For accurate measurement of power density, using GCD profile in a two-electrode system is recommended. This configuration mirrors the actual supercapacitor cell setup and provides a realistic assessment of how quickly the supercapacitor can deliver energy under practical use conditions. The two-electrode system considers the synergy between the two electrodes, crucial for evaluating the real-world performance of supercapacitors, especially in terms of power delivery. There are certain limitations of using CV curves and three-electrode systems for power density measurement. CV measures the response under a continuously varying voltage, which does not accurately simulate the constant current discharge conditions under which supercapacitors are typically used. This leads to an unrealistic assessment of the power density. Similar to the case with energy density, the three-electrode system is not representative of a practical supercapacitor cell. It provides information on the material properties at the electrode level but does not reflect the complete cell behavior. In a real-world application, supercapacitors operate with two electrodes, and the power density calculated from a three-electrode system does not accurately represent the power performance of a two-electrode supercapacitor. A three-electrode setup cannot accurately capture the dynamics between the anode and cathode in a two-electrode system, which is essential for determining the true power density of the supercapacitor. Power density in supercapacitors should ideally be measured using GCD curves in a two-electrode system. This approach offers a more accurate reflection of the supercapacitor's ability to quickly deliver energy, which is essential for many of its high-power applications. Understanding and optimizing power density is key to advancing supercapacitor technology and expanding its use in various fields where rapid energy discharge is critical.

2.2.4. Coulombic efficiency

Coulombic efficiency, often represented by the Greek letter η (eta), is a measure of the charge efficiency of a supercapacitor. It quantifies the ratio of the charge extracted from the supercapacitor during the discharge process to the charge stored in it during the charging process. This parameter is crucial for evaluating the effectiveness and reliability of supercapacitors in energy storage and delivery. Coulombic efficiency helps in assessing the energy losses during charge and discharge cycles, primarily due to internal resistance and side reactions. High Coulombic efficiency is indicative of less energy loss and more stable performance over repeated charge-discharge cycles, which is important for the longevity of the supercapacitor. It provides insights into the operational efficiency of the supercapacitor, which is essential for practical applications where energy conservation and efficient use are critical. Coulombic efficiency (η) can be calculated using equation (5).

$$\eta = \frac{t_d}{t_c} \times 100\% \qquad (5)$$

Where, t_d is the discharging time (sec) and t_c is the charging time (sec) in the GCD profile.

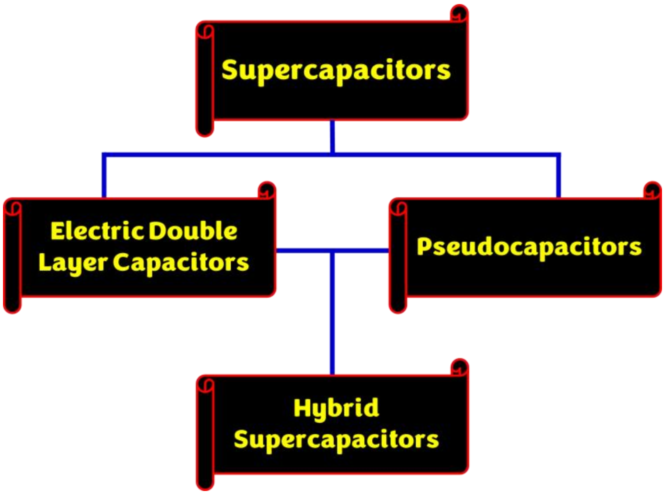
Higher internal resistance can lead to more energy loss as heat during charging and discharging, reducing Coulombic efficiency. Electrochemical side reactions, such as electrolyte decomposition or electrode material degradation, can consume charge without contributing to energy storage, thereby lowering Coulombic efficiency. Faster charge-discharge rates can sometimes reduce Coulombic efficiency due to incomplete charging or increased resistive losses. Operating temperature can also impact Coulombic efficiency, as it influences the internal resistance and reaction kinetics of the supercapacitor. Coulombic efficiency is a vital metric for assessing the charge efficiency and overall performance of supercapacitors. It provides valuable information for optimizing the design and operational parameters of supercapacitors to enhance their efficiency, stability, and lifespan. High Coulombic efficiency is a desirable characteristic in supercapacitors, especially for applications requiring reliable and efficient energy storage and delivery. Understanding these fundamental principles and characterization techniques is crucial for evaluating and improving supercapacitor performance. The ability to store and deliver energy efficiently, as quantified by these parameters, makes supercapacitors a versatile and valuable component in various technological applications, from consumer electronics to renewable energy systems.

2.3. Types of supercapacitors

Supercapacitors are classified mainly into three types, i.e., EDLCs, pseudocapacitors, and hybrid supercapacitors (also known as supercapatteries). The various types of supercapacitors are shown in **Scheme 1**. Each type has its unique mechanism of storing charge, materials used, and specific characteristics.

2.3.1. EDLCs

EDLCs store electrical energy by the electrostatic accumulation of charges at the electrode-electrolyte interface. The charge is stored physically, and there are no chemical reactions involved. Common materials for EDLC electrodes include various forms of carbon like AC, carbon nanotubes, and graphene, which provide a high surface area for charge storage. EDLCs exhibit high power density due to rapid charge-discharge capability, long cycle life with minimal degradation, and lower internal resistance. Whereas they have lower energy density compared to pseudocapacitors and batteries. Performance highly dependent on the surface area and porosity of the carbon materials used. In CV, EDLCs typically show rectangular-shaped curves, indicating ideal capacitive behavior. In GCD, EDLCs exhibit almost triangular charge-discharge profiles [77-80].



Scheme 1. Types of supercapacitors.

2.3.2. Pseudocapacitors

Pseudocapacitors store energy through Faradaic (redox) reactions that occur at or near the electrode surface. These reactions are typically reversible and fast. Conductive polymers and transition metal oxides are commonly used electrode materials in pseudocapacitors. They exhibit higher energy density than EDLCs due to Faradaic charge storage, while still maintaining relatively high power density and good cycling stability. Generally, pseudocapacitors have a shorter lifespan compared to EDLCs due to the degradation of active materials from repeated redox reactions [48, 81]. There are some challenges in pseudocapacitors like improving cycle life and stability while maintaining high energy density and developing cost-effective and environmentally friendly electrode materials. CV curves for pseudocapacitors show distinct redox peaks. In GCD, the charge-discharge curves are non-linear, often with plateaus corresponding to the redox processes [82, 83].

2.3.3. Hybrid supercapacitors (Supercapatteries)

Hybrid supercapacitors combine the charge storage mechanisms of EDLCs and pseudocapacitors. One electrode operates via an electrostatic mechanism (like in EDLCs), while the other involves Faradaic reactions (as in pseudocapacitors). A combination of materials from EDLCs and pseudocapacitors, such as AC paired with metal oxides or conductive polymers are used as electrode materials. Hybrid supercapacitors have higher energy density than standard EDLCs, thanks to the combination of capacitive and Faradaic charge storage mechanisms. The exhibit better power density and faster charge-discharge rates compared to traditional batteries [70, 84]. There are certain challenges in hybrid supercapacitors like optimizing the balance between energy and power density. Ensuring compatibility and stability of the different electrode materials. The CV curve of a hybrid supercapacitor may show a combination of rectangular capacitive behavior and redox peaks. The GCD profiles would be a mix of linear and non-linear regions, indicative of both capacitive and Faradaic processes. The choice among EDLCs, pseudocapacitors, and hybrid Supercapacitors depends on the specific application requirements like energy density, power density, cycle life, and cost. Advanced materials research is continually pushing the boundaries of these supercapacitors, aiming to combine the best of EDLCs and pseudocapacitors into efficient, durable, and cost-effective energy storage solutions [85-91].

2.4. Key performance indicators for supercapacitors

In the realm of supercapacitor technology, the evaluation of performance is pivotal for both academic research and practical applications. Key performance indicators (KPIs) such as specific capacitance, cyclic stability, and rate capability are essential metrics that define the efficiency and applicability of supercapacitors. Specific capacitance is a primary metric for assessing the charge storage capacity of a supercapacitor on a per-unit-mass basis. It is defined as the amount of charge stored per unit mass of the electrode material and is typically expressed in Farads per gram (F/g). The specific capacitance is indicative of how much energy a supercapacitor can store. It is directly influenced by factors like electrode material, surface area, pore size distribution, and the electrolyte used. Cyclic stability refers to the ability of a supercapacitor to maintain its performance over repeated charge-discharge cycles. It is a crucial indicator of the longevity and reliability of a supercapacitor. Cyclic stability is typically assessed by subjecting the supercapacitor to thousands or even hundreds of thousands of charge-discharge cycles, while monitoring parameters like capacitance retention and internal resistance. High cyclic stability is essential for applications where the supercapacitor will undergo numerous cycles over its lifetime, such as in energy storage systems and electronic devices. It is affected by the electrochemical stability of the electrode materials, the integrity of the electrode structure, and the quality of the electrolyte. Rate capability is a measure of how quickly a supercapacitor can charge and discharge, reflecting its performance under high-rate operational conditions [92-94]. Rate capability is evaluated by conducting charge-discharge tests at different current densities. A supercapacitor with good rate capability will show minimal decrease in specific capacitance even at high current densities. Rate capability is critical in applications requiring rapid bursts of energy, like in power backup systems and acceleration in electric vehicles [95-97]. It is largely dependent on the electrode material's conductivity, the ion transport in the electrolyte, and the interface between the electrode and electrolyte. These KPIs, specific capacitance, cyclic stability, and rate capability, collectively provide a holistic view of a supercapacitor's performance. They are interrelated and often require a balance; for instance, improving energy density (specific capacitance) might come at the cost of power density (rate capability). Understanding and optimizing these parameters are fundamental in the advancement of supercapacitor technology, guiding the development of more efficient, durable, and application-specific supercapacitors [98-100].

3. Electrode materials for supercapacitors and their properties

In the domain of supercapacitor, the choice of electrode material plays a pivotal role in determining the overall performance and efficiency. Different classes of materials, each with unique properties, have been extensively explored to enhance the capabilities of supercapacitors. This includes carbon-based materials, conductive polymers, metal oxides and hydroxides, MOFs, MXenes, and various emerging and hybrid composite materials. Carbon materials are widely used due to their high surface area, excellent conductivity, and remarkable chemical stability. AC, carbon nanotubes (CNTs), and graphene are prominent examples [101, 102]. AC, with its porous structure, offers high surface area for charge storage. CNTs and graphene, owing to their unique one-dimensional and two-dimensional structures, respectively, provide superior electrical conductivity and mechanical strength, enhancing both the energy and power densities of supercapacitors. Conductive polymers, including polyaniline (PANI), polypyrrole (PPy), and polythiophene, store charge via Faradaic redox reactions, offering higher specific capacitance compared to carbon materials. They are flexible, easy to process, and can be synthesized with a tailored structure for enhanced performance. However, their lower stability and degradation over cycling are challenges that need addressing. Transition metal oxides like RuO_2, MnO_2, and Co_3O_4, and hydroxides such as $Ni(OH)_2$, are used in pseudocapacitors for their high theoretical capacitance and energy density [103-107]. They undergo redox reactions contributing to charge storage. Despite their high capacitance, issues like poor electrical conductivity and cyclic stability limit their standalone use. MOFs, with their highly porous crystalline structures [108-111], offer a large surface area and tunable pore sizes, making them suitable for both EDLCs and pseudocapacitors. MXenes, a class of two-dimensional materials, have emerged recently, exhibiting high conductivity, flexibility, and hydrophilicity. Their layered structure enables fast ion transport, making them promise for high-rate supercapacitors. Research is continuously exploring novel materials like carbon aerogels, carbide-derived carbons, and composite/hybrid materials that combine the advantages of different classes (e.g., carbon materials with metal oxides). These materials aim to synergistically enhance energy density, power density, and cyclic stability, addressing the limitations of individual material classes. The exploration and optimization of electrode materials for supercapacitors is a dynamic and evolving field. Each material class brings its strengths and challenges, influencing the supercapacitor's performance in terms of energy and power density, stability, and rate capability. The ongoing advancements in material science, including the development of novel hybrid materials, are critical in pushing the boundaries of supercapacitor technology towards higher efficiency and broader applicability [112-114].

