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Study on Strategies to Enhance the Energy Density of MXene Supercapacitors

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Abstract: MXene is a newly developed two-dimensional energy storage material in recent years, featuring abundant surface functional groups, fast ion transport channels, and sufficient surface active sites. It has become a current research hotspot, with a large number of studies emerging in recent years. This paper provides a comprehensive review of the application of MXene materials in supercapacitors. First, the working principle of supercapacitors is introduced, and the key factors affecting their electrochemical performance are analyzed. Then, the advantages and disadvantages of various types of electrode materials are summarized in detail, with a focus on the application of MXene materials in supercapacitor electrodes. This includes an overview of the synthesis methods, structural advantages, electrochemical performance, and existing issues of MXene. Finally, strategies and suggestions for improving the performance of MXene electrode materials in the future are proposed. This review provides detailed information and reference value for the application research of MXene materials in supercapacitors.

Keywords: MXene, Supercapacitor, Electrode Material, Electrochemical Performance, Energy Density, Power Density

1. Introduction

With the rapid development of renewable energy, energy storage technology has become one of the primary bottlenecks limiting its application. Traditional electrochemical energy storage systems face challenges such as limited capacity and low charge-discharge efficiency. Consequently, the development of novel energy storage devices has become a critical topic. Supercapacitors have attracted widespread attention due to their fast charge-discharge capabilities, long lifespan, and high efficiency, making them strong contenders as next-generation high-efficiency energy storage devices. Their unique working principle offers extensive research potential, and investigating factors that impact their performance has become a popular area of study. Current research mainly focuses on improving key parameters of supercapacitors, such as conductivity, specific surface area, and specific capacitance, with a primary approach being the optimization of electrode materials. Different electrode materials impart distinct advantages to supercapacitors, making the enhancement of electrode materials essential to improving supercapacitor performance. Beyond traditional materials such as carbon materials, metal oxides, and conductive polymers, MXene, as a novel two-dimensional material, offers significant advantages like abundant surface chemical components, high conductivity, large specific surface area, and excellent tunability. These attributes reveal MXene's immense

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potential as a supercapacitor electrode material. Present research focuses on increasing MXene's specific surface area, modulating its surface functional groups, and combining it with other electrode materials to enhance its performance. However, MXene still faces considerable challenges in practical applications; for instance, its high preparation costs make large-scale production difficult, and its susceptibility to oxidation during operation leads to aging issues. In view of these challenges, this paper analyzes and discusses the potential of MXene in applications and its remaining issues. It provides a comprehensive review of the latest research progress on MXene's application in supercapacitor electrodes and highlights its significance in advancing this field.

2. Working Principles of Supercapacitors and Performance Influencing Factors

Supercapacitors, compared to regular capacitors, possess exceptionally high power density and a long cycle life. As shown in Figure 1, the working principle of a supercapacitor involves storing charge through the formation of an electrochemical double layer. When electrodes are immersed in an electrolyte, charges form opposing double layers at the liquid interface due to Coulomb forces, atomic forces, and intermolecular forces, creating a stable double-layer interface. This process mainly consists of physical adsorption of charges and electrochemical reactions [1]. In this context, charge adsorption refers to the process in which the positive and negative electrode surfaces in the supercapacitor attract electrons and ions to store charge. The electrochemical reaction mainly involves materials like titanium dioxide or conductive polymers as the anode, storing charge through redox reactions [2]. Key factors affecting the performance of supercapacitors include conductivity, specific surface area, pore size distribution, specific capacitance, and wettability. Optimizing these factors can significantly enhance supercapacitor performance.

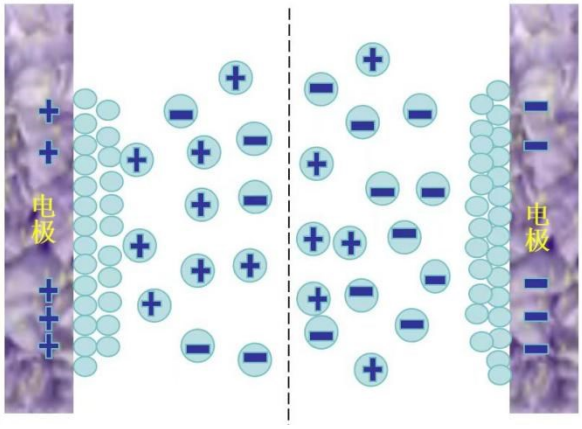

Figure 1: Diagram of Supercapacitor Working Principle.
Note: The dark sections on both sides represent the electrodes.

