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MXenes-Based Supercapacitors: A Review on Energy Storage Devices

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Energy storage and energy-storage devices have been a buzzword for long time as it is one of the essential needs in human life. These devices include mechanical systems, thermal systems, and batteries. These systems embedded with software can monitor the charging and discharging phenomena of energy. In this context, the role of rechargeable batteries needs to be reviewed. Even though novel types of rechargeable batteries are being continuously developed for storage of electricity, more attention and research towards supercapacitors is on the way. Huge number of researchers around the globe is involved in developing supercapacitors with improved performance making them more and more useful. The main aim is to improve their efficiency, energy density, operating voltage, miniaturization, optimization, economy, and environmental acceptance. For the last few years, lightweight and carbon-based novel wearable supercapacitors are developed. High durability, eco-friendliness, being non-volatile and electrostatic mechanism of supercapacitors make them advantageous than conventional batteries. In this regard, advances in microelectronics demand microsupercapacitors (MSCs). The selection of electrode in microsupercapacitor plays significant role in the fabrication. In this selection, MXenes as a family of 2*D* material play a vital role. Very high conductivity and high capacity of charge storage makes MXenes as one of the potential materials for electrodes in microsupercapacitors. This prompts us to review the role of MXenes in microsupercapacitors. This article reviews the recent advances of MXenes-based MSCs with emphasis on their fabrication techniques.

Накопичувачі енергії та пристрої накопичення енергії вже давно стали модним словом, оскільки це одна з найважливіших потреб у житті людини. До таких пристроїв належать механічні системи, теплові системи й акумулятори. Ці системи, вбудовані в програмне забезпечення, можуть контролювати явища заряджання та розряджання енергії. У цьому контексті необхідно переглянути роль акумуляторних батарей. Незважаючи на те, що для зберігання електроенергії постійно розробляються нові типи акумуляторних батарей, все більше уваги та досліджень у галузі суперконденсаторів вже на підході. Величезна кількість дослідників по всьому світу бере участь у розробці суперконденсаторів з поліпшеною продуктивністю, що робить їх все більш корисними. Основною метою є підвищення їхньої ефективности, густини енергії, робочої напруги, мініятюризації, оптимізації, економічности й екологічного сприйняття. За останні кілька років були розроблені легкі та карбонові нові придатні для носіння суперконденсатори. Висока довговічність, екологічність, енергонезалежність і електростатичний механізм суперконденсаторів роблять їх вигіднішими перед звичайними акумуляторами. У зв'язку з цим досягнення мікроелектроніки вимагають мікросуперконденсаторів (МСК). Вибір електроди в мікросуперконденсаторі відіграє значну роль у виготовленні. У цій добірці максени як сімейство 2*D*-матеріялів відіграє життєво важливу роль. Дуже висока провідність і висока ємність накопичувача заряду робить максени одним з потенційних матеріялів для електрод у мікросуперконденсаторах. Це спонукає нас розглянути роль максенів у мікросуперконденсаторах. У цій статті розглядаються останні досягнення МСК на основі максенів з акцентом на технологіях виготовлення їх.

Key words: 2*D* MXenes, microsupercapacitors, energy storage, E-textiles, health monitoring.

Ключові слова: 2*D*-максени, мікросуперконденсатори, накопичувачі енергії, електронний текстиль, моніторинґ здоров'я.

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1. INTRODUCTION

High capacitance value is being one of the top priorities since few decades in electrochemical systems. This could not be achieved earlier, but researchers have proved that supercapacitors could show a new direction in developing electrical energy storage systems [1]. Novel materials, technologies, huge surfaces and minute interelectrode distances are being developed in recent times. Many materials exhibit high pseudo-capacitance achieving large capacitance values as compared to normal capacitors and are called supercapacitors or ultra-capacitors) [2]. Figure 1 shows the image of a supercapacitor.

