Flexible Electrochemical Supercapacitors: A Review

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https://doi.org/10.29072/basjs.20240103

ARTICLE INFO

Keywords

Flexible Supercapacitor, Electrochemical, Electrode materials, Electrolytes, Nanostructure

ABSTRACT

Flexible supercapacitors (FSCs), especially wearable ones, have become a hotspot of research because of their advantages such as energy density, high specific capacitance, good mechanical properties, decent stability, and eco-friendliness. The storage mechanism in FSCs is either a faradaic mechanism in pseudocapacitors (PCs) or a non-faradaic mechanism as in double-layer supercapacitors (ELSCs). This review summarized the research progress for manufacturing electrodes from various types of materials, whereas carbon, metal oxides, or conductive polymers are utilized as electrodes. An overview of what researchers have performed in changing the original shape of materials and combining them to obtain flexible electrodes with good electrochemical performances. The review includes the presence of different types of electrolytes in the FSCs structure. Perspectives on the direction of future development of research in the field of FSCs are also discussed.

Received 16 Apr 2024; Received in revised form 27 Ap 2024; Accepted 29 Apr 2024, Published 30 Apr 2024

1. Introduction

With the rapid development in the present scenario of the global economy, the difficulty of providing a pollution-free environment has become a matter of utmost importance, and this is linked to energy sources. Therefore, the easy availability of traditional fossil energy sources (extraction, available industry, stable price) has made it one of the most common sources of energy generation [1]. One of the disadvantages of fossil energy is the slow renewal of its source, which requires very long years[2]. With the sharp increase in energy consumption in our current era, non-renewable energy sources are no longer fully meeting human needs[3]. Therefore, the need has emerged to search for a source of renewable energy. For instance, solar energy, wind energy, and other renewable type of energy are relatively clean, easy to store and transport, and have applications in all features of life [4]. These energy sources must require efficient electrical energy storage equipment. The aforementioned have made the energy storage topic a hot research area. Storage devices play a vital role in providing a continuous power supply. There are many energy storage devices including lithium-ion batteries, hydrogen fuel cells, and supercapacitors[5-8]. It is known that flexible, wearable and moveable electronics, energy storage systems are the base of many applications e.g., phones, health monitoring systems, computers, and TV screens. Although, flexible lithium-ion batteries have been applied in standards industry and market accessibility[9, 10], their mechanical performances were not sufficient and also limited in size, making it difficult to place them in wearable devices such as personal clothing and watches[11, 12]. Unlike lithium-ion batteries, supercapacitor features fast charge and discharge and good bending and twisting ability, which can be used in wearable devices due to their low cost, environmental friendliness, high stability, and excellent electrochemical performance[13]. Although many studies have shown that the power density of SC is relatively low compared to other devices, it can be further improved by researching new materials to enhance the performance of electrode materials and electrolytic fluids. The researchers aim to build a flexible SC with good size and mechanical properties including bending and torsion. Besides these properties, the SCs must have high capacitance capacity as well as high energy and power densities [14, 15].Supercapacitors have been divided into many types, including what are called self-charging supercapacitors(SCS), color supercapacitors(CS), energy storage supercapacitors(ESS), self-repairing supercapacitors(SRS), and others depending on the use[16]. Flexible supercapacitors (FSCs) and their applications have been broadly considered as energy storage devices. FSCs are different from conventional supercapacitors since FSC electrodes are covered with flexible material, allowing them move in a different direction without affecting their performance [17]. Based on their charge storage mechanism, SCs can be categorized into double-layer electrolytic capacitors (EDLCs) and pseudocapacitors (PC). The EDLC one typically uses a non-faradic electrostatic process to store energy that originates from the charges accumulated at the interface between the electrode and the electrolytic fluid. The PC one uses the faradic redox depending on the interaction process at the electrode/electrolytic interface. In addition, the hybrid supercapacitors (HSC) include the combination of EDLCs and PC [18, 19]. To evaluate the electrochemical performance of the SCs, there are many criteria including the nature of the electrode material and the electrolytic fluid, and also the working environment[20, 21]. Often, the electrode is manufactured in one of two ways. The first way includes using a flexible substrate made of a porous material that can conduct electrical current or a material with high electrochemical properties is grown or cast on top of the flexible substrate[22-24]. About the second axis, the development of electrode material is considered a desirable axiom with a high-performance material strip with a good surface area, while providing highly exposed surface reactive sites[25, 26]. There are various groups of materials with false capacitance such as carbonaceous materials. Many materials have received attention due to the development of materials science[27].This review proposes to highlight the research progress of different types of FSCs in recent years. This review focuses on the terms of defining the basic working principles, analyzing the problems facing research progress at the current stage, proposing some future solutions, and ways to improve their performance.

2. Strategies to prepare the Flexible electrode

It is known that energy storage for any type of supercapacitor, whether flexible or inflexible, occurs through rapid redox interaction between the electrode and electrolyte fluid or through the formation of a non-faradic double layer that collects charges. Therefore, the electrode material is considered one of the important parts that play an important role in determining the performance of a flexible coiled supercapacitor. Researchers may face major challenges in designing electrodes in terms of material cost and capacity efficiency. The material that is manufactured for a flexible supercapacitor must have foldable capability. The material is cast on a flexible substrate to make an electrode used in a flexible supercapacitor[28]. FSCs may depend in general on electrochemical properties, including (cycle life, capacitance, energy density, and power density), and flexibility may include these properties as a property of the electrode materials. To prepare the electrode, the design must take into account the microstructure and some properties specific to the nature of the materials to produce efficient materials. High good conductivity, storage capacity, increased redox activity, and increased interaction between the electrolyte and the manufactured materials, including the electrode. To increase the surface area, the pore size is widely distributed, and small-sized materials known as nanostructured materials are used. According to the storage mechanism in capacitors, superior electronic EDLC materials are classified according to their advantages and disadvantages into transition metal compounds, pseudomaterials[29], carbon materials, and conductive polymers. The thickness of the electrode plays an important role in the occurrence of what is called mechanical strain. The reason is due to the difference in the size of the charged /discharged states, and this causes the electrode to disintegrate quickly and reduces performance. As the thickness increases, the mechanical strain increases[30]. To improve electrochemical performance, strategies for electrode materials through the basic principles of creating various microscopic structures such as structures with dimensions (0D, 1D, 2D, 3D), porous structures, nanostructures, and manufacturing and building various compounds from various materials that have elastic properties. Figure 1 reveals the materials that have been used as electrodes for FCSs.

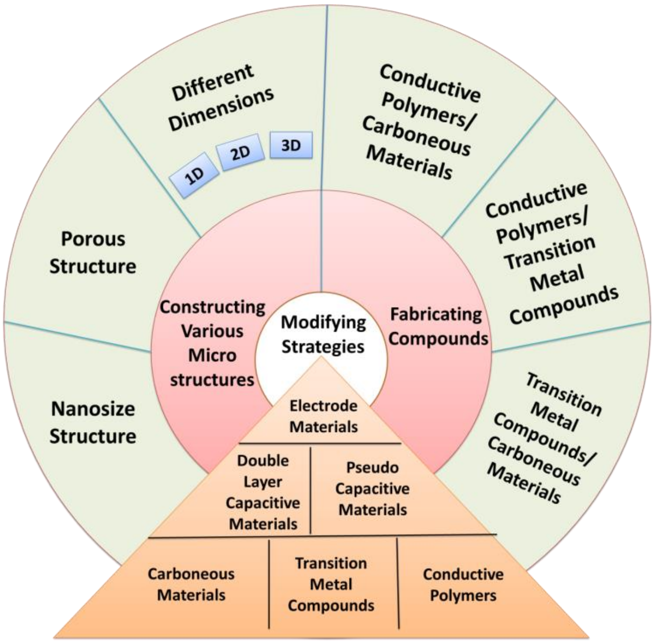

Figure 1: Strategies used to enhance the electrochemical performance of FSCs.

In general, activated carbon is manufactured from organic materials (raw materials), meaning it has a high percentage of carbon that does not contain inorganic materials such as carbon nanofibers, and graphene[31, 32]. Carbon nanotubes (CNTs) and other carbon-based materials which are similar in their structure to honeycombs, are considered good conductors of electrical current. They have good chemical stability and high surface area, as well as mechanical performance and tolerance to high temperatures, making them promising candidates[33, 34].

These materials can be used in a supercapacitor either as flexible or inflexible. However, they have a low capacity compared to other materials, and this affects the energy density, so it may limit their application [35].To improve the capacity to reach the false capacity, the surface of these materials is modified through the manufacture of carbon nanostructures to accelerate the diffusion of ions, whether by surface activation or through exfoliation. To enhance the polarizability and produce additional PC carbon materials are combined with heteroatoms[36]. Rarely, carbon fibers especially elastic textiles are inserted as electrodes due to their low electrochemical properties and small surface area. To convert carbon fiber, it acts as an electrode by creating pyramidal pores, as it has a large capacity compared to carbon fiber[37]. Conductive polymers such as polythiophene (PTh), polypyrrole (PPy), and polyaniline (PANI) provide advantages for electrode materials, including high conductivity, high resistance, ease of fabrication, and high redox activity which are adjustable with a higher operating window compared to some materials due to their low cost and eco-friendly properties[28, 38]. Charge storage occurs through oxidation and reduction reactions by transferring ions into the polymer backbone and releasing them into the electrolyte due to the occurrence of oxidation and reduction reactions in all the electrolyte materials, so it is noticeable that the capacity of conductive polymers is higher than the rest of the capacities of the electrodes of other materials. However, the flexibility of the polymer may reduce the conductivity of the polymer and also reduce the redox activity, and this may cause a deterioration in the stability of the flexible supercapacitor. Conductive polymers are charged depending on the type of ion input (positive or negative)[39].

