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Review

Review on Conductive Polymer Composites for Supercapacitor Applications

Melkie Getnet Tadesse [1,2,*], Abdella Simegnaw Ahmmed [2,*] and Jörn Felix Lübben [1]

[1] Sustainable Engineering (STE), Albstadt-Sigmaringen University, 72458 Albstadt, Germany; luebben@hs-albsig.de
[2] Ethiopian Institute of Textile and Fashion Technology, Bahir Dar University, Bahir Dar 1037, Ethiopia
* Correspondence: melkiegetnet23@gmail.com (M.G.T.); abda0515@gmail.com (A.S.A.)

Abstract: The rising demand for energy storage systems with high power density, rapid charge/discharge capabilities, and long cycle life has pushed extensive research into advanced materials for supercapacitor applications. There are several materials under investigation, and among these materials, conductive polymer composites have emerged as promising candidates due to their unique combination of electrical conductivity, flexibility, and facile synthesis. This review provides a comprehensive analysis of recent advancements in the development and application of conductive polymer composites for supercapacitor applications. The review begins with an overview of the fundamental principles governing electrical conductivity mechanism, applications of conductive polymers and the specific requirements for materials employed for these devices. Subsequently, it delves into the properties of conductive polymers and the challenges associated with their implementation for supercapacitors, highlighting the limitations of pristine conductive polymers and the strategies employed to overcome these drawbacks through composite formation. In this review, conductive polymer composites and their applications on supercapacitors are explored, and their advantages and disadvantages are discussed. Finally, the electromechanical properties of each conductive polymer composite are elaborated.

Keywords: electrically conductive polymers; conductive polymer composite; supercapacitors; energy harvesting; energy storage

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1. Introduction

Polymers are natural or artificial substances that are made up of very large molecules or macromolecules, which are repetitions of simpler chemical building blocks known as monomers. Numerous components of living organisms are made of polymers, such as proteins, cellulose, and nucleic acids. Moreover, they serve as the foundation for products like concrete, glass, paper, plastic, and rubber, as well as minerals like diamond, quartz, and feldspar [1]. Polymers can be categorized as being organic, inorganic, synthetic, natural, thermoplastic, elastomeric, duroplastic, thermosetting, plastic, or fiber. Organic plastics and organic polymers that conduct electricity are known as conductive polymers or, more accurately, intrinsically conductive polymers (ICPs). While polyacetylene, the first conductive polymer, was found in the 1970s, conductive polymers, which are relatively new materials, have undergone extensive research. Conductive polymers (CPs), often known as synthetic metals, are organic polymers that display highly reversible redox behavior and exhibit traits shared by plastics and metals. Researchers were interested in them because they had superior environmental stability compared to standard inorganic materials and had economic relevance, increased stability, decreased weight, higher workability, resistance to corrosion, and excellent electrical conductivity [2–6]. These properties lead researchers to use conductive polymers for supercapacitor applications rather than other applications.

In the pursuit of efficient and sustainable energy storage solutions, supercapacitors have emerged as promising devices with the ability to deliver high power densities, rapid

charge/discharge rates, and extended cycle lifespans [7,8]. Among the various materials explored for supercapacitor electrodes, conductive polymer composites have gained considerable attention due to their unique combination of electrical conductivity, mechanical flexibility, and facile processability [9]. This review aims to comprehensively survey the recent advancements in the development and application of conductive polymer composites for supercapacitor technology. Conductive polymers have been used in supercapacitors due to their unique combination of properties that make them suitable for energy storage applications. Supercapacitors, also known as ultracapacitors or electrochemical capacitors, are energy storage devices that store and deliver energy through the electrostatic separation of charge. Conductive polymers are one of the first choices for supercapacitor applications due to their high conductivity, large surface area, flexibility and processability, and pseudocapacitive properties.

2. Electrical Conductivity Mechanism of Conductive Polymers

The electrical conductivity mechanism of conductive polymers (CP) is not the same as that of conductive material and semi-conductive material due to their electronic properties, which cannot exhibit a standard band theory. The mechanism of conduction is a very important parameter for CP application in numerous areas. The mechanism of electrical conductivity depends on the formation and types of CP. Electrical conductivity could occur only after thermal or photolytic activation of electrons to give them sufficient energy to jump the gap and reach a lower level of the conduction band [10]. The conductive mechanism of conductive polymers involves a unique combination of electronic and ionic conductivity. Unlike traditional metals, where electrons are the primary charge carriers, conductive polymers exhibit a dual charge transport mechanism. The backbone of the polymer chain consists of alternating single and double bonds, allowing for π-electron delocalization along the conjugated structure. The π-electron delocalization enables the formation of a highly conductive pathway, facilitating electronic conduction. Additionally, conductive polymers can undergo reversible redox reactions at the polymer-electrolyte interface, leading to the movement of ions in and out of the polymer matrix. This process, known as doping and de-doping, results in changes in the oxidation state of the polymer, contributing to ionic conductivity.

There are three cases for a conductive polymer to be an electrical conductor. First, the molecule must initially have a linear backbone. Contiguous sp^2 hybridized carbon centers make up the backbone of conductive polymers, which is what this signifies. Each center has a single valence electron that is in an orthogonal p_z orbital to the other three sigma-bonds. A molecule-wide delocalized collection of orbitals is created by the combination of all p_z orbits, and the electrons in these delocalized orbitals have high mobility [11]. The second need is that the molecule has extended conjugation. A continuous array of "p" orbitals that can align to provide a bonding overlap over the entire system is needed for a conjugated system. The condition arises when two systems are "linked together", such as double bonds, and improves the extension of the chemical reactivity that results in an increase in the conductivity of the electrons [12]. Thirdly, the introduction of dopants or charge carriers (either charge carrier's holes or charge carrier electrons) is of great importance. Since the charge created by the dopant is what gives conductive polymers their conductivity, increasing the doping level causes more charges to be created in the polymer, which results in a higher conductivity by reducing inter-particle gaps using metallic fillers for making highly conductive polymer composites. Because the molecules get further apart as the temperature rises, conductive polymer conductivity is also temperature-dependent [13].

Conductive polymers, also known as intrinsically conductive polymers (ICPs), are a class of organic polymers that exhibit electrical conductivity. The electrical conductivity in these polymers arises from the delocalization of π-electrons over the polymer backbone. The most common conductive polymers include but are not limited to poly (3,4-ethylenedioxythiophene)-polystyrene sulfonate (PEDOT-PSS), polyaniline, polypyrrole,

and polythiophene. The electrical conductivity mechanism of conductive polymers can be explained through various concepts:

Doping and De-doping: Conductive polymers typically undergo a process known as doping, where an electron acceptor (dopant) donates electrons to the polymer chain. This introduces charge carriers (positive holes or polarons) into the polymer structure, leading to electrical conductivity. Conversely, de-doping involves the removal of dopant species, resulting in a decrease in conductivity. For example, ethylene glycol can be introduced as a doping element to remove the insulative part of the PEDOT-PSS so that conductivity can be enhanced [14].

π-Electron Delocalization: The backbone structure of conductive polymers contains alternating single and double bonds, which is called conjugation. This conjugation allows for the delocalization of π-electrons along the polymer chain. The delocalized π-electrons can move freely through the polymer structure, thus contributing to the electrical conductivity [15].

Charge Carriers: The charge carriers in conductive polymers are typically polarons or bipolarons, which are charge carriers created through the doping process. Polaron formation involves the movement of electrons within the π-conjugated system, leading to a distortion of the polymer chain and the creation of a charged site [16].

Band Structure: The electronic band structure of conductive polymers plays an important role in their electrical conductivity. The π-conjugated system forms the band structure like semiconductors. The highest embedded molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) entailed in the conduction process [17].

Redox Reactions: Conductive polymers can undergo redox reactions, where the polymer chain itself acts as a redox-active material [18]. In these reactions, electrons are transferred between the polymer and the dopant, leading to changes in the oxidation state of the polymer and, consequently, its electrical conductivity.

Dopant Influence: The choice of dopant significantly influences the electrical conductivity of conductive polymers. Doping introduces charge carriers into the polymer structure, and the type and concentration of dopant can affect the conductivity. Different dopants can lead to p-type or n-type conductivity [19].

Doping induces a change in the morphology of the polymer chain, leading to chain swelling [20]. The increased separation between polymer chains facilitates charge carrier movement and enhances electrical conductivity.

It is important to note that the electrical conductivity of conductive polymers can be tuned by adjusting various factors such as the doping level, type of dopant, polymer morphology, and environmental conditions. This tunability makes conductive polymers attractive for a wide range of applications, including organic electronics, sensors, and energy storage devices.

3. Applications of Conductive Polymers

Conductive polymers are a class of organic materials with unique electrical properties, blending the mechanical flexibility of polymers with the electrical conductivity of metals. Unlike traditional insulating polymers, conductive polymers can conduct electricity due to their conjugated molecular structures, allowing for the delocalization of π-electrons along the polymer backbone. This π-electron delocalization creates a pathway for charge carriers, enabling electronic conduction. Moreover, many conductive polymers exhibit intriguing properties like reversible redox reactions, facilitating ionic conductivity. These characteristics make conductive polymers valuable in a range of applications, including flexible electronics, sensors, actuators, and energy storage devices such as supercapacitors. Ongoing research focuses on enhancing the performance and expanding the versatility of conductive polymers for emerging technological advancements.

In recent years, research based on conductive polymers has played a significant role in advanced applications ranging from optoelectronics to material science. For all intents and purposes, conductive polymers can be described as Nobel Prize-winning materials that

were awarded the Nobel Prize in Chemistry in 2000 [21]. These enhancements have allowed CPs to have practical applications in various fields, including industrial (electromagnetic shielding) EMI shielding, microwave tempering, and the dissipation of static charge [22–24] and biomedical, which enhance the stability, speed, and sensitivity of various biomedical devices [25].