3.1. Carbon-based materials

Carbon-based materials are extensively utilized in supercapacitor electrodes, primarily due to their exceptional properties that align well with the operational requirements of supercapacitors. The high surface area, especially in forms such as AC, is a key feature, often exceeding 1000 m^2/g, which significantly enhances the specific capacitance by facilitating substantial charge accumulation. Additionally, advanced carbon materials like graphene and CNTs exhibit remarkable electrical conductivity, ensuring rapid electron movement within the electrode and contributing to high power density [115-118]. The porous nature of these materials is equally important, providing efficient ion transport pathways essential for swift charge-discharge cycles. This pore structure can be intricately tailored to optimize the interaction with electrolyte ions, maximizing charge storage efficiency. Moreover, the inherent thermal and chemical stability of carbon materials guarantees durability and long-term performance of supercapacitors, particularly under demanding operational conditions. Notably, graphene and CNTs, with their mechanical robustness and flexibility, pave the way for innovative flexible supercapacitor designs [119-123].

The preference for carbon-based materials in supercapacitors is underpinned by their balanced energy and power characteristics. While they are primarily used in EDLCs for electrostatic energy storage, their compatibility with various electrolytes and composite formations extends their applicability to pseudocapacitive mechanisms as well. This versatility is complemented by the environmental friendliness of carbon materials, especially those derived from sustainable sources, making them more attractive from an ecological standpoint. The economic aspect, particularly for AC with its low cost and scalability, is crucial for large-scale application and commercialization of supercapacitors [124-128]. Furthermore, the field of carbon materials is a hotbed of innovation, with ongoing research delving into areas like heteroatom doping, development of hierarchically porous structures, and the creation of 3D carbon networks. These advancements not only enhance the intrinsic properties of carbon materials but also open up new avenues for their application in supercapacitors, ensuring that they remain at the forefront of energy storage technology development.

Biomass-generated AC has exceptional supercapacitor efficiency because of its distinct structural characteristics and large surface area, especially when obtained from economical and sustainable sources like jute sticks [129-137]. Through a two-step pyrolysis method, as seen in **Figure 1a**, Shah et al. were able to effectively manufacture AC derived from jute sticks, which resulted in an outstanding surface area of over 2600 m²/g and a nanosheet-like shape (**Figure 1b-d**) [41]. In order to achieve high capacitance in supercapacitors, this enormous surface area is essential. Outstanding performance was found in a thorough electrochemical examination of symmetric supercapacitors using electrodes built on jute stick AC (JAC).

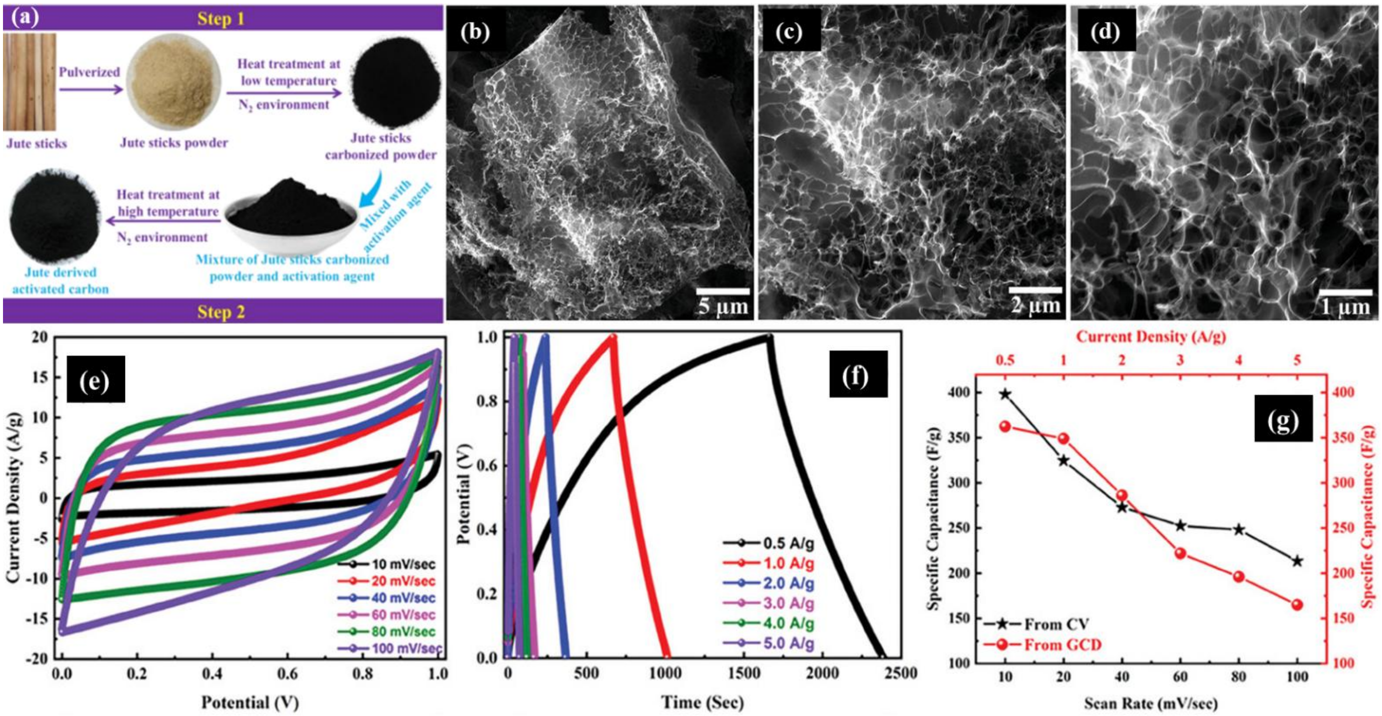


Figure 1. (a) Synthetic procedures for the two-step pyrolysis used to prepare AC obtained from jute sticks. FESEM micrographs (b–d) of the generated AC at various magnifications. CV curves at various scan rates, GCD profiles with varying current densities, and matching specific capacitances of the JAC-based symmetric supercapacitor are shown in (e), (f), and (g). *Reproduced with permission [41]. Copyright 2023, Wiley.*

These supercapacitors were tested in a two-electrode cell design with a PVA/KOH electrolyte utilizing CV, GCD, and EIS. Interestingly, the JAC-2 electrodes from this series showed a distinct rectangular shape in the CV curves (**Figure 1e**), indicating that they were capacitive. GCD profiles (**Figure 1f**), which displayed symmetrical charge/discharge curves indicative of electric double-layer capacitance and superior electrochemical reversibility, further supported this. Because JAC-2 has a large amount of mesopore content and a high surface area, it is better at electrolyte absorption and ion diffusion. This is demonstrated by the longest discharge duration and greatest specific capacitance in GCD experiments. As shown in **Figure 1g**, the JAC-2-based supercapacitors had the greatest specific capacitance values, reaching up to 400 F/g at a scan rate of 10 mV/sec. This image shows the high capacitive nature of JAC-2 as well as the high energy storage capacity that it may potentially have. At a current density of 1 A/g, these supercapacitors demonstrated a remarkable energy density of about 50 Wh/kg, indicating their great potential as materials for next-generation energy storage applications. The thorough electrochemical analysis, which includes a range of characterization methods, solidifies the JAC-based materials, JAC-2 in particular, as superior contenders for high-performance supercapacitors.

In the evolving landscape of supercapacitor technology, a diverse array of biomass-derived carbons and advanced carbon-based materials, including carbon nanofibers (CNFs), CNTs, and graphene, have garnered significant attention due to their exceptional performance in energy storage applications [73, 133, 138-146]. Biomass-derived N-doped carbon and chloride salt-AC are recognized as efficient electrode materials for enhancing the performance of electrochemical supercapacitors [147, 148]. Recent studies have explored various biomass sources, such as coconut shells [149], rice husks [150], and even agricultural waste [14, 151], transforming these into high-surface-area ACs through processes like carbonization and activation [132, 134]. These biomass-derived ACs offer a sustainable, cost-effective, and eco-friendly approach to electrode fabrication, exhibiting excellent electrochemical properties due to their intrinsic porous structures and high surface areas, which are imperative for efficient ion adsorption and rapid charge-discharge dynamics [152, 153]. Simultaneously, engineered carbon forms like CNFs, CNTs, and graphene represent the pinnacle of material innovation in supercapacitors [154-157]. CNFs, with their continuous and interconnected fibrous network, provide a unique combination of electrical conductivity, mechanical strength, and structural stability, enhancing electrode durability and capacitance. Carbon nanotubes, known for their exceptional electrical conductivity and mechanical properties, facilitate electron transport within electrodes and contribute to the overall resilience and efficiency of the supercapacitor. Graphene, a one-atom-thick layer of carbon atoms arranged in a hexagonal lattice, stands out due to its extraordinary electrical conductivity, high surface area, and mechanical flexibility. These properties make graphene an ideal candidate for high-performance supercapacitors, offering low equivalent series resistance (ESR) and high power density. The integration of these advanced carbon materials into supercapacitor electrodes has led to notable improvements in energy density, power density, and cyclic stability, pushing the boundaries of what is achievable in modern energy storage systems. The advancements in these carbon-based materials, from natural biomass derivatives to engineered nanostructures, highlight a significant trend in supercapacitor research: the synergy of sustainability with high-end material science [158, 159]. This convergence not only enhances the performance metrics of supercapacitors but also aligns with global efforts towards environmentally friendly and scalable energy storage solutions. As research continues to refine these materials and uncover new potentials, carbon-based materials are set to remain at the forefront of supercapacitor technology, driving innovations and applications in the field of advanced energy storage.

3.2. Conductive polymers

Conductive polymers have emerged as a significant class of materials in the development of supercapacitors, primarily attributed to their distinct electrochemical properties that are conducive to energy storage applications. These polymers, including prominent examples like PANI, PPy, and polythiophene, exhibit inherent conductivity and a unique ability to undergo reversible redox reactions. This trait is central to their role in pseudocapacitors [98, 160, 161]. The pseudocapacitive behavior of conductive polymers arises from the fast and reversible Faradaic reactions at the polymer-electrolyte interface, which contribute to higher specific capacitances compared to their carbon-based counterparts. Moreover, the ease of synthesis and the possibility of tuning their electrical and physical properties through chemical doping or copolymerization make them versatile for various supercapacitor applications. These polymers can be deposited on various substrates, including flexible materials, allowing for innovative supercapacitor designs, including flexible and wearable energy storage devices.

The exploration of conductive polymers in supercapacitors is driven by several advantages, such as their high theoretical specific capacitance and energy density, which are pivotal in enhancing the overall energy storage capability of supercapacitors. This property, coupled with their relatively low cost and environmental benignity, makes them attractive for sustainable and economical energy storage solutions. However, the application of conductive polymers in supercapacitors is not without challenges. A primary limitation is their cyclic stability; these polymers often suffer from structural degradation over repeated charge-discharge cycles due to swelling and shrinking during the doping/undoping processes. These polymers' mechanical properties and conductivity can also be sensitive to environmental conditions, such as humidity and temperature, which might limit their performance in varying operational environments [162]. Addressing these challenges, recent research in the field has been focusing on the development of composite materials, combining conductive polymers with other materials like carbon nanotubes or metal oxides, to create synergistic effects that enhance conductivity, stability, and mechanical strength. Such innovations are crucial in harnessing the full potential of conductive polymers in the realm of advanced supercapacitor technologies [163].