Transition metal compounds, as another type of pseudocapacitive materials, are based on hydroxides, nitrides, oxides, sulfides, and others. These materials are considered inexpensive, chemically stable, and easy to handle. By changing the synthesis process, it is possible to obtain many different morphologies of individual metal oxides[40-42], since the metal oxide capacitance stores electrical charges in a faradic manner, it must be characterized by high conductivity to transfer electrons without interruption and also have multiple oxidation activity, whether in two or more states. However, their limited cycle life and low capacity are due to their poor electrical conductivity and reversibility [43]. Many electrodes are made of metal oxides that have been studied by researchers such as $NiCo_2O_4$, $NiO_2/Ni(OH)_2$, RuO_2, and others. The capacity of transition metal oxide materials may be higher compared to carbon materials. Although their application in advanced applications may be limited due to their low stability and poor conductivity, nanostructured transition metal oxides can be optimized with Ag quantum dots to enhance the electroactive region, electron-ion mobility, electrical conductivity, and chemical stability, leading to exceptional pseudocapacitive results[44, 45].

3. Types of electrolytes for electrochemical supercapacitor

Increasing the number and accelerating the electron transfer rate are the ways to obtain a high surface area to ensure a good performance rate. This is necessary for the manufacturer to improve the electrode materials used in the formation of a flexible supercapacitor. To enhance the electrochemical performance, different strategies are adopted to increase the surface area of the electrode material and the redox activity. All of these are supposed to be accompanied by increasing the stability of the device and shortening the path of arrival of ions in the case of diffusion into the electrolyte liquid or electrolyte materials. For instance, the electrodes are designed with good morphological structures that have flexible properties and high capacity and energy density[46]. In addition, choosing an appropriate electrolyte that is used in the flexible supercapacitor is considered a necessary factor because it is the main controller in displaying the capacitive properties as it is responsible for the production of ions when dissolution occurs in the solution. It is also responsible for balancing and transferring charges between the two electrodes which are the main factors for the activity oxidation and reduction increases in energy density[47]. The electrolyte must have high potential in terms of high conductivity, good compatibility with the electrode, environmental friendliness, and ideal viscosity. The concentration and size of ions in the electrolyte and their interaction with the electrode materials determine the performance of the electrolyte, i.e. their corrosion behavior and conductivity [28]. There are several types of electrolytes, including (1) aqueous electrolytes, (2) non-aqueous electrolytes, which have two types (organic electrolytes and ionic liquids), and (3) solid-state or solid-state electrolytes, which may include semi-solid electrolytes that have several types (solid/dry polymer electrolytes, polymer gel electrolytes, inorganic solid-state electrolytes, and polycarbonate electrolytes)[48]. One of the advantages of aqueous electrolytes is that they are easy to prepare and inexpensive, as aqueous solution is used as a solvent, and it has good conductivity compared to other electrolytes, which enhances the energy density of the supercapacitor. However, one of the disadvantages of flexible supercapacitors is the potential leakage and narrow temperature range. Aqueous electrolytes are classified into several categories, including alkaline (NaOH, KOH, LiOH), and acidic (H_2SO_4). Due to their high ionic concentration, they may be subject to the Faradaic process[49]. Non-aqueous electrolytes are distinguished by their high specific resistance and good work capabilities. Although they are difficult to manufacture, they have low conductivity, toxic, and expensive due to their large ions.

The performance of the double electrolytic capacitor using them is not high [50]. Many studies conducted by recent researchers include that flexible supercapacitors, in which the electrolyte is in the solid state, have an important role in many applications, especially folding, because they are resistant to leakage, inexpensive, easy to pack, and have good mechanical stability. However, its disadvantage is the low mobility of its ions [51]. Electrolytes may be classified as polymers, with most electrolytes forming solid states, and a few inorganic solids. However, it is difficult to find a solid electrolyte that has characteristics that match the specifications needed for a flexible supercapacitor such as mechanical deformation such as folding, torsion, and other characteristics[52]. Figure. 2 summarizes all types of electrolytes used in the literature.

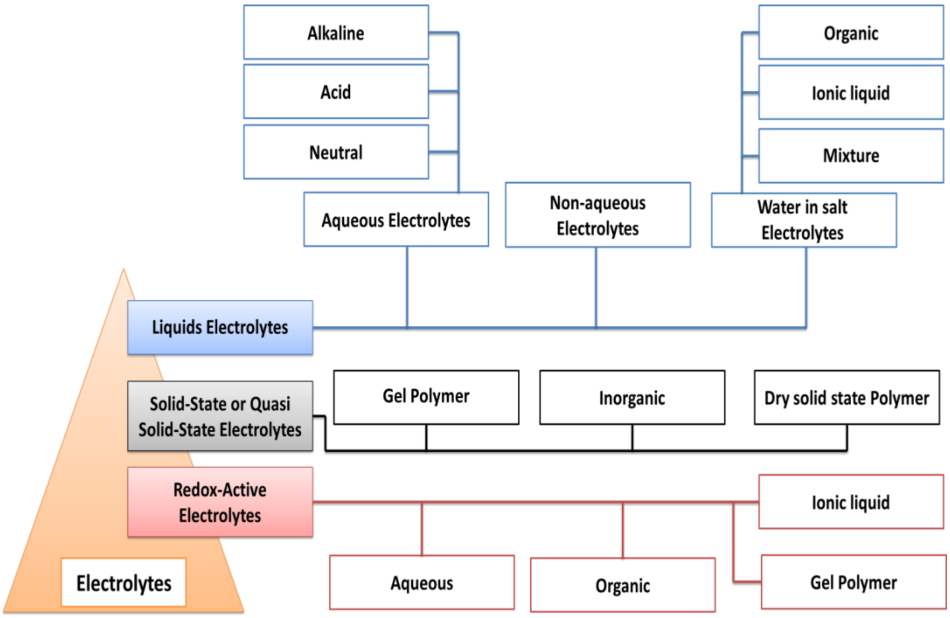

Figure. 2 Different types of electrolytes for electrochemical supercapacitor.

4. Flexible supercapacitors research

4.1 Research progress of carbon family materials based FSCs

In recent years, carbon-based materials have become a hotspot in the field of research because of the advantages they possess, such as large surface area and mechanical stability in flexible supercapacitors, as well as excellent conductivity. Examples of carbon-based materials are graphene and carbon nanotubes (CNTs). Herein we review the research progress of graphene and (CNTs), as well as materials made of activated carbon (AC) and carbon fibers (CFs) will be discussed.

4.1.1Graphene-Based flexible Supercapacitors

Graphene nanocoils (GNSs), composed of two-dimensional graphene sheets, have good elastic properties and a helical microstructure with a large surface area, so they may exhibit mechanical stability in the case of folding, twisting, or other conditions[53, 54]. The study by Jiang et al. [55] discussed the mechanical properties and electrochemical performance of varying aspect ratio GNS. A flexible supercapacitor was prepared using a simple printing technique i.e. electroplating and filling, a membrane electrode enhanced by high aspect ratio GNS. The study has demonstrated in Wade time mechanical durability and high electrochemical performance with little change in capacitance under conditions of mechanical deformation, whether stretching or folding. The SEM shows graphene being wrinkled and interwoven into nanocoils. The flexible supercapacitor is made of an electrode composed of GNS and an electrolyte with an effective leakage property is polyvinyl chloride (PVA)/H_2SO_4. The CV curve measurement shows good performance under different scanning speeds, and the shape of the CV curves is almost rectangular which indicates good capacitive performance. Note the appearance of a perfect triangle with good symmetry when measuring the charge and discharge curves for a current density ranging from 25 to 250 $°C$ at different measurements. It was found that the capacitance values calculated from the CV curves at cm^2 with conditions of 0.025 and 1 mA cm^2 are 10.8 and 4.4 mF. The CV curves did not change significantly when testing rates starting at 0% and ending at 200%. Finally, to measure the stability, which is one of the electrochemical measurements of the supercapacitor, it is used at a rate of 2000 cycles for both expansion rates starting at 0% and then 200%, 100%. Test results for both electrochemical and mechanical properties showed that the flexible supercapacitor's high potential and excellent resistance to mechanical deformation make it an excellent power source for flexible electronics, especially wearable ones[55].

4.1.2 Carbon Nanotube-Based flexible Supercapacitors

One of the graphitic carbon materials is carbon nanotubes (CNTs), which are exploited in many storage systems by incorporating electrolytes that are suitable when used in flexible systems, especially wearable ones. Paul et al. [56] prepared an electrode using the thermochemical vapor deposition method to synthesize CNTs with heterogeneous nitrogen (N) and boron (B) atoms through a pyrolysis process developed on a carbon cloth (CC) substrate. BNCNT-CC was obtained and the electrochemical properties were studied. The SEM images showed the presenting the brush-like BNCNT on the CC substrate covers the entire substrate with a length of 50-150 mm. It has been observed that the O, C, and N atoms are B atoms [57]. The formation of NC-graphite bonds was observed. This may be because in the graphitic carbon lattice, most of the N atoms have been replaced by C atoms[57, 58]. The flexible supercapacitor was prepared from an electrode made of BNCNT-CC, the electrolyte used is PVA/H_2SO_4 gel, and an electrolytic collector made of copper foil was used to make the flexible supercapacitor ideal. The flexible supercapacitor was checked with two tests: first, the electrochemical properties, and second, the mechanical performance, whether deformation, bending or torsion. It was found that the shape of the CV curve was approximately rectangular, at different scan rates starting at 20 mV s^{-1} and ending at 200 mV s^{-1}[59], and this indicates a good capacitive performance of 106.8 mV cm^2. The symmetry of the GCD curve is perfect and is almost triangular, at different current conditions starting at 0.5 mA cm^2 and ending at 20 mA cm^2. This may be attributed to the rapid absorption of ions. The capacitance retained 86.4% of its initial capacity after 5000 cycles when performing the stability test for a flexible supercapacitor with an energy density of (741.8 mWh cm^3). Finally, both CV and stability did not change fundamentally after 100 cycles at a bend angle of 180 for the test, the mechanical performance, meaning that when testing its flexibility, the electrochemical performance did not change significantly under conditions of different angles.

4.1.3 Other carbon materials-Based flexible Supercapacitors

On the other hand, the electrodes of a flexible supercapacitor can be made from environmentally friendly materials and their advantage is that they are widely distributed throughout nature. These materials are extracted from the Tetrapanax papyrus (TC) and their structure is similar to a honeycomb. The study by Hsiao et al. [60] shows the production of a carbon fiber bundle (CFB) as a framework for activated carbon (AC) coating. They used (TC) and (CFB) as raw materials and (AC) as a substrate for current deposition and collector, i.e. the arrangement of the materials is like this: AC-TC-CFB to prepare an electrode, to manufacture a flexible supercapacitor. Fibrous type (FFSC) PVA/H_3PO_4 gel type electrolyte was used to test both electrochemical and mechanical properties. The SEM images of AC-TC-CFB reveal the porous morphology of a large surface area and a honeycomb-like shape (Figure 3).

