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New Advancement of Supercapacitors Technology: a review

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Abstract

Supercapacitors are exciting for researchers due to their ability to store energy very quickly. This makes supercapacitors ideal for applications requiring rapid bursts of power, like regenerative braking in electric vehicles or short-term backup power for electronics. This makes them ideal for low-power devices we wear or carry around (like phones and smartwatches) and even powerful military equipment (like missiles and torpedoes). Scientists are figuring out how to improve SCs by tinkering with the materials they are made from and how those materials interact. These efforts are being done to make SCs a better option than other ways of storing energy, like rechargeable batteries. This article explores the latest findings on how SCs store energy, the materials used to make their electrodes (especially 3D structures that look like paper or fibers), and how they are being used in the real world.

1. Introduction

The Earth's supply of fossil fuels (oil, coal, gas) will run out sooner or later. People will have to rely on renewable energy from the sun, which is the world's primary energy source. As a result, scientists around the world turned to harvesting solar energy for use as electricity, heat, or fuels, the most important of which is electrical energy. To get the most out of the electrical energy generated by solar cells, it must be stored when available and used when not available. Humans discovered their passion for creating devices that store electrical energy, including capacitors and batteries, which they inked with solar cells to create a sustainable system that provides a constant source of energy.

Batteries, the most common electrical energy storage device, provide an adequate level of power and can be used for many different things, both large and small. It also emphasizes the fact that this power is essential for our daily lives. While batteries are excellent at packing of energy into a small space, they're not very good at releasing it quickly as shown in Ragone's plot (Figure 1) which compares how much energy-density storage devices can hold.

Therefore, their application in systems that need fast and high-energy storage is hampered. Conversely, due to their high energy density, excellent durability and fast charge-discharge processes, Supercapacitors are considered a promising alternative to batteries in energy storage systems [1]. A supercapacitor is a device that stores energy using electrochemical processes

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technology that can be divided into several categories based on storage techniques. Additionally, supercapacitors have properties that are halfway between capacitors and batteries. Supercapacitors and their many systems have advanced due to increased research and improvements. Ultracapacitors are another name for supercapacitors that have a higher energy density, and a higher power density compared to ordinary capacitors and batteries. They have good rate capability, cyclic stability, and power density [2].

This review aims to provide a compact rundown of the various concepts of supercapacitors and systems. The discussions of the present review are divided into four sections except for the introduction part. In Section 1, the types of storage mechanisms of supercapacitors and the basic components of a typical supercapacitor device, such as electrode materials, current collectors, electrolytes, and separators, are summarily discussed. In section 3, the majorly utilized electrode materials are generally categorized into three sub-sections: carbon-based materials, transition-metal compounds, and conducting polymers. Section 4 contains the advanced materials of supercapacitors, like MXenes. Finally, the futuristic perspective of high-performance supercapacitors in the domains of design, fabrication, and evaluation techniques towards superior capacitance and stability are summarized in Section 5.
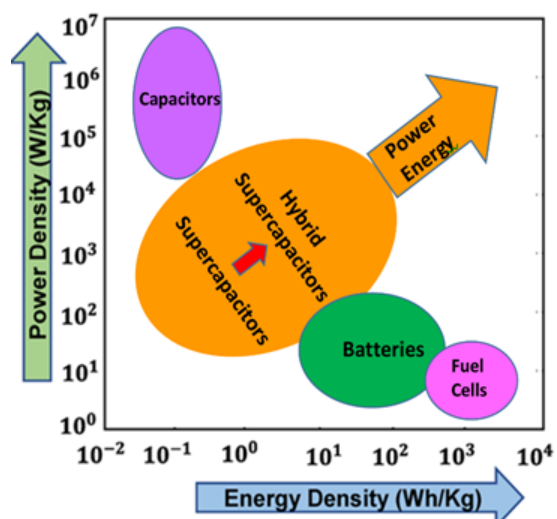

Figure 1. Ragone plot for different energy storage systems [1].

1.1. Differences between batteries and supercapacitors

Although there are some parallels between batteries and supercapacitors, there are several variances. The potential energy in a supercapacitor is stored in an electric field, whereas the potential energy in a battery is stored chemically. Chemical storage technique currently has better energy density than capacitors. The ability of a battery to discharge may be slower than that of a capacitor due to the latency associated with the chemical reaction that transforms chemical energy into electrical energy. Electrical energy can be stored directly above the capacitor electrodes and the capacitor's discharge rate is proportional to the conductivity of the capacitor electrodes. The supercapacitor can discharge and charge faster than a battery due to its energy storage mechanism, however, unlike a battery, which can turn its electrical current on and off, once a capacitor is connected to a circuit, it will rapidly discharge the charge until it is completely drained [3].

Despite their differences, batteries and supercapacitors have several applications in common. Batteries have higher energy density storage, but capacitors have a faster charge and discharge capability (higher power density). Table 1 shows a comparison between Capacitors, Batteries, and supercapacitors [4].

1.2. Supercapacitors: a brief history

Ewald Georg von Kleist created the first capacitor in 1740. Hermann von Helmholtz discovered state capacitive performance for solid-electrolyte interfaces in 1879. Becker, General Electric Corporation, invented the electric double-layered carbon capacitor in 1957. Cleveland's Sohio Corporation manufactured commercial electric double-layered capacitors in 1969. (EDLCs) NEC, a Japanese multinational corporation, renamed the double-layer capacitor "Supercapacitor" in 1978. Conway's team established pseudocapacitance with the increase in energy density in early 1990. Recently, much effort has been focused on other types of supercapacitors with asymmetric electrodes and hybrid systems [5].

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Table 1. Comparison between capacitor, battery, and supercapacitor

	Capacitor	Battery	Supercapacitor
Energy source	Storage of electrostatic charges	Chemical storage	Surface charge storage
Energy density	Very low energy density	High energy density	Low energy density
Power density	High power density	Low power density	Moderate power density
Capacitance	Capacitance present	Non-capacitive faradaic storage	Very high capacitance present
Life cycle	Largest life cycle	Small life cycle due to degradation	Large life cycle

2. Types of Storage Mechanisms

2.1. Mechanisms of non-faradaic storage

For non-faradaic energy storage, there are no electrons or ions transferred into or at the electrode surface. Ions are physically adsorbed on the electrode surface during this process. This mechanism is very reversible since there are no chemical changes that occur during ion adsorption-desorption. Intercalation between pores and non-faradaic charge storage happened in electric double-layered capacitors (EDLCs). There is no other process that took place in the optimally polarized electrode with no transferring of charge between the electrolyte and the electrode contact. Figure 2 shows an electric double-layered generated by the non-faradaic method.