Because of their intrinsic electrical conductivity and special capacity for Faradaic charge storage, which greatly increases the energy density and capacitance, conductive polymers are very effective for supercapacitors. Shah et al. used electrochemical deposition to create PANI, a conductive polymer, which showed exceptional structural and electrochemical performance for supercapacitor applications (**Figure 2A**) [40]. PANI@CC was used as the positive electrode and TAC@CC as the negative electrode in their investigation to create an asymmetric supercapacitor. The separator was only a piece of filter paper saturated in an ionic liquid electrolyte. Selecting an ionic liquid electrolyte was essential as it has a larger operating potential window (OPW) than aqueous electrolytes, which increases the supercapacitor's energy density. As seen in **Figure 2B,a-f**, the electrochemical performance of this asymmetric supercapacitor was exceptional. CV and GCD experiments revealed that OPWs could reach up to 3.0 V and 2.8 V, respectively, which is a notable improvement above traditional aqueous supercapacitor. The quasi rectangular form and redox peaks of the CV curves in **Figure 2B,a** are suggestive of the behavior of a hybrid supercapacitor, which combines characteristics of both pseudocapacitance and EDLC. The discharge curves from the GCD experiments, which were carried out at different current densities within an OPW of 0–2.8 V, showed almost triangular forms (**Figure 2B,b**), indicating a combination of EDLC and pseudocapacitive nature. The asymmetric supercapacitor's strong conductivity was highlighted by its notable minimum IR drop. One important finding was the decrease in areal capacitance as current density increased. This pattern was explained by the restricted diffusion of electrolyte ions at higher rates. Because of the conductivity of PANI and the stability and large surface area of TAC, the special combination of these two materials produced an asymmetric supercapacitor with a high specific capacitance. Together with PANI's excellent electrical and ionic conductivities, TAC's well-designed pore structure allowed for effective ion transport and enhanced charge-transfer kinetics. Using the complementary qualities of PANI and TAC, this asymmetric supercapacitor showed an ultrahigh energy density of 270 µWh/cm^2 at a power density of 1400 µW/cm^2, and it continued to show an energy density of 251 µWh/cm^2 even at a higher power density of 2800 µW/cm^2. Additionally, after 10,000 charge-discharge cycles, EIS studies revealed outstanding cyclic stability, retaining around 95% of its original capacitance. Even after extended cycling, the TAC@CC electrode showed remarkable structural integrity, demonstrating its stability and durability in the ionic liquid electrolyte. The study conducted by Shah et al. yielded results that demonstrate the great potential of conductive polymers, in particular PANI, for the development of high-performance asymmetric supercapacitors. This development has the potential to make considerable progress in the field of energy storage technology.

In the realm of supercapacitor development, the exploration of conductive polymers has intensified, with materials like PANI leading the charge in demonstrating exceptional performance. Recent advancements extend beyond PANI to include a variety of other conductive polymers such as PPy, polythiophene (PTh), and polyacetylene (PA), each contributing uniquely to the enhancement of supercapacitor functionalities.

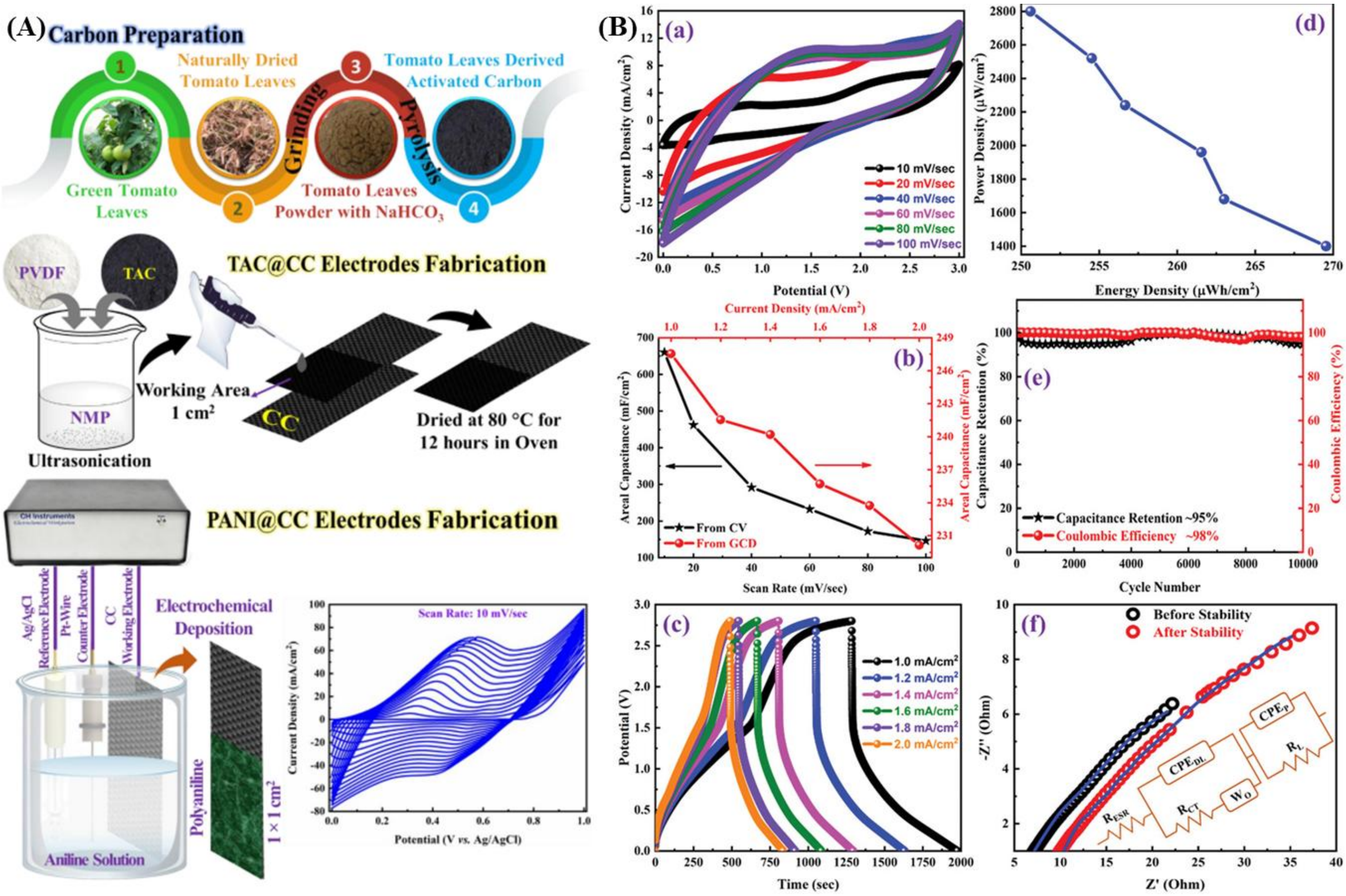


Figure 2. (a) PANI electrochemical deposition synthetic procedures. (B) Electroechemical performances of the generated asymmetric supercapacitor; (a) CV curves recorded at varying scan rates; (b) areal capacitances obtained from CV at varying scan rates and GCD at varying current densities; (c) GCD profiles recorded at varying current densities; (d) Ragone plot; (e) capacitance retention and coulombic efficiency for up to 10,000 cycles; and (f) Nyquist plots with the fitted equivalent circuit diagram prior to and following stability testing of the asymmetric supercapacitor in ionic liquid electrolyte. *Reproduced with permission [40]. Copyright 2023, Wiley.*

These polymers are prized for their inherent electrical conductivity, ease of synthesis, and the ability to undergo rapid and reversible redox reactions, thereby offering significant improvements in energy density and capacitance. The superior performance of these conductive polymers in supercapacitors can be attributed to their pseudocapacitive nature, which involves Faradaic charge storage mechanisms, enabling higher specific capacitances compared to traditional electrostatic double-layer capacitors. Polypyrrole, for instance, is celebrated for its high electrical conductivity and excellent environmental stability, making it an ideal candidate for supercapacitor electrodes. Polythiophene, known for its tunable electrical properties and stability under varying conditions, contributes to the longevity and adaptability of supercapacitors. Polyacetylene, a pioneer in the field of conductive polymers, though less commonly used, offers insights into the potential of conjugated polymer systems in energy storage. These conductive polymers often undergo various functionalization and compositing strategies to further enhance their electrochemical properties. For instance, the incorporation of metal oxides or carbon-based materials with these polymers can lead to synergistic effects, resulting in improved electrical conductivity, ion transport, and overall electrochemical performance. The versatility and tunability of these materials, coupled with their ease of fabrication and potential for scalability, make them highly attractive for future supercapacitor applications. As research continues to uncover novel conductive polymers and optimize existing ones, these materials are poised to play a pivotal role in the advancement of high-performance and efficient supercapacitor technologies.

3.3. Metal oxides and hydroxides

Metal oxides and hydroxides represent a critical category of materials in the field of supercapacitors, particularly in the domain of pseudocapacitors. These materials, including widely studied ones like RuO_2, MnO_2, Co_3O_4, and $Ni(OH)_2$, are known for their ability to undergo Faradaic redox reactions, which contribute significantly to the capacitance. This Faradaic process, involving the reversible oxidation and reduction at the surface or near-surface of the material, allows for higher energy densities compared to purely electrostatic storage mechanisms found in EDLCs. The diversity in the oxidation states of these materials facilitates multiple redox reactions, thereby enhancing the specific capacitance. Moreover, the natural abundance and low cost of certain metal oxides and hydroxides, like MnO_2, make them appealing for cost-effective supercapacitor applications. Their varied crystalline structures and the possibility to engineer their morphology and particle size present additional avenues to optimize their performance in energy storage applications [164].

The exploration of metal oxides and hydroxides in supercapacitors is driven by the pursuit of higher energy densities without significantly compromising power densities. These materials typically exhibit superior specific capacitance and energy density compared to carbon-based materials, making them suitable for applications where higher energy storage is a priority. However, the implementation of metal oxides and hydroxides in supercapacitors is not without challenges. One of the primary limitations is their relatively lower electrical conductivity compared to carbon materials or conductive polymers, which can be a bottleneck in achieving high power densities. Another concern is the cyclic stability; these materials often undergo structural changes during the charge-discharge cycles, which can lead to capacity fading over time. To address these challenges, current research is focused on creating composites or hybrids, combining metal oxides or hydroxides with highly conductive materials, such as carbon nanotubes or graphene. This approach aims to create a synergistic effect where the high energy density of metal oxides/hydroxides is complemented by the high conductivity and stability of carbon materials, leading to supercapacitors that can deliver both high energy and power densities while maintaining structural integrity over prolonged cycles.

Thanks to their superior pseudocapacitive qualities and reversible redox reaction capabilities, which increases specific capacitance and energy density, metal oxides and hydroxides have shown to be very efficient for supercapacitors. In a noteworthy work, Wang et al. made a number of NiCo-layered double hydroxides (NiCo-LDHs) at room temperature (**Figure 3A**) with great success utilizing a novel ammonia diffusion approach [165]. These NiCo-LDHs show outstanding structural and electrochemical capabilities. Wang and colleagues conducted a study utilizing NiCo-LDHs' variable composition and affordability to synthesize and use them as pseudocapacitive electrodes. NiCo-LDHs were synthesized using an innovative ammonia diffusion process that distinguished itself from more traditional, intricate synthesis methods by its simplicity, eco-friendliness, and efficiency. The shape, structure, and elemental content of the produced NiCo-LDHs were studied (**Figure 3B**). When the honeycomb NiCo-LDH was synthesized ideally, it showed an outstanding specific capacitance of 1463.2 F/g at a current density of 1 A/g, as shown by the electrochemical experiments in **Figure 3C**. Interestingly, the material had a capacitance retention of 87.9% after 2000 charge-discharge cycles at a high current density of 20 A/g. It also maintained a rate capacity of 75.2% at that high current density. At a power density of 900 W/kg, the energy density of a hybrid supercapacitor design including NiCo-LDH and AC attained 23.4 Wh/kg, retaining 73.4% of its capacity after 5000 cycles.