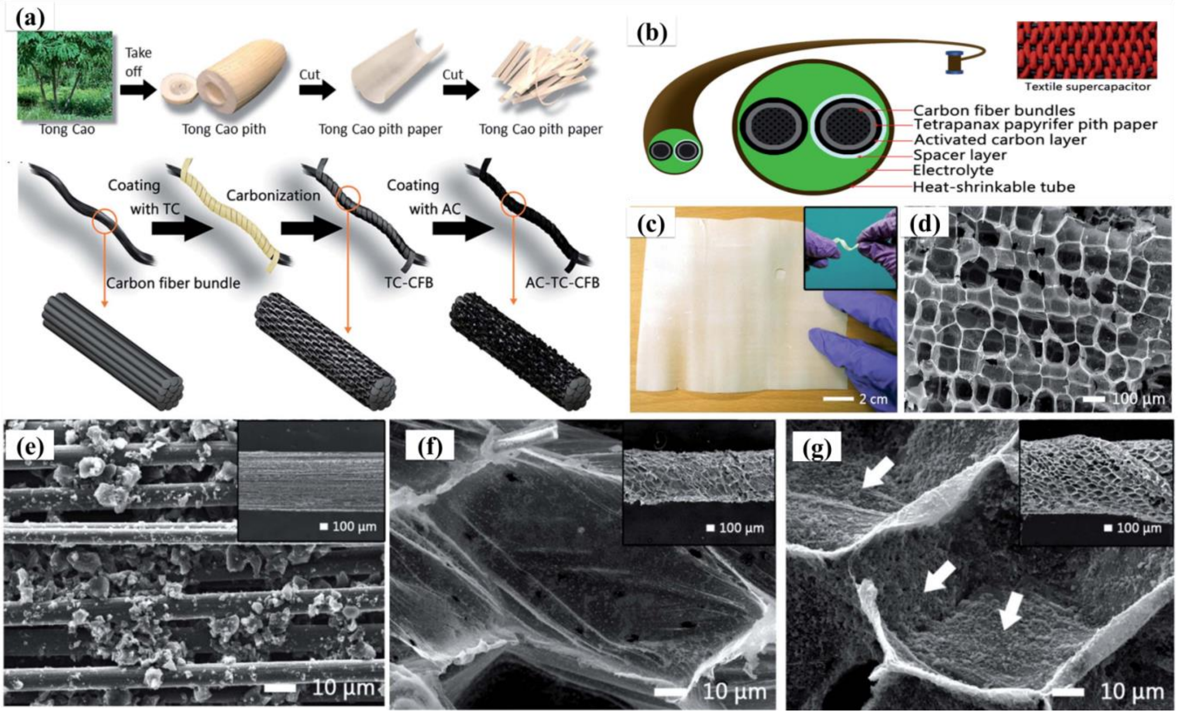

Figure 3: (a) Schematic illustration showing the steps followed to prepare Tetrapanax papyrus, carbon fiber bundle, and framework for activated carbon; (b) graphic diagram of Fibrous type; (c) Photograph shows the Tetrapanax papyrus, the inset shows the Tetrapanax papyrus ribbon; (d), (e), (f) and (g) represent the SEM images of TC, AC-CFB, TC-CFB, and AC-TC-CFB, respectively. This figure is reprinted with permission from ref 60. © 2024 The Royal Society. [60].

The prepared device was conducted in an electrochemical station to obtain the electrochemical measurements. As shown in Figure 4 a, the cyclic voltammetry curves at different scanning speeds starting from 2 mV/s and ending at 200 mV/s are approximately rectangular. The charge/discharge curves at different current densities start at 0.1 A g and end at 12 A g (Figure 4 b) showing good performance with respect to capacity. The stability maintained about 91% of its initial capacity after 10,000 cycles (Figure 4 c) confirming the ability of the device to operate in a long working environment. The flexibility was conducted for the second time with bending 200 times (180). It was found that the tests after bending were identical to those before bending, and this indicates that the capacitor has good bearing capacity even with repeated bending (Figure 4 d). Figure 4 e reveals the specific capacitance as a function of the scan rate while Figure 4 f represents the CV in the cases of bending and unbending.

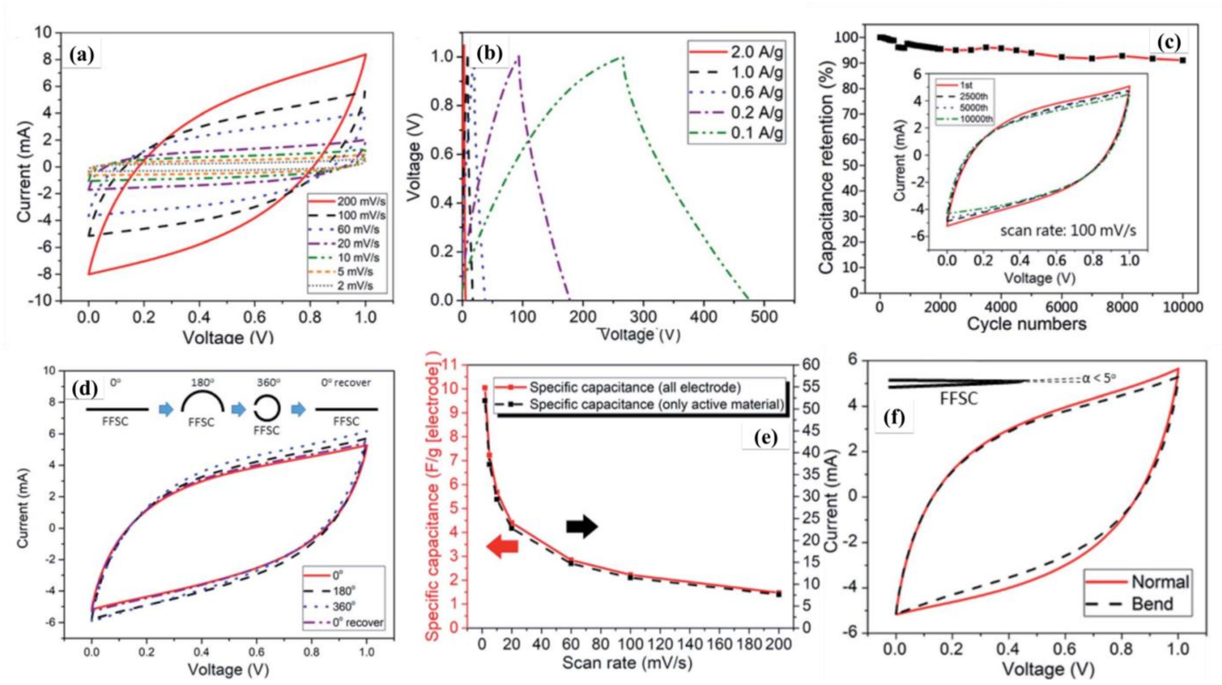

Figure 4: Electrochemical measurements of flexible fibrous supercapacitors using a gel electrolyte: (a) CV curve at different scan rates ;(b) charge/ discharge curves at different energy densities; (c) Capacitance retention after 1000 cycles (scan rate= 100 mV/s) ;(d) CV curves under different bending angles; (e) specific capacitances vs. scan rates ; (f) CV in normal and bending cases. This figure is reprinted with permission from ref 60. © 2024 The Royal Society [60].

Carbon-based materials are used especially in flexible supercapacitors due to their construction structure and performance, and these materials can be combined to form a new material. Table 1 summarizes the surface morphology, specific capacitance, and energy density of carbon materials and their types, including activated carbon, carbon nanotubes, graphene, and other compounds consisting of one or more carbon materials that were studied.

Table 1. Summary on the electrochemical performance of carbon materials for supercapacitors.

Electrode material	Electrolyte type	Energy density	Specific capacitance	Electrode structure
Graphene [61]	[EMIM][TFSI]/PS PMMA-PS	112.6 l Wh /cm^2	268 m F/g	2D
Graphene/CNT [62]	6 M KOH	23.46Wh /kg	312.6 F/g	1D-1D
rGO fiber [63]	1 M H_2SO_4	7.03 mWh /cm^3	185 F/g	1-D
Graphene paper [64]	PVA/H_2SO_4	15 l Wh/ cm^2	13 F/cm^2	2-D
rGO/CNT [65]	PVA/H_2SO_4	12.3 mWh /cm^3	354.9 F/g	2-D/1-D
Graphite / CNT [66]	PVA/H_2SO_4	13.1 Wh /kg	177 F/g	1-D/1-D
Porous AC [67]	6 M KOH	43.54Wh/ kg	207 F/g	2D Porus

4.2 Research progress of composites of metal oxides and conducting polymers materials as flexible supercapacitor electrode materials

Flexible supercapacitors which themselves are based on metal oxides, as well as conductive polymers, have a high storage capacity and good energy density. This may be due to the capacity of the redox reaction between the electrode materials and the electrolyte while maintaining their stability during almost any mechanical deformation. Metal oxides are usually complex in structure and have many active sites due to a large surface area, which makes them have a large charge capacity.[68, 69]

4.2.1 CuCo-layered double hydroxide-based flexible Supercapacitors

The work by Kumar, L., et al[70] revealed a flexible supercapacitor cell with good electrochemical performance that uses an electrode composed of a nanostructured material that has good properties in storing charges electrochemically, which is layered double hydroxide (CuCo-LDH), by gradually increasing the percentage of copper nitrate 100, 200 and 300 respectively. To know the effect of increasing Cu, it is necessary to balance the ratios between Cu and Co to obtain advanced hollow nanopolyhedrons. These structures have a large surface area compared to other types of materials including the LDH family, and are highly porous. Figure 5 refers to a type, in which the material is poured onto a flexible substrate used in applications related to wearable devices.