2.2. Mechanism of faradaic storage

In Faradaic storage, charge moves from the electrode to the external circuit. This charge transfer is due to a redox reaction happening at the boundary between the electrode and electrolyte,

where reactants and products are stored as reservoirs in the bulk phases surrounding the electrode (Figure 3). The charges in the faradic mechanism are taken away from the electrode instead of being stored there. Pseudocapacitance, a way of storing energy in supercapacitors, involves electron transfer reactions at the electrode-electrolyte interface due to redox processes. The capacitive faradic method produces capacitance in a pseudocapacitor. In batteries, there is also a non-capacitive faradaic mechanism where energy is stored during a redox reaction and phase change. The non-capacitive faradaic mechanism relies on localized electron transfer processes at redox active sites. The broadband evolution is explained by the superposition of overlapping faradic redox molecules [6]. Furthermore, another study revealed that the formation of a charged layer at the electrode-electrolyte boundary is the reason for capacitance, even when there is charge transfer through the faradaic mechanism. Depending on the type of energy storage used, supercapacitors were divided into four types. (a) Electrostatic forces store energy at the electrode and electrolyte interface, (b) faradaic processes are used in pseudocapacitors, (c) an asymmetric supercapacitor utilizes electrodes consisting of two dissimilar materials, (d) a hybrid supercapacitor has a battery-type electrode and another made of carbon.
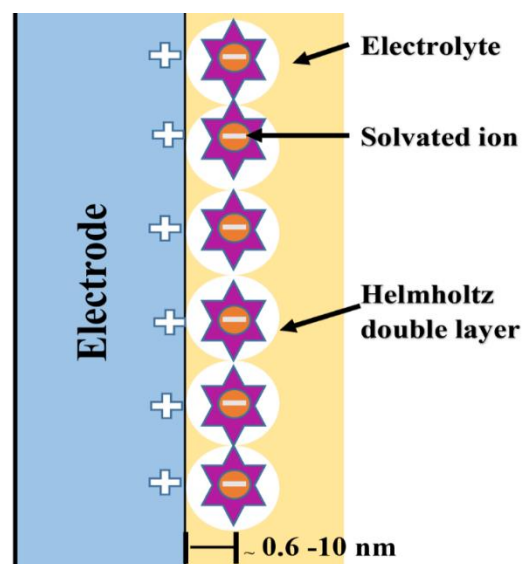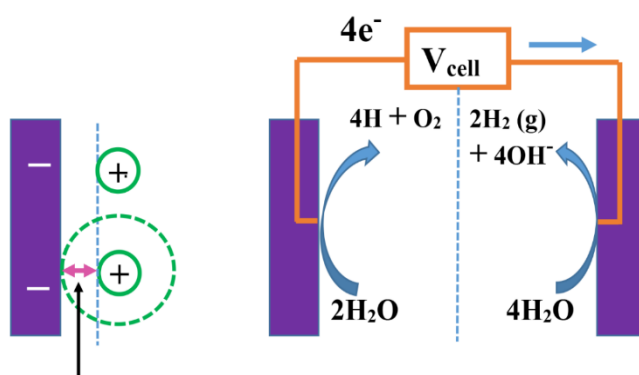

Figure 2. Electrochemical double-layer non-faradaic mechanism.

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Figure 3. Charge transfer in faradaic mechanism.

2.3. Supercapacitors components

The supercapacitor is made up of four major components (Figure 4). The most active components are electrodes and electrolytes, in which the charge storage mechanism is directly involved. The current collector and separator are the other key components.
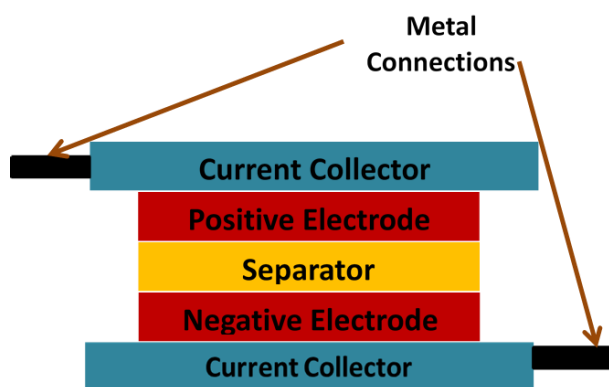

Figure 4. The components of supercapacitor.

2.3.1. Electrodes

Depending on the applied potential, electrodes can produce electric double layers or conduct a redox reaction. Electrostatically, electrochemically, or in combination, the energy was stored. For optimal interaction with the electrolyte and to achieve a greater specific capacitance, the electrode's high surface area is crucial, as well as a rich porosity distribution to boost redox activity and surface area [7]. Electrodes made of nanomaterials were used to achieve a larger surface area. Below are the numerous types of materials utilized in supercapacitor

electrodes. These components were also utilized to make nanocomposite materials by combining them [8].

2.3.2. Electrolytes

When an electrolyte is dissolved in a solution, it produces solvated ions. The electrolyte is primarily responsible for charge transport and balance between the two electrodes. They could increase supercapacitor energy density, increase life cycle, reduce internal resistances, control operating temperature, and reduce self-discharge. The electrolyte must have high ionic conductivity, a Large operating voltage range, and chemical stability, as well as be compatible with electrode materials, have a wide working temperature range, ideal viscosity, be non-toxic, and be inexpensive [9]. The mobility of the ions, their concentration, and the valence of migrated ion charges all affect the electrolyte's conductivity. Ionic conductivity is determined by the number of free ions present. Because the conductivity of different concentrations of the same electrolyte liquid varies [10].

2.3.3. Separators

Ion-conductive membranes and electrical insulators are used as separators. They keep the two electrodes with different polarizations from touching and offer electrical insulation. Ion migration from one electrode to the next is also possible using separators. Chemically inert, they also have a high mechanical strength, an ideal thickness, and a high porosity with superior electrolyte wettability [11]. Examples of separator materials include PVDF, Polypropylene, PTFE, cellulose polymer membranes (cellulose nitrate, cellulose acetate), Nafion, glass fibre, and others.

2.3.4. Current collectors

Large thermal and electrical conductivity, low contact resistance, good electrochemical and chemical stability, low corrosion resistance, and compatibility with the material of the electrode are all desirable qualities of current collectors [12]. It is an electrical conductor capable of collecting electrons from an electrode and transporting them to an external load [13]. In acid-

based electrolytes, corrosion-resistant metal foils such as gold were used; in alkaline mediums, Ni-based materials, carbon-based materials and stainless steel were used; in neutral electrolytes, ITO, Ni, CNT, stainless steel, and titanium oxynitride were used; and in non-aqueous electrolyte, aluminum was used.

