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Review article

Recent advances in synthesis of MXene-based electrodes for flexible all-solid-state supercapacitors

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ABSTRACT

Various energy storage sources have been developed so far, among which supercapacitors are more important for the forthcoming generations due to their small size and portability. Supercapacitors as good alternatives to batteries have recently attracted more attention because they have higher power and excellent charging-discharging rate which is considered as a challenging issue that limits the use of batteries. Supercapacitors also have other advantages over batteries, including higher reversibility and cycle life, lower maintenance costs, and safer electrode materials. MXenes have emerged as a new class of 2D composites in electrode materials for supercapacitors as low-cost and environment-friendly carbides and nitrides. MXenes are suitable inorganic compounds with excellent electrochemical properties and mechanical integrity to improve supercapacitor energy density at a new interval. This review presents new synthesis strategies to prevent the self-accumulation of MXene layers. First, the fundamental working theories of different supercapacitors are outlined. Next, an overview of the electrode material based on MXenes is outlined, and the latest solutions for increasing the active sites and improving the ion transfer rate have been collected. Hybridization and doping of MXenes change the properties of the composite, leading to a transformation in the structure and an increase in the capacitance. Furthermore, the utilization of double-transition metal MXenes solves challenges such as structural destruction and short life spans in multiple charge-discharge cycles. Then evaluation of the new MXene-based electrode materials in all-solid-state supercapacitors has been summarized. Finally, an overview of the latest developments in the creation of all-solid-state flexible supercapacitors as well as our predictions for future lines of inquiry is provided.

KEYWORDS

Supercapacitors
Electrode
Synthesis
MXene
Energy storage

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1. Introduction

Following the increasing consumption of fossil fuels and environmental problems, finding new energy sources has become a challenging issue [1]. The rapid development of countries has doubled the fuel consumption problems [2]. Therefore, many efforts have been made to design and manufacture energy storage devices with easy transportation and zero-emission of pollutants in the form of electrical-based. In this regard, reliable energy storage methods including supercapacitors with high power density are an attractive topic. A

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supercapacitor or ultracapacitor is a type of capacitor that has a much larger capacity than ordinary capacitors to accumulate electric charge. Supercapacitors an alternative energy storage technology, consist of two conductive parallel metal plates with different materials (called electrodes), creating a larger energy storage area [3]. These two plates are immersed in an electrolyte gel or liquid including positive and negative ions, which separate these ions when a voltage is applied to the two capacitor electrodes. For this reason, one of the other names of supercapacitors is an electric double-layer capacitor. Supercapacitors have the ability to charge and discharge hundreds of thousands of times with the most minor drop in performance and quality, long life cycle, low charge and discharge time, easy maintenance, and high heat tolerance or low temperature up to -40 °C, it is popular among researchers of universities and industry [4]. Unlike resistors, which waste energy in the form of heat, ideal capacitors store energy without loss [5].

Although the supercapacitor is an electrochemical device, it does not use any chemical reaction to store electrical energy [6]. This means that the supercapacitor is effectively an electrostatic device that stores electrical energy in the form of an electric field between its two conductive electrodes [7], as shown in Fig. 1. The electrodes, both sides of which have a coating, are made of graphitic carbon in the form of activated conductive carbon, carbon nanotubes, or carbon gel [8]. A paper membrane with tiny pores called a separator keeps the electrodes apart but allows positive ions to pass through while blocking electrons [9]. Both the paper separator and the carbon electrodes are immersed in the liquid electrolyte, and a cover made from zeolite, alumina, etc. between them acts as a current collector, establishing an electrical connection between the two ends of the capacitor [10]. The current collectors act as electrical connections between the electrode and the outward terminals of the supercapacitor [11]. The two-layer structure of carbon electrodes and separator can be very thin, but their effective cross-sectional area when wrapped together will be thousands of m^2. In this case, it is obvious that to increase the capacity of a supercapacitor, we must increase its contact cross-section [12]. This should be done without manipulating the physical size of the capacitor or using special types of electrolytes to increase positive ions to enhance conductivity.

In addition to the electrode, the electrolyte plays a very important role in the electrochemical properties of the supercapacitor. The electrolytes used in the supercapacitor industry can be divided into several different categories: aqueous electrolytes; organic electrolytes; ionic liquids; solid-state electrolytes; and redox electrolytes. All solid-state supercapacitors utilize a solid electrolyte the most promising energy storage devices [13]. Unlike commercial supercapacitors where the separator is used in a liquid electrolyte, in solid-state supercapacitors, polyelectrolyte acts both as an electrolyte and a separator [14]. The safety and flexibility of all-solid-state supercapacitors pave the way for wide-reaching electronic applications [15]. In addition, flexible supercapacitors have great physical, chemical, and mechanical properties that make them extremely attractive [16]. On the other hand, solid-state supercapacitors serve better performance over a wide temperature range (above 100 °C and below 0 °C).

MXenes are a new family of carbide, nitride, and carbonitride materials that have recently been used in fabricating electrode materials, especially for supercapacitors [17]. These compounds are the result of acid exfoliation of MAX phases, which have excellent mechanical and electrochemical properties [18]. So far, various thin-layer carbides such as $Ti_3C_2T_x$, Nb_2CT_x, V_2CT_x, etc. have been prepared that accept a variety of hydrophilic terminal groups, called T [19]. Although 2D MXenes have attracted much attention due to their fast and easy fabrication process, they suffer from some defects such as atomic vacancies and inactive edges due to acidic production conditions [20]. At the same time, the low specific capacity limits their practical applications in electronic devices [21].

Therefore, the present review focuses on the new synthesis procedure of MXene-based composites to overcome defects of electrodes in all-solid-state supercapacitors. However, methods for MXenes preparation are available in the literature; here is a brief description of how to fabricate single metal and bimetallic MXene nanosheets. We provide new references for the design and improvement of MXene-based composites in electrode materials for energy storage. Then, the application of MXenes and their composites have been analyzed in supercapacitors along with electrochemical performance. Finally, an overview of the latest developments in the creation of all-solid-state flexible SCs as well as our predictions for future lines of inquiry is provided.