Supercapacitors play a significant role in new generation electronic devices and systems. Carbon and its allotropes play a vital role in supercapacitors in view of their thermal stability, chemical stability, mechanical strength, good conductivity, high electron mobility, wide range of temperatures, large surface area and morphology.

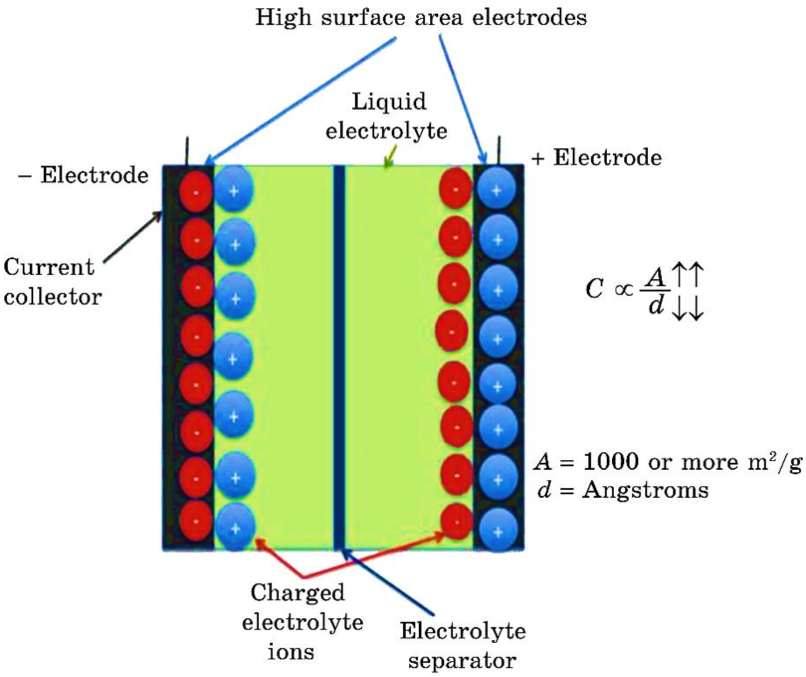

Fig. 1. Supercapacitor (courtesy [3]).

Figure 2 shows the classification of supercapacitors.

Depending on the mechanism involved in energy storage, supercapacitors are of two types: pseudo-capacitors where charge storage is fast due to redox reactions, double-layered capacitors where charge storage depends on electrostatic principles [3, 4]. If an electrode material exhibit either one or both mechanisms mentioned above, hybrid capacitors can be formed with them. Figure 3 shows different types of supercapacitors.

High-power rapid charging and discharging make supercapacitors significant in energy harvesting from renewable energy sources, power, industrial control, transport, consumer electronics, defence, medical, communications, electric and hybrid vehicles [6–9].

2. MATERIALS FOR SUPERCAPACITORS

Supercapacitors are widely used for energy storage mainly due to their environment friendly nature, huge number of charge, discharge cycles and durability with less maintenance [10]. However, as compared to battery they have low energy density. This drawback force researchers towards new materials and technologies. In this context, 2D nanomaterials such as graphene, fullerene, and carbon