3. Nanocomposite Supercapacitor Electrodes

The electrode materials affect the performance of various types of supercapacitors. Various materials and nanocomposites have recently been employed to create supercapacitor electrodes. Solid materials combining multiple phases, where at least one phase is incredibly tiny, on the order of nanometers, are called nanocomposites. Supercapacitors typically use three main kinds of nanomaterials for their electrodes: Nanostructured carbon, nanoparticles of metal oxides, and conductive polymers.

3.1. Supercapacitors electrode based on nanocomposite of carbon structures

Carbon materials are important for supercapacitors because they contain many types of carbon and are plentiful in nature. Carbon's thermal and chemical durability, as well as its high electrical conductivity, make it ideal for making low-cost supercapacitors. Furthermore, certain of its forms and structures are porous and have a large specific surface area [14]. The development of the EDL at the electrode-electrolyte interface is essential for energy storage carbon electrodes. As a result, the large surface area provides enough space for charges to accumulate, increasing both specific capacitance and energy density [15].

Carbon structures are classed as 0-dimensional (0-D), 1-dimensional (1-D), 2-dimensional (2-D), and 3-dimensional (3-D) materials based on their dimensionality. From 0-D carbon quantum dots (QDs) to 1-D carbon nanotubes, from 2-D graphene nanosheets to 3-D carbon spheres, these carbons structures display a uniform nanostructure unit. Moreover, there

is carbon with no clear structure unit, as activated carbon [16]. EDLC electrode is mainly produced by these nanostructures [17].

3.1.1. Activated carbon

In any dimension, three-dimensional materials are materials that can exist in any size, not just the nanoscale. This category includes graphite, activated carbon, carbon black, and carbon fibers [18, 19]. For EDLC electrodes, activated carbons are typically employed. Although the available surface area influences EDLC performance, besides surface area, other properties of the electrode material, like the arrangement and size of its pores, its overall shape, the presence of certain chemical groups on its surface, and how well it conducts electricity, all play a major role in how well an EDLC performs [20]. The electrochemical stability of activated carbon with a strong oxygen functional group but a small surface area is favorable. Scientists are looking for the ideal pore size in a material based on the size of ions it needs to interact with. They also want to find better ways to control the variety of pore sizes during the synthesis process. Research suggests that pores around 0.4 to 0.7 nanometers work well for watery electrolytes (aqueous), while slightly larger pores, around 0.8 nanometers, might be better for electrolytes based on organic solvents. The connection between pore diameter and ion diameter was proven in certain works by obtaining a maximum specific capacitance of Cs. Carbon compounds with functional groups on their surfaces may improve faradaic redox processes, resulting in a 5–10% improve in Cs [21]. We can turn various starting materials rich in carbon, like fossil fuels, coke, and even things like wood or coconut shells, into activated carbon using two different methods: chemical or physical activation [22]. However, controlling the pore size distribution of activated carbons formed through the oxidation reaction in the presence of water vapour and/or other chemicals is difficult. The template technique, in which a hard template (that should be removed using a post-treatment) or soft template (that decomposes during the synthesis process) is utilized, is one of the most effective

techniques for obtaining regulated pore architectures of carbons [23].

3.1.2. Graphene

Since its discovery in 2004, graphene has gained increasing attention as an electrode candidate for optimum supercapacitors, and many viable ways to synthesize it have been devised and perfected. Due to its remarkable properties, including superior electrical conduction, resistance to chemical degradation, and large surface area, a recently discovered one-atom-thick layer of carbon holds promise for use in energy storage devices [24]. This material is desirable due to its flexibility, low ion resistance, strong mechanical properties, and high surface area, graphene electrodes are frequently used in flexible supercapacitors. However, this film suffers from reversible stacking and agglomeration, which limits its application. Because of these limited ion transport issues, battery efficiency suffers. The solution consisting of conductive polymer and transition metal oxide was used to address this problem [25].

3.1.3. Carbon nanotubes CNTs

One-dimension carbon nanomaterials are carbon nanotubes (CNTs). Since first discovered in 1991, CNTs have captured significant scientific interest [26]. Due to their exceptional properties for strength, conduction, and structure, carbon nanotubes are essentially rolled-up sheets of graphene arranged in a seamless, hollow cylinder. They are essentially divided into two categories based on their structure: Single walled carbon nanotubes (SWCNT) and multi-walled carbon nanotubes (MWCNT). Because of their high electroconductivity, MWCNTs have been widely used as a conductive additive for secondary battery electrodes. CNTs have a unique structure, a narrow size distribution in the nanoscale range, a large surface area that is easily accessible, low resistance, and good stability [26–27]. The characteristics of CNTs suggest their potential as polarizable electrode materials. Due to their distinct characteristics, both SWCNTs and MWCNTs have been investigated as electrochemical supercapacitor electrodes. In an

aqueous solution, pure CNTs had a specific capacitance of 20–100 F g^{-1}, according to previous research [27]. The hydrophobic feature and the small specific surface area are to blame for the low capacitance. The presence of functions in the CNT matrix has been shown to significantly improve the specific capacitance. Adding a hydrophilic group also enhances the wettability of the aqueous electrolyte on the CNT surface. The electrochemical behavior of SWCNTs in terms of their electronic structure, their wide potential window in organic electrolytes, and their excellent formation ability in the absence of electrochemically active materials are all highlighted as important characteristics of designing high-performance capacitor devices.

Carbon nanofibers CNFs

Researchers are excited about using carbon nanofibers (CNF) in supercapacitors. This is because CNFs are long, thin structures (one-dimensional) with a large surface area, good electrical properties, and resist breaking down over time. Chemical vapor decomposition, electrospinning, biomaterial pyrolysis, template-assisted solvothermal, and other methods can be used to manufacture high-quality CNF [28]. The electrospinning method is commonly used to generate CNF due to its cost-effective approach, ease of management, and ability to produce a wide range of CNF diameters ranging from a few microns to 100 nm. The synthesis is usually a two-stage process, with the first step consisting of nanofiber pre-synthesis and the second step consisting of carbon fiber carbonization. The insertion of a catalyst during the activation or graphitization process, or the use of more than two polymers, might alter the morphology of carbon fibers. The introduction of redox-active materials can improve electrochemical capacitance, while the incorporation of CNT and metal nanoparticles can improve electrical conductivity [29].