In addition, CPs use a wide range of molecules that can be applied or used as dopants for the application of electromagnetic shielding and microwave absorption, [26,27] static electricity dissipation, heating with high thermal conductivity using conductive polymer composites (CPCs) containing carbon or metallic particles, and [28–32] membrane and biosensor materials [33–36]. Conducting redox polymers such as polypyrrole or polyaniline are actually used for intelligent corrosion protection [37–39].Furthermore, conductive polymers such as polyaniline (PANI), polypyrrole (PPy), and poly (3,4-ethylenedioxythiophene or PEDOT), and polythiophene (PTh) derivatives were used in textile sensors and actuators for various areas namely in medical textiles, protective clothing, touchscreen displays, flexible fabric keyboards, as well as biosensors [40–43].

4. Types of Conductive Polymers

Conductive polymers (CPs) frequently have a linear backbone made up of conjugated monomers that repeat as their basic structural element. Examples of CPs include polyacetylene (PAC), polypyrrole (PPy), polyaniline (PANI), and polythiophene (PTh) [44]. There are two main categories of conductive polymers, which are described below.

The first type of conductive polymer is known as intrinsically conductive polymers (ICPs), and the second type is called extrinsically conductive polymers (ECPs). ICPs consist of electroactive long-range conjugated polymers, including polyanilines, polypyrrole, and polythiophenes and their conjugation [45–47]. These conjugated polymers are composite materials with dimensions in the nanometer range, in which a polymer binds metallic flakes that resemble a conductive filler [48]. The intrinsically conductive polymers were further divided into two types: conjugated conductive polymers such as PEDOT, polyacetylene, polyaniline, and polypyrrole and doped conductive polymers. Partial oxidation (p-doping) may provide the necessary charge carriers because the majority of organic polymers lack inherent charge carriers. Polythiophene and polyaniline falls in this category of doping conductive polymers. To further enhance the conductivity of the conductive polymers, conductive materials such as carbon black, carbon fibers, aluminum flakes, stainless steel fibers, metal-coated fillers, and metal particles can be incorporated to the conductive polymers. Conjugated polymers, oligomers, and a variety of metals can all be used to create carbon nano composite (CNCs). The second classification of CPs consists of extrinsically conductive polymers, also referred to as polymers with conductive components. These conductive polymers ingredients are a collection of polymers whose backbones naturally transfer charge, making the polymer itself conductive [49]. The chemical bonds that form an unpaired electron per carbon atom in the polymer's backbone are what cause the conductivity. The orbits of subsequent carbon atoms overlap in the sp^2p_z hybridized state, where the carbon atoms are bonded, allowing for the delocalization of the electrons throughout the polymer chain [50]. Figure 1 depicts the classification of conductive polymers.

4.1. Intrinsically Conductive Polymer

An intrinsically conductive polymer (ICP) refers to a type of polymer that inherently exhibits electrical conductivity without the need for external doping or treatment. These polymers are unique in that they possess inherent electronic properties due to their specific chemical structure, which allows for the movement of charge carriers along the polymer backbone. The most common types of intrinsically conductive polymers include polyaniline, polypyrrole, and polythiophene. Intrinsically conductive polymers are exciting materials since they combine the advantages of both plastics and metals; however, due to their electrical characteristics, stability, and process ability, their applicability is limited [51,52]. Intrinsically conductive polymers have the potential to be used in a wide

range of unique applications, combining the benefits of both polymers and metals. They are synthetic polymers made of common substances, including C, S, N, O, and H. They have low density and high mechanical characteristics like other polymers, and they are conductive like metals. Additionally, it is feasible to modify the electrical structure and properties of conductive polymers by altering the synthesis conditions, the doping species and degree, and the chemical structure of the conjugated polymer chains. They display remarkable electrochemical behavior as well. The transparent electrodes of optoelectronic devices, such as light-emitting diodes (LEDs), solar cells, and detectors, can be made of intrinsically conductive polymer films. Due to the presence of a conjugated π-electron backbone, these polymers exhibit electronic properties such as low-energy optical transmission, low ionization potential, and high electron affinities. These unique properties make these materials suitable for applications such as thin transistors, organic light-emitting diodes, sensors, supercapacitors, organic solar cells, and electrochromic displays [53].

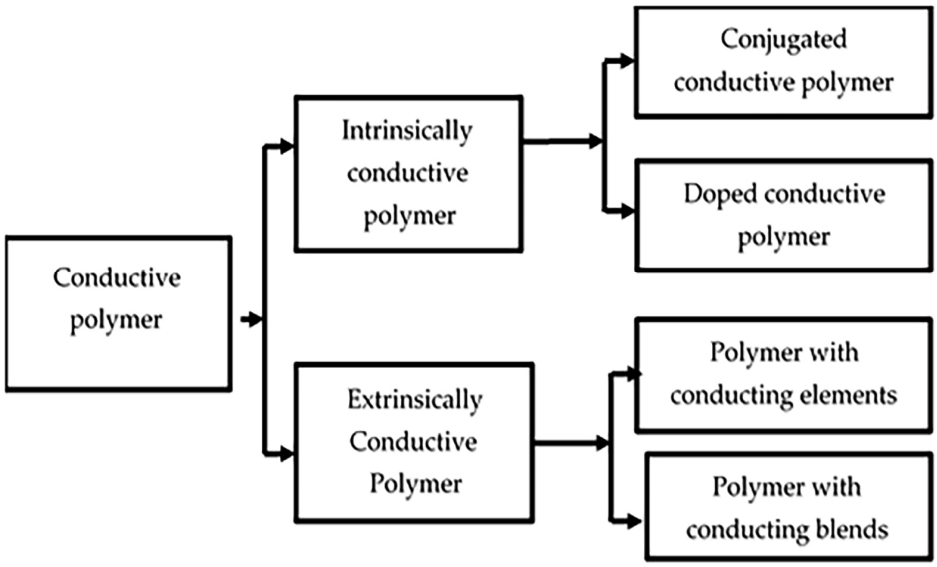

Figure 1. Classification of conductive polymers.

Key characteristics and features of intrinsically conductive polymers include:

Conjugated Structure: ICPs have a conjugated structure with alternating single and double bonds along the polymer backbone [54]. This conjugation creates a delocalized π-electron system that facilitates electronic conduction.

π-Electron Delocalization: The presence of conjugated double bonds allows for the delocalization of π-electrons over an extended molecular orbital system. This delocalization enables the movement of charge carriers, contributing to electrical conductivity.

Charge Carrier Formation: In their neutral state, intrinsically conductive polymers can act as insulators. However, when oxidized or reduced, charge carriers such as polarons or bipolarons are formed within the polymer structure, leading to electrical conductivity [19].

Polymerization: The synthesis of intrinsically conductive polymers often involves chemical polymerization methods. The polymerization process is crucial for achieving the desired conjugated structure and electronic properties [2].

Tunable Conductivity: The electrical conductivity of intrinsically conductive polymers can be tuned by modifying their chemical structure, controlling the degree of polymerization, or introducing specific functional groups [54,55]. This tunability makes them suitable for various applications.

Applications: Intrinsically conductive polymers find applications in a range of electronic devices, including organic solar cells, organic light-emitting diodes (OLEDs), sensors, actuators, and flexible electronics [52]. Their unique combination of electrical conductivity and processability makes them attractive for emerging technologies.

Environmental Stability: Some intrinsically conductive polymers exhibit environmental stability, allowing them to withstand exposure to air and moisture. However, the stability can vary depending on the specific polymer and its chemical composition [56].

In general, intrinsically conductive polymers (ICPs) are a specialized class of polymers that inherently possess electrical conductivity without requiring additional doping or external factors. These polymers typically feature a conjugated structure along their molecular backbone, allowing for efficient charge transport. One notable example of an intrinsically conductive polymer is polyacetylene, which is recognized for its high conductivity and role in pioneering the field of conductive polymers. Other examples include polypyrrole, polythiophene, and polyaniline. These polymers exhibit a range of electrical and electrochemical properties, making them valuable in applications such as organic electronics, sensors, and conductive coatings. Researchers continue to explore and engineer intrinsically conductive polymers to unlock their full potential in various technological advancements. Polymers such as polyacetylene, polyaniline, polypyrrole, and PEDOT-PSS are some types of ICPs.

4.2. Extrinsically Conductive Polymer/Conductive Polymer Composites

Conductive polymer composites (CPC) are primarily made of insulating polymer matrices and conductive fillers with high electrical conductivity. The conductive fillers operate as carriers for the conductive fillers' transfer into the polymer composites [57]. Due to their light weight, ease of manufacturing, chemical resistance, high conductivity, and tunable electrical properties, conductive polymer-based composites have recently gained popularity in both academic research and industrial applications [58]. Conducive polymers can be manufactured in a variety of structural configurations with a variety of design configurations and functionalities for the polymer components as well as the electrically conductive component. Conductive polymer composites refer to materials that combine conductive polymers with other substances to enhance their properties. Conductive polymers are a class of polymers that can conduct electricity. They exhibit unique electronic, optical, and electrochemical properties, making them attractive for various applications, such as sensors, actuators, electronic devices, and more. However, pure conductive polymers often have limitations in terms of mechanical strength, processability, and stability.