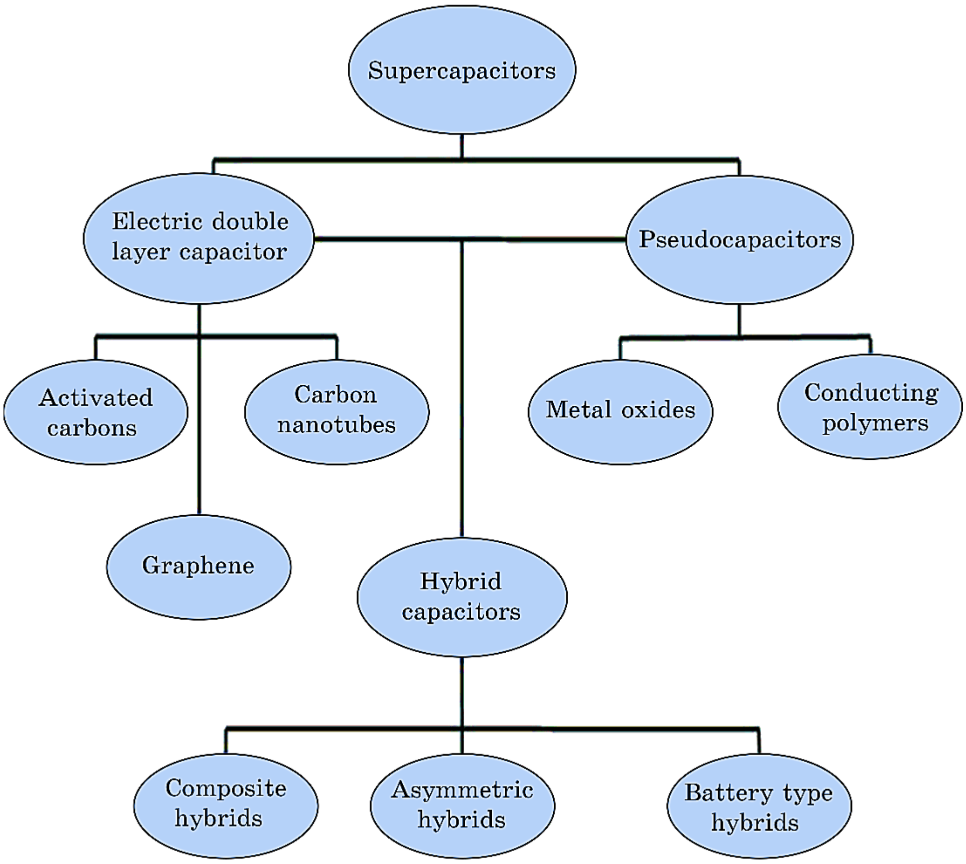

Fig. 2. Classification of supercapacitors (courtesy [3]).

nanotubes are used as appropriate electrolytes [11]. In this journey, usage of copper minerals, chalcosine [12] and coveline [13] was taken up. In addition, perovskite oxides based on lanthanum, strontium, and cerium, *etc.* are being researched [14]. In order to improve further the efficiency of supercapacitors, MXenes were used.

Increased demand for flexible and smart wearable energy micro-devices force researchers towards design and development of micro-energy storage devices. Since microbatteries have limited life and power density, best alternative to them are microsupercapacitors (MSCs) despite lower energy density. Still stability and fast charge/discharge cycles MSCs are preferred [15]. MSCs may be of regular sandwich type or plane interdigital pattern type, in which the second one offers better performance [16, 17]. Various 2*D* materials such as graphene, MXenes with excellent electronic, optical, mechanical, physical and chemical properties make them significant