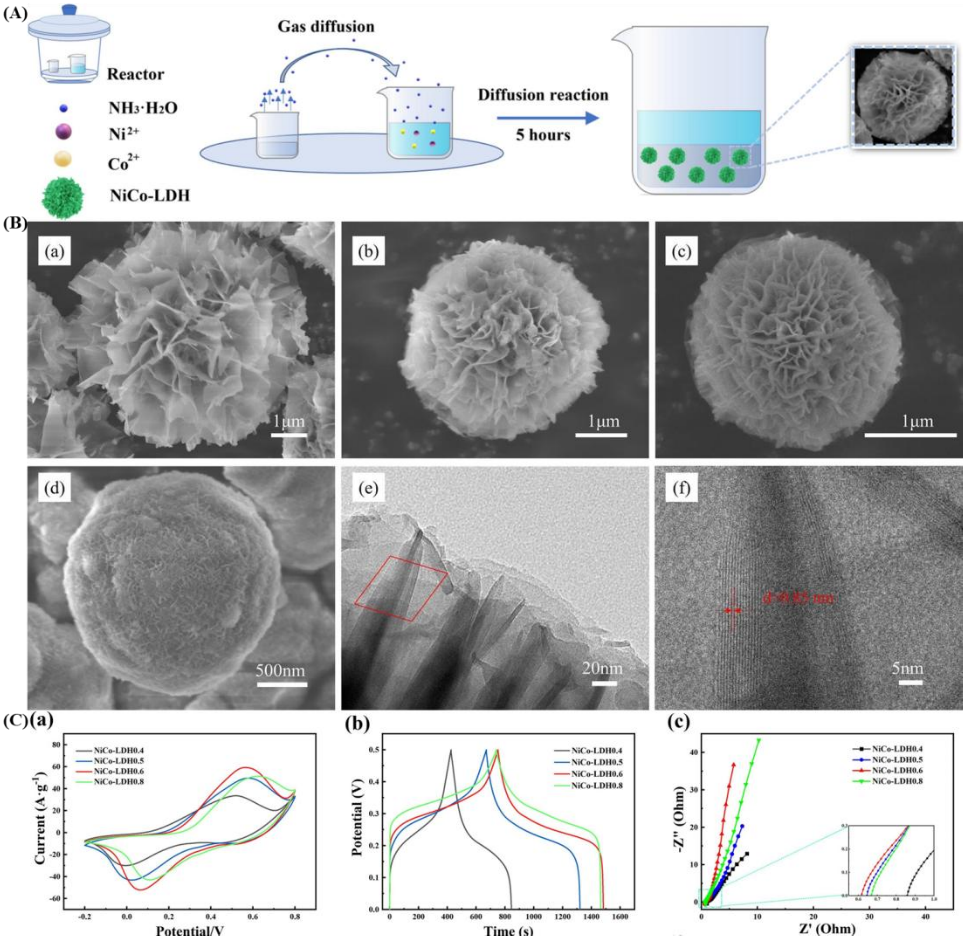


Figure 3. (A) Schematic illustration showing how NiCo-LDHs arise. (B) TEM pictures of (e) NiCo-LDH0.6 honeycomb edge, (f) HRTEM image of NiCo-LDH0.6 with distinct grain boundaries, and (a) SEM images of (a) NiCo-LDH0.4, (b) NiCo-LDH0.5, (c) NiCo-LDH0.6, and (d) NiCo-LDH0.8. (C) (a) NiCo-LDH electrode CV curves at 30 mV/s, (b) NiCo-LDH electrode GCD curves at 1 A/g, and (c) NiCo-LDH electrode EIS curves. *Reproduced with permission [165]. Copyright 2024, Elsevier.*

The synthesis process of NiCo-LDHs was a critical aspect of this study. Wang et al. utilized ammonia water as a direct precipitant in a novel approach, countering the conventional challenges of fast reaction speeds and non-uniform particle sizes. By harnessing the easy evaporation and diffusion properties of ammonia in a controlled reaction environment, they achieved homogeneous particle synthesis at room temperature. This method's mild reaction conditions and scalability potential make it particularly suitable for industrial production. The resultant NiCo-LDHs displayed varied morphologies, with the NiCo-LDH0.6 showing a unique nanoflower-like structure with tightly packed nano lamellae, leading to a maximum specific surface area of 132.83 m^2/g. Electrochemical analyses of these NiCo-LDHs further substantiated their superior performance. CV curves of the electrodes revealed paired redox peaks, indicative of the reversible redox reactions of Ni^{2+}/Ni^{3+} and Co^{2+}/Co^{3+} ions, with the NiCo-LDH0.6 electrode exhibiting the largest peak intervals, signifying the highest specific capacity [166-168]. GCD spectrograms corroborated these findings. EIS analyses demonstrated that NiCo-LDH0.6 had the fastest ion diffusion speed in the electrolyte and the lowest charge transfer resistance, attributes ascribed to its well-defined honeycomb structure and large specific surface area. These results collectively highlight the remarkable potential of NiCo-LDHs, especially the NiCo-LDH0.6 variant, in enhancing the performance of supercapacitors, paving the way for advanced energy storage solutions with high specific capacitance, energy density, and stability [65, 81, 169].

In recent years, the exploration of metal oxides and hydroxides for supercapacitors has gained considerable momentum, with several materials demonstrating excellent performance due to their unique electrochemical properties. Among these, RuO_2, MnO_2, Co_3O_4, and $Ni(OH)_2$ have stood out as prominent examples. These materials are highly favored in the realm of supercapacitors for their ability to facilitate rapid and reversible redox reactions, thereby contributing to enhanced pseudocapacitance. For instance, RuO_2 is renowned for its exceptional electrical conductivity and high capacitance, making it an ideal candidate for high-performance supercapacitors. MnO_2, on the other hand, is valued for its natural abundance, low cost, and environmental friendliness, despite having lower conductivity, which is often mitigated through composite formulations or nanostructuring. Co_3O_4 has emerged as a promising material due to its multiple oxidation states, which enable diverse redox reactions, thus enhancing its capacitive performance. Similarly, $Ni(OH)_2$ is recognized for its high theoretical capacitance and is often utilized in hybrid supercapacitors to exploit its pseudocapacitive properties. These metal oxides and hydroxides exhibit a range of properties such as high surface area, optimal pore distribution, and structural stability, which are crucial for efficient charge storage and rapid charge-discharge cycles. Recent advancements have focused on nanostructuring these materials, doping with other elements, and creating composites with carbon-based materials to further improve their conductivity, stability, and overall electrochemical performance. The exploration of these metal oxides and hydroxides is driven by the need for supercapacitors with higher energy densities and longer life cycles, making them crucial components in the ongoing development of advanced energy storage systems. As research continues to optimize these materials, metal oxides and hydroxides are set to play an increasingly significant role in the future of supercapacitor technology, offering promising avenues for enhanced energy storage solutions.

3.4. Emerging and hybrid composite materials

The evolution of supercapacitor technology is increasingly pivoting towards emerging and hybrid materials, including composite materials, MOFs, and MXenes, owing to their promising properties and potential to overcome the limitations of conventional materials. Composite materials, created by combining two or more distinct materials, leverage the synergistic properties of each component, such as the high conductivity of carbon materials with the high capacitance of metal oxides or conductive polymers. This combination enhances overall performance, achieving a balance between energy density, power density, and cyclic stability. MOFs, known for their highly porous crystalline structures, have garnered attention due to their vast surface areas and customizable pore sizes, which are ideal for efficient charge storage and ion transport. MXenes, a relatively new class of two-dimensional materials, exhibit excellent electrical conductivity, mechanical flexibility, and a hydrophilic nature. Their layered structures provide rapid electron and ion transport pathways, making them particularly suitable for high-rate supercapacitors [170-173].

The exploration of these emerging and hybrid materials is primarily driven by the quest to transcend the energy and power limitations of traditional supercapacitors. Composite materials, for instance, address the trade-off between energy and power densities by combining the best of both worlds – the capacitive behavior of conductive polymers or metal oxides and the high conductivity of carbon materials. MOFs, with their tunable composition, offer the flexibility to tailor the electrochemical properties of the supercapacitor, enhancing its performance and efficiency. Similarly, MXenes, with their unique two-dimensional structure, contribute to the development of supercapacitors that exhibit both high capacitance and excellent rate capability. However, these advanced materials also bring challenges, particularly in terms of scalability, cost, and integration into existing supercapacitor manufacturing processes. Ensuring the long-term stability and reliability of these materials under operational conditions remains a critical area of research. The focus on addressing these challenges and the continued exploration and optimization of these materials is pivotal in advancing supercapacitor technology to meet the growing demands for efficient, high-performance energy storage solutions [174, 175].

Various emerging and hybrid composite materials, including telluride-based materials, $ZnO\text{-}CeO_2$ nanocomposite, and Fe_3O_4, are playing a pivotal role in the development of advanced and efficient supercapacitors, significantly enhancing their performance and energy storage capabilities [47, 176-178]. The ability of emerging and hybrid materials to combine several beneficial qualities, such increased surface area, higher electrical conductivity, and enhanced redox activity, is making them more and more effective for supercapacitors. Higher power densities and enhanced energy storage capacities are the results of these synergistic properties. The synergistic benefits of $MnCO_2O_4/Co_3V_2O_8$ composite nanomaterials, which were created via a unique secondary hydrothermal and calcination technique, were documented by Yu et al. in their breakthrough study [179]. As seen in **Figure 4**, these materials exhibited remarkable structural and electrochemical characteristics, rendering them extremely appropriate for use as high-performance supercapacitor electrodes.

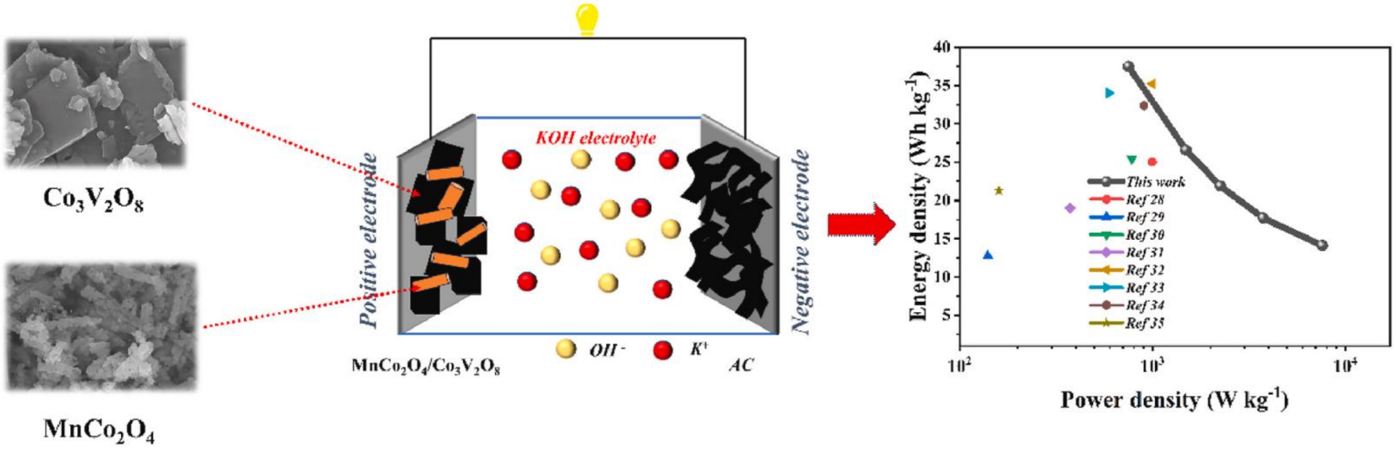


Figure 4. Diagram illustrating the $MnCO_2O_4/Co_3V_2O_8$ composite nanomaterials' synergistic impact for high-performance supercapacitor electrodes. *Reproduced with permission [179]. Copyright 2024, Elsevier.*

The $MnCO_2O_4/Co_3V_2O_8$ composites combine the battery-like charge storage behaviour of $MnCO_2O_4$ with the special intercalation/delamination reaction of $Co_3V_2O_8$, a pseudocapacitive material [180].

