crystals

MDPI

Review

An Overview of Active Electrode Materials for the Efficient High-Performance Supercapacitor Application

Bharathi Arumugam [1], Gopiraman Mayakrishnan [2], Suresh Kumar Subburayan Manickavasagam [3], Seong Cheol Kim [1,*] and Ramkumar Vanaraj [1,*]

[1] School of Chemical Engineering, Yeungnam University, Gyeonsan 38541, Republic of Korea; bharathi250698@gmail.com
[2] Nano Fusion Technology Research Group, Institute for Fiber Engineering (IFES), Interdisciplinary Cluster for Cutting Edge Research (ICCER), Shinshu University, Tokida 3-15-1, Ueda 386-8567, Nagano, Japan; gopiramannitt@gmail.com
[3] Department of Chemistry, Dhanalakshmi Srinivasan Engineering College (Autonomous), Perambalur 621212, India; smsureshkumar2020@gmail.com
* Correspondence: sckim07@ynu.ac.kr (S.C.K.); srirams27@gmaiil.com or ramkumar@yu.ac.kr (R.V.)

Abstract: Recent energy research focuses on the efficiency enhancement of supercapacitor devices for multipurpose applications. Several materials have been used as electrode materials to achieve the maximum specific capacitance. The present review article concludes with three different types of materials recently used to enhance the efficiency of supercapacitors. The first type involves carbon-based materials for storage and supercapacitor applications. The carbon materials could be obtained naturally and synthesized manually based on need. The additional advantage of carbon material is these materials can be obtained from natural sources. The second type discusses the recent advances in metal oxide materials for high-performance supercapacitors. The metal oxide materials are involved in different types of attachment through the bi-tri metallic bonding, which enhances the specific capacitance. The third type involves recently advanced materials for high energy and power density application. The power and energy density of the materials is enhanced by the surface modification of the materials. In recent days, the MXene and nanocomposite materials seem to be an appropriate material to increase the power and energy density of the device. The modification and surface treatment of respective materials could enhance the specific capacitance of the material.

Keywords: supercapacitor; carbon; metal oxides; MXene; power; energy density

Citation: Arumugam, B.; Mayakrishnan, G.; Subburayan Manickavasagam, S.K.; Kim, S.C.; Vanaraj, R. An Overview of Active Electrode Materials for the Efficient High-Performance Supercapacitor Application. *Crystals* **2023**, *13*, 1118. https://doi.org/10.3390/cryst13071118

Academic Editors: Faxing Wang and Yusuf Valentino Kaneti

Received: 19 June 2023
Revised: 6 July 2023
Accepted: 13 July 2023
Published: 18 July 2023

1. Introduction

The industrial revolution, rising per capita consumption, and population growth have all contributed to a sharp rise in global energy demand [1–4]. Around 80% of the world's energy production comes from conventional or nonrenewable sources like coal and petroleum. Overuse of these fossil fuels harms both the environment and global economies. Burning coal and petroleum-based products releases harmful gases like CO_2, CH_4, and N_2O into the atmosphere, which has detrimental effects on both the environment and human health. Research into using eco-friendly and renewable energy sources has been intense due to the shortage and environmental issues caused by the use of fossil fuels. In response to the increase in energy demand, the global science and technology community is developing technologies and products for converting and storing energy. In order to achieve a balance between economic growth and environmental responsibility, researchers and academics are searching for novel, functionalized advanced nanomaterials [5–7].

According to a power survey, there is insufficient energy storage, which results in the loss of about 30% of the power that is generated. To solve these issues, it is crucial to create high-performance, substantial energy storage systems [8,9]. Pumped hydroelectric storage is one alternative energy storage technology. Compressed air storage is another. Flywheels offer various energy storage options, including hydrogen, thermal, superconducting magnetic, and electrochemical, which are increasingly used in industrial and automotive applications. (ECs) [10,11]. Research on novel storage devices is being encouraged by the fact that batteries are favored for applications that need a high energy density, but electrolytic capacitors are favored for applications that require a low energy density [12]. Future generations of energy storage devices will likely favor supercapacitors because of their superior specific capacitance, long lifespan, safety, quick charge-discharge rate, excellent circulation features, high power density, and low cost [13–17]. In a supercapacitor, the type of electrolyte and electrode surface area determines how much charge can be stored overall, whereas the size of the electrode and the total active mass are the limiting factors for the total charge that can be stored in a battery. Supercapacitors work similarly to regular capacitors. Because their electrodes have a huge surface area, supercapacitors have a larger capacitance (F/g, F/cm^2) than regular capacitors. With a long life cycle of about 500,000 cycles, supercapacitors can store between 10 and 100 times as much energy per unit volume as other rechargeable batteries [9,18]. Supercapacitors offer a wider capacitance range (12,700 F), lower equivalent series resistance (1/10th of electrolytic capacitor), and long-cycle stability [19]. Supercapacitors link conventional capacitors and secondary ion batteries, as shown in Figure 1 [20].

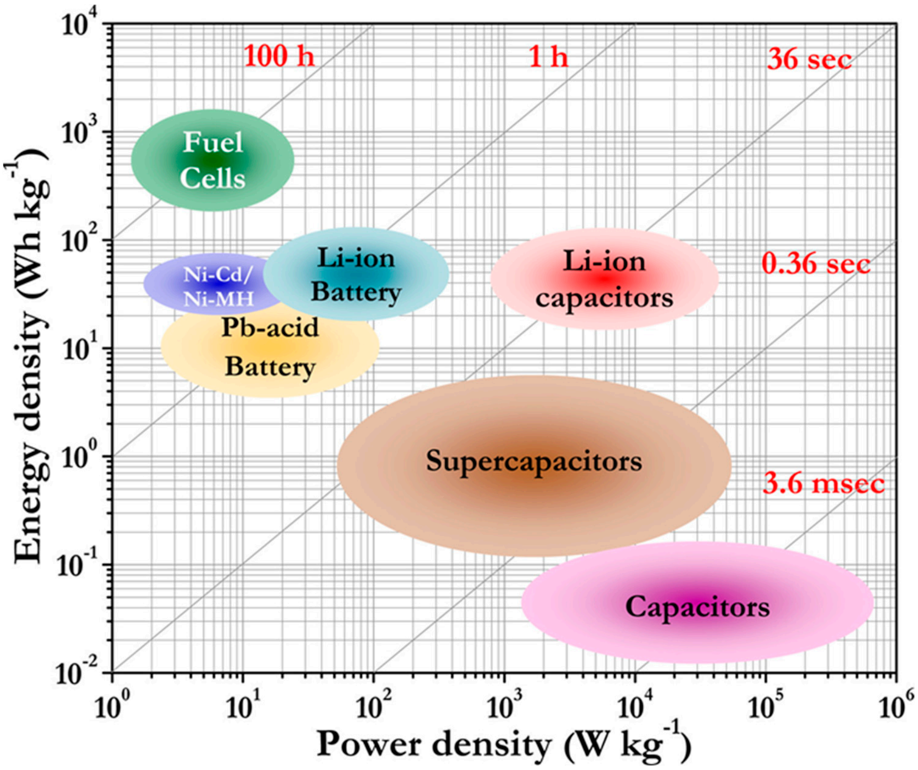

Figure 1. The need for high–power/high–energy electrochemical energy storage devices is demonstrated by the Ragone plot (Reprinted with permission from Ref. [20]. 2014, American Chemical Society).

The energy density of supercapacitors may be improved by the use of new electrolytes and electrode materials. Active materials such as carbon nanotubes, graphene, transition metal oxides, and conducting polymers are examples of such materials. Devices that use supercapacitors are able to make use of a wide variety of electrolytes, including those that are aqueous, non-aqueous, polymer-based, and ionic liquid-based [8]. Active electrode materials significantly impact supercapacitor performance in energy, power density, capacitance, and stability. High-specific capacitance materials like activated carbon or transition metal oxides significantly increase energy density, while high electrical conductivity is crucial for achieving high power density. The typical supercapacitor provides high power density, while the battery devices provide high energy density. The devices would be prepared from various types of materials based on the requirement [16,21,22].

The exact surface area and hole structure of the active electrode material determine its capacitance and stability. The redox properties of the active electrode materials determine the electrochemical performance of supercapacitors. Pseudocapacitive materials, such as metal oxides, hydrogens, or conducting polymers undergo fast and reversible faradaic reactions at the electrode-electrolyte interface, enabling higher energy storage capacities than non-faradaic processes. The choice of active electrode materials affects the specific redox reactions, reversibility of the reactions, and charge transfer kinetics, thereby impacting the overall electrochemical performance of the supercapacitor. Active electrode materials significantly influence the cycling stability and lifespan of supercapacitors. Robust electrode materials with good mechanical stability and chemical resistance are necessary to ensure long-term performance and retain the supercapacitor's capacitance over a large number of cycles.

2. Classification of Supercapacitor Based on the Energy Storage Mechanism

Supercapacitors are made up of four components: two electrodes, an electrolyte, and a separator, all of which are soaked in the electrolyte. As can be seen in Figure 2, supercapacitors may be divided into three distinct categories according to the manner in which they store energy; electrochemical double-layer capacitors (EDLCs), pseudocapacitors, and hybrid systems are all types of electrochemical storage devices. The process that EDLCs use to store energy is predicated on the electrostatic interaction that occurs between ions that are located on the significant specific surface area of active electrode materials and electrolytes. The mechanism that allows pseudocapacitors to store energy is a Faradaic redox reaction that occurs close to the surface between the electrode materials and the electrolyte ions. This reaction is both quick and reversible. Pseudocapacitors typically have a higher capacitance per gram than EDLCs, but their kinetics are slower. This is due to the fact that the process of storing energy in a pseudocapacitor takes place both in the bulk of the electrode materials and on their surface, whereas the charge/discharge process only takes place on the surface of an EDLC. Hybrid EDL and Faradaic processes work together to generate a synergistic combination of EDL and pseudocapacitance, which results in high energy density and better cycle stability in supercapacitors. Supercapacitors are a mix of EDL and Faradaic mechanisms [23].

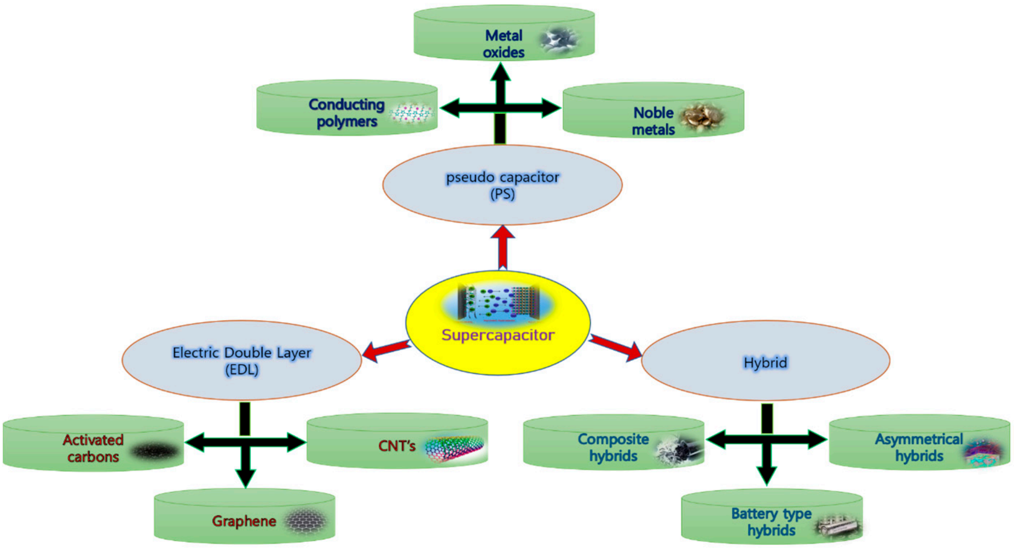

Figure 2. Classification of supercapacitor based on mechanism.

2.1. Electrochemical Double-Layer Capacitors (EDLCs)

EDLCs rely on an electrostatic double layer being formed between the electrode and the electrolyte in order to store energy. When a voltage is placed between two electrodes, ions from the electrolyte are drawn to the electrode surface, forming a double layer of charged species. The storage of energy results from this charge separation. The high-power density of EDLCs makes them capable of delivering and absorbing large amounts of power quickly [24–27]. Rapid charge and discharge rates are made possible by the absence of chemical reactions during charging and discharging, which makes EDLCs ideal for applications requiring quick energy delivery.

In comparison to other energy storage technologies like batteries, EDLCs typically have a lower energy density. The electrode's surface area and the capacitance of the double layer are the main determinants of energy storage. The goal of ongoing research is to create materials and designs that will increase the energy density of EDLCs.

The capacitance of a typical capacitor may be increased by increasing the material's dielectric constant, increasing its surface area, or decreasing its inter-planar thickness [28,29]. However, such an improvement is possible with further modification of the material system and capacitor design. For instance, the material's exceptional electrochemical performance may result from reducing the particle size to the sub-nanometer range. Increasing the capacitance of an electrode material by doping it with metal ions (Fe, Mn, Cr, and Co) improves its ability to conduct electricity. For instance, a capacitor with symmetric electrodes or a working principle based on faradaic reactions has the potential for improved electrochemical performance, making it a supercapacitor. Their non-faradaic electrical processes make them highly reversible, and their cycling stability is exceptional, reaching up to 106 cycles or more. The introduction of ionic, conductive electrolytes has mitigated EDLCs' primary drawback—the scarcity of electrode materials. Carbon content categorizes EDLCs into three broad categories, each of which serves a distinct purpose in the device. The most notable characteristics include carbon nanotubes (CNTs), graphene, and activated carbon [30]. The increased capacitance of these materials may be attributed to their porous nature, which provides a large surface area for the creation of the double layer. EDLCs are able to sustain a great number of charge-discharge cycles because of their high cycling stability. Because no chemical interactions are necessary for energy storage in EDLCs, they are very reliable and durable. Carbon nanotubes (CNTs), graphene, and activated carbon are the most important features.

2.2. Pseudocapacitors

Pseudocapacitance increases as a result of the electrolyte and electrode surface's quick oxidation and reduction [31]. In redox reactions, reactants experience an alteration in the oxidation state. An object is said to be oxidized when an electron has been removed from it, whereas an object has been reduced when an electron has been added. Pseudocapacitors made through this faradaic process have an increased energy density over EDLCs. The size of the electrode's surface areas and the kind of electrolyte used determine how much pseudocapacitance there is. During the Faradaic process, electrons created by the redox reaction are transmitted across the electrolyte-electrode contact, resulting in an increase in electron concentration at the electrode [32,33]. Theoretical metal oxide pseudocapacitance can be calculated using the following formula:

$$\mathrm{C} = \frac{\mathrm{n} \times \mathrm{F}}{\mathrm{M} \times \mathrm{V}}$$

where n represents the average number of electron transfers during the redox process, The Faraday constant is given by F, and the molar mass of the operating voltage window, and V is the metal oxide.

As shown in the following formula, a supercapacitor's energy density (E)

$$E = \frac{CV^2}{2}$$

The formula for calculating a supercapacitor's maximum power density is

$$\mathrm{Pmax} = \frac{V^2}{4R}$$

where R denotes the device's total equivalent series resistance.

Titanium sulfide (TiS_2), ruthenium oxide (RuO), nickel oxide (NiO), and manganese oxide (MnO_2) are all examples of transition metal oxides with high pseudocapacitance. The electrochemical performance of transition metal oxide supercapacitors is considerably improved by their high specific power and electrochemical stability. Due to their low conducting resistance, low bulk, and low cost, as well as their high energy storage capacity and high pseudocapacitance value, conducting polymers have emerged as appealing electrode materials for supercapacitors. Electrode materials might be any metal oxide or noble metal [34], and conducting polymer, including polypyrrole, polyacetylene, poly (phenylenevinylene), polythiophene, polyaniline, etc. However, the life cycle and power density of pseudocapacitors are also shorter. These outcomes are the result of redox reactions occurring in the capacitors.

2.3. Hybrid

To create a hybrid supercapacitor, a single supercapacitor can simultaneously produce EDLC and pseudocapacitance. Hybrid super-capacitors, which can store charges in both Faradaic and non-Faradaic fashions, and are capable of greater energy and power densities and superior cycle stability. For instance, in a single supercapacitor, the non-Faradaic charge-discharge process occurs on one electrode, while the redox reaction takes place on a different electrode. Composite hybrid [35–37], battery-type hybrid [38], and Asymmetric hybrid [39–41] fall under this category.

2.4. Performance Parameters of Supercapacitor Electrode Materials

The main performance indicators of supercapacitors are the specific capacitance (normalized by electrodes mass, volume, or area), density of energy, capacity for power, rates efficiency (retained capacitance at a high current loading), and cycle stability [42]. The energy density and power density of a supercapacitor may be improved by increasing its specific capacitance (C), expanding its working voltage window (V), and decreasing its equivalent series resistance (R). When it comes to EDLC supercapacitors, the electrolyte's stability is the primary limiting factor in the maximum working voltage window (Vm). For supercapacitors based on aqueous electrolytes, Vm is typically 1 V. The creation of non-aqueous electrolytes with high Vm is one of the current research trends in supercapacitors. For instance, an electrolyte based on an ionic liquid can be operated at a Vm as high as 3.5 V [43].