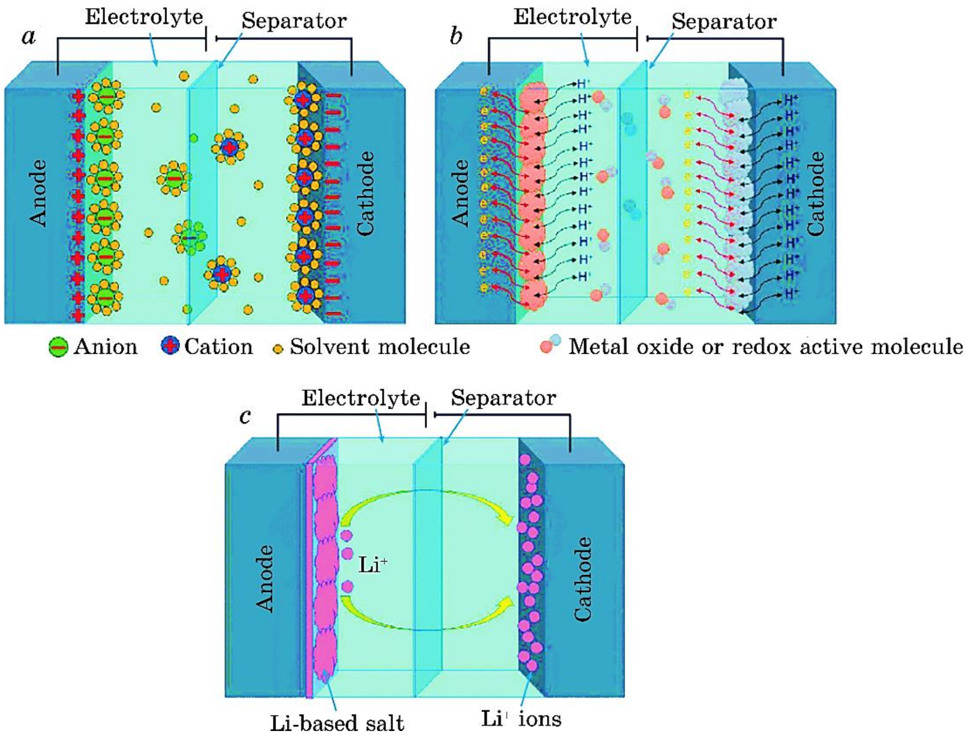

Fig. 3. (*a*) Electrical double layer capacitor (EDLC); (*b*) pseudo-capacitor (PC); (*c*) hybrid supercapacitor (HSC) (courtesy [5]).

in energy storage applications [18]. Even with large conductivity and surface area, carbon based materials lack high energy density [19–24]. Likewise, pseudo-capacitive materials, which have low conductivity [25–28], are used in MSCs. Since the discovery of MXenes in 2011, they have gathered the attention of scientific community for usage in MSCs [29].

2. MXenes: SYNTHESIS AND PROPERTIES

MXenes are synthesized either by selective etching or through chemical vapour deposition. First reports on synthesis of MXene indicated elimination of Al layer from Ti_3AlC_2 (MAX) by using hydrogen fluoride [28]. Exfoliated two-dimensional $Ti_3C_2T_x$ possess morphology similar to 2*D* sheets resembling graphene sheets [30]. Various methods were used to avoid highly acidic HF acid.

Figure 4 below demonstrates the development of MXenes from MAX phases through HF treatment and their compositions in periodic table.