A notable interfacial and synergistic impact is produced by the combination of these several reaction processes, and the product is outstanding electronic conductivity, enhanced electrochemical activity, and high reversible capacity. The synthesized $MnCO_2O_4/Co_3V_2O_8$ nanomaterial's specific capacitance, which peaked at 1460 F/g at a current density of 1 A/g, was very remarkable. Moreover, at a high current density of 10 A/g, the material showed remarkable capacitance retention of 84.61% after 3500 cycles. With $MnCO_2O_4/Co_3V_2O_8$ and AC hybrid supercapacitors, an impressive energy density of 37.5 mAh/g was attained. Even after 28,000 charge-discharge cycles, the electrochemical performance was steady, holding 98.6% of its initial capacity. This work highlights the advantages of mixing various metal oxides and hydroxides to obtain improved performance, demonstrating the promise of employing composite nanomaterials in supercapacitors. With their high specific capacitance and energy density, the $MnCO_2O_4/Co_3V_2O_8$ nanocomposites established a new standard for supercapacitor electrode materials and opened up exciting new possibilities for energy storage technologies in the future.

The realm of supercapacitor technology has recently witnessed a surge in the development of emerging and hybrid materials, leading to significant performance enhancements in energy storage applications. Among these innovative materials, MOFs, MXenes, and various carbon/metal oxide composites have been particularly noteworthy. MOFs, with their highly porous crystalline structures, offer vast surface areas for efficient charge storage and tunable pore sizes for ion transport, making them ideal for high-capacity supercapacitors. MXenes, a family of two-dimensional transition metal carbides and nitrides, have gained attention for their extraordinary electrical conductivity, mechanical flexibility, and hydrophilic nature, which contribute to rapid charge transfer and high power density in supercapacitors. Hybrid materials combining carbonaceous substrates with metal oxides, such as graphene/RuO_2 or carbon nanotube/MnO_2 composites, have shown exceptional results due to the synergistic effects between the high surface area and conductivity of carbon materials and the pseudocapacitive properties of metal oxides. These composites not only enhance the energy density but also improve the cyclic stability and rate capability of supercapacitors. Additionally, research in heterostructured hybrids, such as conductive polymer/MXene or MOF/carbon nanotube composites, has opened new frontiers in supercapacitor development. These materials leverage the distinct advantages of each component, such as the high capacitance of conductive polymers and the structural integrity of carbon nanotubes, resulting in supercapacitors with improved energy storage capacities and operational stabilities [181]. The exploration and optimization of these emerging and hybrid materials are driven by the need for supercapacitors with higher energy densities, longer life cycles, and better rate performances. As research continues to uncover and harness the full potential of these materials, they are set to play a pivotal role in the advancement of supercapacitor technology, offering promising solutions for efficient and sustainable energy storage in various applications.

As we explore the realm of supercapacitor technology, it is imperative to understand the diverse electrode materials that play a crucial role in their performance. **Table 1** provides a comprehensive overview of these materials, including carbon materials, conductive polymers, metal oxides and hydroxides, and emerging hybrid and composite materials, each offering unique properties and applications in the field of energy storage.

4. Electrolytes for supercapacitors and their properties

Electrolytes play a crucial role in the performance of supercapacitors, acting as the medium for ion transport between electrodes during the charge and discharge cycles. The choice of electrolyte significantly impacts the capacitance, energy and power densities, voltage window, and overall stability of the supercapacitor.

Table 1. Comparative overview of electrode materials for supercapacitors.

Electrode Material	Key Properties	Examples	Applications
Carbon Materials	High surface area, excellent electrical conductivity, chemical and thermal stability, flexible structure	AC, CNTs, graphene	EDLCs, flexible supercapacitors
Conductive Polymers	High electrical conductivity, rapid and reversible redox reactions, tunable electrical and physical properties	PANI, PPy, polythiophene (PTh)	Pseudocapacitors, wearable electronics
Metal Oxides and Hydroxides	High theoretical capacitance, multiple oxidation states for redox reactions, good energy density	RuO_2, MnO_2, Co_3O_4, $Ni(OH)_2$	Pseudocapacitors, high-energy density supercapacitors
Emerging Hybrid and Composite Materials	Synergistic properties, enhanced electrical conductivity and capacitance, improved stability, and energy density	MOFs, MXenes, graphene/metal oxide composites	Advanced energy storage systems, high-performance supercapacitors

Aqueous electrolytes, commonly comprising sulfuric acid or potassium hydroxide solutions, are notable for their high ionic conductivity and low cost [182-184]. They contribute to the high power density of supercapacitors but are limited by a narrow operational voltage window, typically up to 1.23V, due to water electrolysis. Organic electrolytes, such as acetonitrile or propylene carbonate with dissolved salts, offer a wider voltage window (up to 2.5-2.7V), leading to higher energy densities. However, they often come with challenges like lower ionic conductivity compared to aqueous electrolytes and higher costs [185].

In the pursuit of enhanced performance, ionic liquids and gel or solid-state electrolytes have emerged as advanced options. Ionic liquids, composed of organic salts that remain liquid at room temperature, are attractive due to their wide electrochemical stability window, non-flammability, and high thermal stability. They enable supercapacitors to operate over a broader voltage range, thereby increasing energy density. However, their higher viscosity and cost compared to traditional electrolytes are aspects that need addressing. Gel and solid-state electrolytes represent a leap towards safer and more flexible supercapacitors. These electrolytes not only eliminate leakage risks associated with liquid electrolytes but also enable the fabrication of flexible and thin-film supercapacitors. Gel electrolytes, which are essentially liquid electrolytes immobilized in a polymer matrix, combine the high ionic conductivity of liquid electrolytes with the mechanical stability of solid electrolytes. Solid-state electrolytes, although offering excellent safety and form factor flexibility, currently face challenges like lower ionic conductivity and interfacial resistance issues. The ongoing research in electrolyte technology, focusing on optimizing conductivity, stability, and compatibility with electrode materials, is integral to advancing supercapacitor performance and broadening their application spectrum [186-192].

4.1. Aqueous electrolytes

Aqueous electrolytes have been a cornerstone in the development of supercapacitors due to their inherent advantages, such as high ionic conductivity, low cost, and environmental friendliness. Predominantly, these electrolytes are composed of water as the solvent with dissolved salts that dissociate into ions, facilitating charge transfer. The most common types of aqueous electrolytes include sulfuric acid (H_2SO_4), potassium hydroxide (KOH), and sodium sulfate (Na_2SO_4). Each of these offers distinct electrochemical properties. For instance, H_2SO_4 is widely used due to its high ionic conductivity and low equivalent series resistance (ESR), leading to superior power capabilities. KOH, on the other hand, is preferred for its high pH stability range and the formation of a quasi-solid electrolyte interface, which enhances cycle life. Sodium sulfate is another viable option, known for its safety and non-toxic nature, though it offers lower conductivity than the other two [193].

The significance of aqueous electrolytes in supercapacitors largely stems from their operational safety and simplicity in handling, making them suitable for a wide range of applications. One of the key properties of aqueous electrolytes is their potential range, which is typically limited to 1.0-1.23V due to the electrolysis of water. This narrow operational window can be a limitation in terms of the energy density achievable with aqueous electrolytes. scope of their application in advanced energy storage systems.

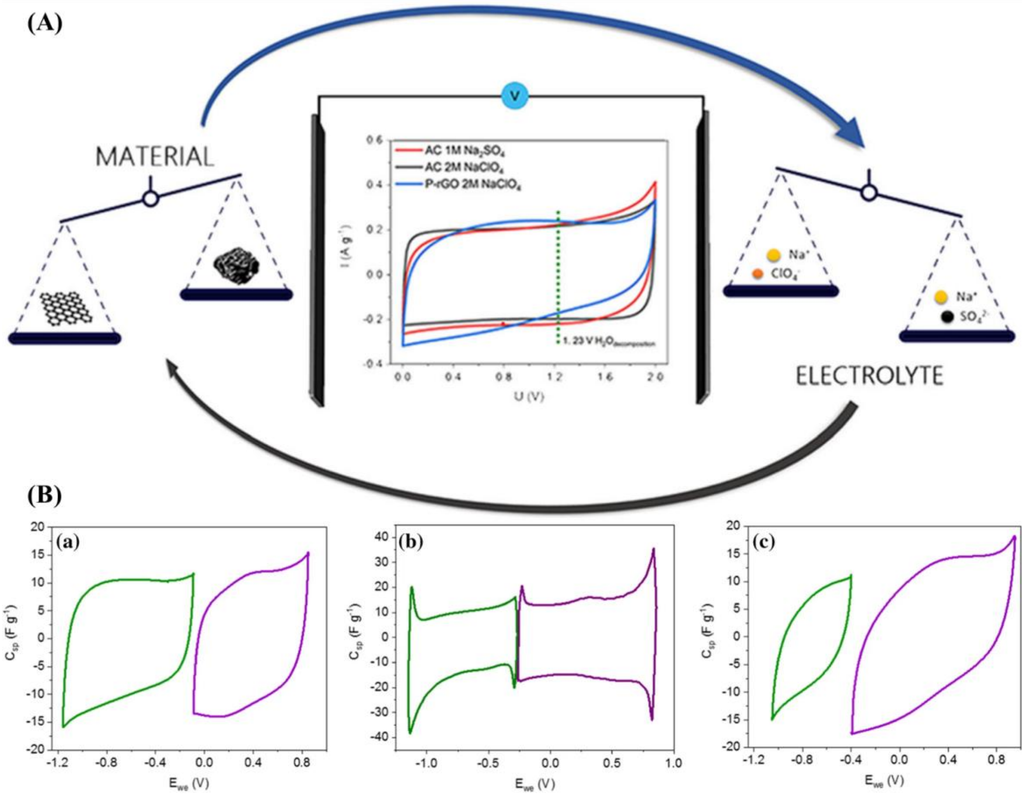


Figure 5. (A) Diagram demonstrating the importance of electrode materials and electrolytes in supercapacitors. (B) The CV curves of AC 1 M Na_2SO_4 (a), AC 2 M $NaClO_4$ (b), and P-rGO 2 M $NaClO_4$ (c) were obtained in the three-electrode arrangement at a scan rate of 10 mV/s. The green and violet lines, respectively, represent the material behaviour for its symmetric counterpart in the voltage range of the positive and negative electrodes. *Reproduced with permission [194]. Reproduced under the term C.C. By 4.0. Copyright 2023, Gajewska et al., Springer Nature.*

However, recent advancements have led to the development of "water-in-salt" electrolytes, which significantly expand the voltage window up to 3V, thereby enhancing the energy density. Despite their lower energy density compared to organic or ionic liquid electrolytes, aqueous electrolytes exhibit faster ion transport and higher power densities. They are also characterized by lower viscosity and easier ion movement, which contribute to reduced internal resistance and excellent charge-discharge rates. Moreover, the environmental benignity and cost-effectiveness of aqueous electrolytes make them highly attractive for large-scale and sustainable supercapacitor applications. The ongoing research in this domain is focused on enhancing the voltage window and stability of aqueous electrolytes while maintaining their inherent advantages, thereby broadening the Supercapacitors benefit greatly from the excellent ionic conductivity and environmental friendliness of aqueous electrolytes. These characteristics significantly improve the device's overall performance and sustainability. Gajewska et al. produced outstanding electrochemical performances in their examination of the influence of carbon material and electrolytes on the electrochemical performance of high-voltage aqueous symmetric supercapacitors, as **Figure 5A** [194] illustrates.