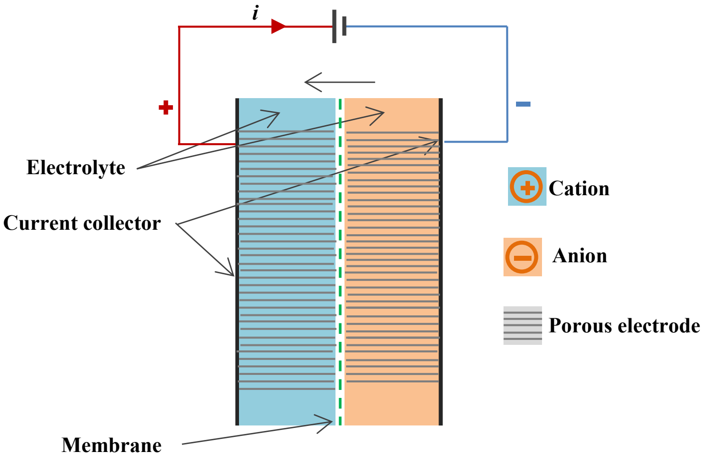

Fig. 1. Schematic representation of a supercapacitor structure.

2. Operating principles of supercapacitors

A supercapacitor, like all capacitors, is an energy storage appliance. Electric energy is stored in the form of charge accumulation in the electrical field between two capacitor plates, and as a result of this stored energy, a potential difference in the form of voltage is established between the two plates [22]. Electric energy is stored between its two plates during charging (current flow from a source connected to the supercapacitor). When the supercapacitor is charged, current will stop flowing from the power supply and the terminal voltage of the supercapacitor will be the same as the power supply voltage. As a result, a charged supercapacitor, even when disconnected from a voltage source, stores electrical energy within itself and supplies the required energy when needed [23].

When the supercapacitor discharges (current flows out of the supercapacitor), the energy stored inside it is converted into electrical energy to supply the connected load. Therefore, the supercapacitor itself does not consume energy, but stores and releases electrical energy when needed. The amount of energy stored in the supercapacitor is proportional to its capacitance [24]. It is obvious that when the supercapacitor is fully discharged, the stored energy will be zero. An ideal supercapacitor does not consume or waste energy, but uses an external charging circuit to store energy in an electrolyte field and then discharges this stored energy when connected to a load. The main disadvantage of supercapacitors compared to batteries is their low energy density [25]. Although the energy density of supercapacitors is still much higher than that of capacitors, it is far behind batteries [26]. Supercapacitors are produced in very small sizes that can have a much higher energy density than batteries. However, the voltage of a supercapacitor is very low, so several capacitors must be connected in series and parallel to make their voltage useful [27].

To increase the energy density in supercapacitors and solve this weakness, a lot of research has been focused on the modification of the materials [28]. Considering that supercapacitors generally include three components: electrode, electrolyte, and substrate, it can be concluded that in order to increase the energy density, each of these parts should be improved and, in short, one of the following should be researched: 1) type of electrode material, 2) structure and morphology of the electrode material, 3) bed, 4) electrolyte, and 5) arrangement of electrodes [29].

Active electrode materials in supercapacitors are classified into the following three categories based on the energy storage mechanism: electrode materials of electric-double layer capacitors (EDLC), pseudocapacitors electrode materials, and hybrid materials [6]. EDLCs consist of two carbon-based electrodes, an electrolyte, and a separator [30]. In EDLCs, charge distribution with a constant nature between electrodes and electrolyte takes place in an electrical double layer, and charges are separated by the Angstrom scale [7]. An EDLC electrostatically or non-Faradaically stores the charges without transfer between the electrodes and electrolyte [31]. Carbon materials such as activated carbon, carbon fibers, graphite, and graphene are widely used in composite materials due to their advantages such as high conductivity, high surface area, corrosion resistance, high-temperature stability, and relatively cheap price [6]. These materials have been used in commercial supercapacitors for years [32]. Currently, activated carbon is a substance that is used more than other carbon allotropes in commercial applications [33]. The mechanism of charge storage in carbon materials is through electrostatic adsorption/desorption known as electric double-layer capacitors. In this mechanism, electrostatic charge accumulation occurs at the electrode-electrolyte interface, which leads to the formation of an electrical double layer. Since the charge transfer process between electrode and electrolyte in this double layer is non-Faradaic, no chemical change occurs in the composition of electrode or electrolyte materials. For this reason, these electrodes are very reversible and thus have a long cycle life.

The pseudocapacitors store charge by the Faradaic mechanism through electrosorption, intercalation process, and redox reactions [34]. Conductive polymers and metal oxides or hydroxides are included in this category. The energy and capacity of these materials are much higher than EDLC electrode materials because of the stored charge in these materials carried out by electrochemical oxidation-reduction reactions at the electrode-electrolyte interface during a short path [35]. Conductive polymers or pseudo-capacitive materials are a group of organic compounds that have the electrical, magnetic, and optical properties of metals, with the general properties of polymers [36]. Conductive polymers display higher specific energy compared to carbon-based EDLCs but expand and contract through charge/discharge, respectively [37].

Hybrid supercapacitors overcome the relative disadvantages of the previous two types because of Faradic and non-Faradic processes usage and thus have a much higher energy density and power than EDLCs and pseudocapacitors [38]. In hybrid capacitors, the advantages of both EDLC and pseudocapacitors systems are used to improve performance and efficiency. Hybrid supercapacitors achieve extraordinary power density and specific capacitance higher than EDLCs without sacrificing the affordability and stability of capacitors [39]. Hybrid supercapacitors are distinguished based on the chosen material for electrodes: 1) asymmetric, 2) battery-type, and 3) composite hybrids [30]. Most hybrid electrode materials are based on composites of carbon materials with metal oxides and hydroxides as well as conductive polymers. The carbon part as the negative electrode facilitates the creation of the electrical double layer and also increases the contact surface of the composite pseudocapacitor material with the electrolyte due to the large surface area. The pseudocapacitor materials in the composite also lead to an increase in the electrode capacity through Faraday reactions [40].