- √ Essential for rate capability and power density is high electronic conductivity;
- √ Increased specific surface area, which controls the specific capacitance;
- √ Low costs for production and raw materials;
- √ Ideal electroactive locations that allow for pseudocapacitance;
- √ High thermal and chemical stability have an impact on cyclic stability;
- √ Rate capability and specific capacitance are impacted by controlled porosity.

These are all the characteristics that an ideal electrode should have [44].

In this review, we have discussed the most recent developments in electrode materials in supercapacitors. This paper describes the three materials used to improve supercapacitor efficiency: materials with high energy and power densities, which include materials made from carbon for storage spaces; metallic oxide materials for top-performance applications, and advanced materials. MXene and nanocomposite materials are suitable for increasing device power and energy density, which makes them perfect for supercapacitor electrodes, as well as methods for producing the materials with appropriate performance.

3. Carbon-Based Materials for the High-Performance Supercapacitor

Researchers are putting in a lot of time and energy into finding new and improved materials to use as solar cells' electrodes and electrolytes. Energy storage electrodes for supercapacitors are typically made out of carbon due to its many desirable properties (low production cost, small size, high availability, large surface area, morphology that can be tuned, intercalation of electrolytes within its high porosity, and high electrical conductivity, to name a few) [45,46]. Post-treatment of carbon materials allows for easy modification of their frameworks and chemical and mechanical properties, hence, further enhancing supercapacitor usefulness and performance. All of these reasons explain why carbon or activated carbon (AC) is the standard active electrode material for supercapacitors today [47,48]. AC is a popular electrode material with a large surface area, good electrical characteristics, and a reasonable price. It can be produced from various carbonaceous materials, either physically or chemically. Physical activation involves heating carbon precursors to high temperatures, while chemical activation uses substances like NaOH, KOH, $ZnCl_2$, and H_3PO_4. AC has well-developed surface areas of up to 3000 m^2 g^{-1} and physiochemical characteristics which differ depending on activation procedures and carbon precursors employed [49,50]. Specific capacitance and specific surface area (SSA) have been found to differ in AC, with a high SSA of 3000 m^2 g^{-1} resulting in a low capacitance. This implies that not all pores are productive when the charge is accumulating. Performance of electrochemical reactions in carbon materials is also influenced by additional factors, such as pore size distribution. Large pore volumes, poor conductivity, and low material density are caused by excessive activation, and these factors lower the energy density and increase power loss [51,52].

Carbon nanotubes (CNTs) have revolutionized the engineering and scientific study of carbon materials due to their unique pore arrangement, mechanical and thermal resilience, and superior electrical characteristics. Through the catalytic decomposition of hydrocarbons, CNTs are created, enabling the creation of nanostructures in different conformations and managing their crystalline structure [49]. They are highly regarded for their electrical conductivity, support for active materials, and high-power electrode materials. Compared to AC, CNTs have a small surface area and a low energy density. Using potassium hydroxide for chemical activation can enhance specific capacitance.

When compared to other carbon materials utilized as electrodes in electrochemical double-layer capacitors, the newly created graphene has the largest specific surface area (SSA), at roughly 2630 m^2 g^{-1} [53–55]. Graphene can reach a capacitance of up to 550 F g^{-1} if the entire SSA is utilized. The fact that both of the graphene sheet's major surfaces are exposed to the outside and are easily accessible to the electrolyte is another advantage of using graphene as an electrode material. Chemical vapor deposition (CVD), micromechanical exfoliation, the arch discharge approach, unzipping of carbon nanotubes (CNTs), epitaxial growth, electrochemical and chemical techniques, and incorporation in graphite are all viable methods for producing graphene [56–58].

3.1. Bio-Derived Carbon

Unlike biomass carbon, which comes from renewable sources like plants, most activated carbon comes from conventional fossil fuels like coal, petroleum, and their derivatives [59–61]. The macroporous, mesoporous, and microporous distributions of biomass, as well as its easily modifiable surface functional groups allow for a more logical three-dimensional hierarchical porous structure. More storage sites for ions and sites for generating pseudocapacitance through redox reactions; both essential in supercapacitors. It has been suggested so far that biomass can be converted into clean renewable energy systems because of a variety of applications [62–64]. Initial research efforts were concentrated on carbon produced synthetically from biomass. Energy storage systems that make use of the structural diversity of carbon extracted from biomass have also been developed. While Toupance places an emphasis on the usage of biomass-based carbon in supercapacitors and hybrid solar cells, Pan is more concerned with the design of biomass-based carbon structures. When presenting carbon derived from biomass or making references to particular applications, these reviews emphasize synthetic techniques. To give a comprehensive overview on synthetic techniques, multiple dimensions, and applications in electrocatalysis, energy conversion, and storage of carbon obtained from biomass is, therefore, still hard and vital.

In addition, biomass is a sustainable and renewable source of carbon, making it a better choice for the environment than more conventional carbon sources like coal and petroleum. Additionally, a variety of natural sources, such as agricultural waste, forestry residues, and municipal solid waste can be easily sourced for biomass (Figure 3). Due to its distinct features and advantageous effects on the economics and the environment, biomass porous carbon is a viable material for high-performance supercapacitors and other energy storage devices. The economic impact of preparing biomass carbon is magnified by the byproduct of biomass oil. Its three-dimensional porous structure provides it with exceptional thermochemical stability and a large specific surface area. It can be readily tailored to specific uses because of its many surface functional groups, which may be altered by adding or removing oxygen, nitrogen, sulfur, or phosphorus. Alkali and alkaline earth metals, which are found in most biomass materials, speed up the activation reaction process and offer an abundance of ion-binding sites for supercapacitors. Therefore, biomass porous carbon is a promising material for use in energy storage and conversion applications [57,58,65,66].

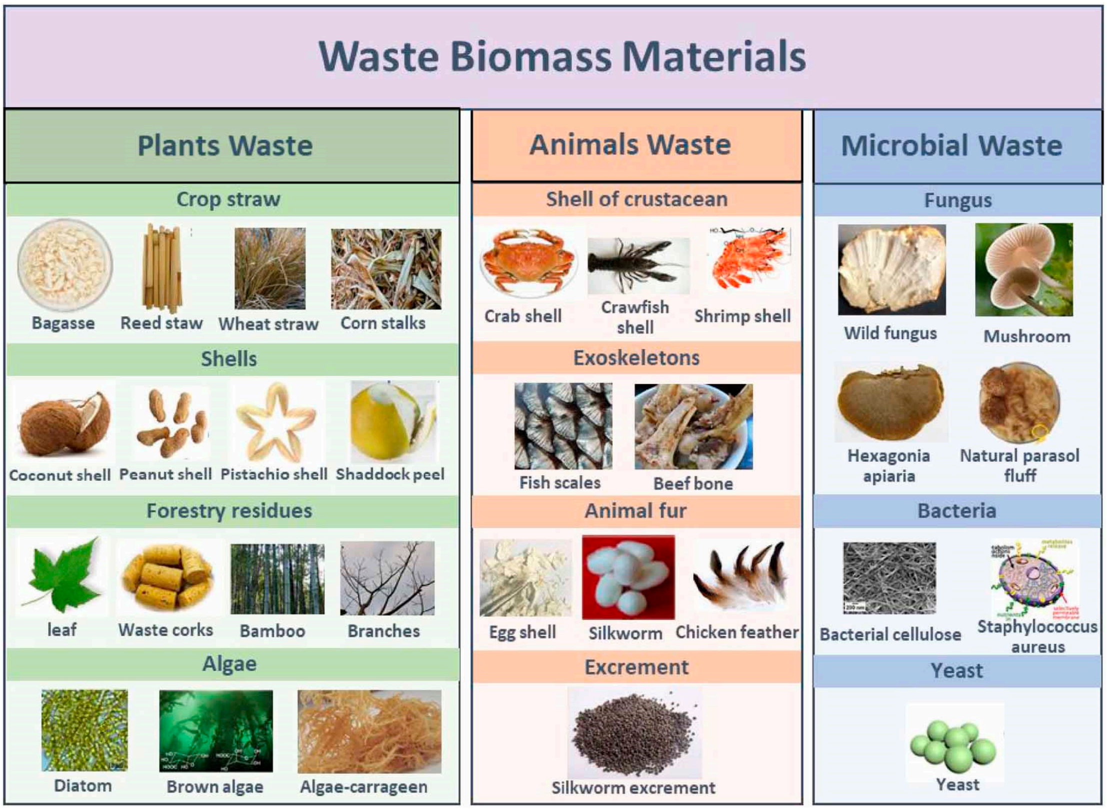

Figure 3. Classification of biomass based on the plants, animals, and microbial waste (Reprinted with permission from Ref. [66]. 2022, Institute of Physics).

Debika Gogoi used porous carbon made from coconut fiber as the anode and a nanocomposite made of $CoFe_2O_4$ nanoparticles (CF) immobilized within the pores of porous carbon (PC) as the cathode, creating a high-performance all-solid-state flexible asymmetric supercapacitor device (Figure 4). The created device had a high energy density of 50.34 Wh kg^{-1} at a power density of 1450 W kg^{-1} and a good cycle life (retention of 91% specific capacitance (CS) after 5000 cycles) [67].

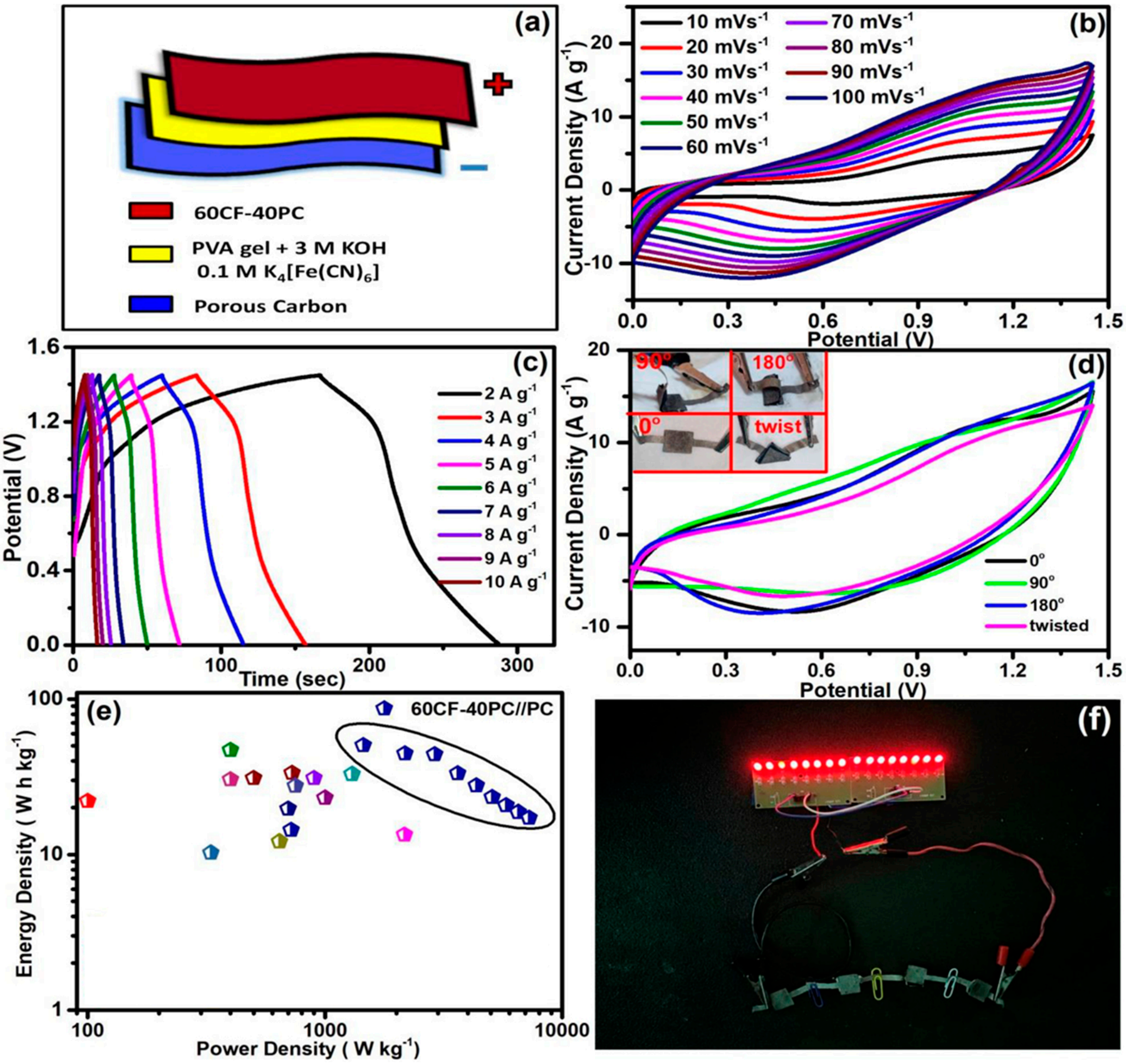

Figure 4. Schematic representation of the supercapacitor device (**a**), and the CV (**b**), GCD (**c**) CV on flexible state (**d**), Ragone plot (**e**), and fabricated device on real–time light; (**f**) application of the prepared material (Reprinted with permission from Ref. [67]. 2022, American Chemical Society).

The successfully fabricated biomass-based carbon and β-$Ni(OH)_2$ hybrid electrode materials (Figure 5i,ii), and the gravimetric capacitance of C–Ni electrode materials reached ~952 F g^{-1} at 1.0 A g^{-1} based on a three-electrode system configuration. The sample can still reach 126 F g^{-1} even at a high current density of 30 A g^{-1} (Figure 5a–f) [68].

The selection of appropriate precursors is one of the most important steps involved in the production of high-performance activated carbon. Tianxiang Jin [69] chose daylily; due to the high levels of nitrogen and phosphorus that it contains, it is suitable for use as a precursor in the production of heteroatom-doped activated carbon (Figure 6i). A straightforward carbonization technique was used to successfully produce a porous carbon derived from daylilies. The resulting carbon had a large specific surface area and a high percentage of heteroatoms. The as-prepared carbon materials exhibited a remarkable specific capacitance of 299.1 F g^{-1} at a current density of 0.5 A g^{-1} and an excellent cycling stability of 99.6% after 4000 cycles at a current density of 1 A g^{-1} (Figure 6A–C). In addition to this, the assembled symmetric supercapacitor demonstrated a high-power density of 598.2 W/kg in 6 M KOH electrolyte while also exhibiting a high energy density of 21.6 Wh kg^{-1} (Figure 6D).

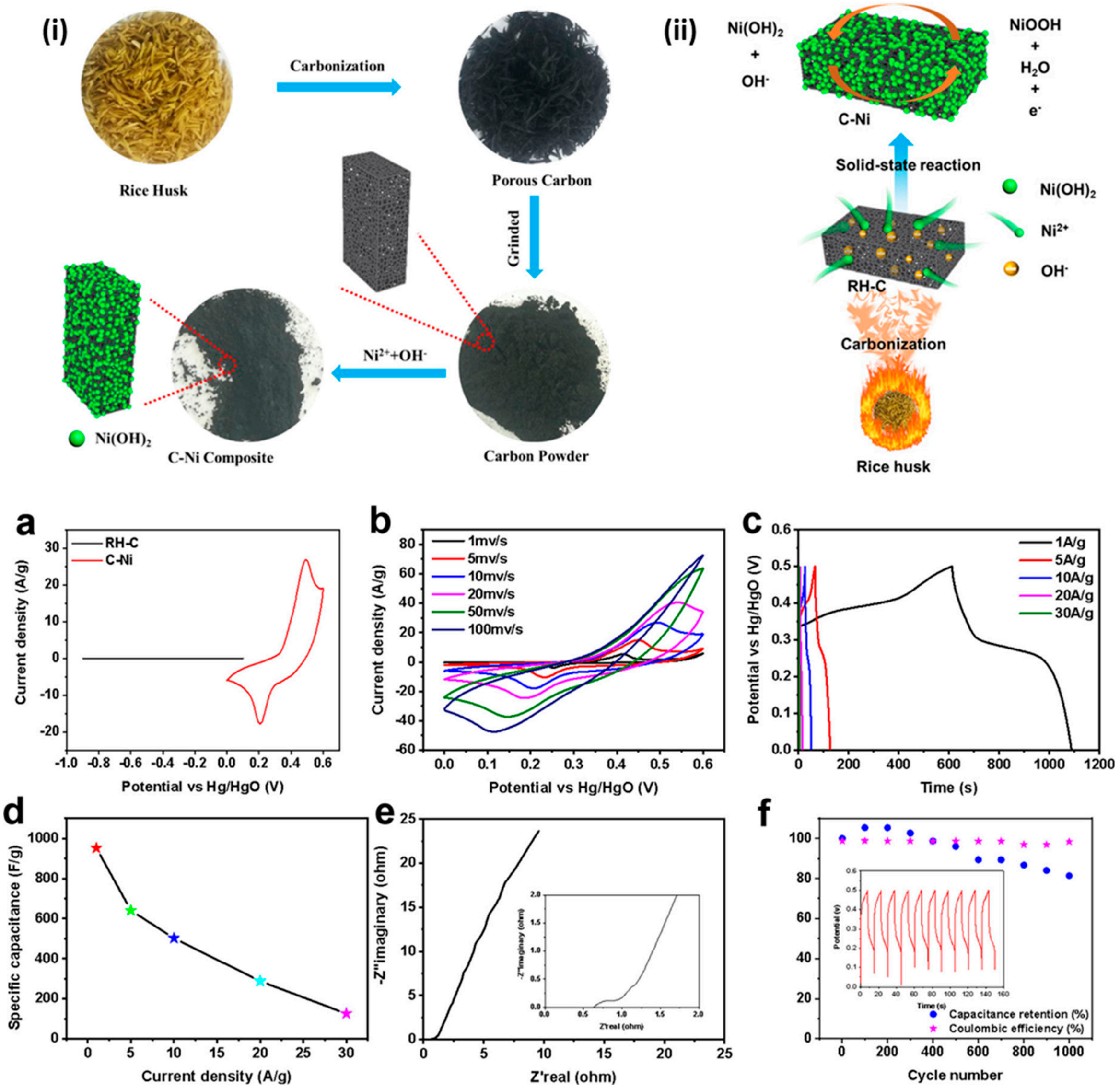

Figure 5. Schematic representation of the bio-derived carbon and its Ni composite (**i**,**ii**), the electrochemical performance of the carbon–nickel composite (**a**–**f**), insert (**f**); the recycle stability test, (Reprinted with permission from Ref. [68]. 2020 American Chemical Society).