3.1.4. Fullerenes

Fullerenes (C60) are a type of carbon with a closed mesh structure formed by a single and double bond between carbon

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atoms [30]. In 1985, Smalley and colleagues discovered fullerene in the mass spectrum of carbon soot. Due to its good conductivity and thermal and chemical stability, carbon soot was created by vaporizing graphite under constant helium gas flow. It has subsequently attracted interest as a filler ingredient in polymeric composites for SC applications. One of the difficulties with fullerene-filled nanocomposites is their limited solubility in organic composites, which causes the fullerene to lump in the matrix. This problem can be solved by modifying fullerene physically or chemically [30].

3.1.5. Carbon quantum dots CQDs

Carbon quantum dots (CQDs) are zero-dimensional carbon nanomaterials with a diameter of less than 10 nm. They have a large specific surface area, low toxicity, chemical stability, and biocompatibility. CQDs made from carbon nanotubes emitted a vivid and colorful photoluminescence. The quantum confinement effect, which is common in C-QDs, is responsible for these remarkable optical qualities. The smallness of the solvated molecules and the pore size influence ion movement in microspores. In other words, when tiny solvent molecules and ions are smaller than the pores, ions have difficulty passing through the restriction and entering the pores. As a result, while increasing microspores can increase specific surface area, it does not always increase specific capacitance (Cs). Mesoporous, on the other hand, is more favorable to fast ion transport, leading to improved electrochemical characteristics. Furthermore, the pore size distribution must be taken into account. The ion transport length can be reduced by using a porous structure with a narrow distribution, which enhances electrode kinetics. As a result, an appropriate pore size and distribution are required for optimal performance [31]. Because of their great charge mobility, carbon dots can be produced via oxidation-reduction processes. One advantage of the composite electrode material is its fast charging time and high power density. However, its ability to store energy (energy density) and store charge per unit mass (specific capacitance) is lower than other materials. This limitation is addressed by combining conductive polymer and transition metal oxide in the composite electrode material.

3.2. Metal oxides nanoparticles

Metal oxides are becoming increasingly popular for use in supercapacitors. This is because they offer several advantages over other materials commonly used for this purpose. Compared to carbon materials, metal oxides can store more energy, and they are more stable and durable than polymer materials when used in electrochemical applications. Metal oxides such as Co_3O_4, RuO_2, MnO_2, V_2O_5, NiO, SnO_2, TiO_2, Fe_3O_4, and $NiCo_2O_4$ are among the most studied ones in supercapacitors due to their environmental benefits, superior theoretical capacitance, and outstanding chemical stability [32]. However, the main drawback preventing supercapacitors from being widely used for storing energy is that they can't hold as much energy compared to their size and weight. Ideally, materials for supercapacitor electrodes should have several features: lots of surface area to maximize the places where energy can be stored, low resistance to allow for fast charge flow, pores that are the right size for ions to move through quickly, and a metal that can change its oxidation state to store even more energy. By making better use of the electrode material and shortening the distance traveled by electrons and ions, these features can improve the specific capacitance, cycle performance, and usable potential range of the device. Metal oxides, unfortunately, have major flaws in the form of low conductivity. As a result, innovative research is required to produce new electrode materials that combine metal oxides and carbon-based materials to provide efficient supercapacitor electrode materials. Metal oxide nanoparticles must be incorporated into carbon nanomaterials such as carbon fibers, medium porous carbons, carbon nanotubes, graphene, and so on. It also increases composite materials' electrical conductivity and mechanical strength [33].

Ruthenium oxides

Ruthenium oxides such as RuO_2 and RuO_2xH_2O, were unique surface redox pseudocapacitive materials. According to

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their good thermal stability, high conductivity, and high thermal stability, ruthenium oxide electrode material for SCs has gotten great attention [32].

Manganese oxides (MnO$_2$ and Mn$_3$O$_4$)

These oxides are gaining significant interest as a material for pseudocapacitive electrodes due to their large energy storage capacity, affordability, plentiful supply, and minimal environmental impact. Wenjing Wang with his teamwork fabricated a novel and flexible Mn$_3$O$_4$ nanosheets/reduced graphene oxide nanosheets (Mn$_3$O$_4$ NSs/rGO NSs) paperlike electrodes have been developed. Flexible Mn$_3$O$_4$ NSs/rGO NSs electrode exhibited a high specific capacitance of 409 F g/1 at a current density of 0.5 A g/1, and superior cycling stability of 92% after 3000 charge-discharge cycles [33].

Iron oxides (Fe$_2$O$_3$ and Fe$_3$O$_4$)

Researchers are interested in iron oxides as potential electrode materials for supercapacitors due to their predicted high capacity, earth-abundance, large operating window in negative potential, non-toxicity, and inexpensive cost. Various single iron metal oxides/hydroxides (Fe$_2$O$_3$, Fe$_3$O$_4$, FeOOH, etc.) and binary metal oxides (CoFe$_2$O$_4$, NiFe$_2$O$_4$, etc.) have been extensively studied as SCs electrodes. A large number of iron-based nanostructures with desirable morphologies, including 0D, 1D, 2D, and 3D architectures, have been developed and exhibited improved electrochemical properties [34].

3.2.1. Pseudocapacitive electrode materials dominated by intercalation

Various metal oxides fabricated to be used as supercapacitor electrodes by intercalation for their pseudocapacitive behavior such as:

Vanadium oxides

Vanadium oxides (VO$_2$, V$_2$O$_5$, V$_2$O$_3$) are extensively used as pseudocapacitive electrode materials. However, due to their weak electrical conductivities (10^{-2} - 10^{-3} S cm^{-1}) and structural instability, vanadium oxides' electrochemical properties were limited [35]. As a result, many strategies have been proposed to enhance the electrochemical properties of vanadium oxide-based electrode materials, such as hybridization with highly conductive materials and structural control.

Tungsten oxide (WO$_3$)

Tungsten oxide (WO$_3$) is regarded as a suitable choice for SCs because of its strong electronic conductivities, environmentally friendly, large theoretical specific capacitance of 1112 F g^{-1}, and low cost [36].

Molybdenum oxides

Due to their excellent capacitance, abundance in nature, affordability, and minimal environmental impact, Molybdenum trioxide (MoO$_3$) and Molybdenum dioxide (MoO$_2$) are considered promising candidates for use in SCs [37].

Niobium oxide (Nb$_2$O$_5$)

The high theoretical capacitance, exceptional structural stability, and intercalation of the pseudoelectrode material of Nb$_2$O$_5$ have aroused interest. To further facilitate the practical application of Nb$_2$O$_5$-based electrode materials in supercapacitors, it is essential to optimize the combination of anode components, such as binders and conductive agents, and the device components such as cathode, electrolyte, membrane, current collector, as well as the fabrication techniques. Concerning the anode materials, the innovative, green, inexpensive, and controllable synthesis methods for preparing Nb$_2$O$_5$-based materials with practical applications should be considered as the top priorities [38].