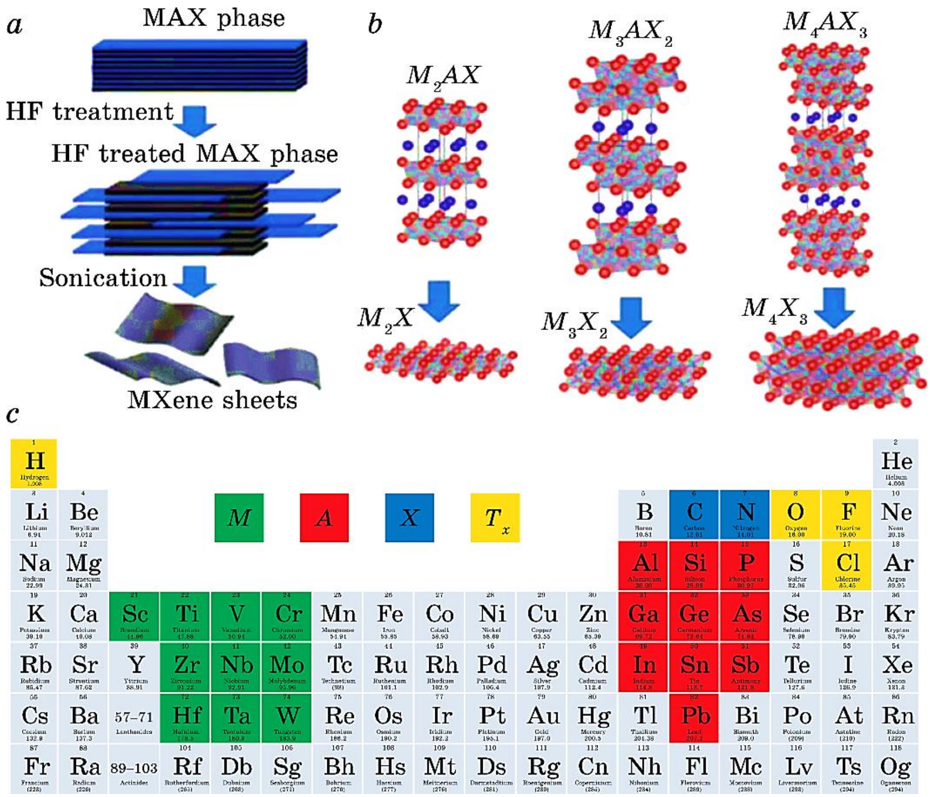

Fig. 4. (*a*) Schematic showing the synthesis of MXene from MAX by HF treatment; (*b*) synthesis mechanism of different order of MXenes by MAX phases; (*c*) compositions of MXene elements in periodic table (courtesy [31]).

Based on the method of synthesis, the characteristics of MXene are altered. It is reported that more than twenty types of MXenes have been synthesized experimentally [32].

MXenes act as significant platforms for supercapacitors in view of their electrical, mechanical properties and their surface morphology. They are classified into metallic, semi-metallic and semi-conducting materials [33]. Usually uncovered, they exhibit high conductivity and their electronic properties strongly depend on morphology and stacking of MXene sheets. It is reported that delaminated MXenes exhibit ultrahigh conductivity of up to 9880 S/cm [34]. Also depending on the synthesis method, a conductivity of 1 000 S/cm with HF etching was reported by MXene. This can be tuned to an extent of 4600 S/cm to 6500 S/cm in case of thick films with delaminated MXene by altering sonication and etching [35].

Mechanical properties of MXenes depend on their specific physical and chemical properties. Many studies on mechanical, electronic and thermal properties of various MXenes have been reported [36–40]. Young's modulus of $Ti_3C_2O_2$ and Ti_3CO_2 were reported as 466 and 983 GPA [41], which correlate with values predicted by simulation [42]. In this context, a study reported a paper film with $Ti_3C_2T_x$/PVA composite of thickness of 5 µm that can withstand approximately 15 000 times of its weight [43] indicating its strong wear. However, surface properties can be modified through surface terminations.

3. MXenes AND THEIR HYBRIDS FOR MICROSUPERCAPACITORS

High metallic conductivity and unique morphology of MXenes make them highly significant in microsupercapacitors. Low cost MXene MSC with 128 S/cm electrical conductivity and 25 mF/cm^2 capacitance in PVA–H_2SO_4 gel electrolyte was reported [44]. They also demonstrated that capacitance increases with thickness of the material. Similarly, MXene based MSC with wafer scale approach using photolithography with more capacitance was reported. This device was capable of converting the output peak voltage from 0.6 V to –0.56 V as compared to a commercial capacitor with 4mF [45]. A $Ti_3C_2T_x$ spray coated glass substrate with inter digital pattern with an areal capacitance of 20 mF/cm^2 at 20 mV/s and ultrahigh volumetric capacitance of 357F/cm^3 at 0.2 mA/cm^2 was fabricated. This was superior to other carbon materials already reported. However, usage of platinum collectors was reported to increase an areal capacitance to 27.3 mF/cm^2 [46]. Recently a semi-transparent MXene film was used with micropatterns of various transparency levels. With increase of 50% transparency, an areal capacitance increased by almost fifteen times from 19 µF/cm^2 to 283 µF/cm^2. At the same time, increase in resistance from 0.8 kΩ to 2 kΩ with increase in coating cycle has been observed [47]. Double-sided MSCs with MXene ink of 7.2 V potential were fabricated. Sharp rise in capacitance was observed with decrease of inter spacing between MXene electrodes. Such device with an inter electrode gap of 10 µm offered huge volumetric capacitance of 308 F/cm^3 at 5 mV/s and 96.4% efficiency above 10 000 cycles too [48]. Series connected MSCs achieved a high potential of up to 2.4 V [49]. Fabrication of MSCs with MXene as negative and MXene–MoO_2 film as positive electrode was reported. This technique included vacuum filtration of films followed by laser cutting of interdigital patterns as shown in Fig. 5 [50].

