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Research Progress of Tin Dioxide Materials for Supercapacitors

Tian Han

School of Chemistry and Molecular Engineering, East China University of Science and Technology, Shanghai, China
19816380564@163.com

Abstract. This paper provides a complete, logically clear and well-structured review of the research progress of tin dioxide (SnO_2) materials in the field of supercapacitors. First, it systematically and rigorously introduces the intrinsic physical and chemical properties of SnO_2 as an n-type semiconductor material, namely high theoretical specific capacitance, abundant redox active sites, excellent thermochemical stability and wide working potential window, which naturally leads to its application basis as a pseudocapacitive electrode material. Then, the main optimization strategies of SnO_2-based materials are fully summarized. More importantly, the paper makes an excellent comparative analysis of the differences in electrochemical behaviors of SnO_2 electrodes in different electrolyte systems such as aqueous, organic and solid-state electrolytes, and naturally transitions to the positive significance of device designs such as flexibility and miniaturization for the practical application of SnO_2-based supercapacitors. Finally, the author objectively and prudently points out various problems encountered in the practical application of existing SnO_2 materials: the actual specific capacitance is far lower than the theoretical value, the volumetric energy density still has room for improvement, and the structure is prone to fatigue during long-term cycling. On this basis, the future development direction and application prospects are proposed. It can be regarded as an excellent model combining theory and practice, and thus provides a solid theoretical reference and direction guidance for the research and development of high-performance SnO_2-based supercapacitor electrode materials.

Keywords: Tin dioxide, Supercapacitors, Materials

1. Introduction

With the increasingly severe global energy crisis and prominent environmental problems, developing efficient and clean energy storage technologies is of direct and great significance for achieving the "dual carbon" goal. Supercapacitors have attracted great attention in portable electronics, electric vehicles, smart grids and other fields due to their advantages of high power density, fast charge-discharge rate and long cycle life. Electrode materials are the fundamental factor determining supercapacitor performance. Therefore, transition metal oxides have naturally become

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the research focus in this field for their abundant redox active sites and large pseudocapacitance capacity [1, 2].

Tin dioxide (SnO_2), as a typical n-type transition metal oxide, has advantages of abundant raw materials, simple preparation, good chemical stability and high theoretical specific capacitance brought by the reversible conversion of Sn^{2+}/Sn^{4+}, which shows great application potential in the field of supercapacitors [3]. By discussing the energy storage mechanism of SnO_2-based electrodes, this paper focuses on clarifying the synergy rule of electric double layer capacitance and pseudocapacitance, as well as the specific effects of different regulation methods on energy storage performance (specific capacitance, rate performance, cycle life), including micro-nano structure regulation (nanosheets, nanofibers, nanoparticles) [4], heteroatom doping (Fe, Cu, Zn, Mg, Mn, Al, etc.), composite with highly conductive carbon materials (carbon nanotubes, graphene, carbon cloth) [2] and MXene, oxygen defect engineering, and conductive polymer coating [5]. The specific mechanisms of each strategy to improve the poor intrinsic conductivity of SnO_2, inhibit volume expansion during charge and discharge, and enhance cycling stability are analyzed in detail.

This paper systematically reviews the research progress of SnO_2-based materials in supercapacitors, analyzes their intrinsic characteristics, optimization strategies and energy storage mechanisms, and prospects the future development direction, so as to provide reference for the research and development of high-performance SnO_2-based electrode materials.

2. Supercapacitors and energy storage systems

Supercapacitors (electrochemical capacitors) are power devices that store energy based on the principles of electric double layer and pseudocapacitance. They have fast charge-discharge rate, high power density and excellent cycling durability, and are gradually developing into a core technology for integrating renewable energy into power grids.

2.1. Introduction to supercapacitors

Supercapacitors, also known as electrochemical capacitors, are new energy storage devices between traditional capacitors and batteries. They have advantages of high power density, fast charge-discharge speed and long cycle life, but their energy density is generally lower than that of batteries. Thus, they naturally become an ideal choice for fast charge-discharge or instantaneous high-power output scenarios, such as electric vehicle acceleration, power grid frequency regulation and renewable energy grid connection. According to the energy storage mechanism, supercapacitors are mainly divided into three categories [6, 7]: 1. Electric double layer capacitors: They store energy through adsorption/desorption of electrolyte ions on the electrode surface, which is a pure physical process with extremely fast response and ultra-long cycle life. Typical electrode materials are high specific surface area carbon materials. 2. Pseudocapacitors: They store energy through fast and reversible redox reactions on the electrode surface. Their capacitance is much higher than that of electric double layer capacitors, and the process involves Faradaic reactions. Common materials include transition metal oxides and conductive polymers. 3. Hybrid capacitors: They combine electric double layer electrodes with pseudocapacitive electrodes or battery-type electrodes, taking into account the merits of both high energy density and high power density capacitors. According to different electrode materials, they can be divided into carbon electrodes, metal oxide electrodes, conductive polymers and so on.