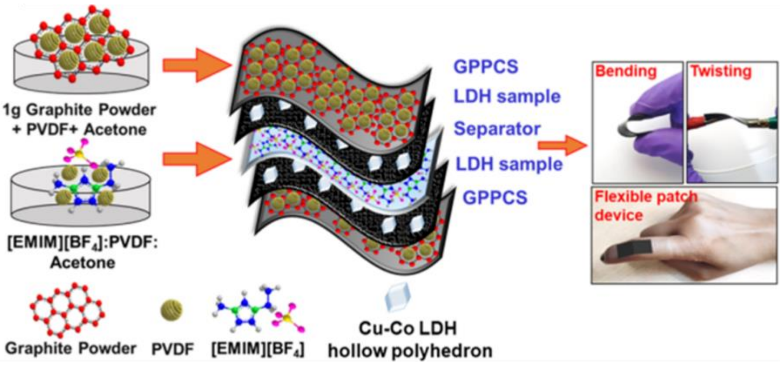

Figure 5: Fabrication of a flexible device based on CuCo-Layered Double Hydroxide and its application in wearable patch devices[70]. This image is reprinted with a permeation from ref. 70@ ACS publication.

Figure 6 displays the surface morphology (FESEM) and crystal structure (XRD) measurements for the prepared CuCo-LDH. The electron microscope image shows that CuCo-LDH is a symmetrical polyhedron. Note that the -OH ions are effectively stored between the two layers, and this may lead to a reduction in redox activity when the ions spread faradically[71]. While the electrode shows a voltage of about 2.5 volts. It may be the best type among many materials included in the LDH family.

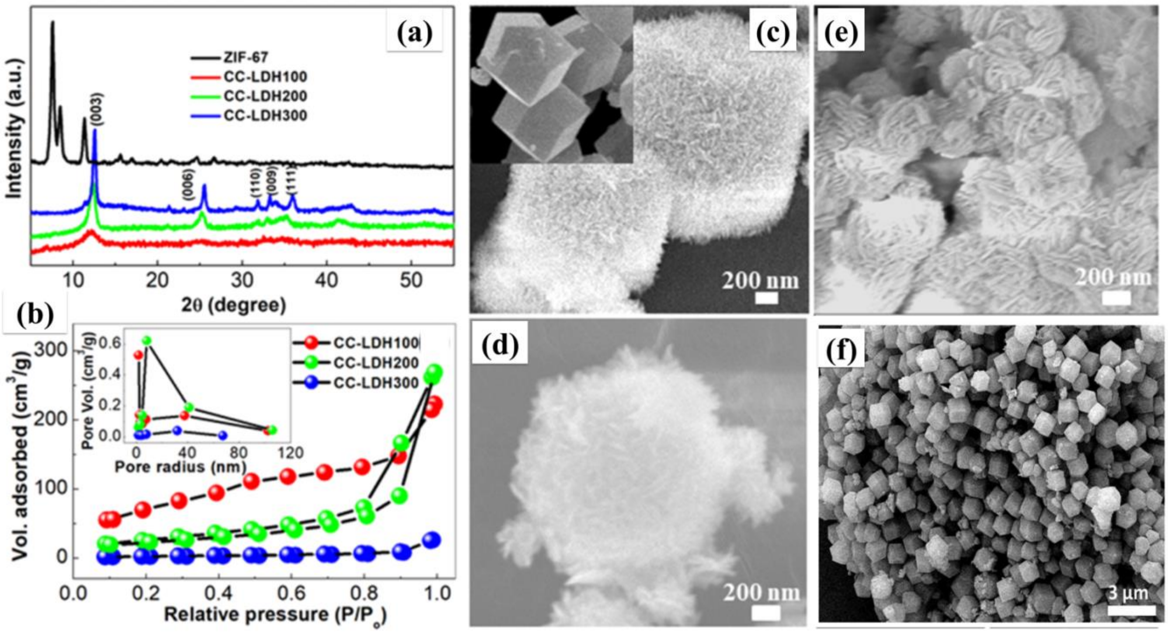

Figure 6: (a) XRD patterns of the as-prepared CuCo -LDHX samples with different LDH mass (X=100, 200, and 300 g) and zeolitic ZIF-67 sample; (b) The BET surface area for CuCo -LDH samples (The pore size curves in the inset). HR-FESEMH images of CuCo -LDH samples [70]. This image is reprinted with a permeation from ref. 70@ ACS publication.

The electrochemical properties of the factory electrode were studied using two types of electrolytes (1) [EMIM][BF4], due to the adhesive property the electrode materials are placed on GPPCS, and then the electrolyte is replaced with (2) 1.0 M KOH, keeping the components unchanged. When examining the previously mentioned samples with different percentages of copper nitrate, it was found that the CuCo-LDH100 sample is the best in terms of specific capacitance and circuit impedance, as well as both power and energy density. Figure 7, the CV diagram, shows the maximum area under the curve, as well as the oxidation and reduction peaks. This indicates the capacitive nature of the material, after selecting the sample whose electrochemical properties will be studied, which is CuCo-LDH100. Now, the electrode will be flexible with different folding angles starting from 0^O and ending at 180^O when the electrolyte is 1.0 M KOH, as it was found that the values of the specific capacitance (Csp) starting from 0 are 662 F g^{-1} and ending at 180 are 200 F g^{-1}, with a current density that may be as large as 12 to 40 A g^{-1}. After performing a charging and discharging process on it to determine the stability of the cell, it was found that the stability reached 90% after 20,000 cycles. The enormous retention of specific capacity may be attributed to the porous nature that characterizes the material CuCo-LDH, which acts as a store for the ions generated from the electrolyte. With a voltage window between 0 to 0.55V, the highest value of both power and power density is 27.81 (W h / Kg) and 7635 (W /Kg), respectively[70].

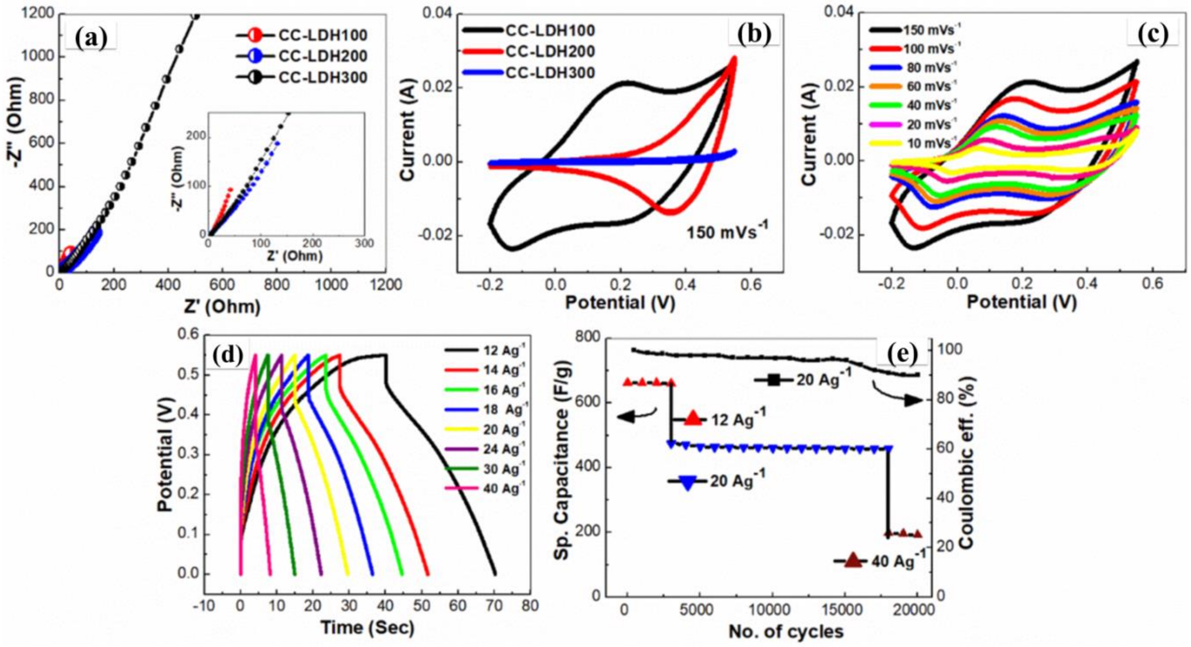

Figure 7: The electrochemical performance of CuCo -LDHX samples using KOH as electrolyte: (a) The Electrochemical impedance plots (the inset shows the high-frequency area); (b) CV curves at 0.15 V/s vs Ag/AgCl; (c) CV curves of CuCo -LDHX at a scan rate ranging from 0.01 to 0.15 V/s; (d) Charge/discharge at different current densities from 12 to 40 A g^{-1} ; (e) specific capacitance and Coulombic eff. as a function of cycle numbers to examine the stability of CuCo -LDHX samples [70]. This image is reprinted with a permeation from ref. 70@ ACS publication.

The electrochemical measurements were performed onto the same CuCo -LDHX samples using [EMIM][BF4] electrolyte instead of KOH. This electrolyte has high conductivity and good mechanical stability when subjected to deformation, whether folding or twisting, without leakage from the cell. Figure 8 shows the measuring CV for the flexible cell. It is noticeable that there is a decrease in the area of the curves compared to the previous cell when the cell is folded at different angles, starting from 0^O and ending at 180^O. The reason for this may be attributed to the obstruction felt by the electrolyte ions when mechanical deformation during the folding[72]. After calculating the specific capacitance values at different folding angles starting from 0^O and ending at 180^O, it was found that the calculated values of 0^O were 244 F/g, while they reached 11.2 F/g at 180^O. The stability of the cell was checked after performing a series of charging/discharging processes, and it was found that the stability rate reached 89% after 10,000 cycles. With a voltage window between 0 to 2.5 V, the highest values for both power and energy density are 52.89 (Wh / kg) and 2766 (W/kg), respectively. Due to the property of not leaking or evaporating the electrolyte, using the new electrolyte is considered a positive point which makes it useful in flexible applications[73].