Titanium oxide (TiO$_2$)

Anatase, brookite, rutile, and TiO$_2$ (B) are some of the polymorphs (bronze). Because of its lower density and more porous structure, TiO$_2$(B) has gotten great attention as an electrode material for intercalation-dominated pseudocapacitors [39].

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3.2.2. Battery-type materials

Electrode materials have ever long been confused between battery-type materials and the traditional pseudo-capacitive material. The main difference between them is that the battery type involves phase change behavior, while the pseudocapacitive does not exist crystallographic phase change during the electrochemical process.

Nickel oxide (NiO)

Nickel oxide has a large theoretical specific capacitance of 2584 F g^{-1} (with a voltage range of 0.5 V), great thermal and chemical stability, and is environmentally friendly. It has attracted significant research interest and is known to be one of the promising options for SCs [40]. The key to storing charge in a nickel oxide pseudocapacitor electrode lies in the reactions happening at the surface between the electrode and the electrolyte. These reactions include both electron transfer (redox) and the formation of an electrical layer, with redox reactions playing a bigger role. The performance of the electrode is ultimately determined by how well nickel oxide reacts in the alkaline electrolyte.

Cobalt oxide (Co_3O_4)

A key advantage of this material is its low cost, ease of fabrication, non-toxic, environmentally friendly nature, and most importantly, high theoretical capacitance, Co_3O_4 is regarded as a highly promising material for supercapacitors (3560 F g^{-1}) [41].

Copper oxide (CuO)

CuO has been considered a suitable material for supercapacitor electrodes because of its non-toxicity, low cost, abundance, and ease of fabrication in many nanoscale forms, as well as good electrochemical performance [42]. CuO is classified as a battery-type material based on its reaction mechanism.

Binary transition metal oxides

Binary metal oxides comprise one transition metal ion and one metal ion that is either electrochemically active or inert. They have been employed as electrode materials for supercapacitors because they have substantially better electrical conductivity and electro-capacitive activity compared with single metal oxides. With their large specific capacitance and good rate capability, spinel cobaltite (MCo_2O_4; M = Mn, Zn, Ni, Cu, etc.), binary metal oxides are a viable choice for supercapacitors [43]. Spinel cobaltite, on the other hand, combines the benefits of both metal ions [44]. $NiCo_2O_4$ is the most prevalent pseudocapacitance material among the numerous spinel cobaltites.

3.2.3. Transition metal oxides (TMOs) -based composites for SCs

According to their response mechanism, composite materials are divided into two categories. The first method involves combining metal oxides with carbon-based compounds or conductive polymers. The "hierarchical structure" is a sort of composite material that combines one metal oxide substance with one metal-based material. In addition, the hierarchical structure may inhibit the aggregation of electrically active elements, which would result in increased particle size and mass loading. As both components have large capacitance, the hierarchical structure may be acceptable for practical use to generate high power densities.

Carbon-based composite

In electrical double-layered capacitors (EDLCs), energy is stored for carbon materials through electrostatic attraction between ions in the electrolyte and the electrode surfaces. Unlike batteries, there are no chemical reactions involved during charging or discharging [45]. This storage system is mainly responsible for quick charging and discharging, resulting in high power density and super cycling stability. As a result, enormously attractive qualities such as pore size distribution, high surface area, lightweight, conductivity, and compatibility

with the other materials distinguish carbon materials as prospective TMO composite pairing materials.

Metal oxides/carbon nanotubes (CNTs)

Because of the high energy density of metal oxides, composites present a viable method for practical applications. CNTs have great chemical and physical stability, a little gravimetric density, and a high electric conductivit (105 S cm^{-1}), making them a good mesoporous matrix for metal oxides. The produced MnO_2/CNT has a higher volumetric energy density, excellent cycling stability, and exceptional flexibility [46].

Metal oxides/graphene

Most metal oxides, including MnO_2, Co_3O_4, and V_2O_5, have low electrical conductivity and are hence high bandgap semiconductors, limiting their practical application. Graphene is thought to be a monolayer sheet of graphite [46]. The term 'graphene' is usually broadened to be multilayer in current work [47]. It contains a bilayer and few-layer but is less than 10 layers, which is also utilized to express graphene. Graphene's properties are determined by the arrangement of its atoms, even though it only contains one element. The material is attractive due to its many appealing properties, including a large surface area, ease of processing, and excellent electrical performance, metal oxides/graphene composites are widely used as an ideal electrode material [48].

Metal oxides/other carbon materials

Researchers are focusing on porous carbon/metal oxide hybrids for supercapacitors for their high surface areas and strong electrical conductivity. To enhance the durability and conductivity of metal oxide micro/nanostructures, carbon coating, specifically core-shell nanostructures, has been created as protective and electrically conductive layers [49].

Metal oxides/conducting polymers

Conducting polymers are a type of organic polymer with a conjugated bond structure that allows them to conduct electricity. Due to their large energy density, low cost, environmentally friendly nature, and reversible Faradaic redox characteristics, conducting polymers have piqued the interest of several supercapacitor applications. Polyaniline (PANI) and polypyrrole (PPy) are two extensively used supercapacitor conducting polymers that have been identified as a suitable candidate for metal oxide composites due to their high conductivity, flexibility, and ease of production [50].

Metal oxides /metal compounds composites

Due to the synergetic effect, Supercapacitors that need both high energy storage and fast power delivery work best when their electrodes are built using a layered structure that combines metal oxides with other metal-containing materials. Wei et al., 2014 [51] advanced 3D ZnO-NiO mesoporous structures for supercapacitors. The research of metal oxide nanostructures will pave the way for the next generations of flexible supercapacitors to achieve acceptable power densities at large power densities.

3.3. Conducting polymers (CPs)

Conducting polymers such as PANI, polyindole (PIn), PPy, and polythiophene (PTh) have been increasingly popular in the field of supercapacitors in recent years. There has been also significant progress recently due to their high capacitances and ability to resist multiple charge-discharge cycles if designed properly [50].

3.3.1. Polyaniline (PANI)

PANI is considered a promising conducting polymer due to its low cost, ease of manufacture, outstanding thermal and electrical conductivities, environmental stability, and wide range of applications, [52]. The fundamental advantage of employing PANI is its substantially larger capacitance than some other conducting polymers because the reversible redox process of PANI can remove up to one electron for each of two monomer units [53].