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2.2. Energy storage system of supercapacitors

A supercapacitor device is mainly composed of electrode, electrolyte, separator and current collector. The core of energy storage lies in the positive and negative electrodes. According to the three charge storage mechanisms mentioned above, different types of materials are usually combined in practical supercapacitor applications to give full play to their respective advantages.

There are two main configurations of common energy storage devices: Symmetric supercapacitors are the simplest type, using exactly the same electrode material for both positive and negative electrodes, and carbon materials are commonly used for both poles. They feature extremely long cycle life but relatively low energy density. In KOH electrolyte, it shows an amazing ultra-long service life with a capacity retention rate of over 100% after 100,000 cycles, making it an ideal choice for high-power and long-endurance demands [8].

Asymmetric/hybrid supercapacitors are the clearest and most mature mainstream development trend at present. Different materials are used for positive and negative electrodes to achieve a good balance between "high energy" and "high power", so they are generally designed as an "energy-type" positive electrode and a "power-type" negative electrode [9]. More importantly, the negative electrode (for power and life guarantee) is specially used to provide fast charge-discharge capability and ensure long cycle life, so high specific surface area activated carbon (AC) is commonly used. The greatest advantage of hybrid devices is that they skillfully and reasonably combine the advantages of two energy storage mechanisms: the battery-type positive electrode improves the overall energy density, while the carbon negative electrode naturally ensures fast charge-discharge and long cycle life. Therefore, this is also the most widely used and effective strategy to improve supercapacitor performance at present [10, 11].

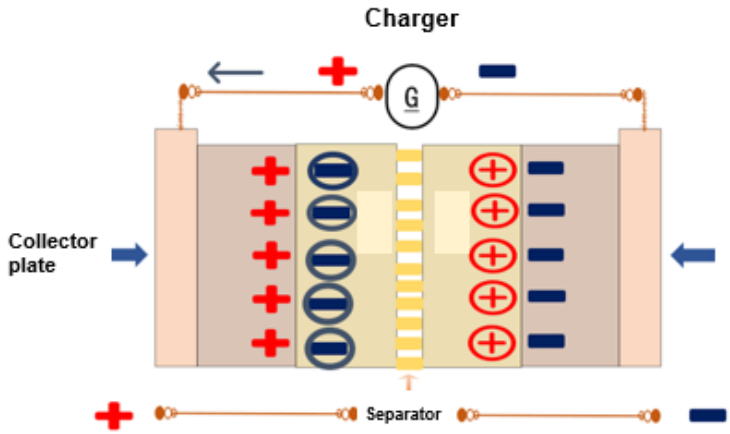

Figure 1. Schematic diagram of supercapacitor

3. Energy storage principles of supercapacitors

3.1. Electric double layer energy storage principle

The electric double layer energy storage principle is the most basic and core pure physical energy storage mechanism of supercapacitors. Its working process completely relies on electrostatic adsorption and ion desorption at the electrode-electrolyte interface. During charging, positive and negative ions in the electrolyte migrate to the surfaces of the positive and negative electrodes

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respectively under an applied electric field, and arrange closely on the electrode material surface to form an extremely thin charge layer. Meanwhile, a diffusion layer forms in the solution, and the two together constitute a stable electric double layer structure to store electric energy. Ions with opposite charges also form an extremely thin charge layer in the solution. During discharging, when the external circuit is connected to a load, electrons flow directionally, and the adsorbed ions desorb rapidly from the electrode surface and return to the bulk electrolyte freely. Since the whole process does not involve any chemical reaction or phase transition and completely relies on physical electrostatic force, it has extremely high reversibility and millisecond-level response speed. Its capacitance mainly depends on the effective specific surface area (A) of the electrode material and the electric double layer thickness (D). According to the electric double layer capacitance formula $C=\varepsilon_r\varepsilon_0 A/d$, porous carbon materials with high specific surface area and reasonable pore size distribution are ideal electric double layer electrodes [11, 12]. It is precisely this pure physical adsorption mechanism that endows electric double layer capacitors with extremely high power density and ultra-long cycle life of up to hundreds of thousands of cycles, but their energy density is usually relatively low [1].