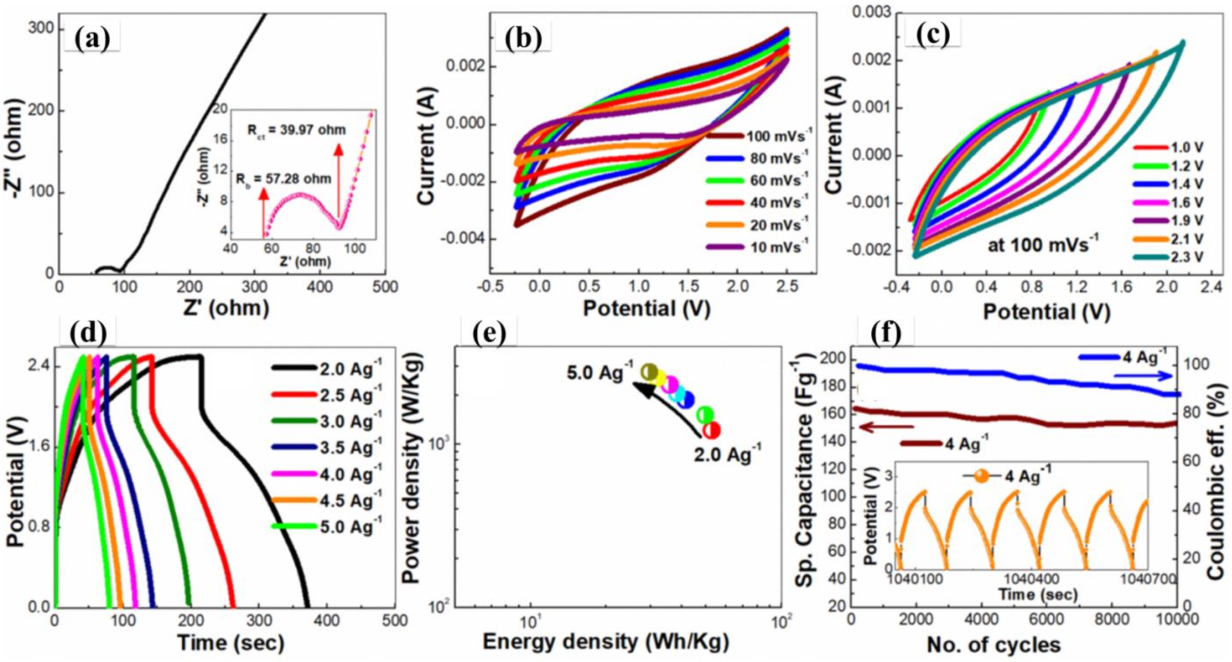

Figure 8: The electrochemical properties of CuCo -LDHX samples using [EMIM][BF4] as electrolyte: (a) EIS measurements of the flexible cell, the inset showing the high-frequency area of Nyquist plot; (b) CV curves of flexible cell at different scan rates from 0.01 to 0.1 V/sec; (c) CV curves measured at a fixed scan rate (0.1 V/sec) and voltages from 1.0 to 2.3 V; (d) Charge/discharge plots at different current density from 2.0 to 5.0 A/g ; (e) Ragone plot ; (f) The stability of the sample after 10000 cycles (the inset shows the charge/discharge in the case of bending)[70]. This image is reprinted with a permeation from ref. 70@ ACS publication.

4.2.2 CNT /Polypyrrole composite based flexible Supercapacitors

It is clear that in recent years, conductive polymers have been the subject of hot research whether used alone or in combination with other materials such as carbon-based ones because of their tensile strength that may exceed the tensile strength of both metal oxide materials and carbon-based materials themselves. Wang et al. [74] reported the preparation of a *CNT /Polypyrrole composite*. The morphology results indicate that the carbon nanotube has a well-bonding mechanism which may help in accelerating the transmitted ions [75]. The *Polypyrrole* (PPy) was tightly bonded to the CNT fiber network after coating on the CNT surface, and the film surface was smooth. The electrode may have high conductivity, and thus this process may improve both electrochemical and mechanical properties. It was found that the CNT decomposed after 5 minutes, while the CNT/PPy remained without decomposition after more than 30 minutes. Figure 9 displays the morphological and structural properties of the CNT electrode and CNT/PPy.

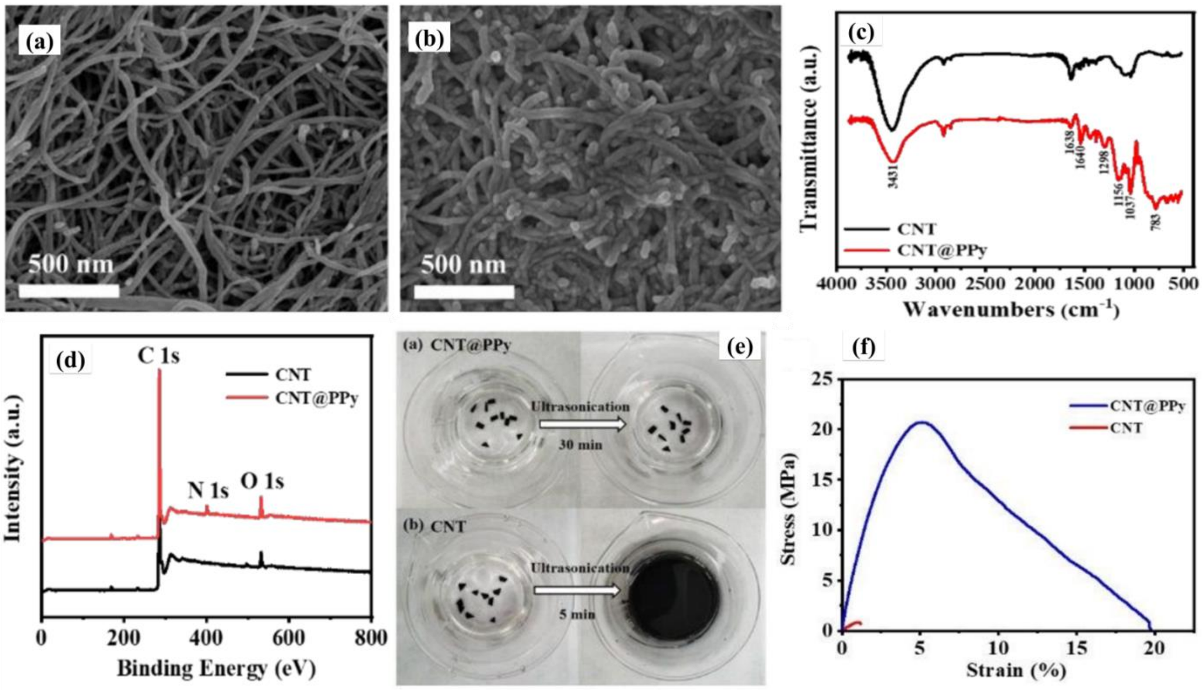

Figure 9: SEM images of (a)CNT and (b) CNT/PPy composite electrodes; (c) Fourier-transform infrared spectroscopy of both CNT and CNT/PPy; (d) XPS spectra of the CNT and CNT/PPy; (e) photographic image of CNT and CNT/PPy-100; (f) Stress-strain plot for CNT and CNT/PPy-100 [74]. This image is reprinted with a permeation from ref. 74@2024 The Royal Society.

The electrochemical properties and the flexibility of the supercapacitor of the prepared electrodes were evaluated using PVA/H_2SO_4 gel as the electrolyte. The electrochemical measurements started with CV at different scanning speeds starting at 0.002 V/s and ending at 0.02 V/s. The shape of the curves is approximately rectangular, and this indicates good storage capacity. As for the charge/discharge test, the shape of the curves was almost triangular at different current densities starting at 2 mA cm^2 and ending at 20 mA cm^2 [76], which indicates that the device has excellent capacitive properties with 72.2% of its initial capacity retained. When repeating the previous tests with bending angles of 0, 90, and 180, it is noted that both the CV and charge/discharge curves did not change significantly compared to the curves before bending, which indicates that the loudness is ideal under conditions of mechanical deformation. Figure 10 shows the electrochemical properties of the CNT/PPy composite in flat and flexible conditions.

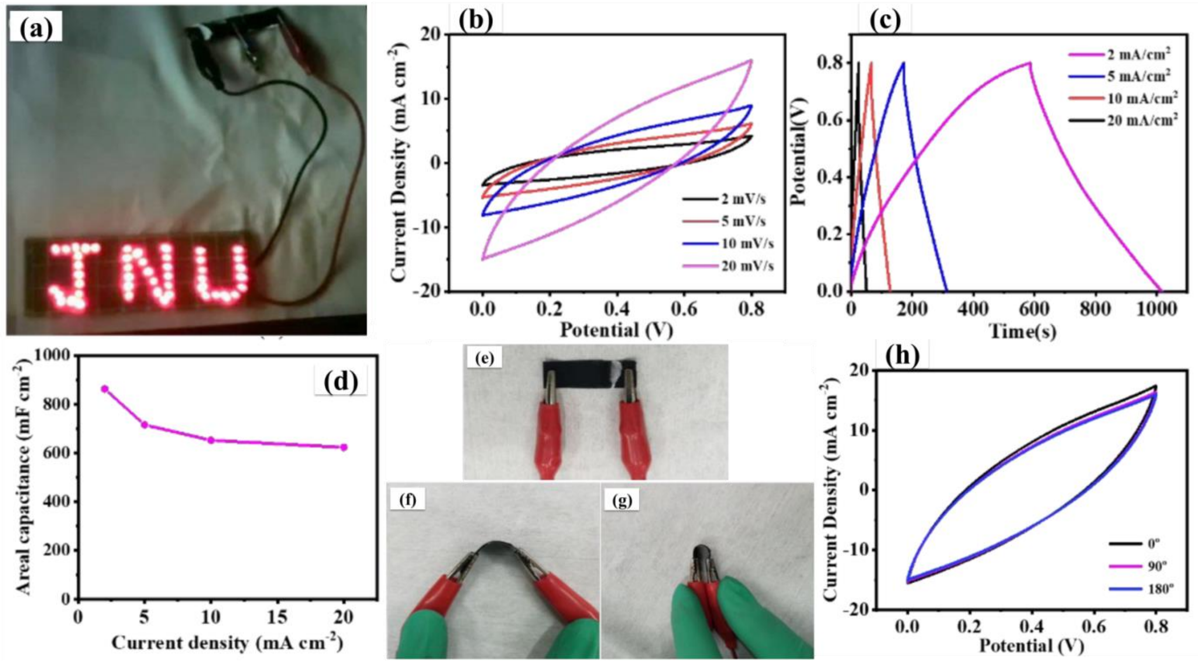

Figure 13: (a) Photograph of the CNT/PPy electrodes connected to red LEDs, (b) CV curves at different scanning voltage rates, (c) charge/discharge test different current densities, (d) The specific capacitances as a function of voltage rates; Photograph of the supercapacitor in (e) flat condition, (b) 90°, and 180° bending status. (h) CV curves in both flat and bent cases [74]. This image is reprinted with a permeation from ref. 74@ 2024 The Royal Society.