To increase the energy storage capacity of environmentally friendly devices, Gajewska et al. addressed the issue of boosting the operating voltage of aqueous supercapacitors over the electrolyte breakdown limit in their study. Using both synthetic P-rGO and commercial AC, the scientists investigated the effects of surface chemistry and textural features on the electrochemical performance of high-voltage aqueous supercapacitors. In a symmetric cell design, the study compared two types of materials: highly porous AC and heteroatom-rich P-rGO with lower porosity. An electrolyte of 2 M $NaClO_4$ was utilized in this setup. It operated within a broad 2.0 V voltage window. Additionally, the performance of the AC-based cell was evaluated in a 1 M Na_2SO_4 solution, illustrating differences in performance between sodium-based electrolytes. The results demonstrated that a porous structure and a significant heteroatom contribution, which enhances electrode hydrophilicity, are necessary to achieve high specific energy density values. The built-in supercapacitors showed an amazing specific energy density of up to 13 Wh/kg at a power density of 30 W/kg. In order to determine potential electrode windows, the study also included CV measurements utilizing a three-electrode design. The CV curves for AC in 1 M Na_2SO_4 and P-rGO in 2 M $NaClO_4$, which depict electric double-layer capacitance and pseudocapacitive behaviour, respectively, showed semi-rectangular shapes (**Figure 5B**). In 2 M $NaClO_4$, the AC showed discrete redox peaks at the curve's edges and rectangular shapes, indicating electrolyte breakdown at certain voltages. Gajewska et al. concluded that in order to obtain successful functioning in supercapacitors, careful consideration of both material properties (such as resistivity, surface chemistry, and porous structure) and electrolyte type is required. Outstanding charge storage properties were shown by the symmetric devices based on AC electrodes in 1 M Na_2SO_4, including a maximum specific capacitance of 94 F/g at 0.2 A/g and an energy density of 13 Wh/kg. The study's findings showed that symmetric arrangements with neutral aqueous electrolytes might be extremely effective energy storage devices with widespread applications in sustainable energy solutions.

Recent advancements in supercapacitor technology have seen a burgeoning interest in various aqueous electrolytes, which are being increasingly recognized for their excellent performance in energy storage applications. Notable among these are electrolytes based on lithium sulfate (Li_2SO_4), potassium hydroxide (KOH), and zinc sulfate ($ZnSO_4$), each bringing distinct advantages to supercapacitor functionality. Li_2SO_4, for instance, is appreciated for its ability to enhance the ionic conductivity of the electrolyte, thereby improving the charge-discharge efficiency of supercapacitors [195]. KOH, a traditional choice in aqueous electrolytes, continues to be favored for its high ionic mobility and compatibility with a wide range of electrode materials, contributing to high power densities and fast charge transfer in supercapacitors. Zinc sulfate ($ZnSO_4$) has emerged as a promising electrolyte, particularly in hybrid supercapacitors, due to its stable operating window and the added benefit of contributing to the redox reactions at the electrode surface. These aqueous electrolytes, with their relatively low decomposition voltages, offer a safer and more environmentally benign alternative to organic electrolytes, while still providing commendable electrochemical performance. Recent studies have also explored innovative approaches such as water-in-salt electrolytes, where a high concentration of salt in water extends the electrochemical stability window, pushing the boundaries of energy density achievable with aqueous electrolytes. Additionally, the use of hybrid aqueous electrolytes, combining different salts, has shown potential in tailoring the electrolyte properties to suit specific electrode materials and supercapacitor configurations. Such developments are crucial in the pursuit of high-performance, cost-effective, and eco-friendly supercapacitors. The continuous evolution and optimization of aqueous electrolytes are poised to significantly contribute to the advancement of supercapacitor technology, catering to the growing demand for efficient and sustainable energy storage solutions.

4.2. Organic electrolytes

Since organic electrolytes offer long operating voltage windows and high energy densities, they have become an important part of the supercapacitor technological development. These electrolytes provide a wider range of electrochemical stability than their watery counterparts since they are made up of an organic solvent and a dissolved salt. Propylene carbonate (PC), ethylene carbonate (EC), and acetonitrile (ACN) are common solvents that are utilized; each gives the electrolyte a special set of characteristics. For example, ACN is used because of its high dielectric constant and low viscosity, which promote quick ion movement and maximize power density. The electrolyte's thermal stability is enhanced by solvents with higher boiling points, such as PC and EC. Salts that breakdown into ions in the solvent to facilitate charge transfer processes are known as quaternary ammonium salts, such as tetraethylammonium tetrafluoroborate ($TEABF_4$) or tetraethylammonium hexafluorophosphate ($TEAPF_6$) [51, 196-199]. The longer potential windows of organic electrolytes, which are frequently as high as 2.5–2.7 V, are essential for increasing supercapacitors' ability to store energy. According to the relationship $E = 1/2\ CV^2$; this greater range translates into higher voltage operation, which in turn results in enhanced energy density. But throughout this wide voltage range, the electrochemical stability of the electrolyte and the electrode materials is what determines the supercapacitor's operating stability. Although organic electrolytes have larger energy densities than aqueous electrolytes, they frequently have poorer ionic conductivities, which can affect power performance. In addition, there are other problems that must be resolved, including increased expense, flammability, and environmental concerns in comparison to aqueous electrolytes. Despite these obstacles, there is still a strong market for organic electrolytes in applications like electric cars and portable gadgets that need large amounts of energy storage. The goal of this field's ongoing research is to create novel organic solvents and salt combinations that will increase the electrolytes' performance qualities while lowering prices, expanding their potential window, and improving their safety profile. This advancement is essential to the supercapacitor technology's future scalability and range of applications [200, 201].

When compared to aqueous electrolytes, organic electrolytes have larger energy densities and broader operating voltage windows, which greatly improve the performance of supercapacitors. Li et al. reported on the creation of an organic electrolyte that is inherently non-flammable for supercapacitors that operate over a wide temperature range and have outstanding electrochemical capabilities [202]. Li and colleagues conducted a unique study whereby they introduced a binary electrolyte based on non-flammable triethyl phosphate (TEP) to solve safety problems related to standard organic electrolytes. When paired with 1-butyl-3-methylimidazolium tetrafluoroborate ($EMIMBF_4$), this electrolyte showed enhanced ionic conductivity and decreased viscosity because TEP effectively broke the hydrogen link in $EMIMBF_4$ ion pairs [203]. The researchers looked at the interactions inside this unique electrolyte using nuclear magnetic resonance (NMR), Raman spectroscopy, and Fourier transform infrared spectroscopy (FTIR). This electrolyte was coupled with carbon nanosheet electrodes to produce an efficient symmetric supercapacitor that functioned well across a broad temperature range (−20 °C to 80 °C) [12, 64, 99, 204]. **Figure 6a-d** shows the remarkable capacitance retention of 86.5% after 10,000 cycles, low self-discharge rate, and high energy density of 52.1 Wh/kg and 19.7 kW/kg that this supercapacitor produced. After prolonged storage, the stability and homogeneity of the electrolyte were confirmed by $EMIMBF_4$'s solubility in TEP. The greater non-flammability of the TEP-based electrolyte over traditional AN and PC-based electrolytes was demonstrated by the flammability tests, which enhanced the supercapacitor's safety profile. After optimizing the ionic conductivity and viscosity of the TEP-based electrolyte at various concentrations, it performed comparably to PC-based electrolytes and exhibited greater conductivity than pure EMIMBF4. **Figure 6e** illustrates how the TEP-based electrolyte is suitable for supercapacitor applications across a wide temperature range since this optimized conductivity was maintained even at below-freezing temperatures. The results of the study by Li et al. show that EMIMBF4/TEP is a potential non-flammable electrolyte that offers the best overall performance in terms of safety, stability, cost, energy, and power densities in addition to a wider temperature range at which supercapacitors may be used. The creation of safe, effective, and adaptable supercapacitors for a variety of applications has advanced significantly because of this effort.

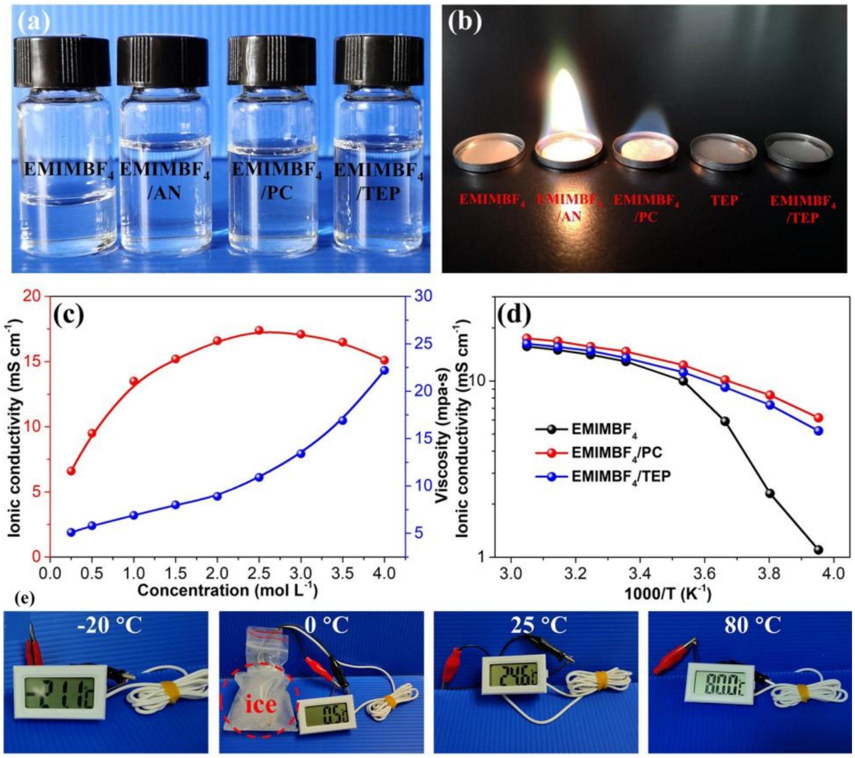


Figure 6. (a) Digital images of the electrolytes $EMIMBF_4$, $EMIMBF_4$/AN, $EMIMBF_4$/PC, and $EMIMBF_4$/TEP. (a) Glass fibre separators soaked in various electrolytes and TEP solvent were put through an ignition test. (c) At room temperature, $EMIMBF_4$/TEP's ionic conductivity and viscosity in relation to salt concentration. (d) Ionic conductivity values of $EMIMBF_4$, $EMIMBF_4$/PC, and $EMIMBF_4$/TEP electrolytes as a function of temperature. (e) A digital thermometer that operates correctly at various temperatures thanks to a symmetric supercapacitor based on the $EMIMBF_4$/TEP. *Reproduced with permission [202]. Copyright 2023, Elsevier.*

The discovery of several organic electrolytes, each with outstanding performance properties, has greatly aided in the growth of supercapacitor technology. Prominent among these are electrolytes based on acetonitrile (AN), ethylene carbonate (EC), and propylene carbonate (PC), which have proved crucial in improving supercapacitors' ability to store energy. Propylene carbonate enhances the charge-discharge efficiency by facilitating greater ion mobility due to its low viscosity and high dielectric constant. To construct electrolytes that offer a compromise between strong ionic conductivity and large electrochemical stability windows, which are essential for obtaining high energy and power densities, ethylene carbonate is frequently employed in conjunction with other solvents, such as dimethyl carbonate (DMC). Acetonitrile is used as a solvent in high-performance electrolytes because of its low boiling point and good solvating qualities, which promote quick ion movement and low internal resistance. Additionally, recent studies have concentrated on the development of mixed solvent systems, in which salts such as tetraethylammonium tetrafluoroborate ($TEABF_4$) or lithium hexafluorophosphate ($LiPF_6$) are mixed with solvents like PC and EC to create electrolytes that provide improved operating safety and thermal stability. Furthermore, the investigation of new additives and co-solvents has resulted in the development of electrolytes that enhance the voltage window and cycle stability, two more supercapacitor performance factors. These developments in organic electrolytes are essential to the ongoing progress of supercapacitor technology, providing viable routes to energy storage systems that are highly effective, long-lasting, and adaptable. The significance of novel organic electrolytes in supercapacitors will rise in importance as the need for more durable and dependable energy storage systems increases, propelling the shift to more environmentally friendly and highly effective energy storage technologies.