3.2. Synthetic Derived Carbon

Porous materials are essential in various industries, including biomedicine, energy, adsorption, separation, and catalysis. Traditional porous materials, such as metal oxide, zeolite, carbon, mesoporous silica, and polymer are amorphous, porous solids with irregular pores and ill-defined structures. Crystalline porous solids known as zeolites have high stability, a periodic structure, and a natural acidity, are widely used in industrial adsorption and catalysis. However, since there are so many different kinds of pore architectures, managing the distribution of acidic sites is a difficult task. The study of porous materials as a whole has been substantially advanced as a result of metal-organic frameworks, often known as MOFs. MOFs are a kind of organic-inorganic hybrids and crystallized porous substance [70–72]. MOFs are fascinating materials due to their high porosity and atomiclevel structures. However, its limited applicability in processes involving large species and diffusion control is due to the pores' narrowness. Interest in extending classic MOF-based carbon materials for use in energy storage applications was prompted by the invention of hierarchically porous MOFs (HP-MOFs), composite materials, and analogues (Figure 7).

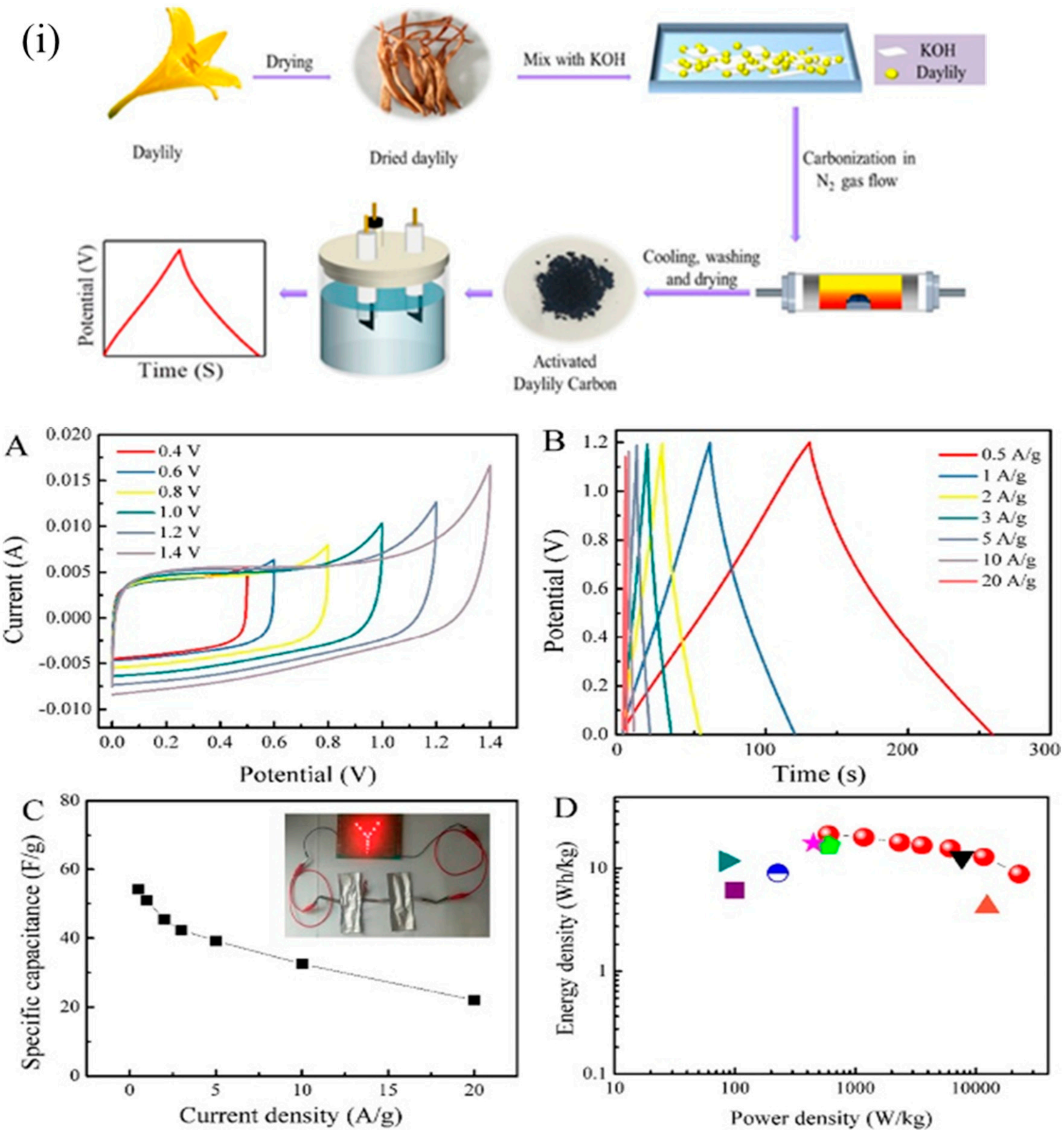

Figure 6. The schematic illustration of the biomass derived carbon from daylily flower (**i**), and the electrochemical behavior of the activated carbon material (**A**–**D**) (Reprinted with permission from Ref. [69]. 2022, American Chemical Society).

Rahul R. Salunkhe [73] made MOF-derived porous carbon (Figure 8(1,2)) and the uniformity of the large-sized NPC particles was good, and their average size was 1 mm. SEM images with high magnification display polyhedral shapes and symmetric geometry (Figure 8a–d). The obtained carbons are ZIF-8 morphologically authentic. The polyhedral shapes of small NPC particles make it simple to adjust particle size by altering reaction conditions. The ZIF-8-derived NPC performed well with a maximum capacitance of 251 F g^{-1} at 5 mV s^{-1}, 10.86 W h kg^{-1} specific energy, and 225 W kg^{-1} specific power. This affordable method may generate next-generation energy storage NPC materials with enhanced pore topologies [74].

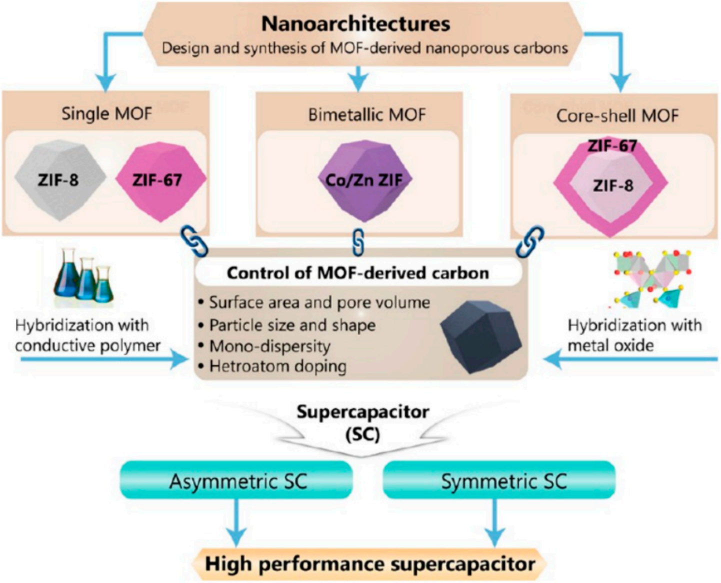

Figure 7. Various NPC Development Methods from Nanoarchitectured MOFs for Supercapacitor Applications (Reprinted with permission from Ref. [28]. 2016, American Chemical Society).

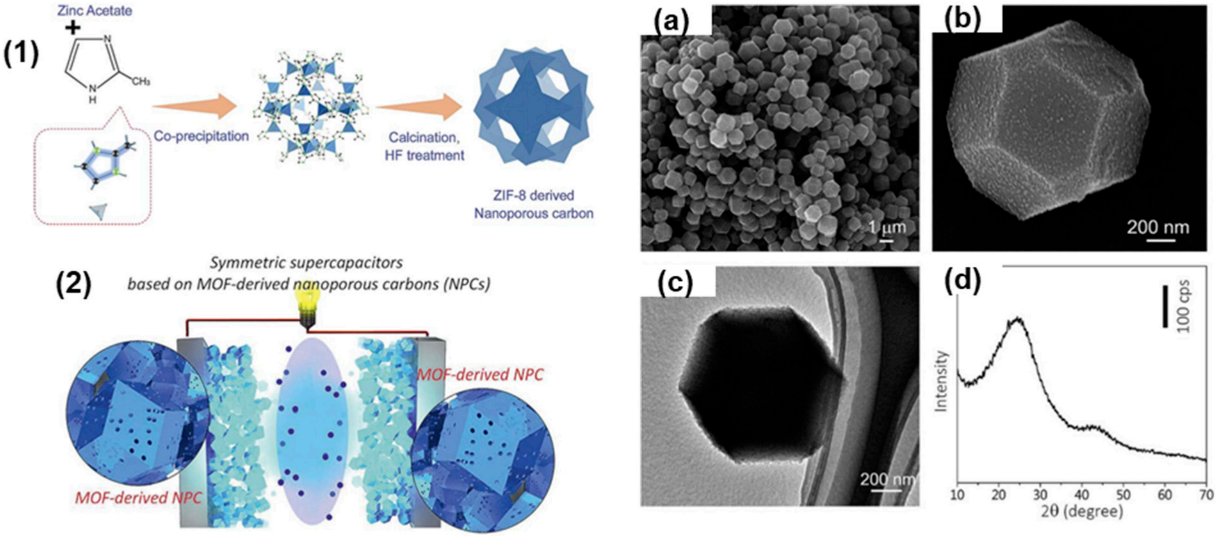

Figure 8. Synthesis and calcination of ZIF–8 (**1**), schematic representation of supercapacitor electrode device (**2**), the surface morphology images FE–SEM (**a**,**b**), HR–TEM (**c**) and the P–XRD results (**d**) of the MOF–derived carbon material (Reprinted with permission from Ref. [73]. 2014, Royal Society of Chemistry).

Jing Yu used KOH to create nitrogen-doped porous carbons (NPCs) from mela-mine-urea-formaldehyde resin. High-performance supercapacitors with superior electrochemical performance were created by adding KOH to improve specific surface area and pore structure. The best sample had a large specific surface area of 2248 $m^2 g^{-1}$ and a high N content, resulting in a specific capacitance of 341 F g^{-1} under 1 A g^{-1} and a retention of 92% after 5000 cycles. A symmetrical solid-state supercapacitor with NPC electrodes exhibited the highest energy generation of 9.60 W h kg^{-1} at 1 A g^{-1} (Figure 9a–e). NPCs offer potential benefits for high-performance supercapacitors and other energy storage technologies [75].

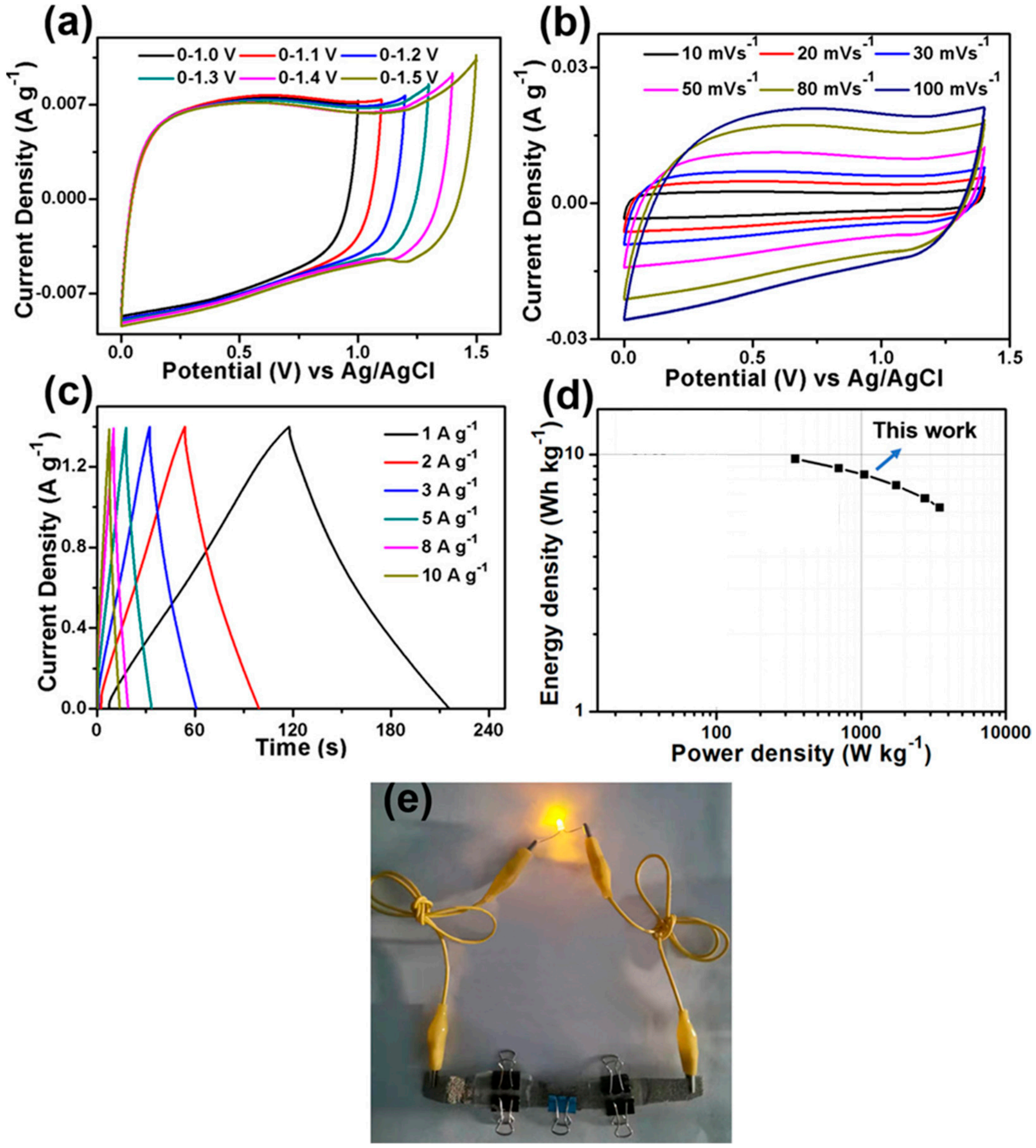

Figure 9. The symmetrical supercapacitor device structure and the CV (**a**,**b**), GCD (**c**), Ragone plot (**d**), and the real–time lighting application (**e**) of the NPCs–2–700/NPCs–2–700.

The use of extremely porous carbon black (CB), carbon Nano horns, carbon nanotubes, graphene, carbon nanoflowers, and their composites in supercapacitors is currently under study. However, low specific capacitance results from pure electrostatic charge accumulation, such as graphene's 200 F g^{-1} in aqueous electrolytes. Short cycle life remains despite composites increasing specific capacitance through the redox process. By incorporating heteroatoms like nitrogen and oxygen into porous carbon composites, specific capacitance may be increased without being noticeable. The advantages of carbon, such as their high conductivity and wide specific surface are maintained. Heteroatom-doped carbon materials can be prepared using chemical vapor deposition [76], post-production processing [77], and direct carbonization [78], with direct carbonization being the preferred method due to its cost-effectiveness. Table 1 lists capacitive data for carbonaceous precursors, including synthetic and natural sources like polyindole, polyaniline, and polyacrylonitrile (Figure 10a–f).