Often, metal oxides and polymeric materials are combined with carbon materials to form composite materials that have good electrochemical performance and excellent stability when any mechanical deformation occurs in FSCs. Table 2 presents some types of supercapacitors manufactured from composite materials in recent years.

Table 2. Summary on the electrochemical performance of composite materials for supercapacitors.

Electrode material	Current collector	Electrolyte	Energy density	Capacitance	Morphology
PANI/graphene paper /MnO2 [77]	MnO_2	PVA/H_2SO_4	5 mWh/cm^3	874 mF/cm^2	3D/ 2D /2D
Copper foam/rGO [78]	Copper foam	PVA/H_3PO_4	11.25Wh/kg	84 F/ g	3D/2D
PANI/UiO66 (MOF) [79]	CC	PVA/H_2SO_4	78.8Wh/ kg	647 F/g	3D/3D
Polyimide/MWCT [80]	-	$1MH_2SO_4$	0.50 mWh/ cm^2	194 mF/ cm^2	3D/ID
Porous carbonized cotton/ZnO/CuS [81]	Ag	PVA/KOH	0.27 Wh/ cm^2	380 F/ g	0D/0D/0D
Graphene/ZnAl-LDHs[82]	-	3 M KOH	2 Wh/ kg	139.6 F/ g	2D/2D
Graphene /V_2O_5[83]	-	1 M KOH	12.53 Wh/ kg	3515 F/ g	2D/0D

5. Conclusions and outlooks

FSCs have great research interest in many fields especially smart wearable clothing and medical care and their comprehensive properties can be determined by choosing the type of electrode. Many researchers have manipulated the origin of the electrodes adding carbon materials to other materials, i.e. pseudo materials, whether by adding metal oxides or conductive polymers. Additional research on the composites for FSCs electrode have been performed in order to determine the mechanism of ionic diffusion and electronic transferring between electrodes. Further combination or modification procedures can lead to reach better performance of FSCs and therefore the new kinds of flexible electrode materials are greeted to be joined to the current state of art. The change is not limited to the electrodes, but the shape of the electrolyte and the separators, as the electrolyte evolved into a mixed gel of PVA and other electrolytes after the aqueous solution was strongly alkaline or acidic. This may lead to improved electrochemical performance of FSCs. Many FSCs have practical prospects in various fields, yet they are limited in cost, production, and size. Therefore, researchers must make every effort to solve this problem facing FSCs.

References

[1]C. Zou, Q. Zhao, G. Zhang, B. Xiong, Energy revolution: From a fossil energy era to a new energy era, Natural Gas Industry B., 3 (2016) 1-11, https://doi.org/10.1016/j.ngib.2016.02.001

[2]X. Shen, Y. Li, T. Qian, J. Liu, J. Zhou, C. Yan, J.B. Goodenough, Lithium anode stable in air for low-cost fabrication of a dendrite-free lithium battery, Nat. Commun., 10 (2019) 900, https://doi.org/10.1038/s41467-019-08767-0

[3]Y. Zhang, H.-l. Gao, X.-d. Jia, S.-w. Wang, J. Yan, H.-w. Luo, K.-z. Gao, H. Fang, A.-q. Zhang, L.-z. Wang, NiMoO4 nanorods supported on nickel foam for high-performance supercapacitor electrode materials, J. Renew. Energy., 10(2018) 054101, https://doi.org/10.1063/1.5032271

[4]Z. Wang, R. Tan, H. Wang, L. Yang, J. Hu, H. Chen, F. Pan, A metal–organic-framework-based electrolyte with nanowetted interfaces for high-energy-density solid-state lithium battery, Adv. Mater., 30 (2018) 1704436, https://doi.org/10.1002/adma.201704436

[5]B. Zhu, X. Wang, P. Yao, J. Li, J. Zhu, Towards high energy density lithium battery anodes: silicon and lithium, Chem. Sci., 10 (2019) 7132-7148, https://doi.org/10.1039/C9SC01201J

[6]S. Wang, S.P. Jiang, Prospects of fuel cell technologies, National Science Review., 4 (2017) 163-166, https://doi.org/10.1093/nsr/nww099

[7]M. Kadhim, M. Mahdi, J. Hassan, A.S. Al-Asadi, Photocatalytic activity and photoelectrochemical properties of Ag/ZnO core/shell nanorods under low-intensity white light irradiation, Nanotechnology., 32 (2021) 195706, https://doi.org/10.1088/1361-6528/abe3b3

[8]A.S. Al-Asadi, L.A. Henley, M. Wasala, B. Muchharla, N. Perea-Lopez, V. Carozo, Z. Lin, M. Terrones, K. Mondal, K. Kordas, Aligned carbon nanotube/zinc oxide nanowire hybrids as high performance electrodes for supercapacitor applications, J. Appl. Phys., 121(2017) 124303, https://doi.org/10.1063/1.4979098

[9]A. Kamišalić, I. Fister Jr, M. Turkanović, S. Karakatič, Sensors and functionalities of non-invasive wrist-wearable devices: Sens. Rev., 18 (2018) 1714, https://doi.org/10.3390/s18061714

[10]Y. Han, L. Dai, Conducting polymers for flexible supercapacitors, Macromol. Chem. Phys., 220 (2019) 1800355, https://doi.org/10.1002/macp.201800355

[11]J. Rao, N. Liu, Z. Zhang, J. Su, L. Li, L. Xiong, Y. Gao, All-fiber-based quasi-solid-state lithium-ion battery towards wearable electronic devices with outstanding flexibility and self-healing ability, Nano Energy., 51 (2018) 425-433, https://doi.org/10.1016/j.nanoen.2018.06.067

[12]A. Yadav, B. De, S.K. Singh, P. Sinha, K.K. Kar, Facile development strategy of a single carbon-fiber-based all-solid-state flexible lithium-ion battery for wearable electronics, ACS Appl. Mat. Interf., 11 (2019) 7974-7980,https://doi.org/10.1021/acsami.8b20233

[13]H. Yang, S. Kannappan, A.S. Pandian, J.-H. Jang, Y.S. Lee, W. Lu, Graphene supercapacitor with both high power and energy density, Nanotech., 28 (2017) 445401, https://doi.org/10.1088/1361-6528/aa8948

[14]X. Wu, Y. Xu ,Y. Hu, G. Wu, H. Cheng, Q. Yu, K. Zhang, W. Chen, S. Chen, Microfluidic-spinning construction of black-phosphorus-hybrid microfibres for non-woven fabrics toward a high energy density flexible supercapacitor, Nat. Commun., 9 (2018) 4573, https://doi.org/10.1038/s41467-018-06914-7

[15]L .Xu, Y. Zhao, J. Lian, Y. Xu, J. Bao, J. Qiu, L. Xu, H. Xu, M. Hua, H. Li, Morphology controlled preparation of ZnCo2O4 nanostructures for asymmetric supercapacitor with ultrahigh energy density, Energy., 123 (2017) 296-304, https://doi.org/10.1016/j.energy.2017.02.018

[16]Y. Zhang, C.-r. Chang, X.-d .Jia, Q.-y. Huo, H.-l. Gao, J. Yan, A.-q. Zhang, Y. Ru, H.-x. Mei, K.-z. Gao, Morphology-dependent NiMoO4/carbon composites for high performance supercapacitors, Inorg. Chem. Commun., 111 (2020) 107631, https://doi.org/10.1016/j.inoche.2019.107631.

[17]J.-H. Choi, C. Lee, S. Cho, G.D .Moon, H. Chang, H.D. Jang, High capacitance and energy density supercapacitor based on biomass-derived activated carbons with reduced graphene oxide binder, Carbon., 132 (2018) 16-24, https://doi.org/10.1016/j.carbon.2018.01.105

[18]E. Lim, C. Jo, J. Lee, A mini review of designed mesoporous materials for energy-storage applications: from electric double-layer capacitors to hybrid supercapacitors, Nanoscale., 8 (2016) 7827-7833,https://doi.org/10.1039/C6NR00796A

[19]N.R. Chodankar, H.D. Pham, A.K. Nanjundan, J.F. Fernando, K. Jayaramulu, D. Golberg, Y.K. Han, D.P. Dubal, True meaning of pseudocapacitors and their performance metrics: asymmetric versus hybrid supercapacitors, Small., 16 (2020) 2002806, https://doi.org/10.1002/smll.202002806

[20]Y. Huang, M. Zhong, F. Shi, X. Liu, Z. Tang, Y. Wang, Y. Huang, H. Hou, X. Xie, C. Zhi, An intrinsically stretchable and compressible supercapacitor containing a polyacrylamide hydrogel electrolyte, Angew. Chem.,Int. Ed. Engl., 56 (2017) 9141-9145, https://doi.org/10.1002/anie.201705212

[21]L. Yu, G.Z. Chen, Ionic liquid-based electrolytes for supercapacitor and supercapattery, fchem ., 7 (2019) 272, https://doi.org/10.3389/fchem.2019.00272

[22]Y. Zhang, H.-X. Mei, H.-l. Gao, Q.-y. Huo, X.-d. Jia, Y. Cao, S.-w. Wang, J. Yan, H.-w. Luo, J. Yang, Metal oxide modified (NH4)(Ni, Co) PO4· 0.67 H2O composite as high-

performance electrode materials for supercapacitors, Inorg. Chem. Commun., 112 (2020) 107696, https://doi.org/10.1016/j.inoche.2019.107696

[23]H. Wang, H. Zhou, M. Gao, Y.-a. Zhu, H. Liu, L. Gao, M. Wu, Hollow carbon spheres with artificial surface openings as highly effective supercapacitor electrodes, Electrochim. Acta., 298 (2019) 552-560, https://doi.org/10.1016/j.electacta.2018.12.070