2.1. Conductivity

Conductivity refers to the ability of a material to conduct electric current. In the field of capacitors, it is mainly reflected in the transfer and loss of electrical energy during the capacitor's charge and discharge processes. The stronger the conductivity, the less current loss during operation, thereby improving the efficiency of the capacitor in use, which in turn affects its lifespan and a range of other performance characteristics. The conductivity of different materials varies significantly. Traditional materials, such as carbon materials, possess excellent conductivity, which is why carbon materials remain a key component in capacitor manufacturing to this day. However, with the advancement of research, various emerging materials have been developed, many of which have made breakthroughs in conductivity. For example, the newly developed conductive polymer materials in recent years offer

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better conductivity and lower internal resistance, making them perform better than activated carbon materials [3]. Additionally, conductivity is not only determined by the material itself; the working environment and capacitor structure also influence this factor. Therefore, while searching for materials with superior conductivity, exploring more rational structures that adapt to specific working environments is also a key area of research. It is clear that the influence of conductivity is multifaceted, involving material factors and beyond, making the study of conductivity crucial for enhancing capacitor efficiency.

2.2. Specific Surface Area

Specific surface area refers to the total surface area per unit mass of a material and is one of the key indicators for evaluating catalysts, adsorbents, and other porous materials such as asbestos, mineral wool, diatomaceous earth, and clay minerals in industrial applications. Generally, porous materials with a large specific surface area and high activity exhibit strong adsorption capacity. As mentioned earlier, the working reaction process of supercapacitors mainly involves adsorption and electrochemical reactions, highlighting the importance of "adsorption" in the field of supercapacitors. The adsorption performance of the materials constituting the supercapacitor electrodes significantly impacts the capacitor's performance. Since electrodes serve as reaction sites, the more electrons and ions they can adsorb, the faster the reaction rate, and consequently, the more energy can be stored. For example, activated carbon, one of the most classic materials, is characterized by its large specific surface area due to its porous structure, and improving this property has been one of the main research directions in recent years. There are many factors influencing specific surface area, and the situation can be quite complex. In some cases, materials with low specific surface area outperform those with high specific surface area. This discrepancy between theory and practice is primarily due to the limitation of the electrode microstructure, which prevents the electrolyte from fully infiltrating the electrode material, resulting in the incomplete utilization of the material's specific surface area. This issue is commonly observed in carbon material electrodes, and typically, heat and chemical treatments are used to modify their properties by increasing surface area and porosity [4]. Based on this, exploring more methods to enhance specific surface area and improve the utilization of its properties is of significant importance for the advancement of supercapacitors.

2.3. Pore Size and Distribution

During the adsorption and chemical reaction processes, electrolyte ions must be stored in the pores on the electrode surface, providing active sites for reactions or catalysis. Among different pore sizes, micropores play a role in energy storage, while mesopores provide channels for rapid ion migration. The proportion of micropores and mesopores within the pore structure is thus a critical indicator of electrode material performance, especially for carbon-based materials, as the combination of both generally yields optimal results. High-performance materials also require a uniform pore size distribution to create a suitable reaction environment. To measure this, surface pore distribution analyzers are often used when selecting electrode materials. Additionally, mass transfer resistance within the carbon pores contributes significantly to internal resistance when carbon electrodes are immersed in solution, which can affect conductivity and, in turn, influence capacitor efficiency. Pore size notably impacts ion diffusion and behavior; not all pores can act as reaction sites, as they must match in compatibility [3]. Therefore, controlled pore-size materials have become a hot research area, such as carbon materials derived from liquefied larch, which allow for tunable morphology and pore structure [5]. This adaptability greatly enhances material suitability, and further exploration of these characteristics shows promising prospects for supercapacitor applications.