3. MXene-based electrode materials

In addition to the type of electrode material, its structure and morphology are also effective on supercapacitor properties, because supercapacitor's mechanisms are based on superficial phenomena and the microstructures of the electrodes determine their electrochemical efficiency [13]. Active sites for electrochemical reactions, transport/permeation pathways, electrolyte access, and kinetics are the most important factors that affect the electrochemical performance of supercapacitors [15]. These factors strongly depend on the surface area, morphology, and wall thickness of electrode materials [41].

Various carbon materials are used to make an advanced supercapacitor such as activated carbon [42], graphene [43], bio-based carbon aerogel [44], reduced graphene oxide (RGO) [45], g-C_3N_4 [46], etc. to facilitate ion diffusion and charge transfer in the supercapacitors. Carbides are also a group of materials derived from carbon whose pores can be adjusted and are characterized by micropores [47]. It is notable that the capacitance is affected by the carbon material's characteristics. Furthermore, edge orientation exhibits a greater affinity for functional

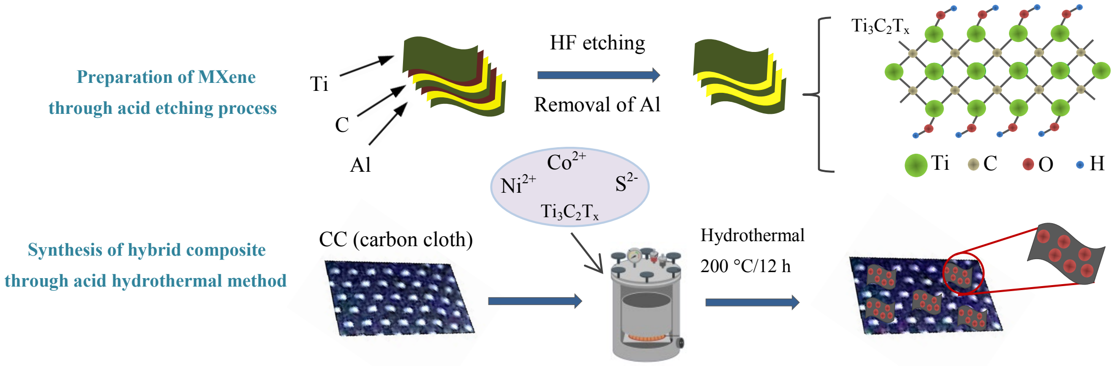

Fig. 2. Representation of the detailed synthesis route of in situ growth metal sulfide on MXene electrodes via a facile hydrothermal method.

groups than basal grouping, which enhances electrode wetting [47]. Generally, large micropores and cone-shaped pores gather more ions with low energy density losses after charge redistributions [48].

4. Recent developments in the MXene-based electrode materials

To date, several MXenes have been utilized as electrodes. The hydro/solvothermal method is known as one of the most useful techniques for the synthesis of nanocrystals depending on the solubility of the raw materials in the solvent [49]. Hence, the coupling of $Ti_3C_2T_x$ and $MnCo_2S_4$/$NiCo_2S_4$ on activated carbon cloth has been followed via a simple hydrothermal procedure by Pathak and Rout [50]. Hybrid composites are attractive candidates for flexible supercapacitors because of their reasonable energy density, low capacitive losses, and long lifetime [51]. Fig. 2 represents the detailed synthesis route of in situ growth metal sulfide on MXene electrodes via a facile hydrothermal method. In another study, the microstructures of hydrothermally as-prepared $Ti_3C_2T_x$ crystalline have revolutionized the charge storage efficiency in hybrid supercapacitors with S,N-rGO (reduced graphene oxide) as the electrode material [52]. Fabrication of MXene-based composites in 3D porous nanostructures is a useful technique to enhance supercapacitor properties by increasing porosity and areal surface and improving ion transport rates [53]. Yang et al. have etched the MAX phase with Lewis acid Fe^{3+} to fabricate $Ti_3C_2T_x$ with in-situ loaded Fe_2O_3 nanostructures. The N,S co-doped $Ti_3C_2T_x$/rGO 3D aerogels have been successfully prepared through the redox self-assembly method under hydrothermal conditions by thiourea. The uniform distribution of $Ti_3C_2T_x$ in the aerogel prevents the accumulation of rGO and creates a strong connection between them. The XPS results of the samples reveal an increase in the hydrophilicity and pseudocapacitance of the composite, which is attributed to the formation of N-C/S-C bonds and changes in the electronic structure of adjacent carbon atoms. In comparison with Liu et al. research on S,N-rGO@MXene [54], the optimized supercapacitor designed with Fe^{3+}-$Ti_3C_2T_x$/N,S-rGO electrode and PVA-KOH gel electrolyte shows excellent compatibility and stability in extreme environments (-20 to 60 °C).

Like other two-dimensional materials, MXenes also suffer from self-assembly and aggregation problems due to van der Waals forces, which reduce active sites and limit ion transport [55]. To overcome these defects, the use of noble metal atoms between MXene nanosheets has been proposed [56]. Noble metal nanoparticles with large specific surface areas have the highest metal conductivity and can improve the

Fig. 3. Schematic illustration of the preparation process of V_2O_5/$Ti_3C_2T_x$ films.