[24]Y. Zhang, C.-r. Chang, X.-d. Jia, Y. Cao, J. Yan, H.-w. Luo, H.-l. Gao, Y. Ru, H.-x. Mei, A.-q. Zhang, Influence of metallic oxide on the morphology and enhanced supercapacitive performance of NiMoO4 electrode material, Inorg. Chem. Commun., 112107697 (2020) , https://doi.org/10.1016/j.inoche.2019.107697

[25]K.-B. Wang, Q. Xun, Q. Zhang, Recent progress in metal-organic frameworks as active materials for supercapacitors, EnergyChem., 2 (2020) 100025, https://doi.org/10.1016/j.enchem.2019.100025

[26]X. Li, X. Yang, H. Xue, H. Pang, Q. Xu, Metal–organic frameworks as a platform for clean energy applications, EnergyChem., 2 (2020) 100027, https://doi.org/10.1016/j.enchem.2020.100027

[27]T. Zhou, C. Wu, Y. Wang, A.P. Tomsia, M. Li, E. Saiz, S. Fang, R.H. Baughman, L. Jiang, Q. Cheng, Super-tough MXene-functionalized graphene sheets, Nat. Commun., 11 (2020) 2077, https://doi.org/10.1038/s41467-020-15991-6

[28]M. Kumar, P. Sinha, T. Pal, K.K. Kar, Materials for supercapacitors, Handbook of Nanocomposite Supercapacitor Materials II: Performance., (2020) 29-70, https://doi.org/10.1007/978-3-030-52359-6_2

[29]K.D. Verma, P. Sinha, S. Banerjee, K.K. Kar, Characteristics of electrode materials for supercapacitors ,Handbook of nanocomposite supercapacitor materials I: characteristics., (2020) 269-285,https://doi.org/10.1007/978-3-030-43009-2_9

[30]S. Kumar, R. Nigam, V. Kundu, N. Jaggi, Sol–gel synthesis of ZnO–SnO 2 nanocomposites and their morphological, structural and optical properties, J. Mater. Sci.: Mater. Electron., 26 (2015) 3268-3274, https://doi.org/10.1007/s10854-015-2826-5

[31]A.S. Al-Asadi, Q.M. Hassan, A.F. Abdulkader, M.H. Mohammed, H. Bakr, C. Emshary, Formation of graphene nanosheets/epoxy resin composite and study its structural, morphological and nonlinear optical properties, Opt. Mater., 89 (2019) 460-467, https://doi.org/10.1016/j.optmat.2019.01.078

[32]M.H. Mohammed, A.S. Al-Asadi, F.H. Hanoon, Electronic structure and band gap engineering of bilayer graphene nanoflakes in the presence of nitrogen, boron and boron nitride impurities, Superlattices Microstruct., 129 (2019) 14-19, https://doi.org/10.1016/j.spmi.2019.03.012

[33]L. Miao, Z. Song, D. Zhu, L. Li, L. Gan, M. Liu, Recent advances in carbon-based supercapacitors, Mater Adv., 1 (2020) 945-966, https://doi.org/10.1039/D0MA00384K

[34]D. Yu, S. Zhai, W. Jiang, K. Goh, L. Wei, X. Chen, R. Jiang, Y .Chen, Transforming Pristine Carbon Fiber Tows into High Performance Solid-State Fiber Supercapacitors, Adv Mater., 27 (2015) 4895-4901, https://doi.org/10.1002/adma.201501948

[35]Z. Song, L. Miao, L. Li, D. Zhu, L. Gan, M. Liu, A robust strategy of solvent choice to synthesize optimal nanostructured carbon for efficient energy storage, Carbon., 180 (2021) 135-145,https://doi.org/10.1016/j.carbon.2021.04.078

[36]L. Ma, Z. Bi, W. Zhang, Z. Zhang, Y. Xiao, H. Niu, Y. Huang, Synthesis of a three-dimensional interconnected oxygen-, boron-, nitrogen-, and phosphorus tetratomic-doped porous carbon network as electrode material for the construction of a superior flexible supercapacitor, ACS Appl. Mater. Interfaces., 12 (2020) 46170-46180, https://doi.org/10.1021/acsami.0c13454

[37]Y. Han, Y. Lu, S. Shen, Y. Zhong, S. Liu, X. Xia, Y. Tong, X .Lu, Enhancing the capacitive storage performance of carbon fiber textile by surface and structural modulation for advanced flexible asymmetric supercapacitors, Adv. Funct. Mater., 29 (2019) 1806329, https://doi.org/10.1002/adfm.201806329

[38]B. Kadem, R.F. Alfahed, A.S. Al-Asadi ,H.A. Badran, Morphological, structural, optical, and photovoltaic cell of copolymer P3HT: ICBA and P3HT: PCBM, Optik., 204 (2020) 164153, https://doi.org/10.1016/j.ijleo.2019.164153

[39]Q. Wu, Y. Xu, Z. Yao, A. Liu, G. Shi, Supercapacitors based on flexible graphene/polyaniline nanofiber composite films, ACS Nano., 4 (2010) 1963-1970, https://doi.org/10.1021/nn1000035

[40]M. Hu, C. Cui, C. Shi, Z.-S. Wu, J. Yang, R. Cheng, T. Guang, H. Wang, H. Lu, X. Wang, High-energy-density hydrogen-ion-rocking-chair hybrid supercapacitors based on Ti3C2 T x MXene and carbon nanotubes mediated by redox active molecule, ACS Nano., 13 (2019) 6899-6905,https://doi.org/10.1021/acsnano.9b01762

[41]A.S. Al-Asadi, L.A. Henley, S. Ghosh, A. Quetz, I. Dubenko, N. Pradhan, L. Balicas, N. Perea-Lopez, V. Carozo, Z. Lin, Fabrication and characterization of ultraviolet photosensors from ZnO nanowires prepared using chemical bath deposition method, J. Appl. Phys., 119 .(2016) 084306, https://doi.org/10.1063/1.4942653

[42]R. Fakher Alfahed, A.S. Al-Asadi, H.A. Badran, K.I. Ajeel, Structural, morphological, and Z-scan technique for a temperature-controllable chemical reaction synthesis of zinc sulfide nanoparticles,Appl. Phys. B., 125 (2019) 1-11, https://doi.org/10.1007/s00340-019-7154-7

[43]B.-T. Liu, X.-M. Shi, X.-Y. Lang, L. Gu, Z. Wen, M. Zhao, Q. Jiang, Extraordinary pseudocapacitive energy storage triggered by phase transformation in hierarchical vanadium oxides, Nat. Commun., 9 (2018) 1375, https://doi.org/10.1038/s41467-018-03700-3

[44]X. Zhang, Q. Fu, H. Huang, L. Wei, X. Guo, Silver-quantum-dot-modified MoO3 and MnO2 paper-like freestanding films for flexible solid-state asymmetric supercapacitors, Small., 15 (2019) 1805235, https://doi.org/10.1002/smll.201805235

[45]A. Tyagi, S. Banerjee, J. Cherusseri, K.K. Kar, Characteristics of transition metal oxides, Handbook of Nanocomposite Supercapacitor Materials I: Characteristics., (2020) 91-123, https://doi.org/10.1007/978-3-030-43009-2_3

[46]Y. Wang, X. Wu, Y. Han, T. Li, Flexible supercapacitor: overview and outlooks, J. Energy Storage., 42 (2021) 103053, https://doi.org/10.1016/j.est.2021.103053

[47]K.D. Verma, S. Banerjee, K.K. Kar, Characteristics of electrolytes, Handbook of Nanocomposite Supercapacitor Materials I: Characteristics., (2020) 287-314, https://doi.org/10.1007/978-3-030-43009-2_10

[48]N. Kanaujiya, N. Kumar, Y. Sharma, G.D. Varma, Probing the Electrochemical Properties of Flower Like Mesoporous MoS_2 in Different Aqueous Electrolytes, J. Electron. Mater., 48 (2019) 904-915, https://doi.org/10.1007/s11664-018-6801-9

[49]B. Pal, S. Yang, S. Ramesh, V. Thangadurai, R. Jose, Electrolyte selection for supercapacitive devices: a critical review, Nanoscale Adv., 1 (2019) 3807-3835, https://doi.org/10.1039/C9NA00374F

[50]C. Zhong, Y. Deng, W. Hu, J. Qiao, L. Zhang, J. Zhang, A review of electrolyte materials and compositions for electrochemical supercapacitors, Chem Soc Rev., 44 (2015) 7484-7539, https://doi.org/10.1039/C5CS00303B

[51]B.E. Francisco, C.M. Jones, S.-H. Lee, C.R. Stoldt, Nanostructured all-solid-state supercapacitor based on Li2S-P2S5 glass-ceramic electrolyte, Appl. Phys. Lett., 100 (2012) 103902, https://doi.org/10.1063/1.3693521

[52]S.B. Aziz, T.J. Woo, M. Kadir, H.M. Ahmed, A conceptual review on polymer electrolytes and ion transport models, J. Sci.: Adv. Mater. Devices., 3 (2018) 1-17, https://doi.org/10.1016/j.jsamd.2018.01.002

[53]Q. Chang, L. Li, L. Sai, W. Shi, L. Huang, Water-Soluble Hybrid Graphene Ink for Gravure-Printed Planar Supercapacitors, Adv. Electron. Mater., 4 (2018) 1800059, https://doi.org/10.1002/aelm.201800059

[54]H. Cao, X. Zhou, C. Zheng, Z. Liu, Metal etching method for preparing porous graphene as high performance anode material for lithium-ion batteries, Carbon., 89 (2015) 41-46, https://doi.org/10.1016/j.carbon.2015.03.003

[55]S. Jiang, X. Zhou, H. Xiao, W. Chen, X .Xu, Z. Liu, Robust and durable flexible micro-supercapacitors enabled by graphene nanoscrolls, J. Chem. Eng., 405 (2021) 127009, https://doi.org/10.1016/j.cej.2020.127009

[56]R. Paul, A.K. Roy, BN-codoped CNT based nanoporous brushes for all-solid-state flexible supercapacitors at elevated temperatures, Electrochim. Acta., 365 (2021) 137345, https://doi.org/10.1016/j.electacta.2020.137345.