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2.4. Specific Capacitance

Specific capacity refers to the energy released per unit mass or volume. This concept can be divided into two categories: one is mass-specific capacity, which refers to the amount of charge that can be released by a battery or active material per unit mass; the other is volume-specific capacity, which refers to the amount of charge that can be released by a battery or active material per unit volume. According to the energy density calculation formula ($E = CU^2/2$, where U is the voltage range and C is the specific capacity), it can be seen that materials with higher specific capacity contribute to greater energy density in capacitors. Specific capacity is determined by the intrinsic properties of the material. Generally, it can be improved by modifying the composition of the electrode materials or altering their crystal structure and microstructure. Traditional methods calculate specific capacity based on hydroxides and oxides, but in recent years, a method based on cations has been proposed, which better aligns with the latest developments in colloidal ion electrode technology. To date, among various materials, ruthenium dioxide (RuO_2) is considered an excellent electrode material for pseudocapacitive supercapacitors. It can react under both aqueous and non-aqueous conditions, and its hydrated form, $RuO_2 \cdot nH_2O$, can also participate in redox reactions within the electrolyte. The theoretical specific capacity of this electrode material is 1360 F/g [6]. As an extremely important parameter, specific capacity provides significant reference value when selecting electrode materials.

2.5. Electrolyte Wettability

Different materials use different electrolytes, and their compatibility is essential for optimal performance. Selecting an electrolyte that best matches and thoroughly wets a specific material is an unavoidable focus in supercapacitor development. Surface wetting of the electrode is a dynamic process that involves the diffusion and mass transfer of electrolytes on the electrode surface and within its pores. Wettability speed directly influences the effective contact area between the electrode and electrolyte, thus affecting the reaction level on the electrode surface. Interfacial tension between the electrolyte and electrode material impacts how the electrolyte wets the electrode pores; capillary action can promote electrolyte entry into narrow pores, but excessive interfacial tension hinders wetting. Some hydrophobic materials resist electrolyte penetration, and pore size and distribution also influence wetting behavior; excessively dense or small pores can block electrolyte infiltration. To address this, surface modification or functionalization can increase hydrophilicity, while applying an electric field can alter the interfacial properties between the electrolyte and electrode material, especially under high-voltage operation. Additionally, wetting is often accompanied by endothermic or exothermic reactions, with temperature changes affecting electrolyte surface tension and thereby impacting wettability. In summary, achieving optimal wettability is essential for enhancing supercapacitor performance and requires balancing factors such as electrode materials, electrolyte properties, and structural design. Systematic research on wetting mechanisms will support the informed selection and design of electrode and electrolyte materials.

3. Analysis and Discussion of Various Electrode Materials

Currently, the primary types of traditional electrode materials for supercapacitors include carbon materials, metal oxides, and conductive polymers.

3.1. Carbon Materials

Carbon materials are the most fundamental, offering excellent electrical conductivity and chemical stability. Based on the double-layer principle, the double-layer capacitance on the electrode surface is approximately 20 $\mu F/cm^2$. Using activated carbon with a specific surface area of 1000 cm^2/g as the

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electrode material can yield a specific capacitance of 200 F/g. However, its drawback is evident: since the reaction only occurs on the material's surface, carbon electrodes exhibit relatively low specific capacitance. Given this limitation, strategies to enhance the energy storage performance of double-layer capacitors focus on the development of porous carbon materials with high specific surface area and low internal resistance, as well as modification research on carbon-based materials. For example, Geng et al. conducted preliminary activation of carbonized coconut shells under high-temperature CO_2, followed by secondary activation with H_2O at 850°C, resulting in expanded original pores and a significant number of new pores. Electrolyte ions could then quickly diffuse and migrate within the developed pore structure, increasing the mass-specific capacitance of porous activated carbon from 140 F/g to 240 F/g at a current density of 0.5 A/g, while also significantly enhancing cycling stability [1]. Moreover, glassy carbon, carbon nanotubes, and other materials are also receiving considerable attention in supercapacitor research. Sullivan M. G. used anodic oxidation to modify glassy carbon electrodes, achieving a volumetric capacitance of 100 F/cm³. Chunming Niu et al. developed a capacitor with carbon nanotubes that exhibited a specific capacitance of 40 F/g and maintained 49 F/g at 100 Hz, a frequency response much higher than activated carbon's 1 Hz, demonstrating superior frequency response characteristics [4]. Currently, methods to improve the performance of carbon materials without altering their composition primarily include thermal and chemical treatments to increase surface area, porosity, and functional group density.