Extrinsically conductive polymers are blended polymers obtained by solvent mixing or melt mixing, and the blends are responsible for electrical conductions [59]. These types of conductive polymers can be obtained by polymerizing an electrode surface (anode) that has been coated with a non-conductive polymer. These conductive polymers receive their conductivity from the presence of externally introduced components. For example, when carbon black or some metal oxides or metallic fibers are introduced into the no-conductance polymer, the polymer becomes electrically conductive [60]. The three most important conductive fillers are carbon (carbon black (CB) and carbon nanotubes (CNTs)), metal powders and their compounds (indium tin oxide (ITO) and aluminum zinc oxide (AZO)), and ICPs (PPy and PANI). ECPs have special properties such as good electrical and thermal conductivity, corrosion resistance, and good mechanical properties [41]. However, they have much lower conductivity values than the ICPs but have equal mechanical properties. Additionally, these types of polymers have conductive polymer composites (CPC) structures, which can be manufactured as binary composites, ternary composites, and quaternary composites that are composed of CP and other materials, elements, parts, or divisions, including metal materials, non-metal materials, organic materials, and polymer composites. Therefore, depending on the filler conductive elements, the conductivity of the CPCs varied. Conductive polymer composites can be produced using conductive polymers with carbon-based materials, graphene composites, CNT, and metal oxides. The metal oxides used as fillers in the composites with conductive polymers for applications of supercapacitor were, namely, ruthenium oxide (RuO_2), manganese dioxide (MnO_2), nickel oxide, metal hydroxides, sulfides, and phosphides, which can be formed either as binary nanocomposites or ternary nanocomposites. Beside conductive polymer composites,

EDLC supercapacitor electrode materials were made from conductive polymers with metal oxide or hydroxide and metal sulfides (metal oxides such as tin oxide (SnO_2), vanadium oxides (V_2O_5), cobalt monoxide (CoO), etc.), and the electrochemical properties were investigated [61].

Conductive polymer-based composites are synthesized via a variety of mechanisms. As a result, the development of conductive polymer composites depends on their intended use. The physical, electromechanical, and chemical properties and applications of the as-obtained conductive polymer-based composites would be significantly influenced by the design and structure of the composites, interfacial adhesion between the conductive polymer and other components, and the synthetic strategies [62]. Electrosynthesis with an insulating polymer present is one of the developments of CPC [63,64]. Another method for making CPC is to encase textile fibers in conductive materials. Additionally, conductive polymer nanocomposites are made by adding secondary nanoparticles via the electropolymerization of monomers, such as aniline, pyrrole, and 3,4-ethylenedioxythiophene [65,66], as well as by the use of dopants with a variety of functionalities, such as di-sulfide biotin [67,68]. Moreover, by integrating noble metal nanoclusters, such as platinum nanoclusters, during oxidative polymerization [69], a conductive polymer composite can be developed. Therefore, the developed conductive polymer composites can exhibit multifunctional features, increased mechanical performances, and processability because of the synergistic interaction of several components. Due to the above reasons, conducting nanocomposites (CNCs) has attracted a lot of interest in the possibility of using them to create materials suitable for electrocatalysis, microelectronics, and chemical sensors. Also, conductive polymer composites exhibit multifunctional features, increased mechanical performance, and processibility because of the synergistic interaction of several components. Moreover, conductive polymers are used in a variety of fields, including electromechanical sensors [70], gas sensors [71,72], transducers [73], electrostatics sensing device corrosion protection [74], energy storage [75], supercapacitors [76], and biosensors [77].

5. Conductive Polymer Composite for Supercapacitor Application

Conductive polymer composites have gained significant attention for supercapacitor applications due to their unique combination of electrical conductivity, flexibility, and ease of processing. Supercapacitors, also known as electrochemical capacitors or ultracapacitors, are energy storage devices that bridge the gap between traditional capacitors and batteries, offering high power density and fast charge/discharge capabilities. Here's how conductive polymer composites are relevant to supercapacitor applications:

Conductive Polymer Selection: Polyaniline (PANI), polypyrrole (PPy), and polythiophene (PTh) are commonly used conductive polymers for supercapacitor applications due to their good electrical conductivity and electrochemical properties [78]. Selecting the appropriate conductive polymers will benefit the researcher in getting high-performance supercapacitor applications.

Composite Structure: Conductive polymers are often combined with other materials, such as carbon-based materials (carbon nanotubes, graphene, carbon black) or metal oxides, to form composite structures [79]. Carbon dots and conductive polymers can be synthesized using various polymerization techniques and can be combined to compensate for their demerits to be used for various applications. The combination of a conductive polymer with a conductive filler enhances the overall electrical conductivity and capacitance of the composite.

Enhanced Electrochemical Performance: The composite structure allows for improved charge storage capacity and faster electron transport, leading to enhanced electrochemical performance compared to pure conductive polymers [79]. This system is used to facilitate the production of high-efficiency supercapacitors.

Flexibility and Mechanical Strength: Conductive polymer composites can provide the flexibility needed for flexible and wearable supercapacitor applications. The addition of flexible substrates or matrices enhances the overall mechanical strength of the composite.

Synthesis Methods: Various methods, including in situ polymerization, solution casting, and electrochemical deposition, are employed to synthesize conductive polymer composites for supercapacitors. The choice of method depends on the specific properties desired for the application.

High Surface Area: Incorporating high-surface-area materials, such as carbon nanotubes or graphene, into the composite structure increases the available surface area for charge storage, contributing to higher capacitance [80].

Cycling Stability: Researchers focus on improving the cycling stability of conductive polymer composites to ensure long-term reliability in supercapacitor applications.

Several researchers have conducted and studied the methods of synthesis and capacitive performance of the recent research progress of CPC based on binary composite, ternary composite, and quaternary composite for supercapacitor applications. Supercapacitors are crucial energy storage and recycling tools with high power densities, quick charge and discharge rates, and extended cycle lives that address the pressing problems of energy scarcity and environmental pollution for the sustainable development of humanity [81–84]. In addition, the energy gap between batteries and traditional solid-state and electrolytic capacitors is currently filled by supercapacitors, which may store electrical energy by double-layer charging, faradaic processes, or a combination of the two. Therefore, due to these qualities, it has drawn a lot of attention for industrial applications. Furthermore, the CPC supercapacitor was able to give bursts of high power rather than large amounts of energy since the quantity of energy stored is often tiny and may be delivered instantly [85].

Shokry. A et al. [86] fabricated a binary composite supercapacitor electrode based on poly(3-hexyl-thiophene-2,5-diyl)(P3HT), single-walled-carbon-nanotubes (SWCNTs) nanocomposites with different ratios onto a graphite sheet as a substrate with a wide voltage window in non-aqueous electrolyte. According to the findings obtained, the P3HT/SWCNT nanocomposite has a higher specific capacitance than its individual components. Microporous structures that facilitate ion diffusion and electrolyte penetration in these pores contributed to the nanocomposite's high electrochemical performance. However, the voltammograms obtained do not clearly show the anodic peaks, nor do they clearly show a small reduction peak attributable to the reduction of the film deposited on the electrode. In addition, the research study by Akbar Mummoorthi, Abdul Rehman [87,88] developed and characterized a novel tertiary composite supercapacitor, which was carried out by the introduction of S-PANI [89], which was made to synthesize 3-dimensional MnO_2/S-PANI/P-RGO composite aerogels at different scan rates and α-Fe_2O_3/NiO/rGO. These composites were used as a high-performance freestanding cathode for SCs applications and were utilized to create high-performance supercapacitors with improved electrochemical performance using an environmentally friendly hydrothermal self-assembly process. The result shows that the electrochemical performance of ternary composites has been recognized because of their well-designed, unique architecture, which provides a large surface area and synergistic effects among all three constituents. The 3D MnO_2/S-PANI/P-RGO composite supercapacitor had a specific capacitance of 571 Fg^{-1} at a current density of 1 $Ag^{-1,}$ along with a specific capacity of 413 Fg^{-1} at 40 Ag^{-1} and a retention capacity of 72.6%. In addition, the Fe_2O_3/NiO/rGO composite supercapacitors had an energy density of 35.38 W h kg^{-1} at a power density of 558.6 W kg^{-1} and retained a 94.52% capacitance after 5000 cycles at a 1 Ag^{-1} current density, as shown in Figure 2.

From the results, it was discovered that every single MnO_2/S-PANI/P-RGO cyclic voltammetry (CV) curve had a rectangle shape with a pair of redox peaks in the voltage range of 0.2–0.6 V, implying that MnO_2/S-PANI/P-RGO composite electrodes have not only double-layer capacitance but also possess pseudocapacitance characteristics (Figure 2A–C). The result also shows he greater the specific capacitance of MnO_2/S-PANI/P-RGO composite aerogels (Figure 2D). Additionally, using a hydrothermal method, Zichen Xu et al. [90] created a ternary composite supercapacitor from zinc sulfide/reduced graphene oxide (ZnS/RGO) and doped it with various conductive polymers (PANI, PPy, PTh, and PEDOT) via in situ polymerization. He then examined the capacitor's capacitance performance and

reliability. According to the results, the PANI ZnS/RGO ternary electrode composite has the best cycle stability and capacitance performance, with values of 160% at 1 Ag^{-1} and 1045.3 Fg^{-1} after 1000 loops and power densities of 18.0 kW kg^{-1} and 349.7 W h kg^{-1}. High-energy power supercapacitors will use the ZnS/RGO/PANI electrode due to its improved performance, such as its fast charge and discharge ability, which is very important for electric energy storage devices. Thus, supercapacitors and batteries have been widely employed in electric cars and portable electronic devices that require safe, trustworthy technology.