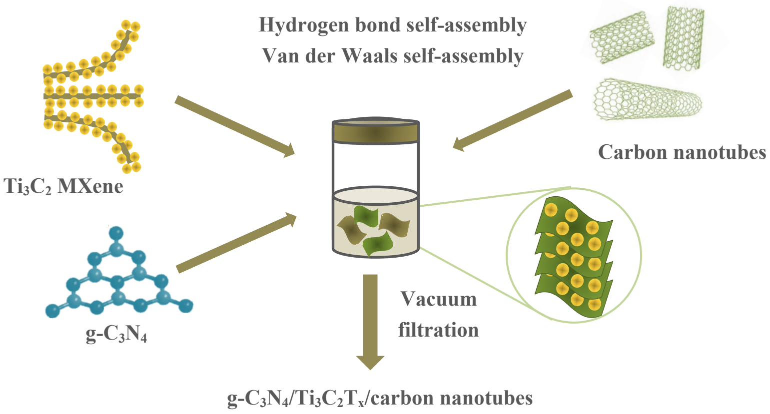

Fig. 4. Schematic illustration of $g\text{-}C_3N_4/Ti_3C_2T_x$/carbon nanotubes through self-assembly.

electrochemical properties of MXenes [57]. On the other hand, it is expected that nanoparticles of noble metals further increase the supercapacitor's performance [58]. Moreover, the wide potential window and reversible charge/discharge capability of noble metal oxides as an auxiliary pseudocapacitive compound boost the energy density of the MXenes-based supercapacitors. For example, carbon nanofibers/$Ti_3C_2T_x$ coated by amorphous RuO_x has been prepared by hydrothermal approach to create a binder-free flexible electrode [59]. As we previously reported, the hydrothermal method has the advantage of being able to control the morphology of nanocrystals immersed in the solvent [60]. Bare MXene/carbon nanofibers have smooth surfaces and interwoven composite fiber strands that maintain the shape of the electrode during the hydrothermal process. The transmission electron microscopy of the synthesized samples indicated partially nanocrystalline and amorphous regions. Furthermore, the added Ti_3C_2 improves energy storage and the hydrophilicity of the electrode is maintained, which is a requirement for the coating during the hydrothermal process. Electrochemical tests on the composite electrode have shown superior energy density (8.5 $Whkg^{-1}$) with a two-fold increase in specific capacity compared to bare MXene/carbon. The super-long cycle of life (99% after 10,000 cycles) is attributed to the structural distortions and mechanical strain of the amorphous RuO_x [61].

The ultrasound method in materials synthesis is an easy and popular technique where ultrasonic waves (>20 kHz) are used for agitation resulting in homogenization. For example, V_2O_5 nanofibers and LiF/HCl etched $Ti_3C_2T_x$ nanosheets have been prepared by ultrasonication route [62]. The $V_2O_5/Ti_3C_2T_x$ composite films as self-supporting materials have been synthesized via vacuum-assisted filtration in a centrifuge tube at last (Fig. 3). The previous studies indicate the weakness of MXenes due to strong accumulation and self-accumulation, which causes the electrolyte not to penetrate into the layer [63]. Vanadium oxides can effectively reduce the degradation of the electrochemical performance of MXene nanosheets [64]. Nanoparticles of vanadium oxides with high specific surface area have shown a better development perspective in the field of energy production and storage [65]. In addition, it has various advantages such as layer structure for mobile guest species [66], low toxicity and cost-effectiveness and a high voltage window, which makes it superior to other metal oxides [67]. Furthermore, MXene composites have more metal redox-active surface sites and short ion diffusion paths [68].

Self-assembly is a common synthesis method of comprising disordered and separate components into a highly ordered structure or pattern [69]. This method gives special properties to materials by creating specific interactions between components, which are the result of spontaneous local interactions. This process has been successfully used

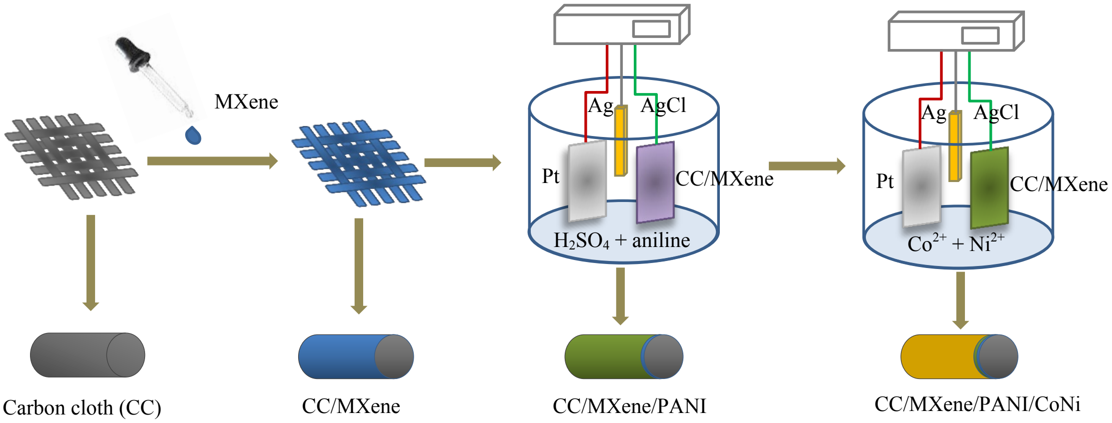

Fig. 5. Schematic diagram of CC/MXene/PANI/CoNi electrode preparation process.

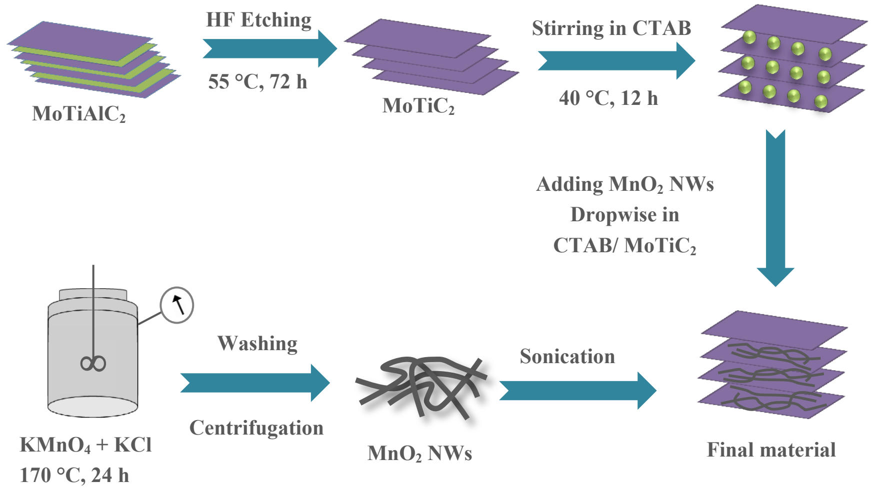

Fig. 6. Schematic synthesis routs of hybrid Mo_2TiC_2/MXene MnO_2 heterostructures.