3.2. Metal Oxides

Unlike the double-layer electrostatic adsorption reaction in carbon materials, metal oxides can store more charge when used as supercapacitor electrode materials, showing higher specific capacitance. For instance, metal oxides represented by RuO_2 exhibit Faradaic pseudocapacitance at the electrode/solution interface, which is far greater than the double-layer capacitance generated by activated carbon. However, these materials also have limitations, including inherently low electrical conductivity and structural rigidity, which negatively impact the reversibility of redox reactions and the structural stability of the electrode. Additionally, the high cost of metal oxides poses challenges for production. Therefore, the focus for improving these materials is on increasing specific surface area and identifying more affordable metal oxides. Current studies have shown that the combination of multiple materials is a feasible approach. Previous research found that $Pb_2Ru_2O_{6.5}$ delivers a discharge energy density greater than 5 Wh/kg, while composite oxides such as $WO_3 \cdot xH_2O/RuO_2$ and $Na_{0.37}WO_3 \cdot xH_2O/RuO_2$ reach a specific capacitance of up to 560 F/g with 50% RuO_2. Amorphous $Ru_{1-y}Cr_yO_2 \cdot H_2O$ composite oxides exhibit a specific capacitance of 840 F/g. Composite oxides with RuO_2 and other oxides (e.g., MoOx, VOx, TiO_2, SnO_2), prepared sequentially through sol-gel methods, have also achieved high specific capacitance while reducing RuO_2 usage [4].

3.3. Conductive Polymers

Conductive polymers such as polyaniline (PANI), polypyrrole (PPy), polythiophene, and poly(3,4-ethylenedioxythiophene)-polystyrene sulfonate (PEDOT) have been widely utilized as active materials in electrochemical energy storage systems. These polymers enable energy conversion and storage through reversible redox reactions of organic groups within their structures or through reversible ion doping/deintercalation reactions with ions from the electrolyte, providing good electronic conductivity and high specific capacitance, often two to three times that of carbon materials. Polyaniline, for example, offers excellent electronic conductivity, reasonable cost, and ease of integration with other materials, making it a research focus [6]. Generally, conductive polymers as electrodes fall into two categories: directly prepared polymer electrodes and polymer-derived hard carbon electrodes via high-temperature pyrolysis. The former can be further classified into three

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capacitor structures: one n-type doped and one p-type doped conductive polymer, two different p-type doped conductive polymers, and two identical p-type conductive polymers. Among these, the first structure has clear advantages, including high energy storage capacity, high conductivity, and optimal utilization of electrolytes. The other pyrolysis-derived product, hard carbon, forms disordered single-layer carbon atoms, retaining the original carbon ring structure and thus providing high charge storage capacity [4]. The main drawback of conductive polymers is their relative instability; their organic structures experience shrinkage and expansion during reactions, leading to volume changes that can damage the material structure during charge-discharge cycles. To address this, mainstream solutions involve combining conductive polymers with carbon materials, with carbon nanotubes supporting the internal structure to prevent structural changes [6]. This area of research remains a major focus.

Each of the three major material types has its advantages and limitations: carbon materials have good conductivity but limited specific surface area; metal oxides offer high specific capacitance but poor conductivity; and conductive polymers provide decent conductivity but poor cycle stability. Considering these pros and cons, composite materials and novel carbon materials have become research hotspots. Among them, MXene has recently emerged as a promising material.

4. Strategies for Enhancing Energy Density of MXene Electrode Materials

MXene, or two-dimensional transition metal carbides/nitrides, is a type of two-dimensional layered material. Due to its graphene-like properties and structure, it is named MXene. Figure 2 presents a schematic diagram of the structure of MXene. In 2011, Ti3C2Tx was successfully synthesized for the first time by chemical exfoliation of Ti3AlC2 using hydrofluoric acid. As of now, more than 100 types of MXene materials have been developed [7]. As a two-dimensional material, MXene has several advantages, including rich surface functional groups, high conductivity, and a large specific surface area. Additionally, because its surface atoms are exposed as a monolayer material, its properties can be easily modulated using surface chemical modifications and element doping methods. Its unique structure also endows MXene materials with high stability, bending strength, and excellent electronic and magnetic properties. In terms of preparation, the precursor of MXene is the MAX phase, from which the A-layer atoms located in the middle layer can be exfoliated to obtain MXene. The MAX phase contains metallic bonds, ionic bonds, and covalent bonds, with metallic bonds existing between M and A layers and ionic and covalent bonds between M and X layers. The metallic bonds are relatively weaker in comparison, so in high-temperature environments, A atoms diffuse and detach. At this point, appropriate methods can be used to etch away the A-layer atoms to obtain MXene, with fluoride etching methods being commonly used [8]. Currently, significant progress has been made in the research on MXene as a supercapacitor electrode material, and its performance has surpassed many other materials. However, issues such as the strong corrosiveness of the synthesis process, poor conductivity of certain MXene materials, and impurities in the synthesized MXene still need to be addressed. The following sections will classify and discuss strategies for improving the performance of MXene electrodes.