By using the technique of vacuum filtration, thick sheets of MXene were used to develop films with conductivity up to $1.25 \cdot 10^5$ S/m for flexible microsupercapacitor. Likewise, MSCs with interdigital pattern having 340 mF/cm^2 areal capacitance and 183 F/cm^3 volumetric capacitance with energy density of 12.4 mWh/cm^3 and

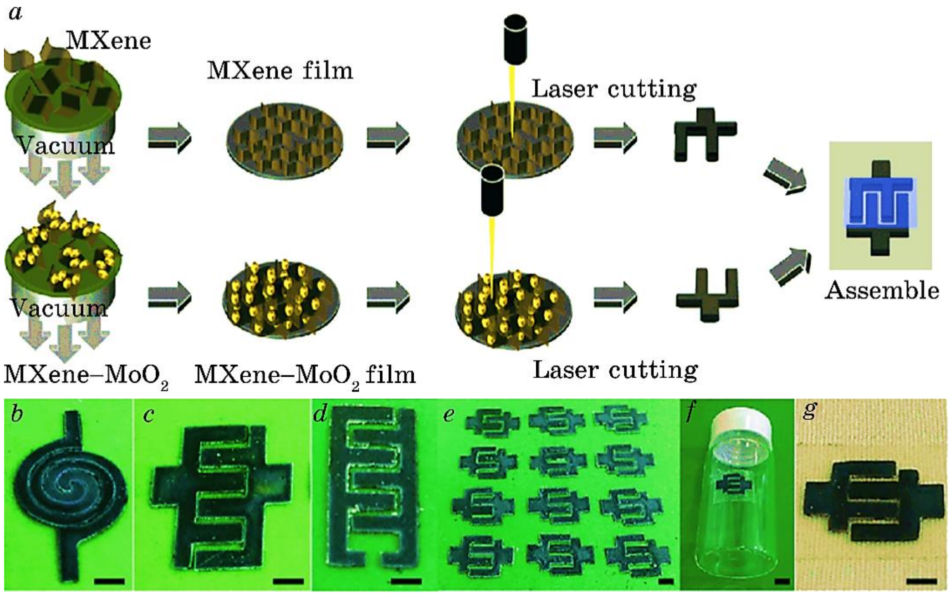

Fig. 5. (*a*) Fabrication process of MXene//MXene–MoO_2–MSCs with different shapes such as spiral (*b*), parallel inter digital fingers (*c*, *d*), twelve parallel inter digital MSCs integrated on one paper (*e*), and MSCs transferred onto glass substrate (*f*) and cloth substrate (*g*) (courtesy [50]).

power density of 218 mW/cm^3 are fabricated [51]. In addition, paper based MXene electrodes with high conductivity and areal capacitance of 23.4 mF/cm^2 at 0.05 mA/cm^2 was fabricated. In continuation, fabrication of electrodes in series as well as parallel was taken up to achieve required capacitance [52].

MSCs based on MXenes and CNT with fast ion diffusion and high conductivity were reported. They achieved this by fixing a gap of 500 nm between interdigital fingers and obtained areal capacitance of 317.3 mF/cm^2 at 50 mV/s. Decrease in gap increased an areal capacitance and energy density attributed to improved rate of ionic transfer [53, 54]. Fabrication of a three dimensional MXene/*r*GO aerogel MSC was reported. Figure 6 show the fabrication process, in which the device was embedded with polyurethane for adhering to external damage. This device exhibits an areal capacitance of 34.6 mF/ cm^2 at 1 mV/s with excellent recovery of electronic and mechanical properties even after full breakdown [55, 56].