Unfortunately, if PANI is employed in extreme settings, structural disintegration may occur. As a result, polymer-based composites with increased thermal stability, mechanical

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characteristics, and multiple functional locations on the polymer backbone have been produced by incorporating diverse materials including zeolites, metal, metal sulphide, carbon nanotubes, and metal-organic framework (MOFs) into the polymer backbone [54].

3.3.2. Polyindole (PIn)

PIn and its derivatives display both poly(paraphenylene) and polypyrrole characteristics at the same time [55]. It's very active in electrochemical reactions and withstands high temperatures. Affordable, stores energy efficiently, is simple to produce and is eco-friendly are just a few of the benefits [56]. In addition, PIn also has a slower hydrolytic breakdown than PANI, and PPy demonstrates that it excels at storing energy for extended periods. PIn is a desirable material in the creation of supercapacitors with superior performance and extended lifespans because of all these features. However, large-scale implementation is still a long way off because of two significant flaws: weak cycle stability (due to the flimsy backbone) and low electrical conductivity [57]. Several research projects have recently been undertaken to address these issues. In acidic electrolytes, however, many PIn derivatives with a high electron drawing group show exceptional ability to conduct electricity and a substantial value of specific capacitance [58].

3.3.3. Polypyrrole (PPy)

PPy stands out as a very promising material (p-type conducting polymer) for building high-performance pseudocapacitors. Attributable to its unique combination of properties: excellent conductivity, rapid charging and discharging, resilience to heat, affordability, and the ability to store significant energy [59]. Despite its simple synthesis and impressive ability to transfer electrons. PPy remains the most studied conducting polymer. This is a testament to its valuable properties, making it a promising candidate for various battery and supercapacitor applications. Unlike traditional capacitors, PPy's inherent faradaic storage allows it to hold a larger charge.

3.3.3.1. PPy and composites

Wu et al., 2015 [60] create a new core–shell polypyrrole/graphene oxide (PPy/GO) nanocomposite of homogeneous PPy nanospheres by in-situ polymerization technique. A continuous core–shell composite architecture was produced by consistently growing 70 nm PPy nanospheres on GO sheets. Evaluating the performance of the composite electrodes in a 1 M H_2SO_4 electrolyte, a higher specific capacitance of about 370 F g^{-1} compared to 216 F g^{-1} for pristine PPy, at 0.5 A g^{-1} in the voltage range from 0.2 to 0.8 V. The PPy/GO composite electrodes showed 91.2 % of capacitance retention across 4000 CV scanning over 100 mVs^{-1}. In addition to materials like graphene and carbon nanotubes, researchers can also use composites combining polypyrrole (PPy) with activated carbon (AC), graphite films, or carbon cloth for various applications [35, 61, 62].

3.3.4. Polythiophene (PTh)

PTh is a five-membered heteroaromatic ring that is highly stable from both an electrochemical and environmental standpoint. The presence and prolonged π-conjugated system in PTh give promising characteristics that include high conductivity, optical, electrochemical, thermoelectrical, and electrochromic properties. In particular, the chain length of conjugation in the PTh polymer backbone affects its optical properties. That is, PTh shows a significant red shift in their absorption properties with the increase in the conjunction chain length. Mainly, the tunable properties of PTh could allow its effective utilization in electrical systems. Poly(3,4-ethylenedioxythiophene) is a strong electrical conductivity, low redox potential, as well as high thermochemical stability, among the various varieties of polythiophene that exist [63].

3.3.4.1. Polythiophene/carbon nanocomposites

Several studies have explored supercapacitor electrodes made by combining polythiophene (PTh) with carbonaceous materials, aiming to enhance functionality. One such example is the work by Fu et al., 2012 [64] created a nanocomposite

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electrode (PTh/MWCNT) by electrochemical polymerization in an ionic liquid electrolyte. The nanocomposite electrode outperformed with a specific capacitance of 110 Fg^{-1} at a scan rate of 60 mV/s. Furthermore, the supercapacitor had a remarkable level of stability, retaining 90% even after 1000 cycles. Zhang et al., 2014 [65] prepared PTh/MWCNT nanocomposite using an electrochemical polymerization process with a thickness of 2–3 nm on the MWCNTs. With good electron transfer rates and low resistance, the nanocomposite had a specific capacitance of 216 Fg^{-1} over a constant current density A/g. Wang et al., 2013 [66] manufactured PTh/MC composite, electrochemically forming CPs comprising PTh on the surface of monolithic core-like porous carbon (MC) and fabricating a range of composite electrode materials. Due to the large specific surface area of MC, the specific capacitance for the PTh/MC could achieve 720 Fg^{-1} at 0.5 A/g, and the composite could retain 79.4 percent of its initial value after 1000 cycles at a current density of 1.0 A/g. Hür et al., 2013 [67] created three types of active electrodes using PTh, poly(3-methylthiophene) (PMeT), and PEDOT-coated pencil graphite electrodes (PGE) using cyclic voltammetry and investigated their supercapacitor capabilities. When the scan rate was 10mV/s, the specific capacitance of PTh/PGE was 1503.20 m Fg^{-1}, which was lower than the other two composites but was more stable than PMeT/PEG and PEDOT/PEG. Kim et al., 2015 [68] investigated the influence of graphite oxide as a chemical oxidant on the construction of reduced graphite oxide-CPs such as PANI, PPy, and PTh. They found that graphite oxide can be used as a chemical oxidant for a variety of CPs, including PTh, with a simple and quick fabrication approach. Alabadi et al., 2016 [69] demonstrated the synergistic effect of microstructure and doping in G by designing and preparing G-TB and GO-TB, and they found that in-situ polymerization of conjugated polymer poly [(thiophene-2,5-diyl)-co-(benzylidene)] (PTCB) on graphite and GO sheets resulted in G-TB and GO-TB, respectively. The GO-TB composite demonstrated good electrochemical performance in the form of lower equivalent serial resistance, remarkable

capacitance, and energy density of 296 Fg^{-1} and 148 W h/kg, respectively, as well as strong cycle stability and ionic conductivity, according to the results. PTh/carbon material composites have better electrochemical performance and cyclic stability than single components due to a synergistic effect, and these properties are influenced by several factors including the effective deposition amount of PTh on carbon materials, the type and microstructure of carbon materials, the synthesis route, and so on. Melo et al., 2017 [70] investigated PTh and graphene composites with various mass proportions for use as supercapacitors. They used cyclic voltammetry to assess the electrochemical behavior of these composites and specific capacitance curves were constructed from the results. The behavior of the different composites and graphene to faradaic impedance spectroscopy was also investigated. Based on these findings, it was shown that a 1:1 mass composite of graphene and PTh had a greater specific capacitance than graphene alone. In comparison to the solitary polymer, the use of graphene in that proportion also improved cyclic stability. This composite material's high specific capacitance (365 Fg^{-1} at 1 Ag^{-1}) suggests that it could be used as an electrode material for supercapacitors. The supercapacitor capabilities of the resultant materials were evaluated and compared. The best composite's specific capacitance could achieve 365 Fg^{-1} at a discharge current density of 1 Ag^{-1}, which is significantly higher than 2006 conductive PTh, the mixture may allow for the creation of low-cost, high-performance electrode materials for supercapacitors.