in the preparation of $g-C_3N_4/Ti_3C_2$ for several applications [70]. The incorporation of MXenes into $g-C_3N_4$ creates an interlayer conductive network and facilitates charge transfer [70]. According to Li and coworker's theory, the chemical bond between nitrogen and hydrogen atoms acts as a bridge for electronegative adsorption of MXene via interfacial self-assembly [71]. For instance, in the recent research of Zhang et al. [72], it has been shown that self-assembled $g-C_3N_4/Ti_3C_2T_x$/carbon nanotubes as ternary hybrid film has high specific capacity (477 F/g), which can be useful in energy storage devices. The schematic illustration of $g-C_3N_4/Ti_3C_2T_x$/carbon nanotubes through self-assembly is shown in Fig. 4.

Electrodeposition is a useful method of electrolytic deposition that is known as a common surface modification applied to any conductive material [73]. In 2019, Jian et al. [74] fabricated carambola-like polypyrrole/$Ti_3C_2T_x$ film through co-electrodeposition for electrode material. They have used MXene nanolayers as a core polymer for the polymerization of monomer cations. The three-dimensional structure of the composite facilitates electron transfer and ion diffusion which causes excellent electrochemical performance. Polyaniline (PANI) is another applied conductive polymer for electrode materials due to its electrochemical properties, ease of preparation, and low cost [75]. It has been proven that the combination of MXene and $g-C_3N_4$ can compensate for the poor stability of polyaniline [76]. In this regard, MXene/PANI has been added to CoNi layered hydroxide to overcome the large internal resistance of the hydroxide [77]. The preparation process of the carbon cloth/MXene/PANI/CoNi composite is represented in Fig. 5. In the other study by Wu et al. [78], the interconnected porous framework of polymer/carbon cloth has been ensured by MXene. The pulsed electrodeposition method has been used to fabricate 3D hierarchical porous polyaniline (PANI)/MXene/carbon for electrode material. In a recent study conducted by Bi et al. [79], PANI-carbon cloth fabricated by in-situ polymerization has been also utilized as the positive electrode, as well as cellulose nanofiber/$Ti_3C_2T_x$ composite prepared via vacuum filtration as the negative electrode.

Monometallic MXenes face various challenges such as low specific capacity, structure degradation, short cycle life, poor ionic transfer, and low rate, which limit their application in flexible supercapacitors [80]. Double transition metal MXenes behave differently from single metal MXenes with high rate, high specific capacitance, and long cycling

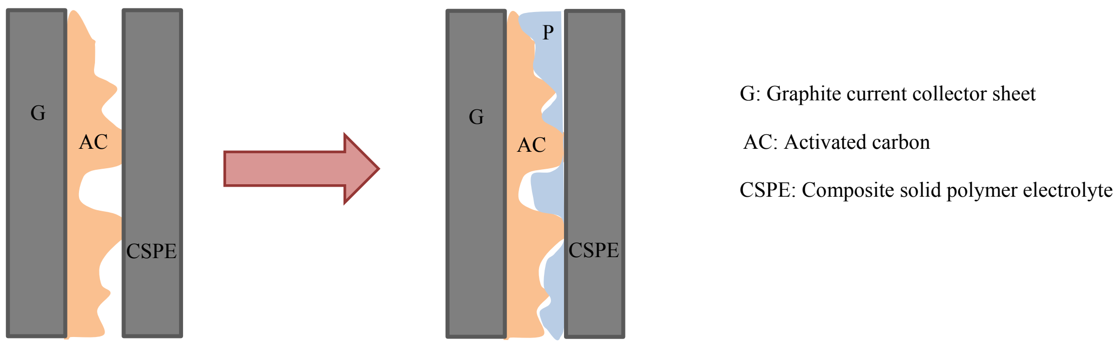

Fig. 7. Schematic diagram of the contact between the electrode and solid polymer electrolyte (P represents polymer)

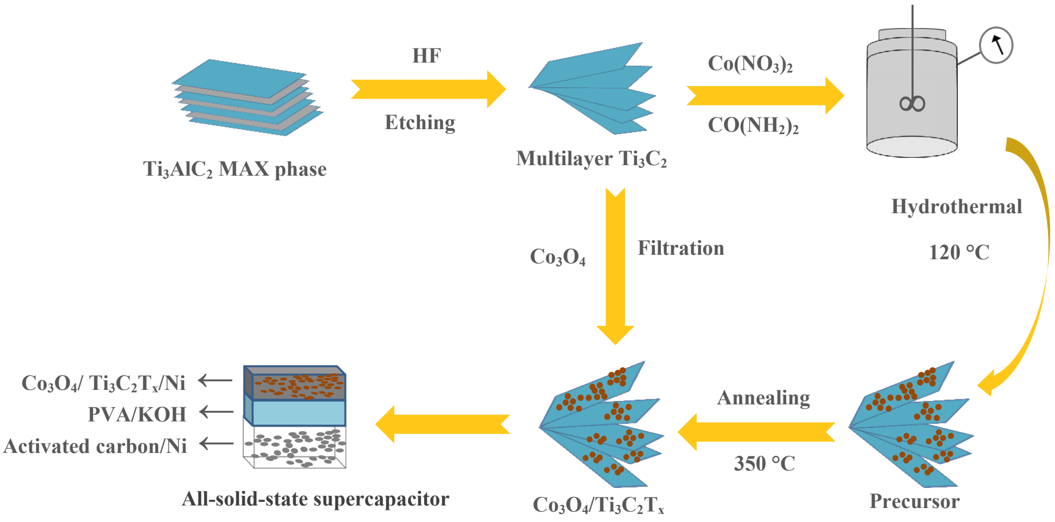

Fig. 8. Schematic illustration of the synthesis of $Ti_3C_2T_x/Co_3O_4$ nanocomposite through vacuum filtration and hydrothermal-annealing methods.