A high performance asymmetric flexible MSC with *r*GO as positive and MXene as negative electrodes achieved a working potential of 1 V with number of bending cycles and 2.4 mF/cm^2 areal capacitance at 2 mV/s indicating MXenes potential for negative electrodes in asymmetric devices with high stability and strong performance [56].

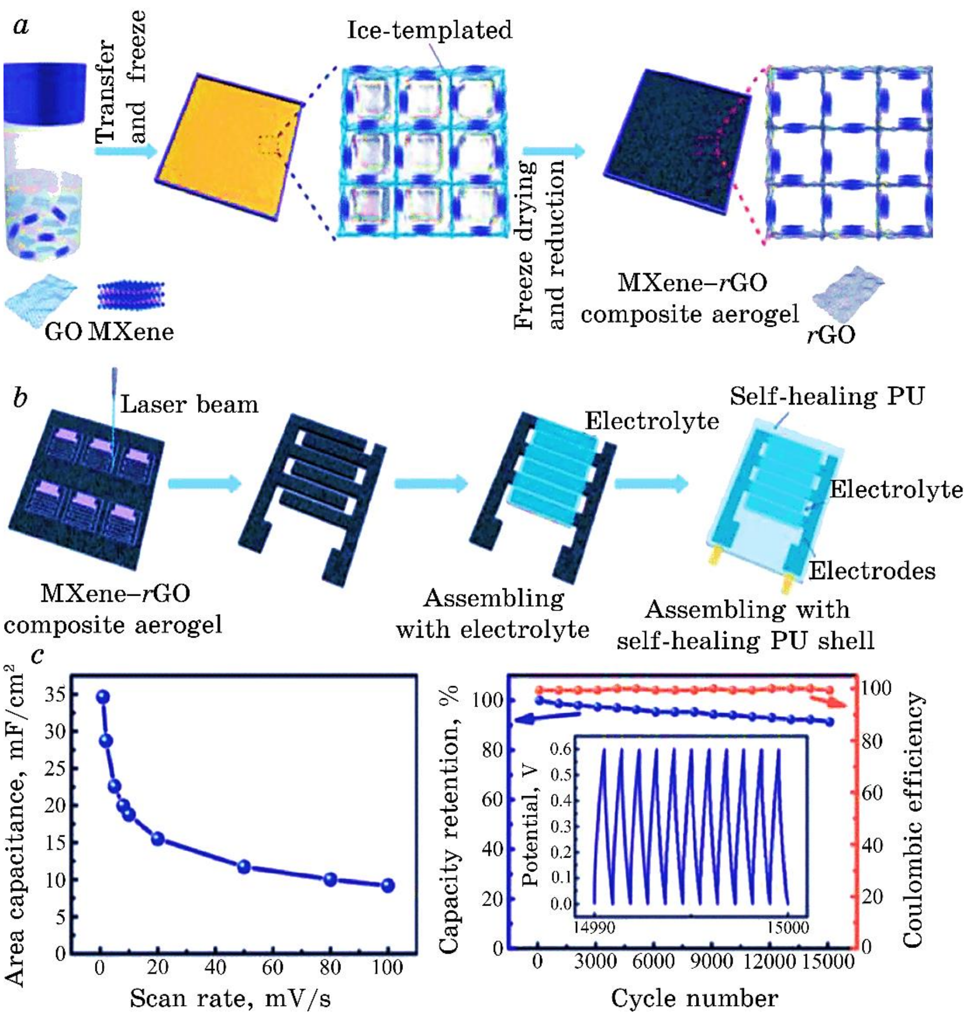

Fig. 6. (*a*) Fabrication of MXene–*r*GO composite aerogels; (*b*) laser cutting of interdigital pattern on MXene–*r*GO composite followed by assembling with self-healing PU; (*c*) plot showing the areal capacitance *vs* scan rate MXene–*r*GO composite; (*d*) cycling stability of MXene–*r*GO composite aerogel MSC at 2 $mA \cdot cm^{-2}$.