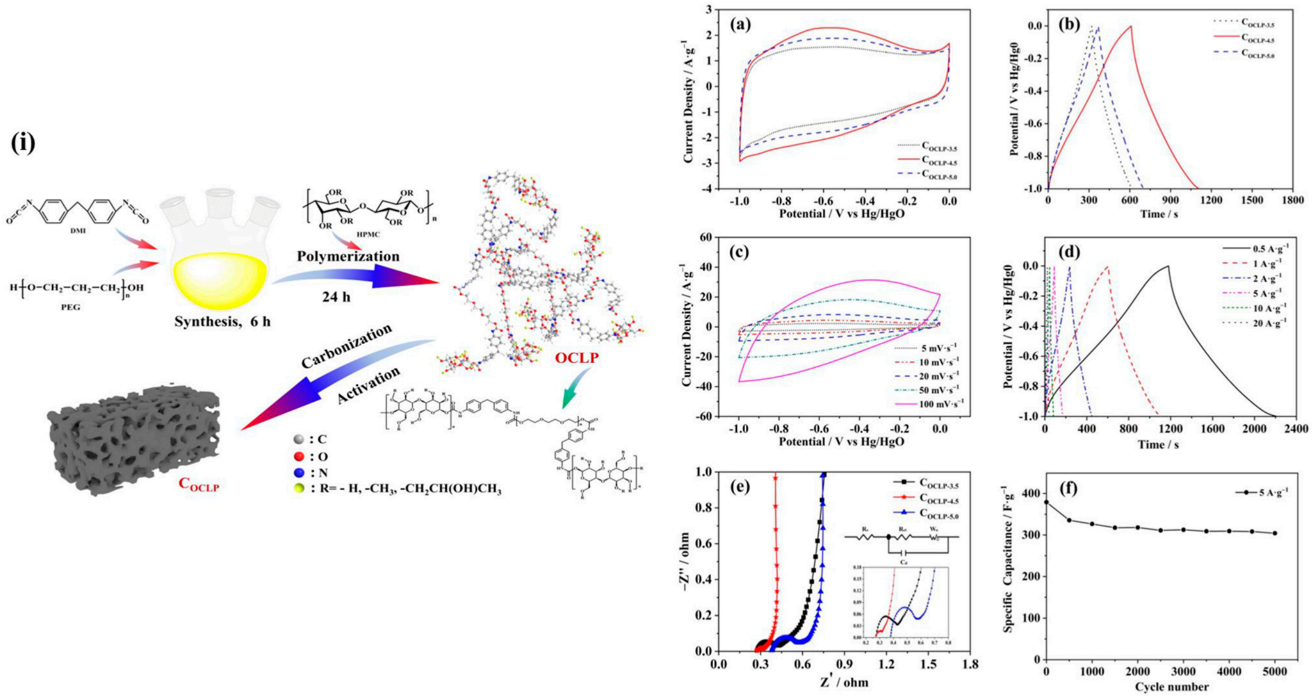

Figure 10. Schematic illustration of the preparation of porous carbon material derived from organic polymer (**i**), the electrochemical performance (CV, GCD, EIS and specific capacitance) of the prepared carbon material (**a**–**f**) [79].

In order to create N/O-doped porous C_{OCLPs} (optimized porous carbon material), Jianhao Lao developed a network-structured organic crosslinked polymer COCLP-4.5, which exhibited exceptional electrochemical performance. The specific capacitance reached 522 Fg^{-1} at 0.5 Ag^{-1} and 309 Fg^{-1} at 20 Ag^{-1} in a three-electrode system. Additionally, the symmetric capacitor attained a power density of 200.0 W kg^{-1} and an energy density of 18.04 Wh kg^{-1}. The C_{OCLPs} benefited from the hierarchical porous carbon formed by organic crosslinked polymers' net structure, as well as from the pseudocapacitance that heteroatoms added.

3.3. CNT and Graphene Based Composites for High-Performance Supercapacitor

The V_2O_5–CNT nanocomposites were used as a negative electrode material for the high-performance supercapacitor applications (Figure 11a–g). The vertically aligned CNTs (VACNT) were prepared by supercritical CO_2 impregnation and the substituent annealing method [80]. The prepared composite material was directly grown of the nickel foam substrate to avoid the use of binding material. The pure VACNT was used as a positive electrode, while the composite was used as a negative electrode material to fabricate the asymmetry supercapacitor device. The neutral Na_2SO_4 was used as an electrolyte solution for the entire analysis. The obtained specific capacitance of the device is 284 F/g at 2 A/g, and the maximum energy and power densities are 32.3 Wh kg^{-1} and 118 Wkg^{-1} [80].

Similarly, the graphene-based nano-composited shows an excellent specific capacitance nature (Figure 11h–n) [81–83]. The reduced graphene oxide (rGO)-based composite material was used as an electrode material for high-performance supercapacitor application. The composite material was directly grown on the surface of the carbon cloth to achieve the binder-free and flexible electrode device structure. The obtained specific capacitance of the device was 451 F/g; another advantage of these composite materials includes better rate performance and recycling stability. The material shows an excellent energy and power densities as 51.11 Wh kg^{-1} and 10 k W kg^{-1}, respectively.

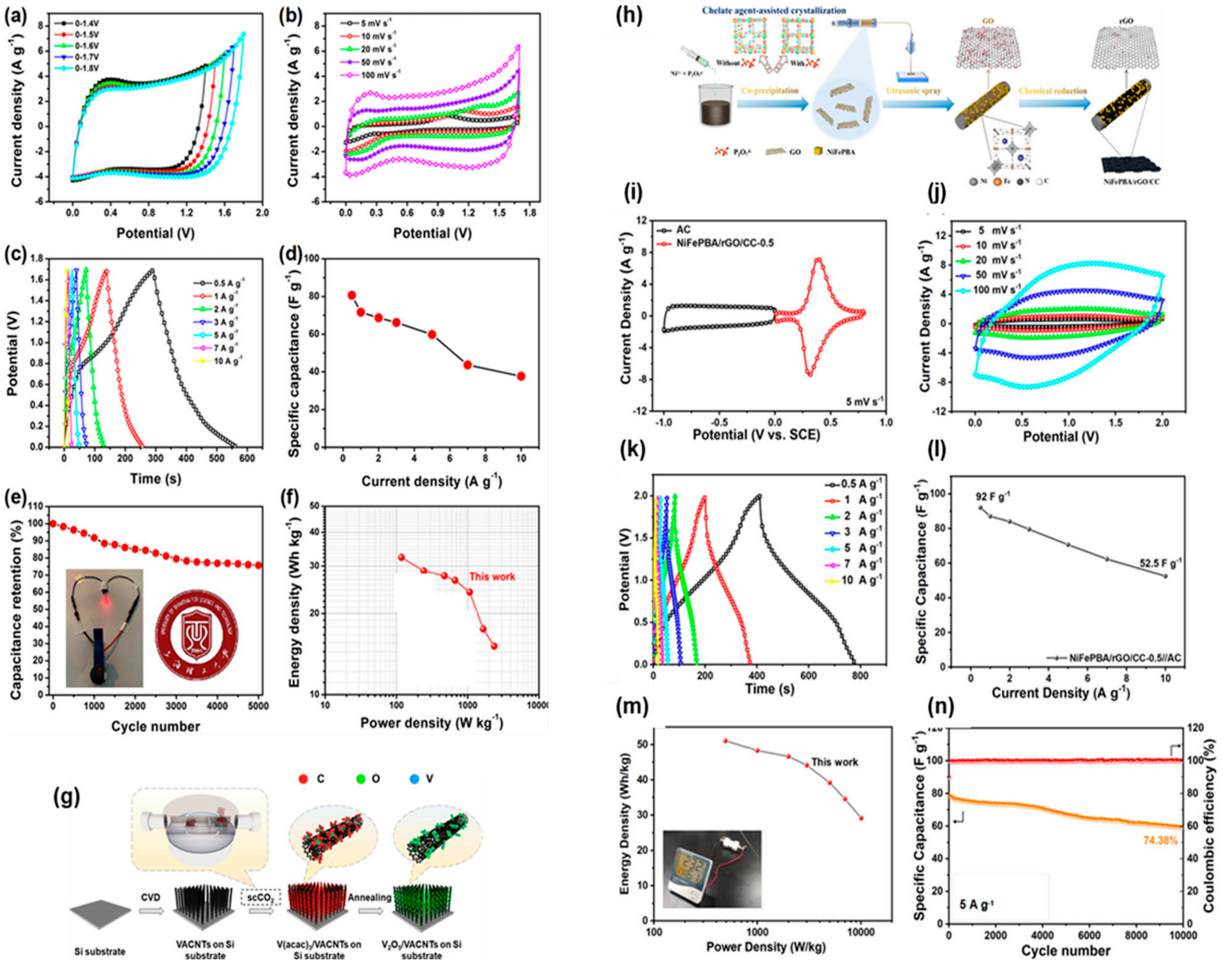

Figure 11. The electrochemical properties of CNT–V_2O_5 nanocomposite materials (**a**–**f**), schematic representation on the preparation of CNT–composite material (**g**) (Reprinted with permission from Ref. [80]. 2021, Elsevier), the synthesis procedure of rGO metal oxide composite materials (**h**) and its supercapacitor characteristic results (**i**–**n**) (Reprinted with permission from Ref. [81]. 2023, Elsevier).

In summary, the carbon-based materials taking a virtual role in the high-performance supercapacitor applications. The major disadvantages of bio-derived carbon materials are lower capacitance with power and energy density. The modification and surface treatment of the carbon materials are very effective for the enhancement of specific capacitance. The many of the carbon materials could be obtained from the natural sources, which is cost effective and environmentally friendly.

4. Recent Metal Oxide Materials on Supercapacitor

The metal oxides-based supercapacitors are very important to the super capacitor applications. The advantages of the metal oxides are as follows, (i) good conductivity, (ii) more stability, (iii) mechanical strength, (iii) high energy and power density. The combination of multi-metal oxide materials may enhance the synergetic effect of the material, which would be modified by the optimization of various metal oxide materials. The multi-metal oxide composition has an advantage to enhance the synergetic effect of the materials; however, it needs to optimize the proper composition by the experimental procedure. Here some of the multi-metal oxide advanced electrode materials for the supercapacitor applications.

4.1. Bi-Metal Oxide

The manganese and molybdenum-based hierarchical $MnMoO_4{\cdot}H_2O@MnO_2$ core-shell Nano arrays were used for the high-performance asymmetric supercapacitor (Figure 12a–n). The bi-metal oxide material of $MnMoO_4{\cdot}H_2O@MnO_2$ was prepared by using a two-step hydrothermal method. To enhance the specific capacitance, the bi-metal oxide material was directly coated on the nickel foam substrate. The FE-SEM and the TEM analysis demonstrated the core-shell nanosheet array structure of the material. The material was confirmed by proper characterization techniques, such as P-XRD, XPS, and elemental mapping analysis. The electrochemical performance of the material studied through the 2,3-electrode system. The 3-electrode system impels the maximum specific capacitance of the material to be 3560.2 F/g at 1 A/g. The obtained result is considerably high in the recently reported material in the supercapacitor area. For the 2-electrode system, the energy and power density of the material are 45.6 Wh k/g and 507.3 W k/g. The synergetic effect of the bi-metal oxide could be the reason for the high efficiency of the material.

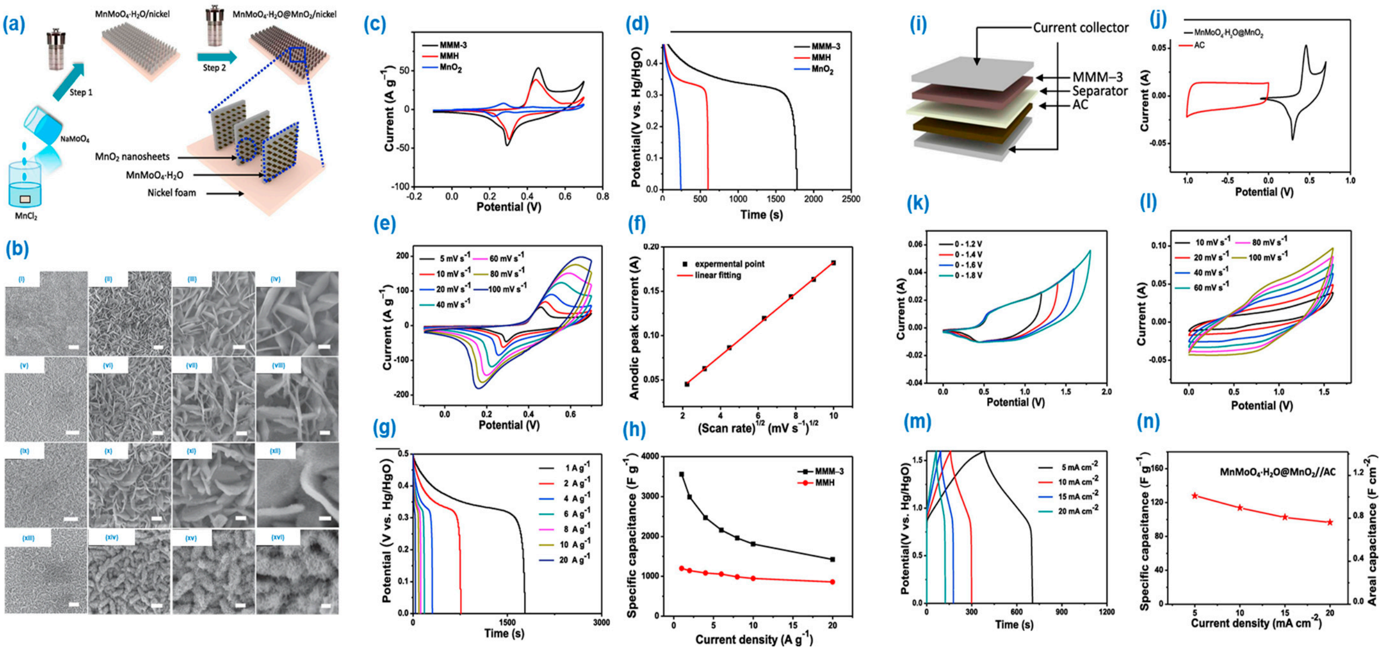

Figure 12. (**a**) Schematic representation on the preparation of $MnMoO_4{\cdot}H_2O@MnO_2$ core–shell Nano arrays, (**b** (**i**–**xvi**)) the FE-SEM image of the prepared metal oxide material on various magnifications, (**c**–**h**) the electrochemical performance of the bi-metal oxide on 3-electrode system, (**i**–**n**) the device structure and the electrochemical analysis of the materials on 2-electrode system (Reprinted with permission from Ref. [84]. 2018, Elsevier).

The hollow structured $NiCo_2O_4$ nano sphere-type material was prepared by modified Stober and hydrothermal method. The material was confirmed by P-XRD, XPS analysis and the surface area was obtained by the BET method. The FE-SEM and HR-TEM analysis suggests the material shows a hollow nanosphere structure with the diameter of ~200–220 nm. The obtained surface area of the $NiCo_2O_4$ is 166.1 m^2/g and the pore size distribution is 3.8–4.3 nm. The electrochemical performance of the material was studied systematically (Figure 13a–l). The specific capacitance of the material was 1229 F/g at 1 A/g with a rate performance of ~83.6%. The material shows specific capacitance of 68.7 F/g, and the attained energy and power density were 21.5 W h/kg, 4500 W/kg. The results suggest the array formation on the nanosphere could enhance the specific capacitance due to the increase in surface area of the material.