4.3. Ionic liquids

Supercapacitors have attracted a lot of attention to ionic liquids, a new class of electrolytes because of their special qualities and improved performance. At room temperature, they are essentially liquid salts made up of inorganic or organic anions and organic cations. The most remarkable feature of supercapacitors is their extraordinarily broad electrochemical stability window, which frequently exceeds 4 V and significantly raises their energy density. One-butyl-3-methylimidazolium hexafluorophosphate ($BMIM-PF_6$) and 1-ethyl-3-methylimidazolium tetrafluoroborate ($EMIM-BF_4$) are examples of ionic liquids that are often utilized. Because of these ionic liquids' excellent thermal stability and lack of volatility, supercapacitors are more reliable and stable throughout a broad temperature range. Supercapacitors' safety profile is further improved by the non-flammability of ionic liquids, particularly when compared to organic electrolytes. In addition, they are more desirable for high-performance applications because to their better conductivity and lower vapour pressure as compared to solid electrolytes [205-208]. The use of ionic liquids in supercapacitors creates opportunities for high-energy storage devices that can function safely in high-voltage and temperature environments. For applications requiring high energy density, such electric cars and renewable energy systems, this wide operating range is essential. Ionic liquid application does, however, come with certain difficulties. Their comparatively high viscosity in comparison to conventional aqueous and organic electrolytes is one of the main issues, as it might hinder ion mobility and, as a result, the supercapacitor's power output. Furthermore, ionic liquids are often more expensive than traditional electrolytes, which might be a barrier for large-scale applications. Ongoing research is concentrated on the synthesis of new ionic liquids with reduced viscosity and cost, as well as on the design of electrode materials that can fully utilize the wide voltage range provided by these electrolytes, in order to optimize the performance of supercapacitors using these electrolytes. These developments are essential to realizing the full potential of ionic liquids and laying the groundwork for the next wave of supercapacitors, which will be highly efficient, secure, and stable [21, 209-212].

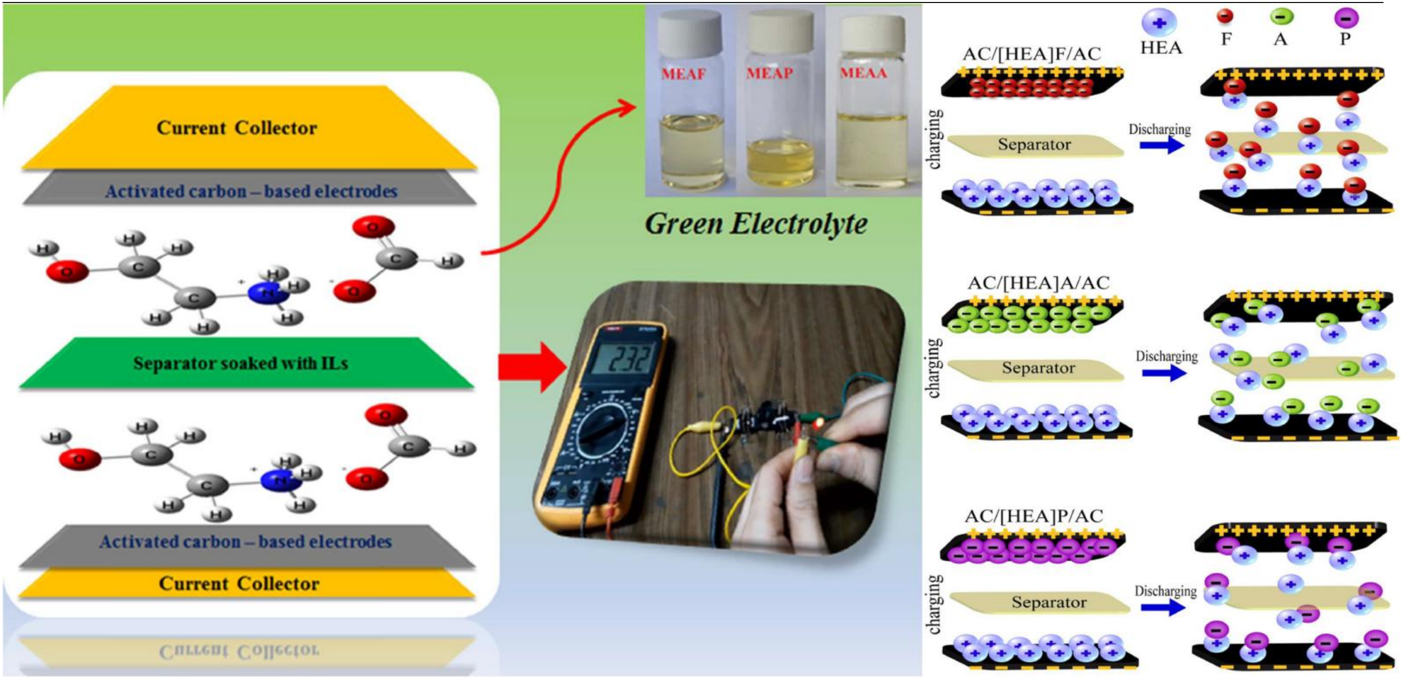


Figure 7. The functionality of ionic liquids based on ethanolamines as innovative environmentally friendly electrolytes for electrochemical energy storage applications and the charge propagation dynamics of AC-based supercapacitors in a two-electrode cell assembly using [HEA]F, [HEA]A, and [HEA]P ILs. *Reproduced with permission [213]. Copyright 2024, Elsevier.*

Supercapacitors benefit greatly from the large electrochemical stability windows and high ionic conductivities of ionic liquids, which also increase the devices' operating safety and energy storage capacity. Excellent electrochemical performances were attained by Mirzaei-Saatlo et al. when they investigated the performance of ethanolamine-based ionic liquids as innovative green electrolytes for electrochemical energy storage applications [213]. **Figure 7** illustrates how Mirzaei-Saatlo et al.'s work presented monoethanolamine-based ionic liquids with formate, acetate, and propionate anions ([HEA]F, [HEA]A, and [HEA]P) as simple, affordable, and environmentally acceptable electrolytes for AC-based supercapacitors. The electrochemical stable potential window (ESPW) and ionic conductivity of these ionic liquids were evaluated, showing encouraging outcomes for supercapacitor applications. EIS, CV, linear sweep voltammetry (LSV), and GCD are examples of electrochemical tests that were performed to assess these ionic liquids' electrolytic performance. It was found that as the size of the ILs' anions shrank, so did the charge transfer resistance (Rct) and solution resistance (Rs). At a scan rate of 10 mV/s, the specific capacitance of the supercapacitors made using [HEA]F, [HEA]A, and [HEA]P ionic liquids were 114, 95, and 73 F/g, respectively. A notable improvement in supercapacitor performance was made by the [HEA]F ionic liquid supercapacitor, which attained a maximum power density of 2941 W/kg and an energy density of 70 Wh/kg. Furthermore, after 5000 charge-discharge cycles, the supercapacitors made with these electrolytes showed outstanding capacitance retention, demonstrating the promise of ethanolamine-based ionic liquids as high-performing and sustainable electrolytes for supercapacitors. The dynamics of charge propagation in these ethanolamine-based ionic liquids were also examined in detail, and the results demonstrated the critical roles that ion mobility, molecule structure, and ion-solvent interactions play in controlling charge transport. The study shed important light on the interactions between ion migration and diffusion in the ionic liquid medium and the processes behind the electrochemical behaviour of the supercapacitors. The superior electrochemical performance and stability of AC-based supercapacitors using these innovative ionic liquids were confirmed by experimental observations and analytical techniques used in the study, opening up a promising path for the development of efficient and environmentally friendly energy storage systems. The use of ionic liquids in supercapacitors creates opportunities for high-energy storage devices that can function safely in high-voltage and temperature environments. For applications requiring high energy density, such electric cars and renewable energy systems, this wide operating range is essential. Ionic liquid application does, however, come with certain difficulties. Their comparatively high viscosity in comparison to conventional aqueous and organic electrolytes is one of the main issues, as it might hinder ion mobility and, as a result, the supercapacitor's power output. Furthermore, ionic liquids are often more expensive than traditional electrolytes, which might be a barrier for large-scale applications. Ongoing research is concentrated on the synthesis of new ionic liquids with reduced viscosity and cost, as well as on the design of electrode materials that can fully utilize the wide voltage range provided by these electrolytes, to optimize the performance of supercapacitors using these electrolytes. These developments are essential to realizing the full potential of ionic liquids and laying the groundwork for the next wave of supercapacitors, which will be highly efficient, secure, and stable.

Ionic liquids (ILs) have been investigated extensively in supercapacitors, and several new formulations have shown impressive performance gains. Prominent examples of ionic liquids with superior electrochemical properties in supercapacitors have been reported. These include 1-ethyl-3-methylimidazolium tetrafluoroborate ($EMIMBF_4$), 1-butyl-1-methylpyrrolidinium bis(trifluoromethanesulfonyl)imide (Pyr14TFSI), and 1-ethyl-3-methylimidazolium thiocyanate (EMIMSCN). Because of its exceptional thermal stability and excellent ionic conductivity, $EMIMBF_4$ is a good choice for supercapacitors that must function in harsh environments. Pyr14TFSI is renowned for having a large electrochemical window and low viscosity, both of which improve supercapacitors' charge-discharge speeds and energy density. Supercapacitors function well at low temperatures and have high capacitance because of EMIMSCN's exceptional solvation capabilities and low melting point, which are a result of its distinct anion composition. Because of their inherent non-volatility, low flammability, and excellent electrochemical stability, these ionic liquids have excelled in supercapacitors, enhancing their overall longevity and safety. Furthermore, the performance of supercapacitors may be further improved by optimizing characteristics including viscosity, ionic conductivity, and interaction with electrode materials by the modification of ILs' chemical composition. Ionic liquids' adaptability in combining with different additives or solvents creates opportunities for the creation of hybrid electrolytes that can solve certain problems like electrode compatibility or operating temperature range. Ionic liquids are becoming more and more important in the creation of safe, reliable, and eco-friendly supercapacitors as research into the material advances. This represents a breakthrough in the field of energy storage technology.

4.4. Gel and solid-state electrolytes

Gel and solid-state electrolytes represent a transformative shift in supercapacitor technology, offering unique advantages in terms of safety, form factor, and mechanical stability. Gel electrolytes, typically formed by immobilizing liquid electrolytes within a polymer matrix, combine the high ionic conductivity of liquid electrolytes with the structural integrity of solid materials. Common polymers used in gel electrolytes include poly(vinyl alcohol) (PVA), polyethylene oxide (PEO), and polyacrylonitrile (PAN), often mixed with traditional aqueous or organic electrolytes [214-216]. Solid-state electrolytes, on the other hand, eliminate the liquid component, and are primarily composed of ion-conducting polymers or inorganic materials. They offer exceptional safety by removing the risks of leakage and flammability associated with liquid electrolytes.