MXenes are also capable of producing textile based energy storage devices with high stability and tunability. Helical shaped MXene/CNT with scaffold hybrid structure reported 19.1 F/cm^3 volumetric capacitance at 1 A/cm^3 in aqueous LiCl electrolyte. Its energy density was of about 2.55 to 1.15 mWh/cm^3 at power density of 0.046 W/cm^3 to 1.82 W/cm^3, which is almost equal to the best performing capacitor [57].

Similarly, fabricated MXene/*r*GO hybrid fibre supercapacitors using wet spinning exhibit high volumetric capacitance of 586.4 F/cm^3 at 10 mV/s. These fibres report high conductivity of $3\cdot10^4$ S/cm whose flexibility can be enhanced by changing the grapheme content [58]. Similarly, fabrication of MSCs by introducing the MoS_2 into MXenes enhanced the electrochemical performance by 60% in comparison to pristine MXene [59]. Self-restacking of MXene layers was taken up by adding RuO_2 nanoparticles in order to improve ion exchange rate. Integration of conductive Ag nanowires into MXene decreases electrodes surface resistance. These strategies achieved MSC with volumetric capacitance of 864.2 $F\cdot cm^{-2}$ at 1 mV/s with 90% of capacitance retention even after 10 000 cycles [60].

PANI/MXene-based film electrodes with an exceptionally high volumetric capacitance of 1167 $F\cdot cm^{-3}$ were reported for the first time [61]. Stretchable MSCs based on MXene/bacterial cellulose composite with high Young's modulus of 15–35 GPa and tensile strength of up to 200–300 GPa were fabricated. Here, bacterial cellulose acts as a gap between MXene sheets preventing re-stacking of MXene flakes [62]. MXene–polymer composite nanofibers as flexible yarn electrodes were synthesized. This was achieved by electro spinning of active material on PET sheets. This device displays high areal capacitance of up to 18.39 mF/cm^2 at scan rate of 50 mV/s, which is better than many other carbon based yarn fibre supercapacitors [63]. In continuation, another group reported similar MSCs such as MXene/PEDOT-PSS-based yarn supercapacitors (YSCs) with 95% capacitive retention after 10 000 cycles, which is found significant in portable electronics [64]. In the same way, fabrication of dual-core yarn supercapacitor (YSC) with *r*GO and MXene hybrid fibres encapsulated with PVA–H_2SO_4 was reported. Its mean diameter was of approximately 500 μm with excellent linear capacitance of 43.6 mF/cm at 20 mV/s [65].

5. CONCLUSIONS

Right from the discovery of MXenes, they have become one of the unique choices for microelectrodes in MSCs for electronics applications. Their excellent properties such as large conductivity, volumetric capacitance makes them well suited for MSCs. However, fabrication of MXene-based MSCs is in the development stage, which needs to be further optimized in terms of material used for electrode, substrates and electrolytes. As per the existing literature, main focus of MXene based MSCs is towards the increase in areal capacitance and power density. Already, it is observed that self-discharging in open circuit condition needs to be attended on immediate basis. As per the earlier reports, this drawback can be rectified through integrating MSCs with solar power cells to enhance long-term charge storage property instead of self-discharging. In order to increase the electrochemical performance, electrolyte selection plays a vital role. Usually, polymer gel electrolyte was used for ion exchange in MXene-based electrodes for microdevices, whose output voltage is low. Hence, alternative is required in order to increase the voltage and stability. Hence, different electrolytes and polymers need to be envisaged, which enhance performance of MSCs. However, ionogel may be one option with high stability in terms of mechanical and thermal than regular gel electrolytes. Expansion of potential may be possible with asymmetric devices for real time applications. Apart from the $Ti_3C_2Ti_x$ (MXene) based MSCs, many MXene materials might be synthesised for better understanding of charge storage mechanism that lead to future MSCs devices.

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