[57]R. Paul, A.A. Voevodin, D. Zemlyanov, A.K. Roy, T.S. Fisher, Microwave-assisted surface synthesis of a boron–carbon–nitrogen foam and its desorption enthalpy, Adv. Funct. Mater., 22 (2012) 3682-3690, https://doi.org/10.1002/adfm.201200325

[58]J. Zhang, Z. Xia, L. Dai, Carbon-based electrocatalysts for advanced energy conversion and storage, Sci. Adv., 1 (2015) e1500564, https://doi.org/10.1126/sciadv.1500564

[59]M. Khalid, R. Paul, A.M. Honorato, H. Varela, Pinus nigra pine derived hierarchical carbon foam for high performance supercapacitors, J. Electroanal. Chem., 863 (2020) 114053, https://doi.org/10.1016/j.jelechem.2020.114053

[60]C. Hsiao, C. Lee, N. Tai, Biomass-derived three-dimensional carbon framework for a flexible fibrous supercapacitor and its application as a wearable smart textile, RSC Adv., 10 (2020) 6960-6972, https://doi.org/10.1039/c9ra07441d

[61]W.J. Hyun, E.B. Secor, C.H. Kim, M.C. Hersam, L.F. Francis, C.D. Frisbie, Scalable, self-aligned printing of flexible graphene micro-supercapacitors, Adv. Energy Mater., 7 (2017) 1700285 -1700285, https://doi.org/10.1002/aenm.201700285

[62]Y. Jia, L. Zhou, J. Shao, Electrochemical performance of flexible graphene-based fibers as electrodes for wearable supercapacitors, Synth. Met., 246 (2018) 108-114, https://doi.org/10.1016/j.synthmet.2018.09.016

[63]S. Chen, W. Ma, Y. Cheng, Z. Weng, B. Sun, L. Wang, W. Chen, F. Li, M .Zhu, H.-M. Cheng, Scalable non-liquid-crystal spinning of locally aligned graphene fibers for high-performance wearable supercapacitors, Nano Energy., 15 (2015) 642-653, https://doi.org/10.1016/j.nanoen.2015.05.004

[64]G.A. Ferrero, M. Sevilla, A.B. Fuertes, Flexible, Free-Standing and Holey Graphene Paper for High-Power Supercapacitors, ChemNanoMat., 2 (2016) 1055-106, https://doi.org/10.1002/cnma.201600228

[65]T. Xu, D. Yang, Z. Fan, X. Li, Y. Liu, C. Guo, M. Zhang, Z.-Z. Yu, Reduced graphene oxide/carbon nanotube hybrid fibers with narrowly distributed mesopores for flexible supercapacitors with high volumetric capacitances and satisfactory durability, Carbon., 152 (2019) 134-143,https://doi.org/10.1016/j.carbon.2019.06.005

[66]C. Yang, Q. Pan, Q. Jia, Y. Xin, W. Qi, H. Wei, S. Yang, B. Cao, Multifunctional microporous activated carbon nanotubes anchored on graphite fibers for high-strength and high-rate flexible all-solid-state supercapacitors, Appl. Surf. Sci., 502 (2020) 144423, https://doi.org/10.1016/j.apsusc.2019.144423

[67]J.R. Rajabathar, M. Sivachidambaram, J.J. Vijaya, H.A. Al-Lohedan, D.M. Aldhayan, Flexible type symmetric supercapacitor electrode fabrication using phosphoric acid-activated carbon nanomaterials derived from cow dung for renewable energy applications, ACS omega., 5 (2020) 15028-15038, https://doi.org/10.1021/acsomega.0c00848

[68]X. Wu, B. Huang, R. Lv, Q. Wang, Y. Wang, Highly flexible and low capacitance loss supercapacitor electrode based on hybridizing decentralized conjugated polymer chains with MXene, J. Chem. Eng., 378 (2019) 122246, https://doi.org/10.1016/j.cej.2019.122246

[69]G. Du, H. Pang, Recent advancements in Prussian blue analogues: Preparation and application in batteries, Energy Storage Materials., 36 (2021) 387-408, https://doi.org/10.1016/j.ensm.2021.01.006

[70]L. Kumar, P.K. Boruah, S. Borthakur, L. Saikia, M.R. Das, S. Deka, CuCo-layered double hydroxide nanosheet-based polyhedrons for flexible supercapacitor cells, ACS Appl. Nano Mater., 4 (2021) 5250-5262, https://doi.org/10.1021/acsanm.1c00628

[71]L. Yu ,B.Y. Xia, X. Wang, X.W. Lou, General Formation of M-MoS3 (M= Co, Ni) Hollow Structures with Enhanced Electrocatalytic Activity for Hydrogen Evolution, Adv. Mater., 28 (2015) 92-97, https://doi.org/10.1002/adma.201504024

[72]A. Zhang, W. Zheng, Z. Yuan, J. Tian ,L. Yue, R. Zheng, D. Wei, J. Liu, Hierarchical NiMn-layered double hydroxides@ CuO core-shell heterostructure in-situ generated on Cu (OH) 2 nanorod arrays for high performance supercapacitors, J. Chem. Eng., 380 (2020) 122486,https://doi.org/10.1016/j.cej.2019.122486

[73]T. Wang, S. Zhang, X. Yan, M. Lyu, L. Wang, J. Bell, H. Wang, 2-Methylimidazole-derived Ni–Co layered double hydroxide nanosheets as high rate capability and high energy density storage material in hybrid supercapacitors, ACS Appl. Mater. Interfaces., 9 .(2017) -15510 15524 https://doi.org/10.1021/acsami.7b02987

[74]S. Wang, Y. Liang, W. Zhuo, H. Lei, M.S. Javed, B. Liu, Z. Wang, W. Mai, Freestanding polypyrrole/carbon nanotube electrodes with high mass loading for robust flexible supercapacitors, Mater. Chem. Front., 5 (2021) 1324-1329, https://doi.org/10.1039/D0QM00649A

[75]H.P. De Oliveira, S.A. Sydlik, T.M. Swager, Supercapacitors from free-standing polypyrrole/graphene nanocomposites, J. Phys. Chem. C., 117 (2013) 10270-10276, https://doi.org/10.1021/jp400344u

[76]C. Sun, X. Li, Z. Cai, F. Ge, Carbonized cotton fabric in-situ electrodeposition polypyrrole as high-performance flexible electrode for wearable supercapacitor, Electrochim. Acta., 296 (2019) 617-626, https://doi.org/10.1016/j.electacta.2018.11.045

[77]N. Song, Y. Wu, W. Wang, D. Xiao, H. Tan, Y. Zhao, Layer-by-layer in situ growth flexible polyaniline/graphene paper wrapped by MnO2 nanoflowers for all-solid-state

supercapacitor, Mater. Res. Bull., 111 (2019) 267-276, https://doi.org/10.1016/j.materresbull.2018.11.024

[78]T. Purkait, G. Singh, D. Kumar, M. Singh, R.S. Dey, High-performance flexible supercapacitors based on electrochemically tailored three-dimensional reduced graphene oxide networks, Sci. Rep., 8 (2018) 640, https://doi.org/10.1038/s41598-017-18593-3

[79]L. Shao, Q. Wang, Z. Ma, Z. Ji, X. Wang, D. Song, Y. Liu, N. Wang, A high-capacitance flexible solid-state supercapacitor based on polyaniline and Metal-Organic Framework (UiO (66-composites, J. Power Sources., 379 (2018) 350-361, https://doi.org/10.1016/j.jpowsour.2018.01.028

[80]D.K. Kim, N.D. Kim, S.-K. Park, K.-d. Seong, M. Hwang, N.-H. You, Y. Piao, Nitrogen doped carbon derived from polyimide/multiwall carbon nanotube composites for high performance flexible all-solid-state supercapacitors, J. Power Sources., 380 (2018) 55-63, https://doi.org/10.1016/j.jpowsour.2018.01.069

[81]S. Zhai, K. Jin, M. Zhou, Z. Fan, H. Zhao, X. Li, Y. Zhao, F. Ge, Z. Cai, A novel high performance flexible supercapacitor based on porous carbonized cotton/ZnO nanoparticle/CuS micro-sphere, Colloids Surf. A: Physicochem. Eng. Asp., 584 (2020) 124025, https://doi.org/10.1016/j.colsurfa.2019.124025

[82]M.A. Al-Hamyd, A.S. Al-Asadi, M.F. Al-Mudhaffer, Preparation and characterization of zinc–aluminum layered doubled hydroxide/graphene nanosheets composite for supercapacitor electrode, Phys. E: Low-Dimens. Syst. Nanostructures., 136 (2022) 115005, https://doi.org/10.1016/j.physe.2021.115005

[83]A.I. Habeeb, A.S. Al-Asadi, Fabrication of hybrid graphene nanosheets/Vanadium (V) oxide nanoparticles composite electrodes for supercapacitor application, Solid State Commun., 360 (2023) 115024, https://doi.org/10.1016/j.ssc.2022.115024

مراجعة حديثة للمكثفات الفائقة الكهروكيميائية المرنة

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المستخلص

أصبحت المكثفات الفائقة و بالخصوص تلك المرنة القابلة للارتداء نقطة ساخنة للبحث بسبب ما تتمتع به من مزايا مثل كثافة الطاقة، والسعة النوعية العالية، والخواص الميكانيكية الجيدة والاستقرار اللائق مع الأخذ في الاعتبار أنها غير مكلفة ولا تلوث البيئة. ان آلية التخزين في المكثف الفائق المرن هي آلية فارادية في المكثفات الكاذبة أو آلية غير فارادية كما هو الحال في المكثفات الفائقة مزدوجة الطبقة. لخصت هذه المراجعة التقدم البحثي في طرق تصنيع الأقطاب الكهربائية من مختلف أنواع المواد سواء الكربون أو أكاسيد المعادن أو البوليمرات، بالإضافة إلى نظرة عامة على ما قام به الباحثون في تغيير الشكل الأصلي للمواد ودمجها مع بعضها البعض، مع التركيز على طريقة تحضير الأقطاب الكهربائية المرنة وأيضاً كلا الأدائين. الكهروكيميائية والميكانيكية، والتعرف على بعض أنواع الإلكتروليتات المختلفة. وتناقش أيضًا وجهات النظر حول اتجاه التطوير المستقبلي للأبحاث في مجال FSCs.