lifetime simultaneously [81]. In a case study, multivalent vanadium atoms were utilized to construct novel $(Ti_{0.5}V_{0.5})_3C_2T_x$ films by Xiao et al. [82]. To fabricate a TiV-based MXene nanosheet, simple etching of $(Ti_{0.5}V_{0.5})_3AlC_2$ powder has been carried out in LiF/HCl for 1 hour at 60 °C. The exfoliated product was washed several times with distilled water and dispersed in tetramethylammonium hydroxide ($N(CH_3)^{4+}$ OH^-) until finally the suspension was obtained with suitable concentration by centrifugation. The as-prepared TiV-based MXene exhibited high performance (387F/g @ 1.0 A/g) and excellent energy/power density (5.60 Wh/kg and 5210 W/kg) in a flexible supercapacitor. In order to overcome the obstacles of $Ti_3C_2T_x$, Wang et al. [83] have synthesized TiNb-based MXene with larger interlayer spacing and more –O terminated surface via etching TiNb-AlC_2 solid solution MAX precursor through a high-temperature self-propagating method. In particular, $Ti_{3-y}Nb_yC_2T_x$ (y=0.1) has been found a promising double metal MXene with excellent specific capacitance of 1015 F/cm^3 at a scan rate of 2.0 mV/s. Moreover, it has been reported superior cycling stability for $Ti_{2-y}Nb_yT_x$ ($0 \leq y \leq 2$) more than its single metal counterparts [84]. Another bimetallic MXene is Mo_2TiC_2, which may open a path for extended research in the field of energy storage [85]. Among the inexpensive metal oxides, MnO_2 has high capacitance but has low theoretical stability and poor electrical conductivity, which is improved with MXenes [86]. In order to prevent the restacking of carbide nanolayers, hybridization with MnO_2 nanowires has been suggested via a facile pillaring technique. Cetyltrimethylammonium bromide (CTAB) has been mixed with etched Mo_2TiAlC_2 powder to construct pre-pillared Mo_2TiC_2 and then combined with manganese oxide nanowires as shown in Fig. 6. This unique heterostructure not only increase the specific capacity but also facilitate electron transport and ion diffusion. Thermogravimetric analysis results of the as-prepared samples exhibited good thermal stability up to 1000 °C with less than 13% weight loss. High entropy MXenes, as a newly emerging family of 2D composites, are expected to be a high-potential electrode material for energy storage [87]. This group of MXenes provides unexpected physiochemical features, and thus they have excellent potential for supercapacitors.

Despite the high capacity of all-solid-state supercapacitors, problems related to the electrode–electrolyte interface adversely affect their performance during long-term cycles [88]. The rapid transport of ions in electrochemical interfaces through the electrode/electrolyte contact is very important in supercapacitors [45]. Although liquid-free supercapacitors have high ionic conductivity, tuning the interface properties is very challenging [89]. In order to approach the ideal behavior of supercapacitors, the unevenness of solid-solid interfaces must be removed. In this regard, Sharma et al. [90] have added a small amount of ion-conducting PEO-$LiClO_4$ (polymer-salt) into the activated carbon-based electrodes. The performance parameters of the supercapacitor that works with the electrode combined with the polymer and a small amount of salt are improved because the mobile salt ions (Li^+ and ClO^{4-}) in the electrode matrix lead to the formation of a smooth interface and maintain continuity at the interface as shown in Fig. 7. In another work, Li et al. have prepared poly(3,4-ethylene dioxythiophene) (PEDOT)/$Ti_3C_2T_x$ heterostructures through an electrodeposition method by $LiClO_4$/acetonitrile/ethylene dioxythiophene solution. This non-aqueous electrolytic bath prevents the oxidation of $Ti_3C_2T_x$ at anode potential while depositing [91]. Adding a coupling polymer causes: 1) facilitates the long-range ionic diffusion, 2) improves the effectual contact area, and 3) decreases the surface polarization. It should be noted that the ratio of salt to polymer at the maximum value of 15:85 increases the conductivity by about four orders [92].

Carbon aerogels are a class of 3D porous carbon materials without structural collapse that provide very attractive supercapacitor applications [93]. Chen and coworkers [94] have suggested vacuum filtration followed by annealing for the preparation of $Ti_3C_2T_x$ MXene flake/nitrogen-doped carbon. The self-supporting composite uses polydopamine nanospheres as spacers while adding porosity to the composite networks. In addition, nitrogen doping can improve conductivity and ionic transfer and provide additional pseudocapacitance properties. Sun et al. [95] have prepared N, O co-doped carbon aerogel via a combination of freeze-drying and carbonization methods from sodium alginate/melamine composite. The symmetric supercapacitor based on carbon aerogel with the highest specific surface area has exhibited excellent performance (440 F.g^{-1} @ 0.5 A.g^{-1}) and great cycling stability. In another research, a composite structural supercapacitor has been manufactured by KOH-activated carbon cloth/solid electrolyte [96]. This novel supercapacitor has provided a power density of 445 mW/kg and a specific capacitance of 88 mF/g at an energy density of 10 mWh/kg. The annealing method along with the hydrothermal technique has also been used for the synthesis of $Ti_3C_2T_x/Co_3O_4$ nanocomposite [97]. The different steps to prepare $Ti_3C_2T_x/Co_3O_4$ nanocomposite are shown in Fig. 8. The asymmetric supercapacitor assembled by this composite has shown high energy density and excellent long-term durability.

Recent MXenes are summarized for capacitor electrodes in Table 1 and compared with similar electrode materials.