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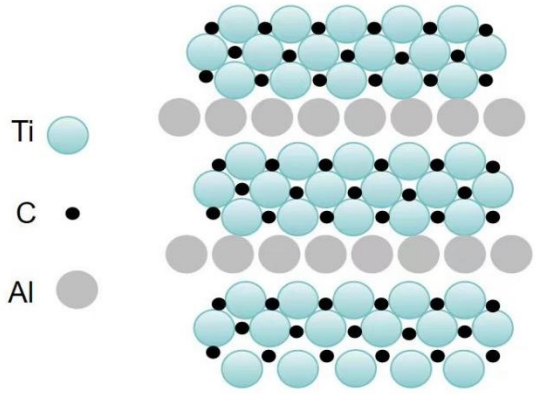

Figure 2: Schematic of MXene Structure

4.1. Increasing the Specific Surface Area of MXene, such as by Optimizing Interlayer Spacing

Enlarging the interlayer spacing exposes more active sites to the electrolyte, facilitating additional redox reactions and broadening ion transport channels, thereby enhancing performance. Various methods exist for optimizing interlayer spacing. For example, inserting large organic molecules between MXene layers, such as ethylenediamine (EDA), which coordinates with N-Ti bonds on Ti_3C_2Tx nanosheets, increases interlayer spacing. The modified material exhibits a higher specific surface area, richer mesoporosity, and improved crystallinity [9]. These inserted organic molecules, referred to as intercalants, fall into two main categories: organic molecules and metal or ammonium cations. EDA is an example of the former. For the latter, the selectively etched MXene surface carries functional groups like -O and -OH, resulting in a negatively charged surface that can spontaneously adsorb metal and ammonium cations. These cations can then insert between MXene layers, adjusting the interlayer spacing [9]. Different cations affect interlayer spacing variably, enabling the use of ion exchange methods to tailor spacing to diverse application needs.

Additionally, self-supporting MXene membranes can be prepared using ice templating and freeze-drying. When MXene is frozen, water molecules between layers form ice templates, significantly increasing interlayer spacing. Introducing carbon nanotubes further introduces water molecules, creating additional ice templates [9].

4.2. Regulating Surface Chemical Composition of MXene to Introduce Favorable Functional Groups

The synthesis method of MXene materials greatly impacts the types and quantities of functional groups on their surface. For example, MXene synthesized through the fluoride salt etching method is rich in mixed functional groups such as -F, -O, and -OH, whereas MXene obtained through hydrothermal-assisted alkaline etching lacks -F but introduces -O and -OH groups due to the aqueous solution used. Electrochemical etching introduces -Cl, -O, and -OH functional groups [9]. Numerous studies demonstrate that MXene's surface functional groups significantly influence its thermal, mechanical, and electrochemical properties. Research shows that materials rich in -O functional groups tend to exhibit better thermodynamic stability and mechanical strength, and the -O groups enhance lithium-ion storage capacity by facilitating additional lithium ion adsorption. In contrast, -F and -OH groups can reduce lithium-ion transport and reaction efficiency, thereby suppressing charge storage capacity. Meanwhile, -S functional groups endow MXene with excellent conductivity and

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charge storage capacity, along with a low diffusion barrier [9]. Additionally, MXene's surface functional groups impact its ability to adsorb metal ions; MXene without functional groups demonstrates higher capacity and electron mobility.