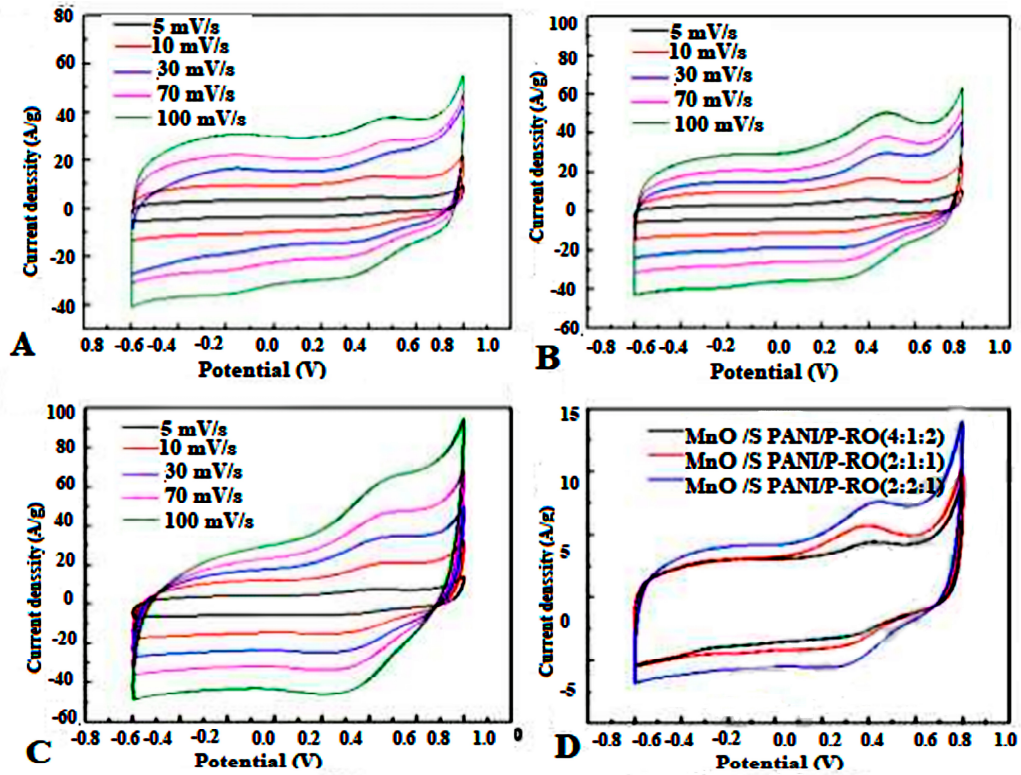

Figure 2. CV curves of (**A**) MnO_2/S-PANI/P-RGO (4:1:2), (**B**) MnO_2/S-PANI/P-RGO (2:1:1), and (**C**) MnO_2/S-PANI/P-RGO (2:2:1) aerogels at different scan rates; (**D**) CV curves of all MnO_2/S-PANI/P-RGO samples at a rate of 10 mV/s [87] Copyright Elsevier, License Number: 5716450597815.

Moreover, the recent advancements in flexible electronic devices, such as flexible displays, curved smartphones, flexible implantable medical devices, and wearable electronic devices, indicate that flexible devices are the first to be developed as an important revolution in the next generation of advanced electronics. Lighter weight, wearability, bendability, environmental friendliness, and reduced costs are advantages of flexible electronic devices compared to conventional electronic devices. To balance and apply the growth of flexible electronic devices, the energy storage mechanism should be developed to be light, thin, and flexible [91].

In the field of flexible supercapacitors, numerous researchers have developed and investigated the electrochemical properties of flexible supercapacitors. For example, Yu Jin Kang et al. [92] investigated flexible supercapacitors based on papers coated with carbon nanotubes and gel electrolytes based on ionic liquids using a drop-dry technique. CNTs were coated on office paper to create flexible electrodes. High power density and energy density performances were demonstrated by this solid-state flexible supercapacitor. Simply considering the mass of active materials, the specific capacitance of the CNT electrodes was 135 Fg^{-1} at a current density of 2 Ag^{-1}. The supercapacitors' maximum power and energy density were 41 Wh kg^{-1} and 164 kW kg^{-1}, respectively [93].

The most popular electrode material for developing flexible supercapacitors and enhancing their electrochemical properties was activated carbon. Since activated carbon

may be made with a large specific surface area and is inexpensive and readily available on the market, nevertheless, the electrical double layer (EDL) could only be charged using the surface area that was accessible for electrochemistry. Both the energy and the power are significantly increased in such a situation. It seems that materials having pseudocapacitance qualities would greatly benefit from the addition of nanotubes as a conducting additive and/or support [94,95]. In addition, a conductive graphene film is produced by different methods such as blade coating, vacuum-assisted self-assembly wet spinning, gel die casting interface, and self-assembly methods for wearable electronic products (such as curved smartphones, smartwatches, laptops, and electronic skins) [96–98].

In recent times, the rapid development of flexible electronics, such as roll-up displays, touch screens, smart electronics, smart textiles, and wearable sensors, is being accelerated by developments in material science and technology as well as the robust consumer demand for portable, thin, flexible electronics and supercapacitors that possess remarkable features of miniaturization, high security, and easy integration [99,100]. Electroactive materials that are flexible and lightweight are increasingly being developed to meet the demands of the expanding electronic sector. A number of methods for flexible supercapacitors have recently been proven, including wire-shaped [101,102], fiber-shaped coaxial supercapacitors [103], smart textiles made of carbon nanotubes (CNTs), and very stretchable fiber-shaped supercapacitors [104–109]. However, because of the high surface area of carbon nanostructures, the capacitance of the electrochemical double-layer capacitors (EDLCs) was low and constrained. Furthermore, several planar, 2D, and 3D printable conductive polymers were employed in the production of flexible supercapacitors. Ghosh et al. [110] conducted a study and produced conducting-polymer hydrogel electrodes that were significantly swollen. By using a straightforward preparation process, the aqueous dispersion of PEDOT-PSS is paradoxically cross-linked into a nanometer-scale conducting network, giving the material its high porosity and swelling capacity. Also, textiles, such as carbon cloth, polymer, and ceramic fabrics, and 1D fibers, including aligned or twisted CNTs and graphene fibers, have been utilized as flexible substrates for CP-based supercapacitors [82,111–113]. The energy density of SCs is greater than that of conventional capacitors; however, the power density of capacitors is greater than that of SCs, as shown in Figure 3.

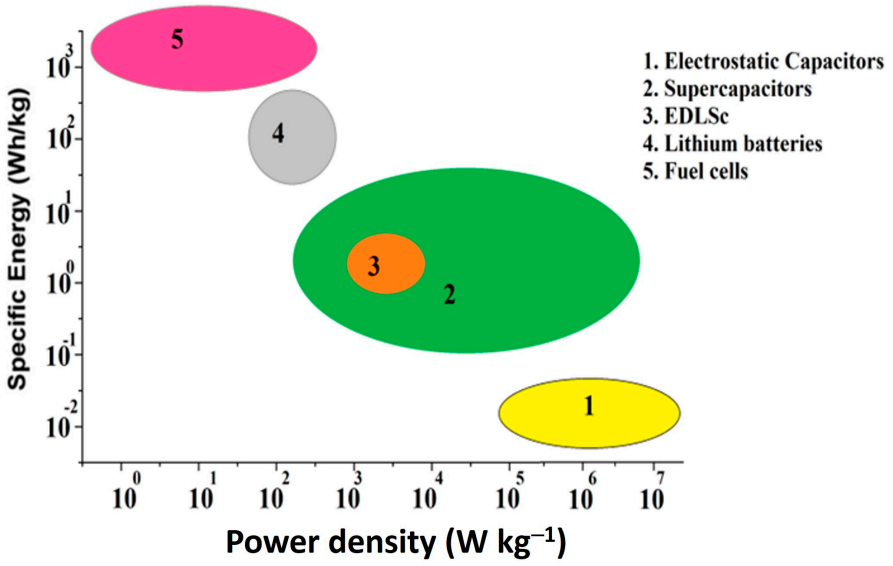

Figure 3. Power and energy density of different energy storage.

Figure 3 shows an illustration of a region plot, which contrasts the overall performance of several energy storage technologies by graphing specific energy against specific power. In that diagram, SCs are positioned between batteries and conventional capacitors to show that they have more specific energy than traditional capacitors. To compete with the efficiency of batteries and fuel cells, the specific energy of SCs must be increased.

6. Types of Supercapacitors

Supercapacitors can be classified into three groups based on their charge and discharge mechanisms [114]. Supercapacitors can be made from different materials based on the type of energy storage required and the application of the capacitance range. To produce supercapacitors, electrode materials are presently available in different forms and different materials, such as carbons, metal oxides, and conductive polymers, as shown in Figure 4.

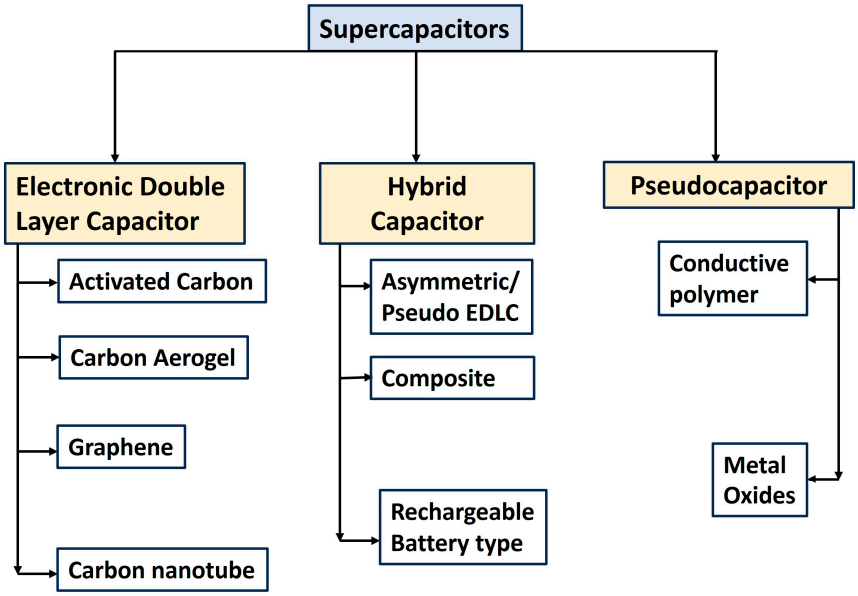

Figure 4. Hierarchical classification of supercapacitors.