Table 1. Comparison of the electrochemical performances of recent all-solid-state supercapacitors.

Electrode	Synthesis method	Energy density	Power density	Electrolyte	Specific capacitance	Capacity retention (after no. of charge/discharge cycles)	Ref.
$MnCo_2S_4/Ti_3C_2T_x$//carbon cloth	Hydrothermal	20 µWh/cm^2	1.24 mW/cm^2		274 mF/cm @ 0.6 mA/cm^2	80% (5000)	[50]
$NiCo_2S_4/Ti_3C_2T_x$//carbon cloth	Hydrothermal	20 µWh/cm^2	1.24 mW/cm^2		206 mF/cm_2 @ 0.4 mA/cm^2	80% (5000)	[50]
$Ti_3C_2T_x$/carbon nanofibers/RuO_x	Hydrothermal	8.5 Wh/Kg	85.8 W/K	PVA-H_2SO_4 gel	279.4 F/g @ 2 mV/s	99% (10000)	[59]
$Ti_3C_2T_x/V_2O_5$	Ultrasonication	20.83 Wh/Kg	374.9 W/K		319 F/g @ 0.5 A/g	83.9 % (8000)	[62]
$Ti_3C_2T_x$/g-C_3N_4/carbon nanotubes	Self-assembly	25.28 Wh/Kg	449.99 W/K		477.4 F/g @ 1 A/g		[72]
Polyaniline/g-C_3N_4/MXene	Electrodeposition	18.8 Wh/Kg	1563 W/K		570 F/g @ 5 mVs^{-1}		[76]
Activated carbon//carbon cloth/MXene/polyaniline/CoNi	Electrodeposition	39 Wh/Kg 27.3 Wh/Kg	399. W/K 2400 W/K	Alkaline	110.94 F/g @ 0.5 A/g	91% (10000)	[77]
Polyaniline/MXene/carbon cloth	Electrodeposition	42.5 µWh/cm^2		Gel	478 mF /cm^2 @ 0.5 mA/cm^2	75.4% (4000)	[78]
$Ti_3C_2T_x$-cellulose nanofiber//PANI-carbon cloth	Self-assembly	30.6 Wh/Kg	1211 W/K	PVA-H_2SO_4 gel	96 F/g @ 5 mA/g	86% (5000)	[79]
$(Ti_{0.5}V_{0.5})_3C_2T_x$	Etching	5.6 Wh/Kg	5210.3 W/K		387 F/g @ 1 A/g	100% (100000)	[82]
$Ti_{0.29}Nb_{0.1}C_2T_x$	Etching			KOH gel	1015 F/cm^3 @ 2.0 mV/s	(84000)	[83]
MnO_2/Mo_2TiC_2	Pillaring				918.6 F/g @ 2 mV/s	83.3% (10000)	[85]
$Ti_3C_2T_x$/N doped carbon	Annealing	23.2 $\mu Whcm^{-2}$	317.7 μWcm^{-2}		942.43 mF/cm^2 @ 1 mA/cm^2	93.7% (10000)	[94]
$Co_3O_4/Ti_3C_2T_x$//active carbon	Annealing	95.9 Wh/kg	630.4 W/kg		240.1 F/g @ 0.1 A/g		[97]
MXene/Ag/cellulose	Self-assembly			PVA-H_2SO_4 gel	505 F/g @ 10 mVs^{-1}	90.9% (500)	[98]
MXene/Graphdiyne nanotube	Hydrothermal-annealing	19.7 Wh/kg	750 W/kg	PVA-H_2SO_4 gel	337.4 F/g 100 mV/s	88.2% (10000)	[99]
MXene/carbon nanotube	Freeze-drying	7.3 Wh/kg	50 W/kg			99% (5000)	[100]
α-Fe_2O_3/MXene aerogel	Freeze-drying	3.61 µWh/cm^2	119.04 µW/cm^2	H_2SO_4	182 F/g @ 1 A/g	81.74% (10000)	[101]
MXene/carbon nanotube aerogel	Freeze-drying				410 mF/cm^2 @ 0.8 mA/cm^2	85% (1000)	[102]
$Ti_3C_2T_x$/acid-treated carbon fiber (ACF)//$NiCo_2O_4$-rGO-ACF	Drop-casting	44.36 Wh/Kg	985 W/Kg		246.9 F/g @ 4.0 mA/cm^2	96.7% (5000)	[103]
Nitrogen-doped $Ti_3C_2T_x$	Decomposition	9.57 Wh/kg	250 W/kg	PVA-H_2SO_4 gel	449 F/g @ 2 mV/s	80.4% (4000)	[104]
$Ti_3C_2T_x$ foam	Chemical foaming	14.1 Wh/kg	250 W/kg	H_2SO_4	426 F/g @ 1 A/g	92% (10000)	[105]

Table 1. Continued.

Electrode	Synthesis method	Energy density	Power density	Electrolyte	Specific capacitance	Capacity retention (after no. of charge/discharge cycles)	Ref.
$Ti_3C_2T_x$/vertical graphene arrays	Plasma treatment-thermal evaporation	13.57 µWh/cm^2	31.07 µW/cm^2		199 mF/cm^2 @ 0.08 mA/cm^2	90% (6000)	[106]
Ti_3C_2/graphite/carbon black/chitin	Casting	3.55 Wh/kg	81.1 W/kg	Biopolymer-based	100 F/g @ 2.5 A/g	85% (10000)	[107]
d-$Ti_3C_2T_x$	Etching- adjusting in ink	207.81 µWh/cm^2	3.74 mW/cm^2		2.337 F cm^{-2} @ 2 mVs^{-1}	93.1% (10000)	[108]
$Ti_3C_2T_x$/polyaniline	Physical mixing-suction filtration	31.18 Wh/kg	1079.3 W/kg		272.5 F/g @ 1 A/g	71.4% (4000)	[109]
$Co_3V_2O_8/Ti_3C_2T_x$//activated carbon	In situ encapsulation	70.2 µWh/cm^2	3.3 mW/cm^2	PVA/KOH	3000 mF/cm^2 @ 8 mA/cm^2	94.5% (10000)	[110]

5. Applications of all-solid-state supercapacitors

Supercapacitors can be used as energy storage devices, like batteries. But unlike batteries, they can have much higher power densities in a short period of time. Supercapacitors are used in many gasoline-hybrid vehicles and fuel-cell-powered electric vehicles because of their ability to rapidly discharge high voltages and then recharge [111]. However, using supercapacitors along with fuel cells and batteries makes it possible to control peak power demand and rapid load changes more effectively. A hybrid combination of the supercapacitor and battery is considered a better option for electrical energy storage [5].