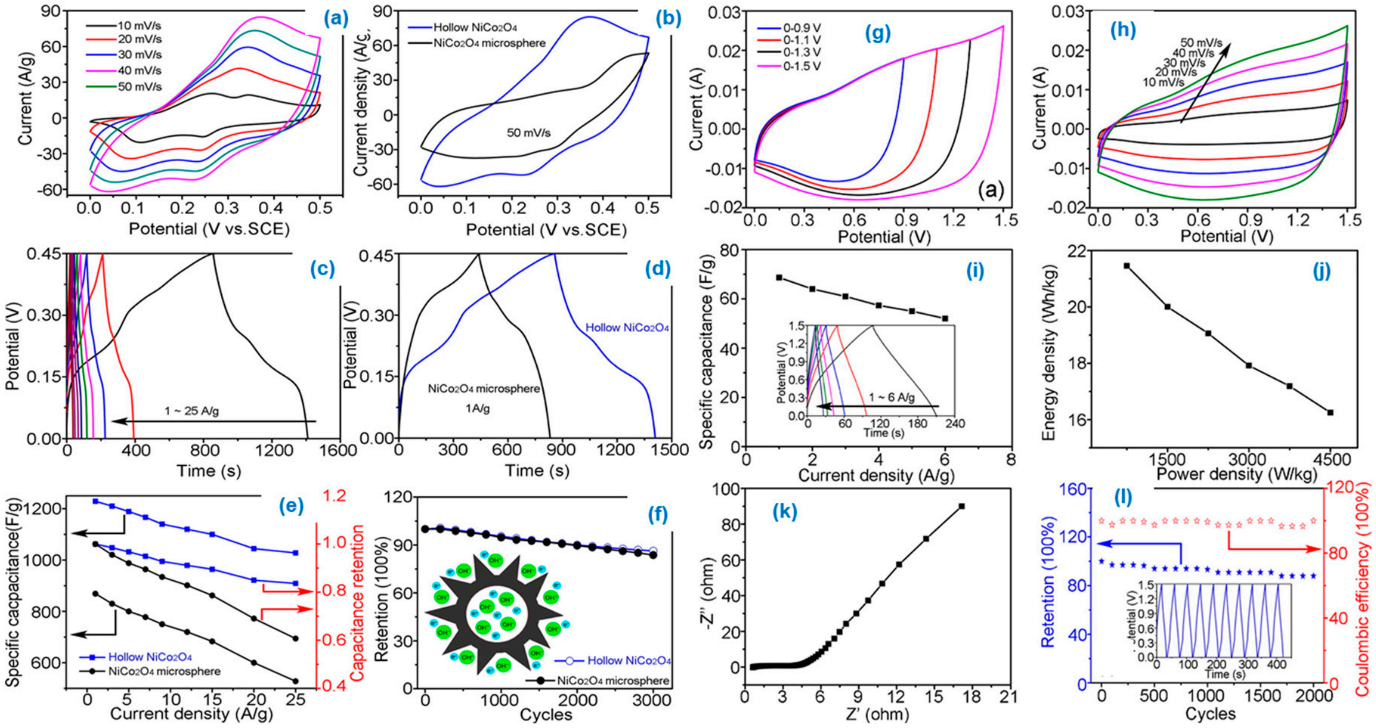

Figure 13. The electrochemical properties of the $NiCoO_4$ on the 3-electrode system (**a**–**f**), and the electrochemical behavior of the $NiCoO_4$ on 2-electrode system (**g**–**l**) (Reprinted with permission from Ref. [85]. 2018, Elsevier).

4.2. Tri-Metal Oxide

The tri-metal oxides-based materials have much interest for battery-type super capacitor applications. These materials show fast charge diffusion and reversible faradaic reactions; hence, they provide high specific capacitance, energy, and power density. Many available metal oxides, such as Co_3O_4, MnO_4, Fe_2O_3, MoO_3, V_2O_5 and ZnO could be applicable for the preparation of tri-metal oxides-based materials. The advantages of theses metal oxide materials include cost effectiveness, and they are environmentally friendly as well.

The Cu-Ni-Co oxides were used to prepare tri-metal oxide materials, and followed by boron doping, were adopted for the high-performance super capacitor application. The FE-SEM analysis shows needle-like morphology was obtained for tri-metal oxide materials, while the agglomerated needle-like morphology obtained for boron-doped metal oxide materials (Figure 14a–o). The obtained specific capacitance of the doped tri-metal oxide is 974 C/g at 2 A/g, and the materials show 700 C/g at 20 A/g, which suggests the material shows a high rate capability. The energy and power density of the material was calculated by using the coin cell method. The rGO was used as a negative electrode, while the prepared material was used as a positive electrode. The obtained energy and power density of the trimetal oxide material was 42 Wh/kg and 926 W/kg. The materials stability and the electrochemical performance of the tri-metal oxide material were increased after the boron doping. The results suggest as the doping of hetero atom onto the metal oxide material could enhance the electrochemical performance of the material.

The Ni-Co-M (M = Mn, Fe, Zn)-based porous trimetallic fluoride materials were prepared as an electrode material for the high-performance asymmetric supercapacitors (Figure 15a–z). The obtained material shows high rate capability and recycle stability. Among the various compositions, the Ni-Co-Zn fluoride shows the highest specific capacitance of 914.4 F/g at 1 A/g with 81.3% capacitance retention at 20 A/g. The fluoride-doped tri-metal oxide material shows ahigh energy density of 49.3 Wh/kg at 800 W/kg. The reason for the high energy density is due to the superior synergetic effect between the metal oxide materials. The obtained fluoride-doped transition metal oxide materials suggest the doping of the halogen group element to the transition metal oxide materials could enhance the specific capacitance of the electrode materials.

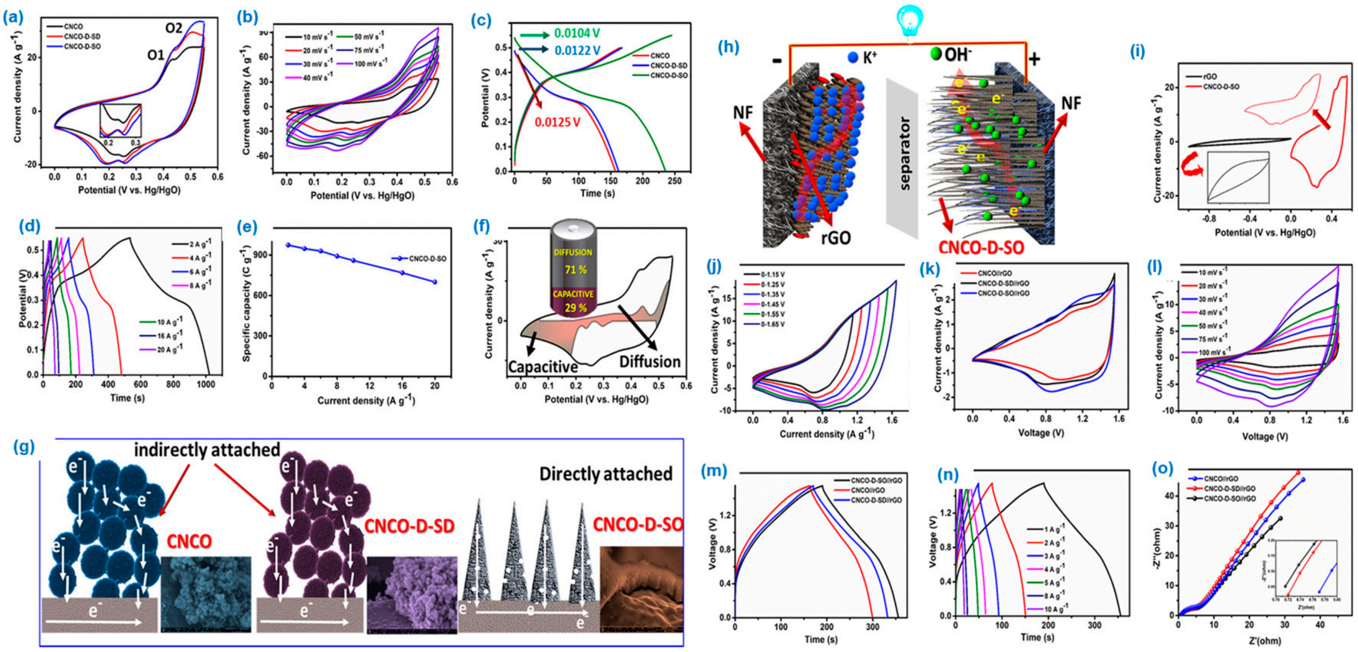

Figure 14. The electrochemical performance of Cu–Ni–Co oxides on 3-electrode system (**a**–**f**), (**g**,**h**) the possible mechanism on charging and discharging of electron on the metal oxide surface, (**i**–**o**) the device fabrication and the electrode performance on 2-electrode system (Reprinted with permission from Ref. [86]. 2021, American Chemical Society).

In summary, the different metal oxide materials possess the ideal candidate for the supercapacitor applications. The conductivity is also expected to be higher than other materials. The morphology and the crystal structure of the metal oxide materials is also important to enhance the efficiency of the supercapacitor device. The doping of different materials and the formation of the core-shell structure enhances the synergetic effect, which further enhances the specific capacitance of the device [87].

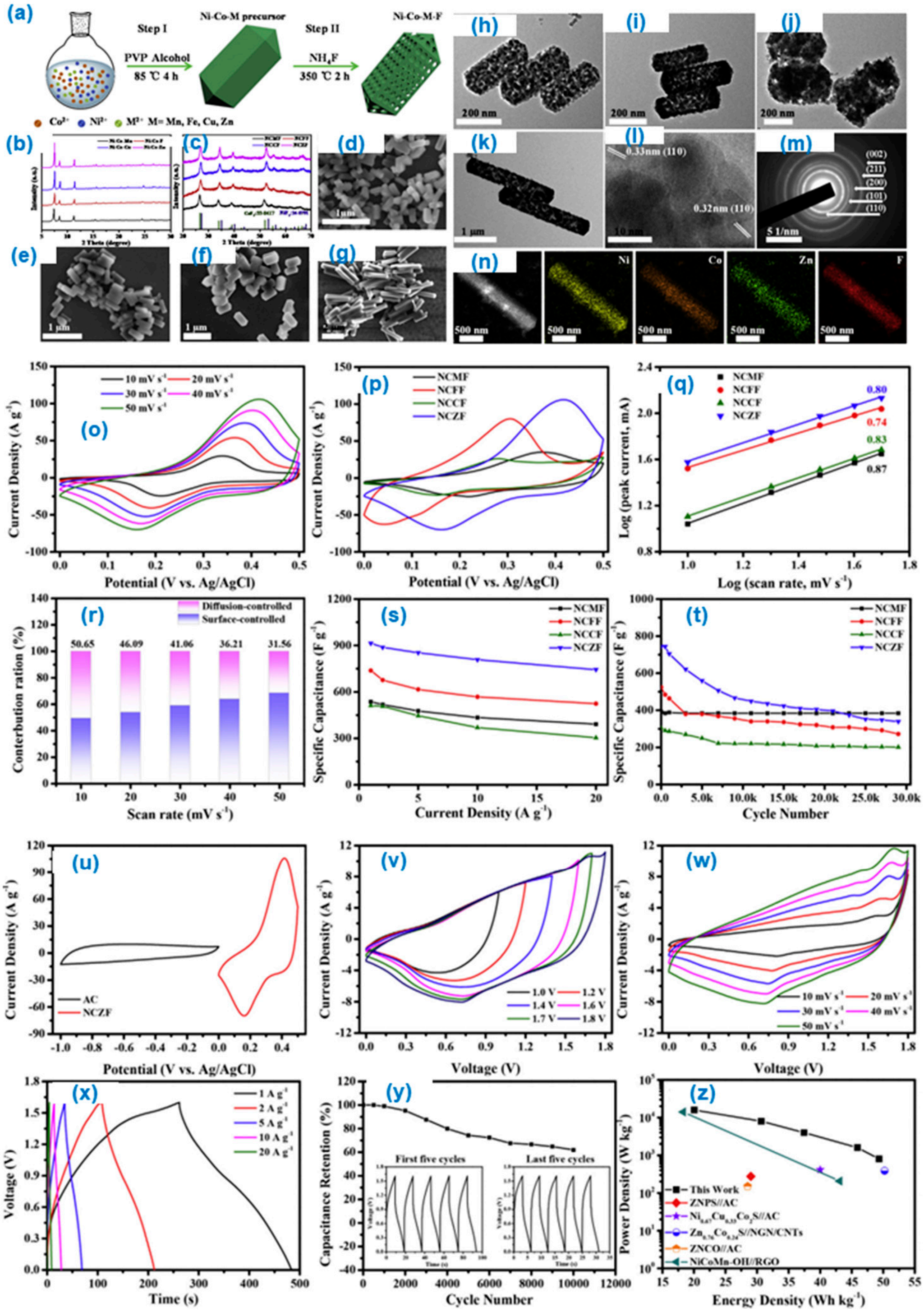

Figure 15. Synthesis of fluoride–doped tri–metallic Ni–Co–M (M = Mn, Fe, Zn) oxide material (**a**), the powder XRD (**b**,**c**), FE–SEM (**d**–**g**), HR–TEM (**h**–**m**) and the elemental mapping (**n**) analysis of the prepare trimetallic oxide material, the electrochemical performance on 2,3–elctrode system (**o**–**z**) of the fluoride–doped trimetallic oxide material (Reprinted with permission from Ref. [87]. 2020, Elsevier).

5. Advanced Materials for the High Power and Energy Density Application

5.1. Carbon Metal Oxide Composites

Due to their substantial surface area, low price, and simplicity of processing, activated carbons (AC) are frequently used as electrode materials. However, their rate and energy storage capabilities are subpar. Pseudocapacitance is a form of capacitor that, although being capable of producing large levels of capacitance, has low power density and unstable cycling due to its weak electrical conductivity. The creation of activated carbons and pseudocapacitive materials is essential for the enhancement of supercapacitors. Recent composite materials showing better capacitance performance and cycle life combine carbon-based materials with metal oxides, hydrocarbons, or conducting polymers. The mechanical strength, electrical conductivity, and electrochemical activity of these composites are all improved. The surface energy of active nanomaterials may be lowered by using a hybrid design. Electrodes made of activated carbon or metal oxide-hydroxides have found usage in a number of contexts.

Greater capacitance may be seen in metal oxides such as RuOx, SnOx, CoOx, NiOx, FeOx, and MnOx because of a phenomenon known as pseudocapacitance. This phenomenon is the consequence of a fast redox reaction that occurs between the oxide and the electrolyte [88]. The estimated capacitances of Co_3O_4 and NiO electrodes are much higher than those of carbon electrodes, coming in at 3560 and 2584 F g^{-1}, respectively [33,89]. However, due to their severe agglomeration, poor electrical conductivity, and short cycle life, are frequently ineffective when used directly in high-performance supercapacitors. As electrodes for supercapacitors, composite materials that are made up of porous carbon and metal oxides have attracted a lot of interest recently. By mixing hierarchical porous carbons with pseudocapacitive metal oxides, these materials improve electrochemical performance. Zheng and colleagues created a binder-free 3D electrode by employing hierarchically porous carbons, Co_3O_4 nanoarrays, and morpho butterfly wing scales. With this electrode, they were able to achieve a specific capacitance of 978.9 F g^{-1} at 0.5 A g^{-1} [90].

Zhiwei Tian [91] reported a manganese oxide/carbon composite with high specific capacitance and conductivity (Figure 16a–i). Utilizing homemade porous carbon and $KMnO_4$ as raw materials, manganese oxides/carbon composites have been created. Due to the Mn-O-C interaction, these composites have high specific metal oxide capacitance and high carbon conductivity. At current densities of 1, 2, 3, 5, and 10 A g^{-1}, the optimized composite electrode exhibits extremely high specific capacitance values of 550, 520, 481, 402, and 318 Fg^{-1}. The completed asymmetric supercapacitor (ASC) device exhibits excellent rate performance at 1600, 2297, 3715, and 6890 W/kg and an energy density of 50.2 Wh kg^{-1} at 800 Wkg^{-1}. After 10,000 cycles, the ASC device still maintained 90.5% capacitance, demonstrating excellent cycling stability. Energy storage uses for these ASC devices could include powering 35 lights and an electronic stopwatch. The material is a promising candidate for manganese/carbon composite supercapacitors, which are anticipated to be produced in large quantities commercially due to their straightforward preparation methods and ultra-high performance.

5.2. MXenes-Based Materials for the High-Performance Supercapacitor Application

MXenes are a group of two-dimensional advanced functional nanomaterials that have been investigated for use in the synthesis of fuel and clean energy, among other applications [8–12]. They have distinct layered structures that enhance electrolyte ion transport in addition to offering active transition metal redox sites on the surface. MXenes are particularly promising as high-performance electrodes for electrochemical capacitors because of qualities like conductivity, aspect ratio, etc. [92]. With aqueous electrolytes like $(NH_4)_2SO_4$ and $MgSO_4$, electric double-layer capacitors (EDLC) have a significant impact on the charge/discharge tests of MXene electrodes. Because hydrated ions fill the interlayer spacing, the EDLC dominates these tests [93,94]. A higher capacitance is seen when H_2SO_4 is used, suggesting that protons are the predominant charge carriers. H_2SO_4's impact on the storage mechanism is still debatable, necessitating additional theoretical research and combined characterization. Redox reactions, intercalation, and underpotential deposition are blamed for pseudocapacitance. Pseudocapacitance is produced when MXene cathodes are coupled to conductive polymer-based anodes while the system is immersed in an H_2SO_4 electrolyte. Intercalation is a method for storing alkali metal cations in supercapacitors based on MXene that make use of organic electrolytes. In spite of the fact that in situ characterization technologies are able to shed light on the structural evolution and chemical circumstances of MXene-based electrodes, there are still a great deal of concerns that have yet to be solved about the dynamics of ions in supercapacitors [95,96].