4.3. MXene Composites

Different MXene materials have unique advantages and drawbacks. Single-material MXene electrodes often exhibit limitations in specific aspects, such as low specific capacity and inadequate self-repair capability. Composite materials effectively address these issues and can be broadly categorized into three types: composites with carbon materials, metal oxides/sulfides, and polymers. Carbon materials, with high porosity, large surface area, and high conductivity, are among the most commonly used electrode materials. Using electrophoretic deposition, binder-free Ti_3C_2Tx/carbon nanotube films can be deposited on graphite sheets, where the addition of carbon nanotubes helps create a stable structure within the electrode, preventing further degradation of MXene materials. Another method involves using Ti_3C_2Tx as a flexible conductive binder to encase activated carbon particles within MXene layers, producing flexible carbon electrodes that build a 3D conductive network to facilitate electron transport. However, due to the inherently low specific capacity of carbon materials, carbon composites alone are ineffective in significantly enhancing the specific capacity of electrode materials [10]. Unlike carbon materials, oxides/sulfides offer high specific capacity and excellent rate performance. Metal oxides, in particular, provide additional pseudocapacitance. Due to the positive charge of metal oxides/sulfides, they can readily combine with the negatively charged MXene through electrostatic assembly. MXene/CoAl-layered double hydroxide heterostructured films are prepared using an electrostatically ordered self-assembly approach; these films exhibit excellent electrochemical performance owing to their unique structural composition and strong interfacial interactions. Additionally, three-dimensional flower-like NiS structures can be hydrothermally grown on MXene, forming a hollow network structure that facilitates ion and charge transfer. Given the limited conductivity of metal oxides/sulfides, current development focuses on leveraging their high specific capacity, with precise structural tuning being a primary research focus [10]. As a standalone electrode material for supercapacitors, polymers often suffer from poor cycling stability due to volume changes during charging and discharging. When combined with MXene, the composite structure prevents MXene agglomeration and simultaneously inhibits polymer expansion. Polymer monomers can spontaneously intercalate between MXene layers; however, as conductive polymers, they still face inherent challenges such as poor conductivity and low cycle stability, making them less of a research focus [10].

5. Future Outlook

MXene shows significant promise as a supercapacitor electrode material. Its unique structure provides it with superior properties, including high specific surface area and conductivity, and, more importantly, its adjustable nature allows for a wider range of applications. This paper summarizes the traditional electrode materials, thoroughly highlighting the unique advantages and characteristics of MXene and exploring its potential applications. Future research should focus on simplifying the synthesis process, reducing costs, and increasing yield. Additionally, researchers should leverage MXene's unique tunability to optimize its electrochemical performance through structural and compositional control, thereby enhancing its practical application value in supercapacitors. Although research on MXene electrode materials has made substantial progress, some issues remain to be addressed.

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5.1. Large-scale Production of MXene.

The corrosiveness of the MXene synthesis process limits its large-scale application, and high material wastage during production keeps the cost of MXene elevated, restricting its use across various fields. A key research direction is finding ways to reduce the corrosiveness of the synthesis process while maintaining or improving the material's electrochemical performance.

5.2. Oxidation Issues with MXene.

Due to its structure, MXene is prone to oxidation, as the Ti atoms on its surface are particularly susceptible to oxidation. This process accelerates as electrons accumulate, making MXene materials prone to oxidation and shortening their service life. Enhancing the thermal and electrochemical stability of MXene is a critical topic. Besides improving working environments and oxygen isolation, MXene's oxidation issue can be alleviated through surface charge regulation. For instance, since MXene layers generally have a negatively charged surface, loading cations onto the surface can help mitigate oxidation [11].

5.3. Compatibility of MXene with Various Electrolytes.

MXene exhibits different properties in various electrolytes, including organic, ionic, aqueous, and solid-state electrolytes, all of which demonstrate strong energy storage capabilities. Organic electrolytes provide a wider voltage window, leading to higher energy density, while in ionic electrolytes, MXene exhibits energy storage capacity suitable for electrochemical supercapacitors. Aqueous electrolytes offer higher ionic conductivity and improved safety. In acidic and alkaline electrolytes, MXene displays varying performance, with sulfuric acid electrolytes showing the highest capacitance [8]. However, MXene materials—and even those synthesized by different methods—demonstrate unique properties in different electrolytes, indicating further research opportunities regarding MXene-electrolyte interactions.

5.4. Application of MXene in Other Energy Storage Devices.

MXene's applications extend beyond supercapacitors and into various energy storage devices. For example, MXene has applications in aqueous zinc-ion batteries, where it serves in the development of cathode materials, zinc anode modification, and electrolyte enhancement. Utilizing the tunable properties of MXene as a cathode can improve discharge energy and zinc storage capacity in these devices. When used as an anode, MXene effectively suppresses zinc dendrite growth, enhancing performance. MXene additives in the electrolyte can further optimize various battery metrics [12]. In summary, MXene holds tremendous potential for various applications and warrants further exploration.

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