3.3.4.2. PTh /metal oxide composites

Metal oxide is an essential electroactive material that can be used to make PTh-based composites. So far, there has been less research on PTh/metal oxide nanocomposites than on PANI/metal oxide and PPy/metal oxide composites. This could be because the specific capacitance of a supercapacitor with a pristine PTh electrode is smaller than that of PANI and PPy. After all, there is no practical way to increase the properties. Recently, some research on PTh/metal oxide composites has been completed, and some progress has been made [71, 72].

However, more research on PTh/metal oxide composites is needed to improve their electrochemical performance.
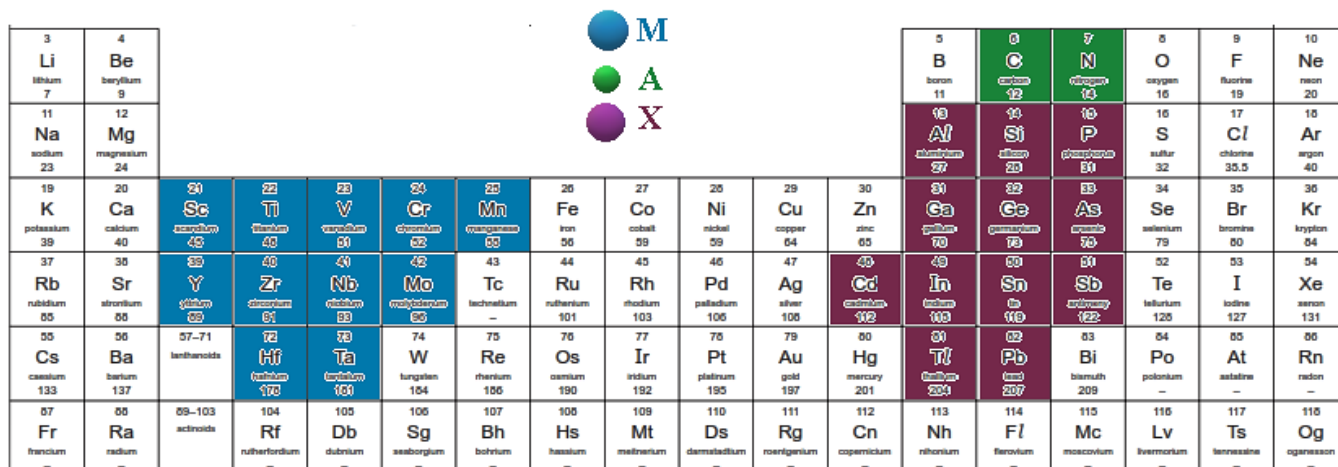
4. Advanced Materials of Supercapacitors

4.1. MXenes

A recent discovery from 2011 by Basroum and colleagues at Drexel University is a new class of 2D materials called MXenes [73]. It is a novel inorganic material with a two-dimensional structure composed of various transition metals nitrides, carbides, and carbonitrides [74]. It has a general formula of $M_{n+1}X_nT_x$, where M represents the transition metal like Ti, V, Cr, Zr, Mo, Nb, Ta, etc, and n=1,2 or 3, while X is carbon or nitrogen and T symbolizes the termination by fluorine, Oxygen or hydroxyl. Mxenes are characterized by their hexagonal crystal structure like the MAX family with the formula of $(M_{n+1}AX_n)$ with the same structure of Mxenes in addition to A symbol of main group elements 13 and 14 of (Al, Si, P, S, Zn, As, Pb, Cd, Sn, Ga, In, Ti, Ga and Ge), which has a combination between the properties of metals and ceramics of thermal stability, thermally conductive, resistance to oxidation, so they can obtain the Mxenes by chemical delamination through etching the A element layer in Max phases of three or quaternary dimensional carbides and nitrides (Figure 5). For example, $Ti_3C_2T_x$ was the first discovered Mxene in 2011 by etching Al from Ti_3AlC_2 in HF solution, in a similar manner, the direction of the research field started to prepare different Mxenes such as V_2C, Nb_2C, Mo_2C and Ta_4C_3 [75-83]. The termination of Mxenes by O, OH improves the hydrophilicity and the electrochemical characterization which improves the reaction of ions with electrolyte and produces the peudocapacitance, also Mxenes have thermodynamic stability. There are three types of Mxenes: pristine, in-plane and out-plane Mxenes. The pristine consists of one single transition metal, as mentioned like TiC_3C_2, but for more improvement of these compounds, the researchers have succeeded in the preparation of Mxenes formed of binary or double transition metal and classified it as in-plane and out-of-plane double transition metal as a multilayer with high thermodynamic stability. The out-of-plane ordered Mxenes (O-Mxenes) are fabricated from layers of two different transition metals represented by M' and M'', where X atoms layers are inserted between layers of M' and M'' to obtain the general formula of $M'_2M''X_2$ or $M_2'M''_2X_3$ with the same hexagonal structure, while the in-plane ordered Mxenes have the formula of $(M'_{2/3}M''_{1/3})_2X$ or M1.33X with ordered vacancies. For more interesting the researchers started to report elements like Sc, W, and Y that had never been used before such as MO_2ScC_2Tz and Cr_2TIC_2 as out-plane ordered Mxenes. Moreover, they produce in-plane Mxenes as $(Mo_{2/3}Y_{1/3})CTz$ by exfoliation method of i-MAX by etching Al layer or removing M element with Al element to form $Mo_{1.33}CTz$ and $W_{1.33}CTz$ compounds [84-88].
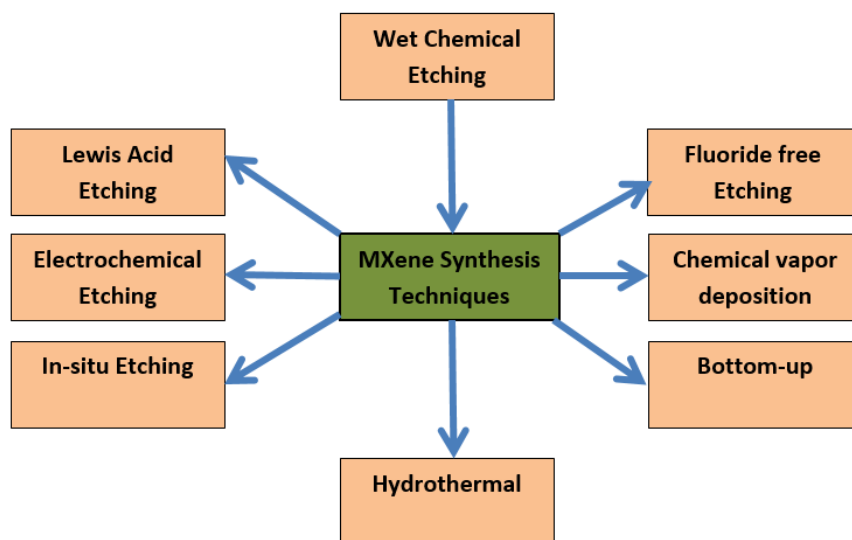
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Figure 5. Elements formed MAX phases Mn+1AXn.