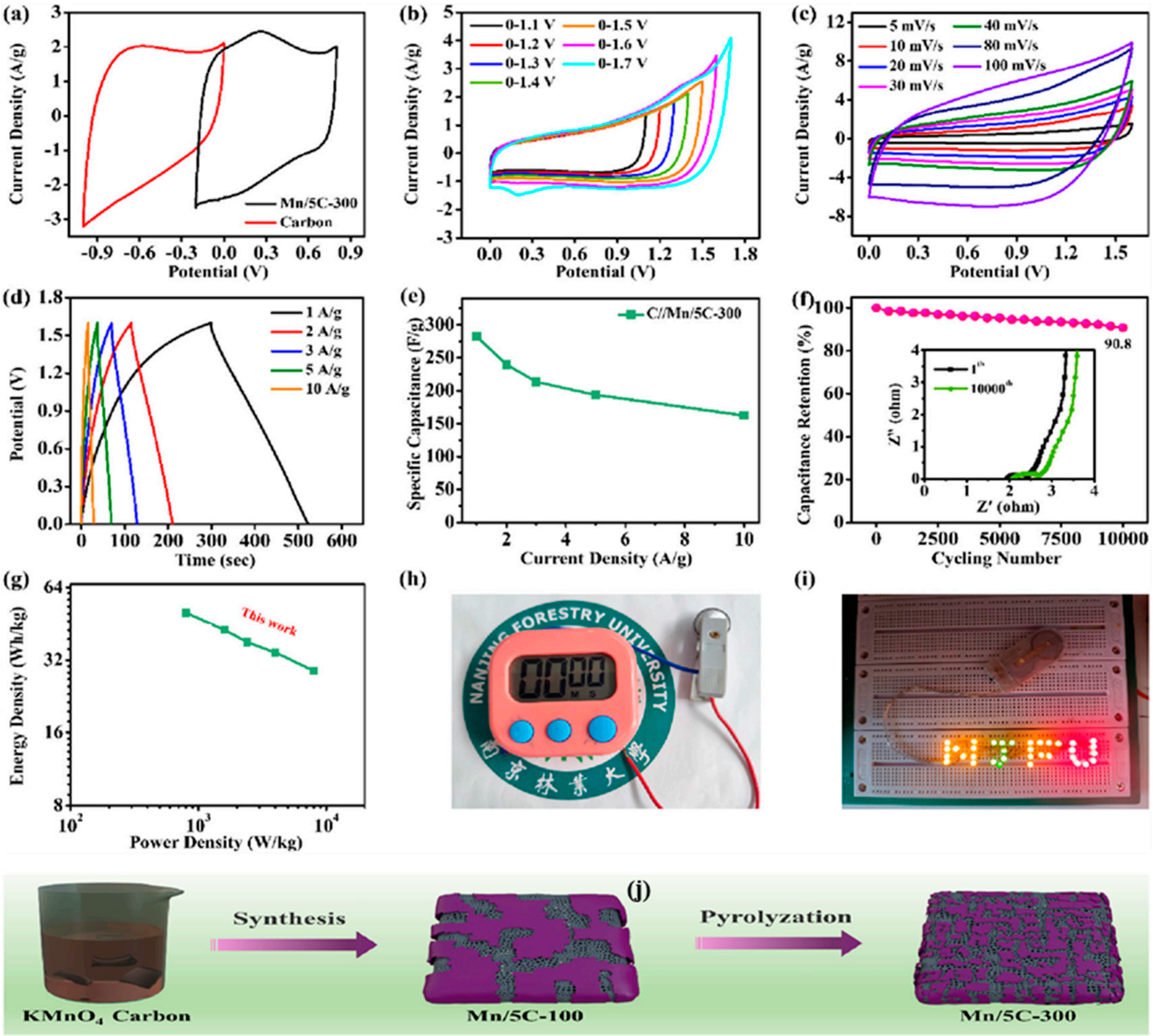

Figure 16. The electrochemical performance (**a**–**g**), the real time application (**h**,**i**) and preparation of the manganese oxide/carbon composite material (**j**) (Reprinted with permission from Ref. [87]. 2023, Elsevier).

MXene-based electrocatalysts require innovative methods for synthesizing monolayer MXenes with large transverse dimensions and thoughtful structural modifications in order to achieve excellent activity and selectivity. Rui Ma [97] developed $Ti_3C_2T_x$ MXene-based supercapacitor electrodes with improved capacitance, rate capability, cyclic stability, and mechanical flexibility. A capacitance of 532.9 F g^{-1} was attained by the P-MXene/CPAQ-A electrode, which was composed of a quinone-amine polymer. Additionally, the rate performance and cyclic stability were both significantly enhanced. After 40,000 iterations, the integration strategy that was optimized resulted in a retention rate of 97.1% (Figure 17a–i). This electrode outperformed pristine MXene and P_{MXene} electrodes in terms of retention and cyclic stability. The optimized techniques produce more active sites, quicker ion accessibility, better chemical stability, and good mechanical flexibility. The P-MXene/$C_{polymer}$ electrodes show capacitance enhancements due to enlarged interlayer spaces, nanostructured carbonized polymers, PMMA sacrificing templates, and intercalated K^+ ions. MXene surface changes include the elimination of fluorine, a reduction in the number of hydroxyl groups, an increase in the number of oxygen groups, intercalation of potassium ions, and the presence of carbonized polymers with electrochemically active groups. Expanded interlayer gaps and porous or hierarchically nanostructured polymers, both of which avoid restacking and speed up ion accessibility, are responsible for the improved rate capability of the material. However, undesirable nanostructures may affect performance at higher scan rates. Improved cyclic stability is likely due to enhanced chemical stability and repressed oxidation.

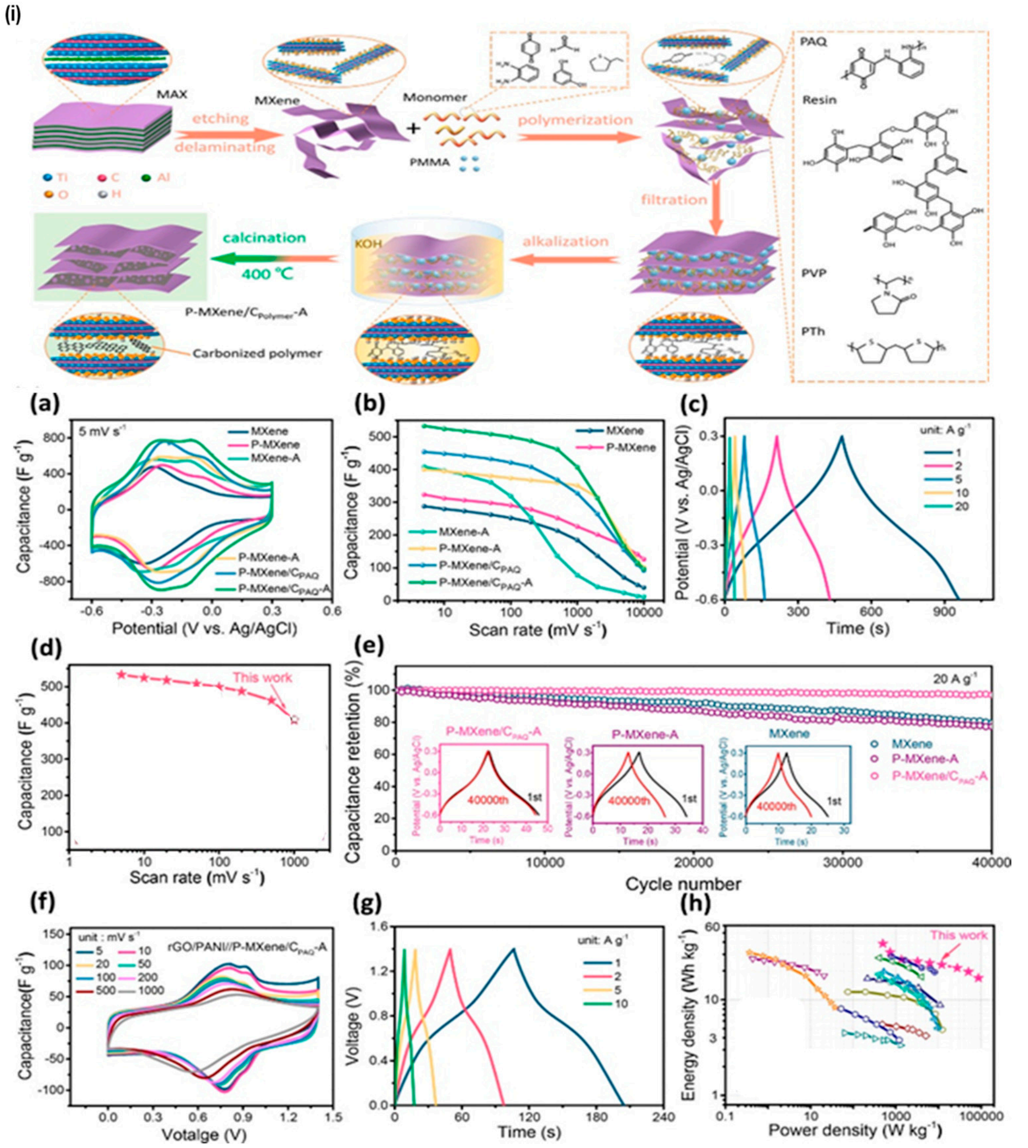

Figure 17. The Schematic representation of the preparation of MXene and the MXene–modified material upon the various process (**i**), the electrochemical analysis results of the rGO/PANI/P–MXene/C (**a**–**h**) with various parameters (Reprinted with permission from Ref. [97]. 2022, American Chemical Society).

Bao Shi [98] successfully synthesized self-supported and vertically aligned Ti_2CT_x MXene nanosheets on the surface of carbon fibers (CF), which required the use of the molten salt method in conjunction with the selective etching method (Figure 18a–h). This technique lowers the risk by using CFs as a flexible substrate and carbon supply rather than collecting powder samples from HF mixes. This eliminates the necessity to collect the powder samples. In addition, the vertical alignment of the Ti_2CT_x MXene helps to eliminate the stacking problem that occurs with standard MXene film electrodes created using the filtering process. This helps to shorten and streamline the ion transport channel, which enhances the performance of zinc-ion hybrid supercapacitors (HSC). Zn-ion high-speed capacitors manufactured from Ti_2CT_x MXene@CF/Zn@Cu@CF have great stability since they retain 90% of their capacity after 10,000 cycles, have an extremely low self-discharge rate of 0.53 mV h^{-1}, and have a high areal capacitance of 380 mF Cm^{-2} at a scan rate of 5 mV s^{-1}. Additionally, these Zn-ion HSCs have outstanding capacity retention [98].

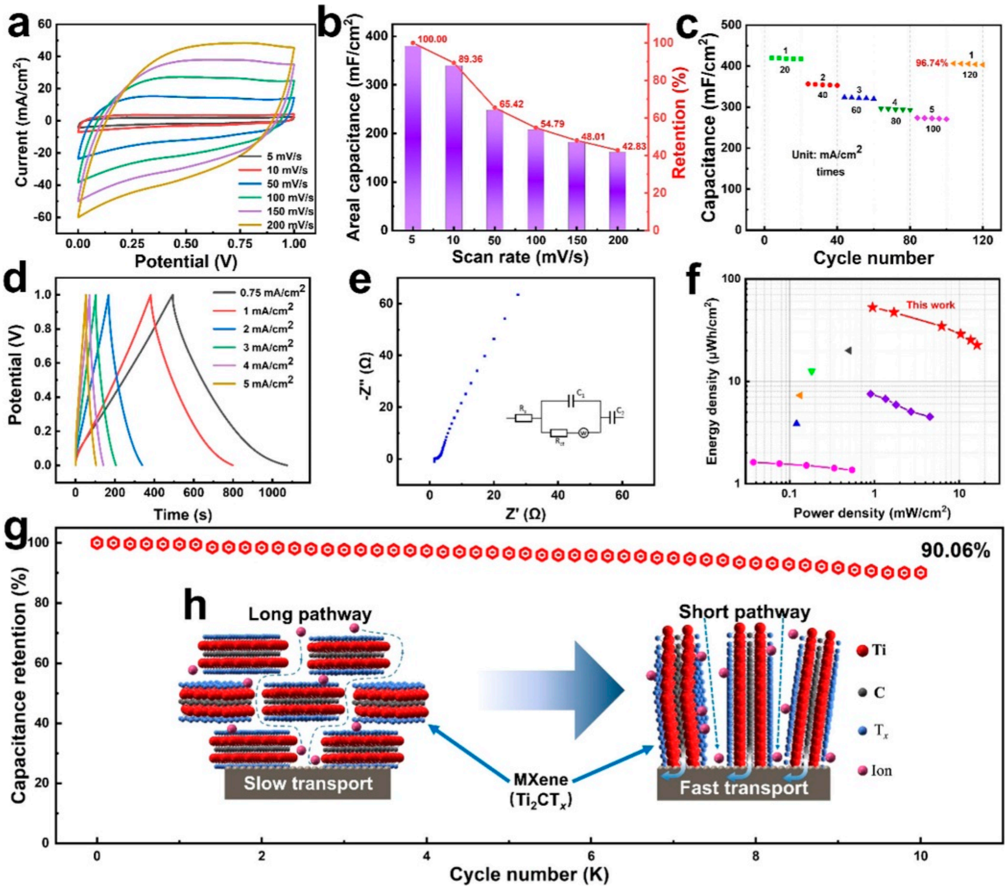

Figure 18. The electrochemical performance of the Ti_2CT_x MXene@CF/Zn@Cu@CF on various analysis (**a**–**g**) and the possible charging and discharging mechanism (**h**) of the prepared material (Reprinted with permission from Ref. [98]. 2023, American Chemical Society).

Overall, the MXenes are new class of materials for the high-performance supercapacitor applications. The MXene-based materials possess the high energy and power density in nature. The reason behind the high performance is the materials show the unique layered-like structure. The complete MXene looks as the combination of the multilayer-like structure similar to the graphite molecule. The each and individual layer behave like an active channel to accommodate the applied electron. The layered structure enhances the surface area, which opens a new window for the charging and discharging of electrons.

Table 1. The electrochemical performance results of some recently reported martials.

Materials	Specific Capacitance (F g^{-1})	Energy Density (Wh kg^{-1})	Power Density (W kg^{-1})	Ref.
Biomass-derived Carbon	258.8 at 1 Ag^{-1}	7.11	125.46	[99]
Biomass-derived Carbon	382 at 1 Ag^{-1}	23.13	300	[100]
polyindole	293 at 0.2 A g^{-1}	15	-	[101]
polyaniline	~290 at 1 A g^{-1}	10.3	-	[102]

Table 1. *Cont.*

Materials	Specific Capacitance ($F\ g^{-1}$)	Energy Density ($Wh\ kg^{-1}$)	Power Density ($W\ kg^{-1}$)	Ref.
polyacrylonitrile	252 at 0.1 A g^{-1}	8.8	-	[103]
Metal Oxide (Cu-doped Co_3O_4 NP)	-	64.1	800	[104]
Metal Oxide (Mn_3O_4 triangular structures)	751.3 at 1 A g^{-1}	91.7	899.5	[105]
Carbon/metal oxide composite (ZnO/NiO@MWCNT)	1988.8	43.59	4000	[106]
Carbon/metal oxide (NiO@Co_3O_4-Activated C)	800.9 at 1.0 A. g^{-1}	136.6	-	[107]
MXene ($Ti_3C_2T_x/V_2O_5$)	−319.1 at 5 A g^{-1}	18.43	603.2	[108]
MXene (Ti_3AlC_2/polyaniline/Co Ni LDH)	1200 at 1 A g^{-1}	399.95	39.33	[109]
CNT-V_2O_5	284 at 2 A g^{-1}	32.3	118	[80]
rGO-NiFe-PBA	451 at 1 A g^{-1}	51.11	10,000	[81]

6. Conclusions

In a comparable form, supercapacitors are examined in detail in this paper, including their structure, operating principles, specifications, materials, and applications. The classification of supercapacitors based on mechanisms was briefly discussed as well. It examines their benefits, contemporary technologies, and suppliers. The article clearly envies the supercapacitor by various types of materials, such as carbon, metal oxide, and advanced materials. The carbon-based materials deal with the environmentally friendly carbon derived from natural sources and synthetic material. The metal oxide materials portion promotes the different multi-metal oxide materials such as bi- and tri-metal oxides for the electrode preparation. The advanced materials conclude the composites of both carbon and metal oxide materials followed by the MXene-based materials. The supercapacitors model, a real-time application, and performance evaluations are also shared in this paper. The article discusses the most recent advances in material technologies, overcoming application challenges, and enhancing the marketability and efficacy of supercapacitors. The composites of carbon-metal oxide and carbon-MXene are expected to be promising composites in the future because of their excellent performance, environmental friendliness, simple manufacturing procedures, and commercial viability.