6.1. Electric Double Layer-Stage Capacitors (EDLCs)

Electric double-layer capacitors (EDLCs) are made mostly of high-surface-area carbon materials such as carbon aerogels, activated carbon, and carbon nanotubes and graphene create capacitors by using electrodes or electrolytes to separate charges, as shown in Figure 5. EDLCs store charge electrostatically using the mechanism of the Helmholtz layer principle [115]. The process for energy storage and release was based on nanoscale charge separation at the electrochemical interface established between the electrode and electrolyte. There are no chemical oxidation-reduction (redox) reactions associated with the non-faradaic charge storage mechanism. EDLCs had relatively long cycle lives because only physical charge transferring occurs [116]. There are a variety of mechanisms for assembling the double-layer for producing the EDLCs supercapacitor, such as dip-coating, electrodeposition, spin-coating, spray-assembly, and chemical bath deposition methods [117–120]. The electromechanical characterization of EDLC supercapacitors has been studied by a variety of scholars [121–126]. The average energy density of EDLCs is about 5 Wh kg^{-1}.

In general, electric double-layer capacitors (EDLCs), also known as supercapacitors or ultracapacitors, are energy storage devices that rely on the principles of electrostatic charge separation. Unlike traditional capacitors, EDLCs store electrical energy through the formation of an electric double layer at the interface between a porous electrode material and an electrolyte solution. The double layer consists of positively and negatively charged ions attracted to the electrode surface without undergoing a faradaic reaction, leading to a purely electrostatic energy storage mechanism. EDLCs offer high power density and fast charge/discharge rates due to the rapid ion adsorption and desorption processes at the electrode-electrolyte interface. The capacitance of EDLCs is typically much higher than that of conventional capacitors, making them well-suited for applications requiring quick bursts of energy. EDLCs find use in various fields, including automotive systems, renewable energy storage, and portable electronic devices, where their long cycle life and ability to deliver rapid bursts of energy are advantageous. Ongoing research aims to further improve

the energy density and overall performance of electric double-layer capacitors for broader applications in energy storage.

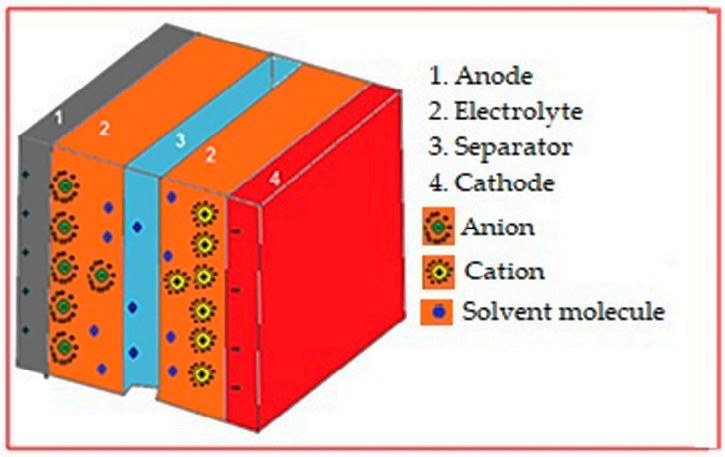

Figure 5. Electrical double-layer capacitors (EDLCs).

6.2. Pseudocapacitors

The second kind of supercapacitor is called a pseudocapacitor. It is primarily made up of conductive polymer and transition metal oxide and is produced by a fast, reversible redox reaction on the surface of an electrode [127–130]. They store energy electrochemically or faradaically, respectively. A pseudocapacitor (Figure 6) is built using high-energy electrode materials based on metal oxides, metal-doped carbons, or conductive polymers in faradaic redox processes [131]. Higher-energy-density supercapacitors are made possible by these electrode materials. Because of this, pseudocapacitors often offer larger energy densities at the expense of slower rates and shorter life cycles than EDLCs.

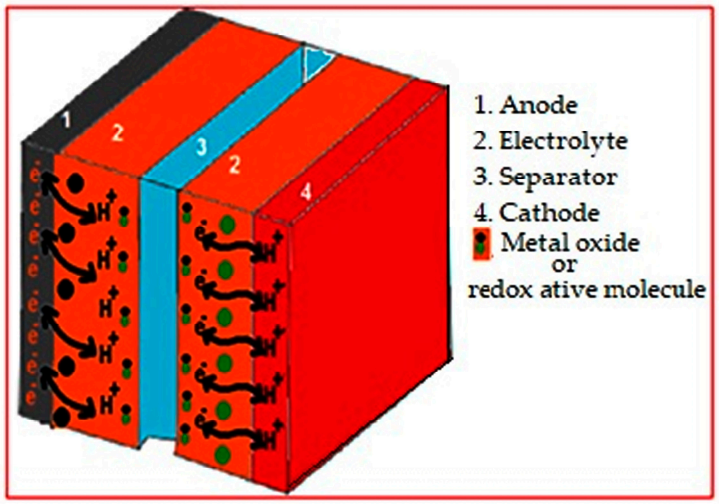

Figure 6. Pseudocapacitor configuration.

Typically, pseudocapacitors can store more charge than EDLCs, but the kinetics of pseudocapacitors are slower than those of EDLCs. This is because the energy storage process in pseudocapacitors occurs in the bulk of the electrode material, while for EDLC, the charge/discharge process only occurs on the surface of the electrode material [132]. Therefore, a two-dimensional layered electrode material with multiple active sites can greatly improve the capacitance and reduce the dynamic effects, which is a promising electrode material for pseudocapacitors. The electrodes are made of metal oxides or

conductive polymers, which show much higher capacitance than EDLCs. A capacitance of 600–1000 Fg^{-1} is attainable.

In summary, pseudocapacitors are a type of electrochemical energy storage device that combines the principles of traditional capacitors with a pseudocapacitive mechanism. Unlike electric double-layer capacitors (EDLCs), pseudocapacitors involve reversible faradaic redox reactions at the electrode–electrolyte interface, leading to additional charge storage beyond the electrostatic double layer. This pseudocapacitance results in enhanced energy density compared to EDLCs. Pseudocapacitors often utilize transition metal oxides, conductive polymers, or other materials that undergo reversible redox reactions, enabling the storage of electrical energy through surface-bound ions. This combination of electrical double-layer capacitance and pseudocapacitance provides pseudocapacitors with higher energy storage capabilities and improved performance in terms of specific capacitance and energy density. Electric pseudocapacitors find applications in various fields, including portable electronics, renewable energy systems, and hybrid electric vehicles. Ongoing research focuses on developing new materials and optimizing electrode designs to further enhance the energy storage performance of pseudocapacitors for advanced energy storage applications.

6.3. Hybrid Supercapacitors

The third type is a hybrid type which is formed by a combination of EDLCs and pseudocapacitors [50]. As the name implies, hybrid supercapacitors combine processes from EDLCs and pseudocapacitors. They store charges by combining electrostatically and electrochemically. A semi-permeable membrane acts as a separator and separates two electrodes from electrical contact in a supercapacitor. An electrolyte solution is used to impregnate the electrodes and separator, allowing ionic current to flow between them while preventing electronic current from discharging the cell [116,133–138]. Current collectors carry electrical current from the electrodes. As a matter of fact, the charge storage mechanisms of the hybrid capacitors (symmetrical and asymmetrical capacitors) are a combination of faradaic and non-faradaic reactions. As a result, the hybrid capacitors provide synergistic effects from both electrodes on the resultant material. A mixture of different types of transition metal oxides, hydrogen oxides, or doped CP is also used to construct the hybrid capacitor, as shown in Figure 7.

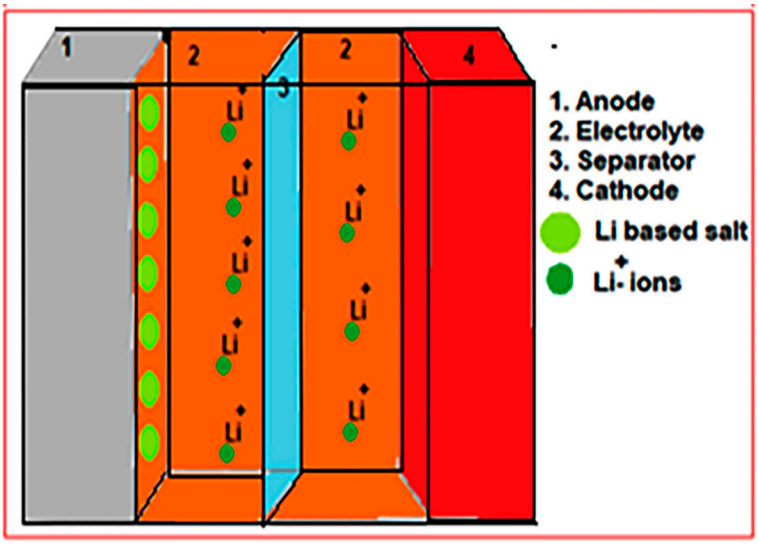

Figure 7. Hybrid supercapacitors schematic configuration.