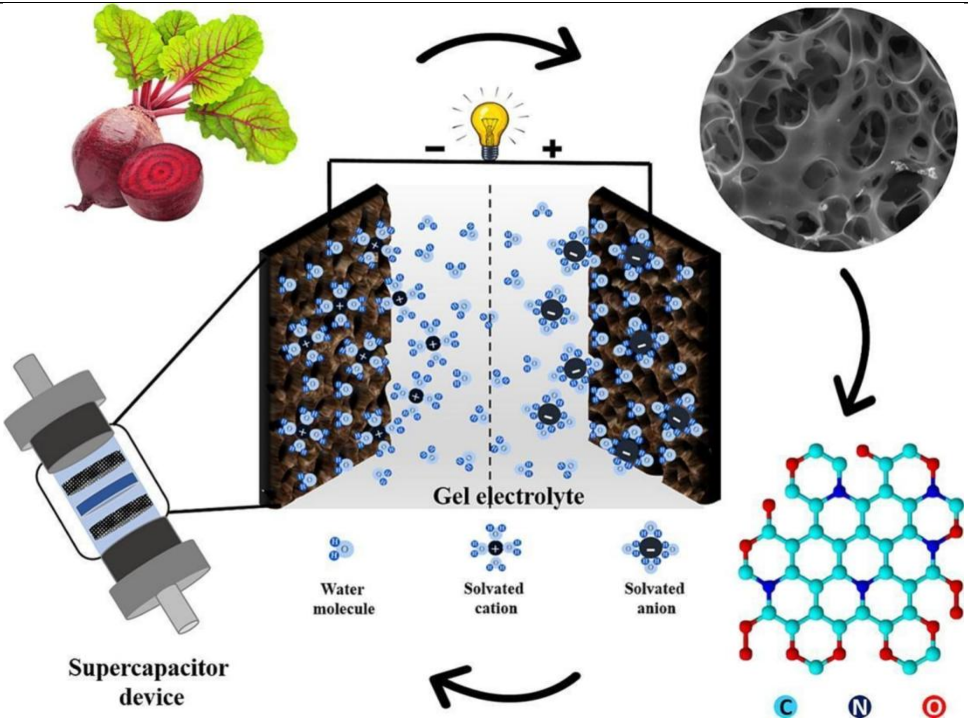


Figure 8. Novel biocompatible gel electrolytes and nitrogen-doped AC produced from biomass for use in solid-state supercapacitors. *Reproduced with permission [221]. Copyright 2023, Elsevier.*

The potential ranges of gel and solid-state electrolytes can vary, but generally, they aim to match or exceed those of their liquid counterparts, often striving for operational windows of up to 3-4 V to maximize energy storage [217-220]. The use of gel and solid-state electrolytes in supercapacitors is important because it makes wearable and flexible energy storage solutions possible, an increasingly popular use in contemporary electronics. Without sacrificing performance, supercapacitors may be included into a variety of form factors, such as bendable and rollable devices, because of the mechanical flexibility and durability of these electrolytes. These electrolytes are also excellent for harsh environment applications because of their improved thermal stability and wide operating temperature range. Notwithstanding, several obstacles persist, namely with ionic conductivity and interfacial resistance in contrast to liquid electrolytes. Gel electrolytes can nonetheless have poorer ion mobility even when they have better conductivity than their completely solid-state counterparts. Solid-state electrolytes, on the other hand, have problems because of their increased interfacial resistance and possible incompatibilities with electrode materials. Thus, the goal of this field's continuing research is to create novel materials and fabrication processes that will improve these electrolytes' ionic conductivity and lower their interfacial resistance. To fully use next-generation supercapacitors, gel and solid-state electrolyte development is essential, especially for applications requiring high safety, flexibility, and integration with innovative electronic devices.

The stability, safety, and flexibility of gel and solid-state electrolytes make them effective for supercapacitors because they allow for the creation of flexible and leak-proof energy storage devices. Excellent electrochemical performances were demonstrated in Selvaraj et al.'s article [221] on the creation of nitrogen-doped AC generated from biomass and innovative biocompatible gel electrolytes for solid-state supercapacitor applications. Using red beetroot (Beta vulgaris L.) as a biomass precursor, Selvaraj et al. synthesized microporous nitrogen-doped AC (NACBR) with an increased specific surface area (SSA) of 2200 m^2/g and a distinct sponge-like porosity (**Figure 8**). In vitro cytotoxic tests revealed this structure to be biocompatible, which increases its use for implanted medical devices. The specific capacitance of the electrode material was enhanced by nitrogen doping in the carbon structure, which also raised its surface wettability and conductivity. In 1 M Na_2SO_4 electrolyte, the NACBR electrode demonstrated a high specific capacitance of 492 F/g at a current density of 1 A/g, according to electrochemical measurements. To build biocompatible solid-state supercapacitors that might power implanted medical devices, the study also investigated the usage of Na_2SO_4, phosphate-buffered saline (PBS), and simulated bodily fluid (SBF) gel-electrolytes. The research's last section emphasized how NACBR's high SSA and hierarchical porous structure enable it to store a tremendous amount of charge. It was determined that the synthesized NACBR was biocompatible, which gives it potential use in biomedical applications. Utilizing the three gel electrolytes, solid-state supercapacitors were constructed, with specific capacitances of 348, 284, and 164 F/g in 1 M Na_2SO_4, PBS, and SBF gel electrolytes, respectively, at 1 A/g. With almost 100% capacity retention over 5000 cycles, these devices demonstrated outstanding cyclic stability. Notably, the supercapacitor with the 1 M Na_2SO_4 gel electrolyte produced a 500 W/kg power density and 12 Wh/kg energy density. The supercapacitors' operating range was increased by the increased electrochemical potential windows seen in the Na_2SO_4 and PBS gel electrolytes. In addition, these devices were shown to be useful in real-world situations by powering an LED. The research conducted by Selvaraj and colleagues makes a noteworthy contribution to the field of biocompatible energy storage. It highlights the potential uses of carbon produced from biomass and gel electrolytes in the creation of solid-state supercapacitors for medical and other flexible electronics applications.

Table 2. Key properties, examples, and applications of various electrolytes in supercapacitors.

Electrolyte Type	Key Properties	Examples	Applications
Aqueous electrolytes	High ionic conductivity, environmental friendliness, low cost, narrow operational voltage window	H_2SO_4, KOH, Na_2SO_4	Standard supercapacitors, low-cost energy storage solutions
Organic electrolytes	Wide operational voltage window, higher energy densities, low flammability, higher cost	Propylene carbonate (PC), ethylene carbonate (EC), acetonitrile (AN)	High-performance supercapacitors, energy storage for advanced electronics
Ionic liquids	Wide electrochemical stability window, high thermal stability, non-volatility, non-flammability	$EMIMBF_4$, $Pyr_{14}TFSI$, EMIMSCN	Supercapacitors for extreme conditions, safer energy storage systems
Gel and solid state electrolytes	Leak-proof, flexible, safer, stable, suitable for flexible and thin-film supercapacitors	PVA based electrolytes, polyethylene oxide (PEO), polymer-ceramic composites	Flexible and wearable supercapacitors, thin-film energy storage devices

The development of several gel and solid-state electrolytes has increased significantly in response to recent developments in supercapacitor technology, and each of these electrolytes has shown impressive performance improvements. Ionic-conducting polymers such as polyethylene oxide (PEO), polymer-ceramic composites, and electrolytes based on PVA are notable examples.

PVA-based electrolytes have gained popularity due to their superior mechanical stability and ion transport capabilities, which make them appropriate for wearable and flexible supercapacitors. They are frequently mixed with acidic or alkaline solutions. High-performance energy storage systems require polymer-ceramic composites with improved ionic conductivity and thermal stability, such as PEO/$LiClO_4$ with alumina fillers [23, 41, 52, 222]. Ionic-conducting polymers, such as PEO, that are doped with lithium salts yield a solid polymer electrolyte that combines the advantages of a solid matrix's structural stability with strong ionic conductivity. The superior performance of gel and solid-state electrolytes in supercapacitors can be attributed to their inherent safety, which reduces the possibility of fire and leakage, flexibility, and thin-film formation capabilities. Furthermore, the lack of liquid components in these electrolytes efficiently resolves the problem of electrolyte evaporation, which extends the supercapacitors' lifespan and operating stability. In order to further increase the operating range and efficiency of supercapacitors, recent advances have also concentrated on hybrid electrolyte systems, which combine the strong ionic conductivity of liquid electrolytes with the mechanical qualities of solid polymers. Supercapacitor technology is evolving towards more efficient, robust, and adaptable energy storage solutions because of the versatility and adaptability of these new gel and solid-state electrolytes. This represents a significant advancement in advanced energy storage systems.

The efficiency and versatility of supercapacitors are significantly influenced by the type of electrolyte used. **Table 2** explains various electrolytes, including aqueous, organic, ionic liquids, and gel and solid-state types, delineating their key properties, examples, and applications. This comparative analysis underscores electrolyte selection's crucial role in optimizing supercapacitors' performance and applicability in various technological contexts.

5. Conclusions and future directions

This review, which concentrated on the characteristics of electrode materials and electrolytes, offered a thorough summary of the state-of-the-art supercapacitor technology as it exists now. We have established a solid grasp of the fundamental factors and important characterization methodologies that drive the performance of supercapacitors. Examining several kinds of supercapacitors, such as hybrid supercapacitors, EDLCs, and pseudocapacitors, has brought attention to the many strategies for energy storage as well as the distinctive benefits that each kind of supercapacitor has to offer. Electrode materials have clarified the contrasting and complementing nature of carbon-based materials, conductive polymers, metal oxides and hydroxides, and new materials such as MOFs and MXenes. The importance of these materials in influencing the energy and power densities, cycle life, and overall efficiency of supercapacitors is emphasized in this study. Likewise, the electrolyte discussion including organic, ionic liquid, gel, and solid-state types emphasizes the crucial role electrolytes play in establishing the voltage range, safety profiles, and operating stability of supercapacitors. Offering a synthesis of current knowledge and a critical analysis of the trends and problems in supercapacitor technology, this review article stands out as an important addition to the area. It directs upcoming developments and applications in energy storage and is a vital resource for academics and business people. Future developments in the field of supercapacitors are expected to be revolutionary. When it comes to electrode materials, the emphasis will probably move to creating more stable and economically viable substitutes with higher energy densities. Novel nanostructures and hybrid compositions will be explored further in the field of carbon-based materials research, while conductive polymers will witness improvements in their mechanical and chemical stability. It is anticipated that electrolyte research will shift towards safer, more ecologically friendly choices with expanded operating windows and enhanced ionic conductivities. Future research on the possibilities of solid-state and gel electrolytes in flexible and wearable electronics seems very promising. Moreover, combining lithium-ion batteries and supercapacitors with other energy storage technologies is a viable way to build hybrid systems that incorporate the best features of both technologies. The rising need for sustainable energy sources and effective energy storage across several industries, such as consumer electronics, grid storage, and the automotive industry, will also propel the development of supercapacitor technology. In summary, the dynamic and developing character of supercapacitor technology is demonstrated by this review work. To guarantee that supercapacitors continue to play a crucial part in the shift towards more effective and sustainable energy storage solutions, it offers an overview of the existing situation and plots a route for future research and development.

Conflicts of Interest

There are no conflicts of interest reported by the authors.

Authors contribution

Syed Shaheen Shah: Conceptualization, Investigation, Validation, Formal analysis, Writing-original draft, Visualization. Md. Abdul Aziz: Supervision, Conceptualization, Project administration, writing-review and editing.

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Data Availability statement

All the data support this article will be provided on request from the first author.

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