Author Contributions: Conceptualization, B.A. and R.V.; methodology, G.M.; software, S.K.S.M.; validation, S.C.K., B.A. and R.V.; formal analysis, G.M.; investigation, S.K.S.M.; resources, S.C.K.; data curation, B.A.; writing—original draft preparation, R.V.; writing—review and editing, G.M.; visualization, B.A.; supervision, S.C.K.; project administration, S.C.K.; funding acquisition, S.C.K. All authors have read and agreed to the published version of the manuscript.

Funding: S.C.K. greatly acknowledges the Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Education (2020R1I1A3052258) for financial support.

Data Availability Statement: The data pertaining to this report can be provided on reasonable request.

Acknowledgments: The author G.M. acknowledges the Shinshu University for the support.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Rashidi, S.; Esfahani, J.A.; Hormozi, F. Classifications of Porous Materials for Energy Applications. *Encycl. Smart Mater.* **2022**, *2*, 774–785. [CrossRef]
2. AL Shaqsi, A.Z.; Sopian, K.; Al-Hinai, A. Review of energy storage services, applications, limitations, and benefits. *Energy Rep.* **2020**, *6*, 288–306. [CrossRef]

3. Grim, R.G.; Huang, Z.; Guarnieri, M.T.; Ferrell, J.R.; Tao, L.; Schaidle, J.A. Transforming the carbon economy: Challenges and opportunities in the convergence of low-cost electricity and reductive CO_2 utilization. *Energy Environ. Sci.* **2020**, *13*, 472–494. [CrossRef]
4. Mohanty, B.; Jena, B.K.; Basu, S. Single Atom on the 2D Matrix: An Emerging Electrocatalyst for Energy Applications. *ACS Omega* **2020**, *5*, 1287–1295. [CrossRef] [PubMed]
5. Inagaki, M.; Konno, H.; Tanaike, O. Carbon materials for electrochemical capacitors. *J. Power Sources* **2010**, *195*, 7880–7903. [CrossRef]
6. Raj, B.; Padhy, A.K.; Basu, S.; Mohapatra, M. Review—Futuristic Direction for R&D Challenges to Develop 2D Advanced Materials Based Supercapacitors. *J. Electrochem. Soc.* **2020**, *167*, 136501. [CrossRef]
7. Miao, L.; Song, Z.; Zhu, D.; Li, L.; Gan, L.; Liu, M. Recent advances in carbon-based supercapacitors. *Mater. Adv.* **2020**, *1*, 945–966. [CrossRef]
8. Béguin, F.; Presser, V.; Balducci, A.; Frackowiak, E. Carbons and electrolytes for advanced supercapacitors. *Adv. Mater.* **2014**, *26*, 2219–2251. [CrossRef]
9. Theerthagiri, J.; Senthil, R.A.; Nithyadharseni, P.; Lee, S.J.; Durai, G.; Kuppusami, P.; Madhavan, J.; Choi, M.Y. Recent progress and emerging challenges of transition metal sulfides based composite electrodes for electrochemical supercapacitive energy storage. *Ceram. Int.* **2020**, *46*, 14317–14345. [CrossRef]
10. Zhang, X.; Zhang, H.; Li, C.; Wang, K.; Sun, X.; Ma, Y. Recent advances in porous graphene materials for supercapacitor applications. *RSC Adv.* **2014**, *4*, 45862–45884. [CrossRef]
11. Huang, S.; Zhu, X.; Sarkar, S.; Zhao, Y. Challenges and opportunities for supercapacitors. *APL Mater.* **2019**, *7*, 100901. [CrossRef]
12. Najib, S.; Erdem, E. Current progress achieved in novel materials for supercapacitor electrodes: Mini review. *Nanoscale Adv.* **2019**, *1*, 2817–2827. [CrossRef]
13. Abdah, M.A.A.M.; Awan, H.T.A.; Mehar, M.; Mustafa, M.N.; Walvekar, R.; Alam, M.W.; Khalid, M.; Umapathi, R.; Chaudhary, V. Advancements in MXene-polymer composites for high-performance supercapacitor applications. *J. Energy Storage* **2023**, *63*, 106942. [CrossRef]
14. Yang, Q.; Li, Z.; Xu, B. Layered Double Hydroxide with Interlayer Quantum Dots and Laminate Defects for High-Performance Supercapacitor. *Adv. Funct. Mater.* **2023**, *2300149*, 2300149. [CrossRef]
15. Ashraf, I.; Ahmad, S.; Dastan, D.; Wang, C.; Garmestani, H.; Iqbal, M. Delaminated N-Ti_3C_2@Ni_3S_4 nanocomposites based high-performing supercapacitor device fabrication. *Electrochim. Acta* **2023**, *442*, 141899. [CrossRef]
16. Vanaraj, R.; Daniel, S.; Haldhar, R.; Asrafali, S.P.; Kim, S.C. Direct growth of TiO_2–MoO_2/MnO_2–MoO_2 on plasma-treated carbon-cloth surface for high-performance supercapacitor and oxygen evolution reaction. *Electrochim. Acta* **2023**, *440*, 141705. [CrossRef]
17. Deng, C.; Hong, X.; Wang, G.; Dong, W.; Liang, B. Research advance of NiCoP-based materials for high-performance supercapacitors. *J. Energy Storage* **2023**, *58*, 106379. [CrossRef]
18. Revankar, S.T. *Chemical Energy Storage*; Elsevier Inc.: Amsterdam, The Netherlands, 2019; ISBN 9780128139752.
19. Gautham Prasad, G.; Shetty, N.; Thakur, S.; Rakshitha; Bommegowda, K.B. Supercapacitor technology and its applications: A review. *IOP Conf. Ser. Mater. Sci. Eng.* **2019**, *561*, 012105. [CrossRef]
20. Aravindan, V.; Gnanaraj, J.; Lee, Y.S.; Madhavi, S. Insertion-type electrodes for nonaqueous Li-ion capacitors. *Chem. Rev.* **2014**, *114*, 11619–11635. [CrossRef]
21. Deng, W.N.; Li, Y.H.; Xu, D.F.; Zhou, W.; Xiang, K.X.; Chen, H. Three-dimensional hierarchically porous nitrogen-doped carbon from water hyacinth as selenium host for high-performance lithium–selenium batteries. *Rare Met.* **2022**, *41*, 3432–3445. [CrossRef]
22. Wei, L.; Deng, W.; Li, S.; Wu, Z.; Cai, J.; Luo, J. Sandwich-like chitosan porous carbon Spheres/MXene composite with high specific capacitance and rate performance for supercapacitors. *J. Bioresour. Bioprod.* **2022**, *7*, 63–72. [CrossRef]
23. Ho, M.Y.; Khiew, P.S.; Isa, D.; Tan, T.K.; Chiu, W.S.; Chia, C.H. A review of metal oxide composite electrode materials for electrochemical capacitors. *Nano* **2014**, *9*, 1430002. [CrossRef]
24. Halper, M.S.; Ellenbogen, J.C. *Supercapacitors: A Brief Overview*; MITRE: McLean, VA, USA, 2006.
25. Jayalakshmi, M.; Balasubramanian, K. Simple capacitors to supercapacitors—An overview. *Int. J. Electrochem. Sci.* **2008**, *3*, 1196–1217. [CrossRef]
26. Kiamahalleh, M.V.; Zein, S.H.S.; Najafpour, G.; Sata, S.A.; Buniran, S. Multiwalled carbon nanotubes based nanocomposites for supercapacitors: A review of electrode materials. *Nano* **2012**, *7*, 1230002. [CrossRef]
27. Choi, H.; Yoon, H. Nanostructured electrode materials for electrochemical capacitor applications. *Nanomaterials* **2015**, *5*, 906–936. [CrossRef]
28. Salunkhe, R.R.; Kaneti, Y.V.; Kim, J.; Kim, J.H.; Yamauchi, Y. Nanoarchitectures for Metal-Organic Framework-Derived Nanoporous Carbons toward Supercapacitor Applications. *Acc. Chem. Res.* **2016**, *49*, 2796–2806. [CrossRef]
29. Tang, J.; Salunkhe, R.R.; Liu, J.; Torad, N.L.; Imura, M.; Furukawa, S.; Yamauchi, Y. Thermal conversion of core-shell metal-organic frameworks: A new method for selectively functionalized nanoporous hybrid carbon. *J. Am. Chem. Soc.* **2015**, *137*, 1572–1580. [CrossRef] [PubMed]

30. Gu, W.; Yushin, G. Review of nanostructured carbon materials for electrochemical capacitor applications: Advantages and limitations of activated carbon, carbide-derived carbon, zeolite-templated carbon, carbon aerogels, carbon nanotubes, onion-like carbon, and graphene. *Wiley Interdiscip. Rev. Energy Environ.* **2014**, *3*, 424–473. [CrossRef]
31. Mohapatra, S.; Acharya, A.; Roy, G.S. The role of nanomaterial for the design of supercapacitors. *Lat. Am. J. Phys. Educ.* **2012**, *6*, 380–384.
32. Beidaghi, M.; Wang, C. Micro-supercapacitors based on interdigital electrodes of reduced graphene oxide and carbon nanotube composites with ultrahigh power handling performance. *Adv. Funct. Mater.* **2012**, *22*, 4501–4510. [CrossRef]
33. Chen, T.; Dai, L. Carbon nanomaterials for high-performance supercapacitors. *Mater. Today* **2013**, *16*, 272–280. [CrossRef]
34. Yan, Y.; Wang, T.; Li, X.; Pang, H.; Xue, H. Noble metal-based materials in high-performance supercapacitors. *Inorg. Chem. Front.* **2017**, *4*, 33–51. [CrossRef]
35. Bao, L.; Zang, J.; Li, X. Flexible Zn_2SnO_4/MnO_2 core/shell nanocable-carbon microfiber hybrid composites for high-performance supercapacitor electrodes. *Nano Lett.* **2011**, *11*, 1215–1220. [CrossRef]
36. Ghosh, D.; Giri, S.; Moniruzzaman, M.; Basu, T.; Mandal, M.; Das, C.K. α MnMoO4/graphene hybrid composite: High energy density supercapacitor electrode material. *Dalt. Trans.* **2014**, *43*, 11067–11076. [CrossRef] [PubMed]
37. Neiber, R.R.; Kumar, J.; Soomro, R.A.; Karkus, S.; Abo-Dief, H.M.; Alanazi, A.K.; Altalhia, A.A.; El-Bahy, Z.M. $NiZnCoO_4/CoWO_4$ hybrid composite with improved electrochemical performance for supercapacitor application. *J. Energy Storage* **2022**, *52*, 104900. [CrossRef]
38. Iro, Z.S.; Subramani, C.; Dash, S.S. A brief review on electrode materials for supercapacitor. *Int. J. Electrochem. Sci.* **2016**, *11*, 10628–10643. [CrossRef]
39. Wu, N.; Bai, X.; Pan, D.; Dong, B.; Wei, R.; Naik, N.; Patil, R.R.; Guo, Z. Recent Advances of Asymmetric Supercapacitors. *Adv. Mater. Interfaces* **2021**, *8*, 2001710. [CrossRef]
40. Zheng, S.; Wu, Z.S.; Wang, S.; Xiao, H.; Zhou, F.; Sun, C.; Bao, X.; Cheng, H.M. Graphene-based materials for high-voltage and high-energy asymmetric supercapacitors. *Energy Storage Mater.* **2017**, *6*, 70–97. [CrossRef]
41. Shao, Y.; El-Kady, M.F.; Sun, J.; Li, Y.; Zhang, Q.; Zhu, M.; Wang, H.; Dunn, B.; Kaner, R.B. Design and Mechanisms of Asymmetric Supercapacitors. *Chem. Rev.* **2018**, *118*, 9233–9280. [CrossRef]
42. Stoller, M.D.; Ruoff, R.S. Best practice methods for determining an electrode material's performance for ultracapacitors. *Energy Environ. Sci.* **2010**, *3*, 1294–1301. [CrossRef]
43. Balducci, A.; Dugas, R.; Taberna, P.; Simon, P.; Plee, D.; Mastragostino, M.; Passerini, S.; Balducci, A.; Dugas, R.; Taberna, P.; et al. High temperature carbon—Carbon supercapacitor using ionic liquid as electrolyte. *J. Power Sources* **2007**, *165*, 922–927. [CrossRef]
44. Kreysa, G.; Ota, K.-I.; Savinell, R.F. *Encyclopedia of Applied Electrochemistry*; Springer: New York, NY, USA, 2014; ISBN 9781441969965.
45. Mahankali, K.; Thangavel, N.K.; Ding, Y.; Putatunda, S.K.; Arava, L.M.R. Interfacial behavior of water-in-salt electrolytes at porous electrodes and its effect on supercapacitor performance. *Electrochim. Acta* **2019**, *326*, 134989. [CrossRef]
46. Liu, X.; Xu, W.; Zheng, D.; Li, Z.; Zeng, Y.; Lu, X. Carbon cloth as an advanced electrode material for supercapacitors: Progress and challenges. *J. Mater. Chem. A* **2020**, *8*, 17938–17950. [CrossRef]
47. Tutlienė, S.; Petrulevičienė, M.; Pilipavičius, J.; Žarkov, A.; Selskis, A.; Stanionytė, S.; Juodkazytė, J.; Vilčiauskas, L. Electrochemical Performance of Sol-Gel Synthesized $NaTi_2$ $(PO_4)_3$—Carbon Composites as Aqueous Na-Ion Battery Anodes. *J. Electrochem. Soc.* **2021**, *168*, 060545. [CrossRef]
48. Lan, G.; Yang, J.; Ye, R.P.; Boyjoo, Y.; Liang, J.; Liu, X.; Li, Y.; Liu, J.; Qian, K. Sustainable Carbon Materials toward Emerging Applications. *Small Methods* **2021**, *5*, 2001250. [CrossRef] [PubMed]
49. Pandolfo, A.G.; Hollenkamp, A.F. Carbon properties and their role in supercapacitors. *J. Power Sources* **2006**, *157*, 11–27. [CrossRef]
50. Syarif, N.; Tribidasari, A.; Wibowo, W. Direct Synthesis Carbon/Metal Oxide Composites for Electrochemical Capacitors Electrode. *Int. Trans. J. Eng. Manag. Appl. Sci. Technol.* **2012**, *3*, 21–34.
51. Lota, K.; Sierczynska, A.; Acznik, I.; Lota, G. Effect of aqueous electrolytes on electrochemical capacitor capacitance. *Chemik* **2013**, *67*, 1138–1145.
52. Lufrano, F.; Staiti, P. Mesoporous carbon materials as electrodes for electrochemical supercapacitors. *Int. J. Electrochem. Sci.* **2010**, *5*, 903–916. [CrossRef]
53. Tamilarasan, P.; Mishra, A.K.; Ramaprabhu, S. Graphene/ionic liquid binary electrode material for high performance supercapacitor. In Proceedings of the 2011 International Conference on Nanoscience, Technology and Societal Implications, Bhubaneswar, India, 8–10 December 2011; pp. 1–5. [CrossRef]
54. Miller, J.R.; Outlaw, R.A.; Holloway, B.C. Graphene electric double layer capacitor with ultra-high-power performance. *Electrochim. Acta* **2011**, *56*, 10443–10449. [CrossRef]
55. Kim, T.Y.; Lee, H.W.; Stoller, M.; Dreyer, D.R.; Bielawski, C.W.; Ruoff, R.S.; Suh, K.S. High-performance supercapacitors based on poly(ionic liquid)-modified graphene electrodes. *ACS Nano* **2011**, *5*, 436–442. [CrossRef]
56. Apriwandi, A.; Taer, E.; Farma, R.; Setiadi, R.N.; Amiruddin, E. A facile approach of micro-mesopores structure binder-free coin/monolith solid design activated carbon for electrode supercapacitor. *J. Energy Storage* **2021**, *40*, 102823. [CrossRef]