In conclusion, hybrid supercapacitors are energy storage devices that combine the features of both traditional electric double-layer capacitors (EDLCs) and electrochemical pseudocapacitors. This hybrid design aims to capitalize on the strengths of both tech-

nologies, offering a balance between high power density and enhanced energy density. Typically, hybrid supercapacitors consist of an electrode with a double-layer capacitor material alongside a redox-active material capable of pseudocapacitance. This combination allows for rapid charge/discharge rates characteristic of EDLCs and additional energy storage through reversible faradaic reactions, leading to increased overall energy storage capacity. Hybrid supercapacitors find applications in various fields, including electric vehicles, renewable energy systems, and portable electronics, where the demand for both high power delivery and improved energy density is crucial. Ongoing research is focused on optimizing electrode materials and designs to further improve the performance and efficiency of hybrid supercapacitors.

7. Application of Supercapacitors

Supercapacitors, also known as ultracapacitors or electrochemical capacitors, have a range of applications across various industries due to their unique characteristics, which include high power density, fast charge and discharge rates, long cycle life, and relatively simple maintenance. Supercapacitors are conveniently positioned between capacitors and batteries in electrical component designs. They have several benefits to be able to store a significant amount of energy as an electrostatic field. Due to their high-power density and small size, they can be employed to store energy for common electrical circuits, possess the capacity to charge and discharge quickly, can be utilized to meet peaks in demand for power, and can provide massive bursts of power for a short period of time. They lack electrochemical reactions, which means they have less operational wear and tear and a longer lifespan. They do not need to be replaced after hundreds of thousands of uses.

Due to their special properties, supercapacitors are used in many different applications, including electronics, thermoelectric, energy storage, and renewable energy in industry, vehicles, and robotics. In addition, supercapacitors are used in wind turbines, cellular base stations, electronic devices, and various industrial processes that require frequent charge/discharge cycles and high-power density [139–141].Due to their advantages over lead-acid batteries, they are now also used in UPS systems, electric cars, and a variety of power electronics applications. In addition, SCs have been used as energy storage devices for voltage regulation in renewable and hybrid energy storage systems for source and grid control [142–147]. SCs support stable power supplies and electronics tools in applications with variable loads and portable speakers. These applications include voltage stabilization, micro grid [148,149], high energy harvesting [150], renewable energy storage [151,152], high energy harvesting [153–155], street lights [156,157], medical applications [158–162], energy recovery in military [163–169], and energy recovery in automobiles and machines [170–176].

7.1. Electronics

The development of flexible electronics relies heavily on conductive polymer composites (CPCs) with excellent conductivity and flexibility. Flexible solid-state supercapacitors with high gravimetric-specific capacitances (80–200 Fg^{-1}) have been created using graphene or carbon nanotube-based thin films; however, these devices typically have relatively low overall or area-specific capacitances (3–50 mF/cm^2). Owing to their extremely low mass loading and extremely thin electrode thickness (usually a few micrometers), they are highly sought-after as a transportable power source for flexible electronics of the future [177]. In addition, Weng, Zhe, [178] fabricated flexible supercapacitors via graphene-cellulose paper (GCP), as shown in Figure 8. The electrical conductivity of the GCP membrane shows high stability, with a decrease of only 6% after being bent 1000 times, as shown in Figure 8B. This flexible GCP electrode has a high capacitance per geometric area of 81 mF cm^{-2}, which is equivalent to a gravimetric capacitance of 120 F g^{-1} of graphene and retains >99% capacitance over 5000 cycles.

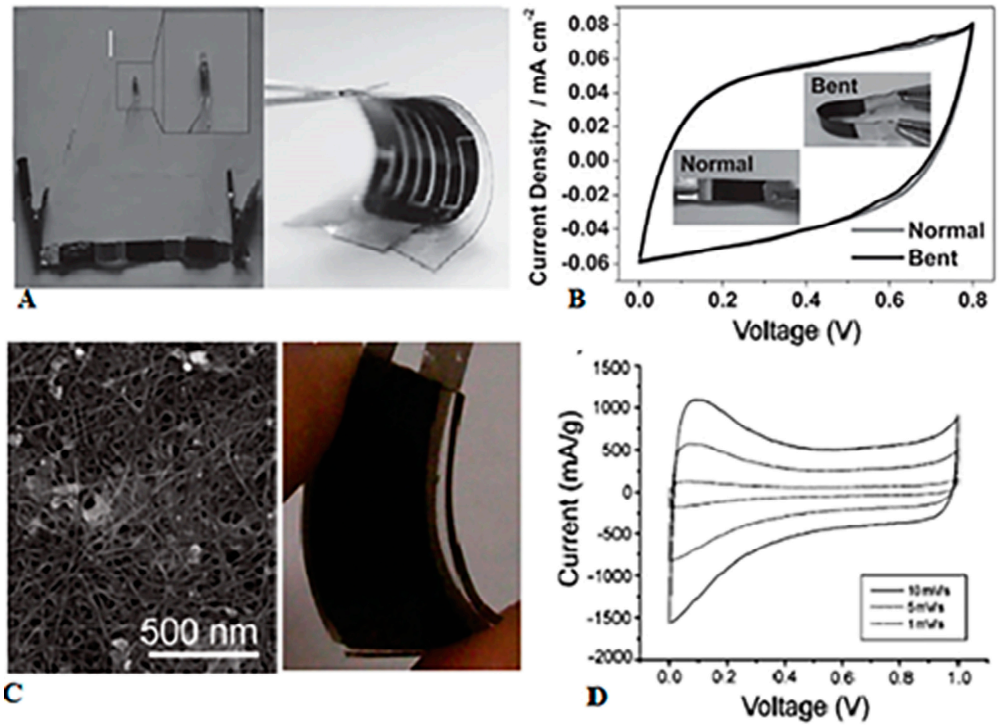

Figure 8. Flexible interdigital graphene cellulose paper SC (**A**), Current density before and after mechanical action (**B**) [178] Advanced energy materials BY—Creative Commons Attribution License. Carbon nanotube-based flexible SC (**C**), and its current density (**D**) ACS publishing, creative common attributes, https://creativecommons.org/licenses/by/4.0/ accessed on 19 December 2023.

The flexible composite polymer supercapacitor was used to manufacture soft electronics using printable single-walled carbon nanotubes and an organic liquid aqueous gel as an electrolyte. The performances of the devices show very high energy and power densities (6 W h/kg for both electrolytes and 23 and 70 kW/kg for aqueous gel electrolyte and organic electrolyte, respectively) [179]. From the result, it can be shown that these types of printable supercapacitors will lead to a new class of entirely printable charge storage devices, allowing for full integration with the emerging field of printed electronics. However, the long-term performance and its durability were not investigated in relation to external factors, namely, environment, mechanical abrasion, moisture, temperature, and pressure. Moreover, the most common application for ICP films with high conductivity is as flexible electrodes or even as flexible transparent electrodes in optoelectronic devices, which are manufactured using transparent and highly stretchable ICP such as poly(3,4-ethylenedioxythiophene) (PEDOT) and its derivatives [180]. The result shows that to acquire a high-quality signal for the long term, the ICP should always have good contact with skin or cells. Thus, they should be self-adhesive and have good stability in the biological environment. There are tremendous opportunities for ICPs in flexible electronics, but much more effort from materials scientists, chemists, physicists, and biologists is needed.

In order to enhance device performance in terms of electrical and mechanical qualities, a series of soft materials for flexible electronics have been developed using CPC for next-generation wearable electronic devices for the application of magnetism, photoluminescence, and electrochemical reactivity [181–183]. From the result, it can be concluded that the developed CPC for the application of wearable electronics and implantable healthcare areas should fulfill the characteristics of being fully biocompatible and/or biodegradable for inorganic elements and exhibiting high electrical conductivity.

7.2. Thermoelectric Application

The need for sustainable development in CO_2 minimization and the use of renewable energy sources and heat management systems is increasing, making the development of novel thermoelectric materials using conductive polymer composites an important topic for researchers in various fields [184]. Thermoelectric systems are particularly consistent, reliable, eco-friendly, and stable power sources, making them efficient in generating power from heat sources with low-temperature gradients in comparison to the ambient temperature. Thermoelectric materials receive increasing attention due to their unique capability of realizing the direct energy conversion between waste and low-grade heat and electricity potentials as sustainable and green technologies [185–187]. In addition, small molecules and polymers, including semiconductor nanomaterials like tellurium (Te), bismuth telluride (Bi_2Te_3), and lead telluride, can be combined with conductive polymers (PbTe). Furthermore, Dimethylsulfoxide (DMSO)-treated composite films of PEDOT:PSS and spherical PbTe could be blended with noble metals such as silver and gold (Au-Ag) nanomaterials and with carbon nanomaterials for the application of thermoelectric [188].

Various researchers studied conductive polymer composite-based thermoelectric for application to wearable thermoelectric devices. For example, researchers [189,190] developed and studied the thermoelectric effects of a conductive composite made from PPy and silver into a thin film through a simple, nature-friendly optical-chemical process. The result shows that, by changing the concentration and size of Ag particles in the PPy substrate, σ was increased from 1.5 S cm^{-1} to 17.3 S cm^{-1}, and κ was decreased to 0.16 W $m^{-1}K^{-1}$. As a result, the thermoelectric potential of this material reached the maximum value of ~7.4 × 10^{-3} at 355 K and exhibited high thermoelectric stability after repeated bending.

The performance of polymer-based thermoelectric generators is measured using Seebeck coefficient and power factor calculations, and the thermoelectric generator is demonstrated as shown in Figure 9.