Figure 6. possible routes for MXenes fabrication.

4.1.1. Fabrication of MXenes

MXenes can be prepared by two main methods, top-down and bottom-up. The top-down method is a chemical etching to remove the reactive layer A element from the MAX phase to form MX layers and finally terminated groups such as O, OH, F, the etching process via HF hydroflouride or using milder alternative salts like HCl/NH_4F, HCl/KF and HCl/NaF [89, 90]. The Al layer could be removed by alkali etching, for example, Ti_3AlC_2 was etched by using tetramethylammonium hydroxide (TMAOH) after the treatment with 20-30 wt % of HF [90]. On the other hand, electrochemical etching was also used for MXenes preparation such as Ti_2CTx was formed by electrochemical etching of Ti_2AlC in dilute HCl aqueous electrolyte and then terminated by O, OH, or Cl [87]. Molten salt etching is another type which obtained in molten fluoride salt at 550 °C using argon. Bottom-up involves techniques like chemical vapor deposition (CVD) and atomic layer deposition

(ALD) [92]. Figure 6 illustrates possible routes for MXenes fabrication.

4.1.2. Properties and applications of MXenes

The reported MXenes showed and characterized by metallic conductivity, and excellent thermal stability, also the termination with oxygen and hydroxyl groups increases the hydrophilicity which increases the interaction with the electrolyte ions, large surface area, Low diffusion barrier, and biocompatibility [93]. Therefore, MXenes compounds could be used in different applications such as biomedical applications, toxic removal applications, sensors, and energy storage devices like lithium or sodium Batteries and supercapacitors. The terminated OH, O, and F groups create negative charges on the surface of the MXenes and lead to the formation of an active surface that can react with positive charges [94].

4.1.3. MXene nanocomposites for supercapacitor electrodes

To improve the electrical, mechanical, and thermal properties, MXenes nanoflakes were combined with monomers. Polypyrole, polyaniline, and polythiophene as poly (3,4-ethylene dioxythiophene (PEDOT) have been involved with MXene by electrochemical deposition or photopolymerization techniques [95]. Adding conducting polymers to MXene as nanocomposite electrodes (NCPs) for electrochemical energy storage improves flexibility and grants pseudocapacitive properties, low cost, and non-toxicity [96]. The conjugated polymers act as an electrochemically active material with a different band gap, charge transportation, and good processability with different structures [97]. Metal oxides with good redox and pseudocapacitive characteristics could be used as composites with MXene to produce electrode material with good electrochemical properties in supercapacitors. MXene/WO_3, [98], MXene /TiO_2, and MXene /SnO_2 NCPs are examples of MXene/ metal oxide NCPs and could be prepared by in-situ growth or ex-situ mixing and self-oxidation. MXene composites based on carbon materials like graphene, carbon nanotubes (CNTs), and carbon nanofibers (CNF_S) are also are also used for supercapacitor electrodes, and as a spacer interlayer between MXene flakes. Adding carbon-based materials to MXene nanocomposite electrodes (NCPs) significantly boosts their conductivity, flexibility, and overall electrochemical performance [99, 100]. Venkateshalu et al., 2020 synthesized V_2NTX MXene for a supercapacitor electrode by AL etching from V_2AlN after immersing it in LiF-HCl salt acid. They obtained a specific capacitance value of 112.8 F g^{-1} at a current density of 1.85 MA/cm^2 [101]. Liang and Zhitomirsky, 2021 reported the preparation of a composite of Ti_3C_2TX-Fe_3O_4 and multi-walled carbon nanotube (CNT) electrodes. They obtained areal capacitance CS of 5.52 F cm^{-2} using a sodium sulfate electrolyte at a concentration of 0.5 molar [102]. Wang et al., 2021 created composites combining polyaniline (PANI) and MXene (V_2C) through a process called in situ polymerization as building blocks of supercapacitors. They designed PANI/MXene as the positive electrode (anode) and active carbon as the negative electrode (cathode). They achieved a capacitance of 337.5 F g^{-1} at a current density of 1 A/g [103]. De et al., 2021 synthesized Ti_3C_2Tx (MXene) based polyindole nanocomposite material to create both symmetrical and asymmetrical supercapacitors. They produced a specific capacitance of 226.5 F g^{-1} at a current density of 2A/g and cyclic stability of 90.5 % for 8000 cycles. However, when used as an anode in an asymmetric device, the composite achieved a specific capacitance of 117 F g^{-1}[104]. Liu et al., 2021 synthesized Ti_3C_2TX@PEDOT composite and PEDOT of poly(3,4-ethylenedioxythiophene) by in-situ polymerization and produced 564 F g^{-1} specific capacitance at 1 A g^{-1} current density [105].

5. Conclusions

Supercapacitors due to their excellent properties are a promising source of energy, especially as alternatives to non-renewable sources. However, the huge potential is accompanied by great responsibilities. Most of the materials used for supercapacitors are carbon-based. The true potential of

supercapacitors can be explored when it is viable for mass production. There are many industrial applications of supercapacitors and production lines dedicated to this. Materials with excellent properties should be scaled up for mass production. A combination of research and production could enable the reaping of the benefits of supercapacitors. However, there's a catch: they can't store as much energy (low energy density) and are expensive to make right now. Researchers are working hard on new materials and designs to overcome the limitations and make SCs the next big thing in power storage. There are some works already being carried out in the direction of flexible supercapacitors. Biocompatible wearable fabric with supercapacitor-integrated devices could be explored for applications such as medical and defense.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

Data will be made available on request.

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