57. Gou, G.; Huang, F.; Jiang, M.; Li, J.; Zhou, Z. Hierarchical porous carbon electrode materials for supercapacitor developed from wheat straw cellulosic foam. *Renew. Energy* **2020**, *149*, 208–216. [CrossRef]
58. Du, J.; Zhang, Y.; Lv, H.; Hou, S.; Chen, A. Yeasts-derived nitrogen-doped porous carbon microcapsule prepared by silica-confined activation for supercapacitor. *J. Colloid Interface Sci.* **2021**, *601*, 467–473. [CrossRef]
59. Zhao, X.Y.; Huang, S.S.; Cao, J.P.; Xi, S.C.; Wei, X.Y.; Kamamoto, J.; Takarada, T. KOH activation of a HyperCoal to develop activated carbons for electric double-layer capacitors. *J. Anal. Appl. Pyrolysis* **2014**, *105*, 116–121. [CrossRef]
60. Zhao, X.Y.; Huang, S.S.; Cao, J.P.; Wei, X.Y.; Magarisawa, K.; Takarada, T. HyperCoal-derived porous carbons with alkaline hydroxides and carbonate activation for electric double-layer capacitors. *Fuel Process. Technol.* **2014**, *125*, 251–257. [CrossRef]
61. Zhang, W.L.; Xu, J.H.; Hou, D.X.; Yin, J.; Liu, D.B.; He, Y.P.; Lin, H.B. Hierarchical porous carbon prepared from biomass through a facile method for supercapacitor applications. *J. Colloid Interface Sci.* **2018**, *530*, 338–344. [CrossRef]
62. Zhao, G.; Yu, D.; Zhang, H.; Sun, F.; Li, J.; Zhu, L.; Sun, L.; Yu, M.; Besenbacher, F.; Sun, Y. Sulphur-doped carbon nanosheets derived from biomass as high-performance anode materials for sodium-ion batteries. *Nano Energy* **2020**, *67*, 104219. [CrossRef]
63. Luo, H.; Chen, M.; Cao, J.; Zhang, M.; Tan, S.; Wang, L.; Zhong, J.; Deng, H.; Zhu, J.; Lu, B. Cocoon Silk-Derived, Hierarchically Porous Carbon as Anode for Highly Robust Potassium-Ion Hybrid Capacitors. *Nano-Micro Lett.* **2020**, *12*, 113. [CrossRef] [PubMed]
64. Jain, A.; Balasubramanian, R.; Srinivasan, M.P. Hydrothermal conversion of biomass waste to activated carbon with high porosity: A review. *Chem. Eng. J.* **2016**, *283*, 789–805. [CrossRef]
65. Gao, F.; Shao, G.; Qu, J.; Lv, S.; Li, Y.; Wu, M. Tailoring of porous and nitrogen-rich carbons derived from hydrochar for high-performance supercapacitor electrodes. *Electrochim. Acta* **2015**, *155*, 201–208. [CrossRef]
66. Huang, Y.; Tang, Z.; Zhou, S.; Wang, H.; Tang, Y.; Sun, D.; Wang, H. Renewable waste biomass-derived carbon materials for energy storage. *J. Phys. D Appl. Phys.* **2022**, *55*, 313002. [CrossRef]
67. Gogoi, D.; Makkar, P.; Das, M.R.; Ghosh, N.N. $CoFe_2O_4$ Nanoparticle Decorated Hierarchical Biomass Derived Porous Carbon Based Nanocomposites for High-Performance All-Solid-State Flexible Asymmetric Supercapacitor Devices. *ACS Appl. Electron. Mater.* **2022**, *4*, 795–806. [CrossRef]
68. Cai, J.; Zhang, D.; Ding, W.P.; Zhu, Z.Z.; Wang, G.Z.; He, J.R.; Wang, H.B.; Fei, P.; Si, T.L. Promising rice-husk-derived carbon/$Ni(OH)_2$ composite materials as a high-performing supercapacitor electrode. *ACS Omega* **2020**, *5*, 29896–29902. [CrossRef]
69. Jin, T.; Su, J.; Luo, Q.; Zhu, W.; Lai, H.; Huang, D.; Wang, C. Preparation of N,P Co-doped Porous Carbon Derived from Daylily for Supercapacitor Applications. *ACS Omega* **2022**, *7*, 37564–37571. [CrossRef] [PubMed]
70. Kreno, L.E.; Leong, K.; Farha, O.K.; Allendorf, M.; Van Duyne, R.P.; Hupp, J.T. Metal-organic framework materials as chemical sensors. *Chem. Rev.* **2012**, *112*, 1105–1125. [CrossRef]
71. Islamoglu, T.; Goswami, S.; Li, Z.; Howarth, A.J.; Farha, O.K.; Hupp, J.T. Postsynthetic Tuning of Metal-Organic Frameworks for Targeted Applications. *Acc. Chem. Res.* **2017**, *50*, 805–813. [CrossRef]
72. Xiao, J.D.; Jiang, H.L. Metal-Organic Frameworks for Photocatalysis and Photothermal Catalysis. *Acc. Chem. Res.* **2019**, *52*, 356–366. [CrossRef]
73. Salunkhe, R.R.; Kamachi, Y.; Torad, N.L.; Hwang, S.M.; Sun, Z.; Dou, S.X.; Kim, J.H.; Yamauchi, Y. Fabrication of symmetric supercapacitors based on MOF-derived nanoporous carbons. *J. Mater. Chem. A* **2014**, *2*, 19848–19854. [CrossRef]
74. Torad, N.L.; Hu, M.; Kamachi, Y.; Takai, K.; Imura, M.; Naito, M.; Yamauchi, Y. Facile synthesis of nanoporous carbons with controlled particle sizes by direct carbonization of monodispersed ZIF-8 crystals. *Chem. Commun.* **2013**, *49*, 2521–2523. [CrossRef]
75. Yu, J.; Fu, N.; Zhao, J.; Liu, R.; Li, F.; Du, Y.; Yang, Z. High Specific Capacitance Electrode Material for Supercapacitors Based on Resin-Derived Nitrogen-Doped Porous Carbons. *ACS Omega* **2019**, *4*, 15904–15911. [CrossRef]
76. Iyyamperumal, E.; Wang, S.; Dai, L. Vertically aligned BCN nanotubes with high capacitance. *ACS Nano* **2012**, *6*, 5259–5265. [CrossRef] [PubMed]
77. Wang, X.; Lee, J.S.; Zhu, Q.; Liu, J.; Wang, Y.; Dai, S. Ammonia-treated ordered mesoporous carbons as catalytic materials for oxygen reduction reaction. *Chem. Mater.* **2010**, *22*, 2178–2180. [CrossRef]
78. Qie, L.; Chen, W.M.; Wang, Z.H.; Shao, Q.G.; Li, X.; Yuan, L.X.; Hu, X.L.; Zhang, W.X.; Huang, Y.H. Nitrogen-doped porous carbon nanofiber webs as anodes for lithium ion batteries with a superhigh capacity and rate capability. *Adv. Mater.* **2012**, *24*, 2047–2050. [CrossRef] [PubMed]
79. Lao, J.; Lu, Y.; Fang, S.; Xu, F.; Sun, L.; Wang, Y.; Zhou, T.; Liao, L.; Guan, Y.; Wei, X.; et al. Organic Crosslinked Polymer-Derived N/O-Doped Porous Carbons for High-Performance Supercapacitor. *Nanomaterials* **2022**, *12*, 2186. [CrossRef] [PubMed]
80. Sun, G.; Ren, H.; Shi, Z.; Zhang, L.; Wang, Z.; Zhan, K.; Yan, Y.; Yang, J.; Zhao, B. V_2O_5/vertically-aligned carbon nanotubes as negative electrode for asymmetric supercapacitor in neutral aqueous electrolyte. *J. Colloid Interface Sci.* **2021**, *588*, 847–856. [CrossRef]
81. Miao, T.; Zhang, J.; Wang, Y.; Fang, K.; Wang, Z.; Zhan, K.; Zhao, B. Composite cathode with low-defect NiFe Prussian blue analogue on reduced graphene oxide for aqueous sodium-ion hybrid supercapacitors. *J. Colloid Interface Sci.* **2023**, *648*, 768–777. [CrossRef] [PubMed]
82. Ren, H.; Zhang, L.; Zhang, J.; Miao, T.; Yuan, R.; Chen, W.; Wang, Z.; Yang, J.; Zhao, B. Na^+ pre-intercalated $Na_{0.11}MnO_2$ on three-dimensional graphene as cathode for aqueous zinc ion hybrid supercapacitor with high energy density. *Carbon N. Y.* **2022**, *198*, 46–56. [CrossRef]

83. Shi, Z.; Sun, G.; Yuan, R.; Chen, W.; Wang, Z.; Zhang, L.; Zhan, K.; Zhu, M.; Yang, J.; Zhao, B. Scalable fabrication of $NiCo_2O_4$/reduced graphene oxide composites by ultrasonic spray as binder-free electrodes for supercapacitors with ultralong lifetime. *J. Mater. Sci. Technol.* **2022**, *99*, 260–269. [CrossRef]
84. Xu, J.; Sun, Y.; Lu, M.; Wang, L.; Zhang, J.; Qian, J.; Liu, X. Fabrication of hierarchical $MnMoO_4 \cdot H_2O@MnO_2$ core-shell nanosheet arrays on nickel foam as an advanced electrode for asymmetric supercapacitors. *Chem. Eng. J.* **2018**, *334*, 1466–1476. [CrossRef]
85. Xu, K.; Yang, J.; Hu, J. Synthesis of hollow $NiCo_2O_4$ nanospheres with large specific surface area for asymmetric supercapacitors. *J. Colloid Interface Sci.* **2018**, *511*, 456–462. [CrossRef]
86. Hussain, I.; Mak, T.; Zhang, K. Boron-Doped Trimetallic Cu–Ni–Co Oxide Nanoneedles for Supercapacitor Application. *ACS Appl. Nano Mater.* **2021**, *4*, 129–141. [CrossRef]
87. Zhou, X.; Dai, H.; Huang, X.; Ren, Y.; Wang, Q.; Wang, W.; Huang, W.; Dong, X. Porous trimetallic fluoride Ni–Co–M (M = Mn, Fe, Cu, Zn) nanoprisms as electrodes for asymmetric supercapacitors. *Mater. Today Energy* **2020**, *17*, 100429. [CrossRef]
88. Cottineau, T.; Toupin, M.; Delahaye, T.; Brousse, T.; Bélanger, D. Nanostructured transition metal oxides for aqueous hybrid electrochemical supercapacitors. *Appl. Phys. A Mater. Sci. Process.* **2006**, *82*, 599–606. [CrossRef]
89. Guo, X.L.; Li, G.; Kuang, M.; Yu, L.; Zhang, Y.X. Tailoring Kirkendall effect of the KCu7S4 microwires towards $CuO@MnO_2$ core-shell nanostructures for supercapacitors. *Electrochim. Acta* **2015**, *174*, 87–92. [CrossRef]
90. Zheng, Y.; Li, Z.; Xu, J.; Wang, T.; Liu, X.; Duan, X.; Ma, Y.; Zhou, Y.; Pei, C. Multi-channeled hierarchical porous carbon incorporated Co_3O_4 nanopillar arrays as 3D binder-free electrode for high performance supercapacitors. *Nano Energy* **2016**, *20*, 94–107. [CrossRef]
91. Tian, Z.; Li, H.; Yang, W.; Jian, S.; Zhang, C.; Jiang, S. Fabrication of manganese oxides/carbon composites for high energy density asymmetric supercapacitor. *Diam. Relat. Mater.* **2023**, *131*, 109582. [CrossRef]
92. Chen, Y.; Yang, H.; Han, Z.; Bo, Z.; Yan, J.; Cen, K.; Ostrikov, K.K. MXene-Based Electrodes for Supercapacitor Energy Storage. *Energy Fuels* **2022**, *36*, 2390–2406. [CrossRef]
93. Sugahara, A.; Ando, Y.; Kajiyama, S.; Yazawa, K.; Gotoh, K.; Otani, M.; Okubo, M.; Yamada, A. Negative dielectric constant of water confined in nanosheets. *Nat. Commun.* **2019**, *10*, 1–7. [CrossRef]
94. Hu, M.; Hu, T.; Li, Z.; Yang, Y.; Cheng, R.; Yang, J.; Cui, C.; Wang, X. Surface Functional Groups and Interlayer Water Determine the Electrochemical Capacitance of Ti_3C_2 T x MXene. *ACS Nano* **2018**, *12*, 3578–3586. [CrossRef]
95. Boota, M.; Chen, C.; Van Aken, K.L.; Jiang, J.; Gogotsi, Y. Organic-inorganic all-pseudocapacitive asymmetric energy storage devices. *Nano Energy* **2019**, *65*, 104022. [CrossRef]
96. Luo, J.; Zhang, W.; Yuan, H.; Jin, C.; Zhang, L.; Huang, H.; Liang, C.; Xia, Y.; Zhang, J.; Gan, Y.; et al. Pillared Structure Design of MXene with Ultralarge Interlayer Spacing for High-Performance Lithium-Ion Capacitors. *ACS Nano* **2017**, *11*, 2459–2469. [CrossRef] [PubMed]
97. Ma, R.; Zhang, X.; Zhuo, J.; Cao, L.; Song, Y.; Yin, Y.; Wang, X.; Yang, G.; Yi, F. Self-Supporting, Binder-Free, and Flexible Ti_3C_2TxMXene-Based Supercapacitor Electrode with Improved Electrochemical Performance. *ACS Nano* **2022**, *16*, 9713–9727. [CrossRef]
98. Shi, B.; Chen, L.; Jen, T.C.; Liu, X.; Li, L.; Chen, A.; Shen, G. Vertical Arrangement of Ti_2CT_x MXene Nanosheets on Carbon Fibers for High-Performance and Flexible Zn-Ion Supercapacitors. *ACS Appl. Nano Mater.* **2023**, *6*, 315–322. [CrossRef]
99. Farma, R.; Indriani, A.; Apriyani, I. Hierarchical-nanofiber structure of biomass-derived carbon framework with direct CO_2 activation for symmetrical supercapacitor electrodes. *J. Mater. Sci. Mater. Electron.* **2023**, *34*, 81. [CrossRef]
100. Palanisamy, R.; Karuppiah, D.; Rengapillai, S.; Abdollahifar, M.; Ramasamy, G.; Wang, F.M.; Liu, W.R.; Ponnuchamy, K.; Shim, J.; Marimuthu, S. A reign of bio-mass derived carbon with the synergy of energy storage and biomedical applications. *J. Energy Storage* **2022**, *51*, 104422. [CrossRef]
101. Wu, J.; Zhou, W.; Jiang, F.; Chang, Y.; Zhou, Q.; Li, D.; Ye, G.; Li, C.; Nie, G.; Xu, J.; et al. Three-Dimensional Porous Carbon Derived from Polyindole Hollow Nanospheres for High-Performance Supercapacitor Electrode. *ACS Appl. Energy Mater.* **2018**, *1*, 4572–4579. [CrossRef]
102. Zhu, T.; Zhou, J.; Li, Z.; Li, S.; Si, W.; Zhuo, S. Hierarchical porous and N-doped carbon nanotubes derived from polyaniline for electrode materials in supercapacitors. *J. Mater. Chem. A* **2014**, *2*, 12545–12551. [CrossRef]
103. Shu, Y.; Maruyama, J.; Iwasaki, S.; Maruyama, S.; Shen, Y.; Uyama, H. Fabrication of N-doped and shape-controlled porous monolithic carbons from polyacrylonitrile for supercapacitors. *RSC Adv.* **2017**, *7*, 43172–43180. [CrossRef]
104. Liu, S.; Kang, L.; Hu, J.; Jung, E.; Zhang, J.; Jun, S.C.; Yamauchi, Y. Unlocking the Potential of Oxygen-Deficient Copper-Doped Co3O4Nanocrystals Confined in Carbon as an Advanced Electrode for Flexible Solid-State Supercapacitors. *ACS Energy Lett.* **2021**, *6*, 3011–3019. [CrossRef]
105. Yesuraj, J.; Vajravijayan, S.; Yang, R.; Nandhagopal, N.; Gunasekaran, K.; Selvam, N.C.S.; Yoo, P.J.; Kim, K. Self-Assembly of Hausmannite Mn_3O_4Triangular Structures on Cocosin Protein Scaffolds for High Energy Density Symmetric Supercapacitor Application. *Langmuir* **2022**, *38*, 2928–2941. [CrossRef] [PubMed]
106. Hariharan, G.; Arunpandiyan, S.; Shanmugapriya, V.; Bharathi, S.; Babu, M.; Selvakumar, B.; Arivarasan, A. Role of various redox additive electrolytes on the electrochemical performances of mixed metal oxide loaded multiwalled carbon nanotube based supercapacitors. *J. Energy Storage* **2023**, *57*, 106178. [CrossRef]
107. Gouda, M.S.; Shehab, M.; Helmy, S.; Soliman, M.; Salama, R.S. Nickel and cobalt oxides supported on activated carbon derived from willow catkin for efficient supercapacitor electrode. *J. Energy Storage* **2023**, *61*, 106806. [CrossRef]

108. Luo, W.; Sun, Y.; Lin, Z.; Li, X.; Han, Y.; Ding, J.; Li, T.; Hou, C.; Ma, Y. Flexible $Ti_3C_2T_x$ MXene/V_2O_5 composite films for high-performance all-solid supercapacitors. *J. Energy Storage* **2023**, *62*, 106807. [CrossRef]
109. Lin, Z.; Li, X.; Li, S.; Li, B.; Ding, J.; Han, Y.; Li, T.; Hou, C.; Ma, Y. Highly flexible, foldable carbon cloth/MXene/polyaniline/CoNi layered double hydroxide electrode for high-performance all solid-state supercapacitors. *J. Energy Storage* **2023**, *64*, 107116. [CrossRef]