Supercapacitors are used in the following cases: 1-computer parts 2-medical devices 3-military devices 4-lasers 5-microwave 6-power supplies 7-security and information systems for backup storage 8-high power LED circuits 9-wind turbines 10-power transmission and power supply networks for network stability 11-UPS 12-supercomputers and sensitive computers 13-electric doors for power outages 14-brake electricity generating systems in new electric cars 15-voltage stabilizer 16-circuits that require capacitors with fast charging and discharging time [112–117].

6. Challenges and future outlook

Solid-state supercapacitors are becoming increasingly prevalent in a wide range of energy storage systems, including wearable electronics, self-powered gadgets, and intelligent medical equipment. The fundamental workings of various solid-state supercapacitors types were systematically discussed in this review, along with current developments in the electrode and electrolyte technologies utilized to create flexible solid-state supercapacitors. A review of the electrodes was conducted, covering MXenes and their performance. These novel and emerging materials have demonstrated exceptional electrochemical performance, making them ideal candidates for the development of high-performance electrodes.

While several remarkable and promising approaches to high-performance flexible solid-state supercapacitors have been developed, the following issues still need to be resolved:

- The majority of flexible solid-state supercapacitors still had lower energy densities than secondary batteries, which limited the wearable and portable devices' ability to operate for extended periods of time. The main goal is to investigate novel pseudocapacitive materials with large working windows and high capacitance.
- Emerging 2D layered materials with promising high performance as electrodes include chalcogenides, MXenes, and transition metal nitrides. However, the durability of these materials is a challenging issue Nevertheless, the durability of these 2D materials was limited by the collapse of one layer due to the collapse of their layers, which need to be fixed in the near future.
- MXene's oxidation is connected to surface flaws that result from the chemical etching procedure. Two strategies may improve the stability of MXenes: (i) elimination of dissolved oxygen using dry nitrogen, and (ii) conditioning of Mxenes at lower temperatures. Therefore, the electrochemical charge storage mechanism of MXene-based electrode materials has been significantly impacted by the aggregation of the synthesized MXene and the impacts of MXenes' structural properties, such as basal spacing, surface chemistry, etc.
- Ion transport kinetics in irregular porous materials remain poorly understood. The ideal model, which is unable to fully capture the reality of the situation, is the foundation for the majority of described methods or simulations. Clear insights into the design of new materials can be obtained from the more intricate mechanisms of charge transport and storage with the aid of in situ characterization techniques.
- The majority of researchers in previous decades have used mass-specific capacitance to characterize the device performance of solid-state supercapacitors. Both the mass of the entire device and the mass of the active materials on the electrodes can be used to compute the specific capacitance. However, since wearable and portable devices are often lightweight, using the volumetric-specific capacitance makes more sense. To adequately explain the performance of the device, the standard parameters should be configured.
- The limited processability and high cost of electrode materials limit the development of large-scale devices for supercapacitor applications. Researching low-cost, readily processed materials can help advance large-scale processing technologies like roll-to-roll (R2R) blade casting.
- The industrial application of MXene into energy storage device electrodes will experience the same challenges as graphene if the colloidal MXenes cannot be avoided to accomplish the achieved and predicted property enhancements. For industrial use, the high

cost of producing MXene must be reduced, which presents a hurdle for further study.

7. Conclusions

With the emergence of new energy vehicles and modern electrical appliances, expanding the use of supercapacitors, improving energy density, reducing costs and environmental effects are some of the important goals of mankind. Supercapacitors are alternative energy storage technology due to their high capacity and low price. Supercapacitors also have other advantages over batteries, including higher reversibility and cycle life, better speed, lower maintenance costs, and safer electrode materials. Supercapacitors have a vast potential market and endless prospects for development. Currently, research centers are focusing on saving the cost of supercapacitors, reducing their risks to the environment, and increasing efficiency. The main disadvantage of supercapacitors compared to batteries is their low energy density. Although the energy density of supercapacitors is still much higher than that of capacitors, it is far behind batteries. Scientific and research centers are working on reducing the cost of supercapacitors and making them economical, reducing the problems they cause for the environment, and increasing their efficiency.

This review focuses on the latest developments in MXene-based electrodes for energy storage applications. We cover basic knowledge of charge storage mechanisms in electrodes, the synthesis process of composites for electrodes, structure and chemical modifications, and electrochemical analysis.

The energy density of MXene-based supercapacitors can be enhanced by: i) using active carbon as a large specific surface area material, ii) hybridization with metal oxide, iii) elemental doping, iv) fabrication of double transition metal MXene, v) incorporating with 3D carbon nanotube, vi) controlling the surface chemistry to enhance interactions.

Regardless of the preparation method of monometallic MXenes, the synthesis method of MXene-based composites has a high variety from hydrothermal to electrodeposition, ultrasound-assisted technique, and annealing.

Overall, the future of MXene-based supercapacitors looks bright in powering advanced electronic devices with the development of cost-effective synthesis methods and improved capacity enhancement techniques.

CRediT authorship contribution statement

Asieh Akhoondi: Writing – original draft, Resources, Supervision.
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Brian Yuliarto: Writing – review & editing.
Hicham Meskher: Writing – original draft, Writing – review & editing.

Data availability

As this is a review article, no new data were generated. All information is publicly available or cited appropriately within the article.

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