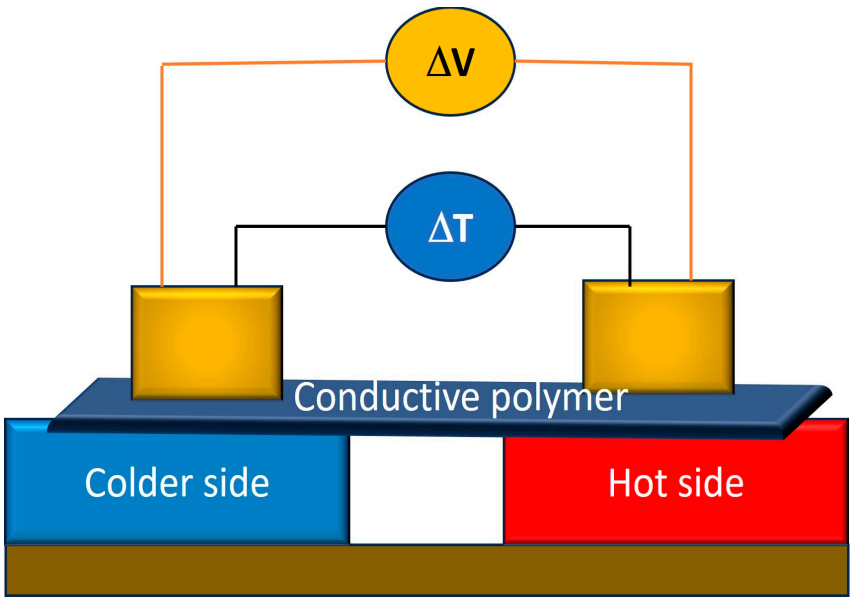

Figure 9. The schematic diagram of the thermoelectric device and its measurement setup.

Seebeck coefficient is calculated as S = ΔV/ΔT (Figure 9) where ΔV is thermovoltage difference and ΔT is designated as temperature difference. Optimum electrical conductivity and Seebeck coefficient have meaningful correlations.

Xinyu Yang [191] studied an ionic thermoelectric supercapacitor, which is manufactured from NaCl-polyacrylamide/Sodium carboxymethyl cellulose (NaCl-PMSC) electrolyte and carbon nanotube-polyacrylamide (CNT-PAM) electrodes that relies on the synergistic functions of thermoelectricity and supercapacitors in the thermoelectric ion gel

electrolyte and high-performance hydrogel electrodes to enhance the ECS performance under a thermal gradient. The result shows that the TE ion gel electrolyte shows the maximum Seebeck coefficient of 17.1 mV K^{-1} by adjusting the Na^+ content in the ion gel. The integrated thermoelectric supercapacitor was capable of outputting a voltage of ~450 mV at ΔT 30 K and storing a charge of ~1.3 mC at ΔT 10 K.

Kyungwhan Yang et al. [192] developed a thermoelectric generator-coupled microsupercapacitors (TEG-MSC) consisting of planar microsupercapacitors linked directly to the thermoelectric pn modules of p-Ag_2Te and n-Ag_2Se nanoparticle thin films. In the TEG-MSC, a Seebeck voltage of 82 mV is generated at a temperature difference of 15.8 K and is rapidly charged with an efficiency of 98%.

7.3. Energy Storage

Supercapacitors made of conductive polymer composites have a lot of potential for fast charging and energy storage. Single supercapacitors may store a lot more energy than a standard capacitor with a solid dielectric. Electronically conductive polymer composites, such as PEDOT-PSS, were used as the electrodes of transparent supercapacitors and as high-capacity energy storage devices [193,194]. Supercapacitors are an example of an energy storage and delivery technology that can store and deliver energy quickly and provide high current for a short period of time. Because of their high specific surface area and excellent energy storage, carbon and carbon-based materials have become more popular as supercapacitor electrode materials [195–199]. From Figure 2, the supercapacitor had high energy storage and a long cycling life. Moreover, the specific capacitance of supercapacitors built of composites of NiMnCo ternary oxide and PEDOT-PSS can reach 1234.5 F/g^{-1} at current densities of 1 A/g^{-1} and an energy density of 51.9 Wh/kg^{-1} at power densities of 275 W/kg^{-1}, respectively [200]. A study by Astha Shrivastava et al. [201] describes the application of supercapacitors for photovoltaic systems in the automotive and avionics sectors. Energy storage devices are constructed in such a way that two electrodes (negative and positive electrodes) are separated by an insulator where an electrolyte is incorporated between the two electrodes (Figure 10).

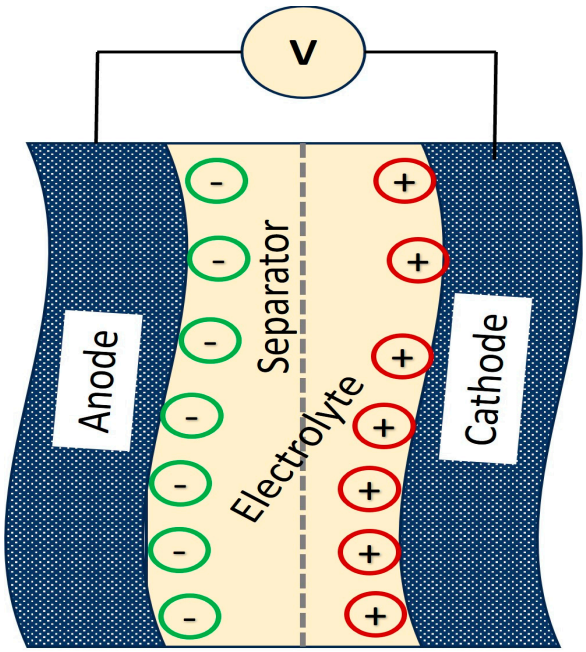

Figure 10. The schematic diagram of energy storage materials (supercapacitors) and their setup.

Energy storage is a critical component of modern energy systems, providing the capability to store and release energy for later use. Various technologies are employed for energy storage, including batteries, pumped hydro storage, flywheels, and capacitors.

These systems play a crucial role in balancing the intermittent nature of renewable energy sources, such as solar and wind, by storing excess energy during periods of high generation and releasing it during times of high demand or when renewable sources are not actively producing [202]. Effective energy storage is vital for enhancing grid reliability, improving the efficiency of energy utilization, and facilitating the integration of renewable energy into the power grid. As the demand for sustainable and resilient energy solutions grows, ongoing research and development in energy storage technologies aim to improve efficiency, decrease costs, and expand the overall capacity of energy storage systems [203].

7.4. Smart Textiles

The recent rapid development of flexible and wearable electronics requires flexible and portable supercapacitors, which are foldable, stretchable, and/or bendable, with smart functions and long-time outdoor operation, as the power source wearable [204–208]. Due to their high flexibility, conductivity, pseudocapacitance, smart characteristics, and moderate preparation conditions, supercapacitors were widely applied. In addition, fiber-shaped supercapacitors have the advantages of being lightweight, highly flexible, soft, low-cost, highly flexible, can be transformed into any shape, and can be manufactured using different approaches [209]. These fiber-shaped supercapacitors were used for wearable applications and are considered to be one of the most promising power source candidates [210–212]. Stretchable electrodes for engineered tissues based on conductive polymer composites can also be created by adding metal or carbon-based nanoparticles to elastomers like polydimethylsiloxane (PDMS) or styrene-butadiene-styrene (SBS) [213]. In addition, Marjan Barakzeh et al. [214] developed and studded a textile-based electrode through modification with reduced graphene oxide (rGO) nanosheets and polypyrrole (PPy) nano spherical particles onto polyethylene terephthalate (PET) fabric. The conductive composites are used to fabricate flexible, all-solid-state supercapacitors using a gel electrolyte that was prepared by dissolving 1 g of PVA in 10 mL of DI water at 90 °C.

Moreover, flexible and wearable electronics have many potential applications in daily life, ranging from body-worn entertainment to protection, sensing, communication, therapy, and electromechanical sensors [215,216]. For instance, a stretchable fiber-shaped synthetic composite with other materials, which include carbon materials and transition metal oxides and hydroxides, was used to manufacture wearable electronics that were stretchable up to 400% without obvious capacitance degradation. These devices had a high-voltage output of 12.8 V and an ultrahigh energy density of 41.1 μWhcm^{-2} at a power density of 3520 μWcm^{-2} [217–219].

According to researchers [220–222], carbon-based yarn/fiber-shaped and planar electrodes with tiny volumes and promising performances have gained a lot of attention as flexible electrodes. These electrodes can be used to fabricate flexible and wearable pseudocapacitors. The materials used to create fiber-shaped electrodes (1DSCs) include metal wires, carbon material-based fiber, cotton fiber, Kevlar fiber, and polymer nanofibers, as shown in Figure 11. This stores electrical energy through electrochemical double-layer capacitance (EDLCs) by reversible adsorption of electrolyte ions at the surface or inside pores of electrodes and pseudocapacitance through surface faradaic redox reactions or ion intercalation at electrode surfaces [223].

In addition to the previously mentioned novel electric double-layer capacitor with a coaxial fiber structure (Figure 11), a wire-shaped EDLC woven into electronic clothes by the well-developed textile technology [224] shows promising applications in a wide variety of fields. However, the high contact resistance between two twisted fiber electrodes has largely decreased the energy storage capability. The result shows that the developed 1D supercapacitor had high electrochemical performance, with a maximum discharge capacitance of 59 F g^{-1} (32.09 F cm^{-3} or 29 μ F cm^{-1} or 8.66 mF cm^{-2}). Further, wire-shaped EDLCs were not stretchable in the straight wire state, so the resulting electronic textiles based on the wire-shaped EDLC will break during mechanical action such as tensile force, bending force, and mechanical abrasion [225]. To overcome this problem, researchers

use a transition-metal dichalcogenide as the active material. For example, Ma et al. [226] used MoS_2, a transition-metal dichalcogenide, as the active material.

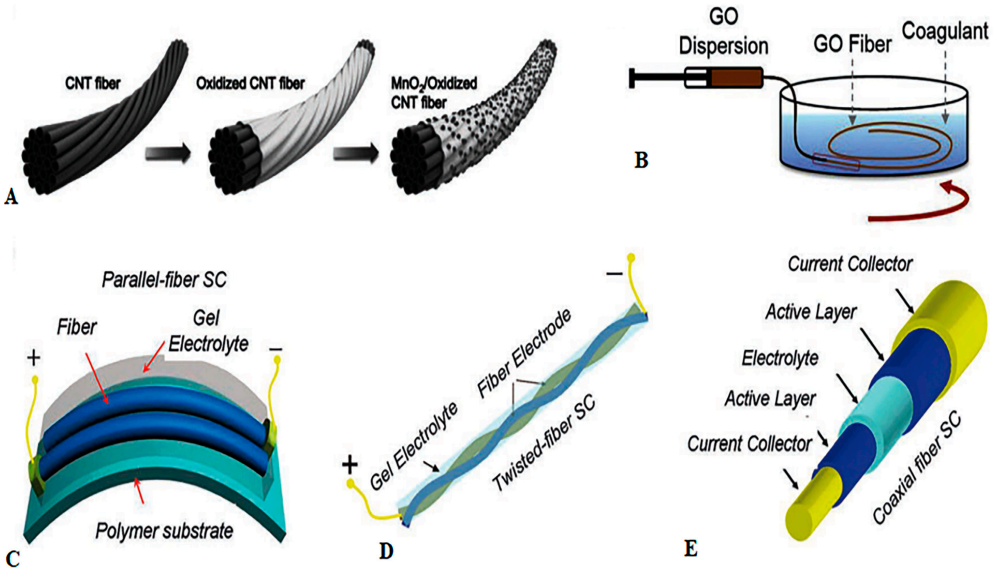

Figure 11. Deposition of nanostructured MnO2 on oxidized CNT fibers (**A**), configurations of 1D SCs, wet spinning of GO nanosheet dispersion into freestanding GO fibers to obtain fiber/yarn electrodes (**B**), parallel (**C**) two-ply (**D**), and coaxial types (**E**) [220] permission on Advanced Material journals: Creative Commons Attribution 4.0 International (CC-BY-4.0).

The materials exhibit significant promise in the creation of flexible supercapacitors due to their huge surface area, high conductivity, favorable mechanical characteristics, and great stability. Unfortunately, carbon-based materials have poor specific capacitance due to the intrinsic EDLC mechanism, which is a major barrier to raising the energy density of supercapacitors. Surface modification was an effective method for raising the energy density of these types of supercapacitors. For example, surface activation and N-doping or S-doping approaches can effectively increase the capacitance of carbon-based materials [227]. Generally, N-doping uses the nitrogen sources of nitrogen gas, ammonium salts, urea, and ammonia gas, whereas S-doping deals with different sulfur sources, including thiourea, benzyl disulphide, sulphates, methionine and sulphuric acid temperatures [228].

7.5. Biomedical

Biomedical implantable devices mostly require micro power sources with tiny sizes and enough power supply [229]. Implantable medical devices have improved the treatment of several chronic diseases. For example, modern cardiac pacemakers can monitor and control the patient's heart function and report critical events to hospital control centers. However, powering such implanted devices with batteries creates problems because these devices must be replaced when the battery is drained [161]. This necessitates that the patient undergoes painful surgery at a significant expense. Therefore, a few researchers are exploring battery-free implantable devices by harvesting energy directly from the human body. Recently, researchers have successfully developed an implantable nanogenerator in a living rat that works by extracting energy from its periodic breathing. This energy was used to power a prototype pacemaker [230–233]. A tiny supercapacitor as an energy source used for medical implants and healthcare monitoring devices was manufactured using different methods, such as by doping poly (3,4-ethylenedioxythiophene): poly(styrene sulfonate) composite with a supramolecular solvent (β-cyclodextrin and citric acid), a soft, self-adhesive conductive polymer [234]. In addition, there was also a biodegradable and nontoxic food material-based supercapacitor for biomedical applications with a primary

battery and biosensors using an implantable triboelectric nanogenerator [235]. Furthermore, one of the key areas of research to address the increasing ecological problems associated with the issue of electronic waste is the development of environmentally friendly and biodegradable electrical components, as shown in Figure 12. This energy storage unit is made up of biodegradable Zn-ion hybrid supercapacitors with an anode made of zinc foil, a cathode made of molybdenum sulfide (MoS_2) nanosheets, and an electrolyte made of ion-crosslinked alginate gel. The high capacitance of 93.5 mF cm^2 and an output voltage of 1.3 V are achieved with this architecture. These types of biodegradable supercapacitors are also highly desirable for biomedical applications such as integrated bioelectronics, which also call for biocompatibility. However, in the case of wireless power transfer, biodegradable supercapacitors, low energy density, short lifetimes due to self-discharge, limited capacity, and limited continuous power supply capability were mentioned as drawbacks.

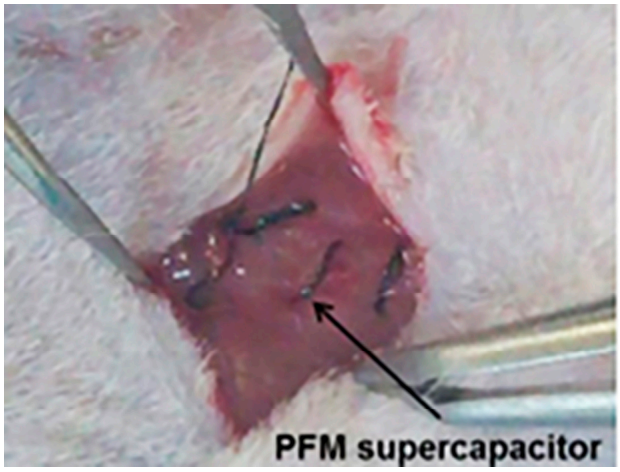

Figure 12. Biodegradable supercapacitors for medical implants [233]. Permission on ACS applied materials creative commons attribution 4.0 international (cc-by-4.0).

8. Challenges for Conductive Polymer Composite Supercapacitors

As previously mentioned, CPC supercapacitors offer many advantages over batteries and fuel cells. Even so, they also face challenges at the current stage of technology. The major disadvantage of conductive polymers when used as supercapacitor electrodes is their poor cycle life [131], low energy density [236,237], high cost [238], high self-discharging rate [239], and challenges regarding the mass production of CNT-based flexible supercapacitors should also be overcome. The fabricating process with high-throughput synthesis should be held on a large scale, allowing commercial application. In addition, insolubility and intractability remain the major challenges obstructing the use of CPs in energy storage; other problems include diffusion issues, cycling, and stability [61]. Furthermore, in current practical applications, flexible supercapacitors still face many challenges in wearable electronics, making them unsuitable for industrial production, in addition to safety issues related to the development of flexible capacitors. Since it is used close to the skin, biocompatible materials are one of the best choices for constructing supercapacitors [240].

Research conducted by S. R. Sivakkumar [241] has shown that conductive polymer composites swell and contract substantially on charge and discharge, respectively. This volume change, or swelling, causes mechanical failure of the electrode under prolonged cycling. Consequently, cycle life is poor compared with carbon-based supercapacitors, which generally only charge via adsorption and desorption of ions (typically giving a few thousand cycles for conductive polymers compared with >500,000 cycles for carbon-based devices). This draw back was solved by Mengting Liu [242], who developed novel non-woven Al_2O_3 nanowire polyvinyl butyral (PVB) membrane separators with highly porous networks that demonstrated tensile strength of >30 MPa, extremely high electrolyte ab-

sorption (>200 wt%), low-to-no swelling behavior, and stable electrochemical performance substantially exceeding that of analogous cells with commercial separators.

9. Outlook, Future Perspectives and Conclusions

Conductive polymers offer various benefits as supercapacitor electrodes. They are flexible, highly conductive, easy to process, have high charge storage, and have a low resistance to electroactive materials to achieve high capacitance performance. Many conductive polymers exhibit high specific capacities and capacitances while being able to deliver energy at a relatively rapid rate. The major disadvantage of conductive polymers, when used as supercapacitor electrodes, is their poorer cycle life than those based on carbon.

Several researchers are currently working on the further development of supercapacitors made of conductive polymer composites. Recent studies indicate that the annual growth rate for the development of reliable, high-capacity supercapacitors will skyrocket over the next few years. Demand for increasingly advanced and electroconductive polymer composite supercapacitors has increased as technology has developed. Flexible conductive polymer composite supercapacitor design and development issues have frequently been solved using unique techniques created by several researchers. This review clearly shows the conductive polymers and composites for supercapacitor applications in various fields. The fabrication and conductivity mechanisms of conductive polymer supercapacitors are explained. Overall, the application of the conductive polymer composite discussed above led to the development of a broad range of flexible, reliable, and high-capacity conductive polymer supercapacitors with full performance. Current challenges remain in ensuring consistent performance, achieving satisfactory energy density, and developing suitable cathode materials and electrolytes. In particular, the electromechanical performance of a conductive polymer-based supercapacitor can store huge amounts of electricity, have a long shelf life without deteriorating, and have extremely low energy waste.

In general, for the future, the design and development of conductive polymer composite supercapacitors should be environmentally friendly and use a sustainable green electrode material that is easily recyclable and has zero waste.

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