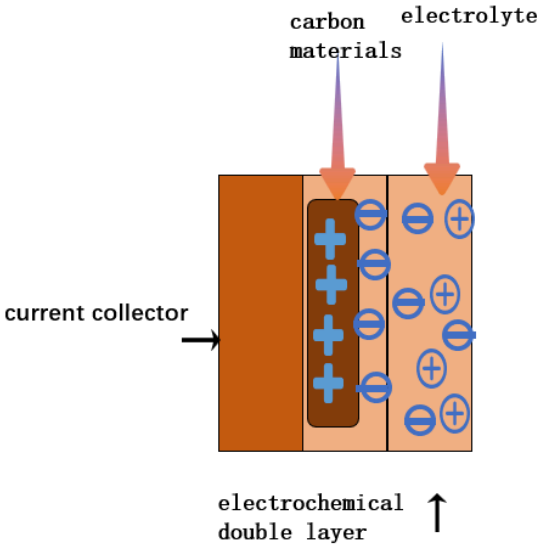

Figure 2. Schematic diagram of double-layer energy storage

3.2. Pseudocapacitance energy storage principle

The pseudocapacitance energy storage principle is based on highly reversible Faradaic redox reactions occurring on or near the electrode material surface, which is essentially different from the energy storage mechanism of pure physical electrostatic adsorption electric double layer capacitance. Its charge storage originates from fast electron transfer of electrode active materials with potential change, thus generating Faradaic charge storage throughout the charge-discharge process. The advantage of pseudocapacitance is that its capacity is significantly higher than that of EDLC, often 10 to 100 times that of EDLC, and it can improve energy density while maintaining high power density. In practical applications, such materials often face a core bottleneck: restricted by poor conductivity in the bulk phase, a large number of active materials cannot effectively contact the electrolyte and participate in reactions, resulting in actual capacitance far lower than the theoretical value. Therefore, the key to releasing pseudocapacitance performance is to improve the

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utilization rate of active materials [9, 10]. The improvement of pseudocapacitance performance largely depends on fine regulation of the nanostructure and electronic conductivity of electrode materials, which can be realized by compounding with highly conductive carbon materials or adopting heteroatom doping. It can effectively improve the inherent insufficient conductivity of transition metal oxides, so as to construct composite electrodes with high energy density and excellent rate performance.

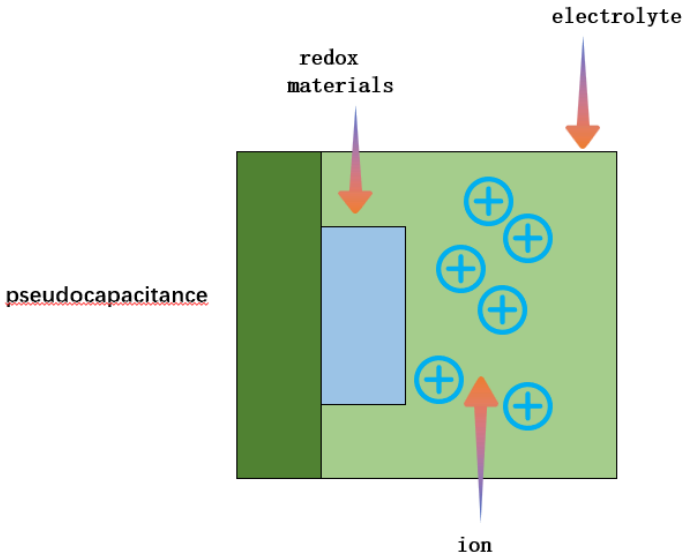

Figure 3. Schematic diagram of redox energy storage

4. Types of supercapacitor electrode materials

4.1. Carbon-based materials

The application of carbon-based materials in supercapacitors can naturally and hierarchically expand from pure carbon electrodes to carbon-based hybrid composite systems. Pure carbon materials (activated carbon, carbon nanotubes, graphene, carbon nanofibers and carbon aerogels) have high specific surface area, excellent conductivity, adjustable pore structure and outstanding cycling stability, so they can realize fast and reversible ion adsorption through the electric double layer capacitance mechanism, and thus become extremely important electrode components in high-power and long-life energy storage devices. However, it goes without saying that the energy density and specific capacitance of pure carbon materials still have obvious limitations (generally lower than 300 F/g). Therefore, the academic community has systematically and rationally developed carbon-based hybrid materials, that is, combining carbon materials with metal oxides, conductive polymers, metal sulfides, carbides, nitrides or biomass-derived carbon to introduce pseudocapacitance contribution and greatly improve the total energy storage capacity. Typical successful examples include carbon/metal oxide systems and carbon/conductive polymer systems, which not only retain the excellent cycling stability of the carbon substrate, but also increase the specific capacitance to more than 500 F/g, and some composite systems even reach 1800 F/g [13]. In addition, carbon-based materials also act as conductive skeletons and mechanical supports for improving the rate performance and structural integrity of metal sulfide or nitride electrodes; biomass-derived carbon provides precursor sources, which is green, low-cost and has natural porosity. Although carbon-based materials have made great progress in the field of supercapacitors, the main existing problems

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must be faced squarely: it is difficult to balance energy density and power density, wide-temperature performance is prone to degradation, and the cost of large-scale preparation is still high. Therefore, the reasonable development direction in the future is to design three-dimensional hierarchical pore structures, carry out heteroatom doping, construct multi-dimensional carbon networks, and reasonably composite with new two-dimensional materials such as MXene [14].

4.2. Conductive polymers

Conductive polymers (polyaniline, polypyrrole, polythiophene, etc.) store charges in supercapacitors mainly through fast and reversible Faradaic redox reactions, so they have typical pseudocapacitance characteristics. Thus, they naturally obtain higher specific capacitance and energy density than pure carbon materials, and also have outstanding advantages such as high intrinsic conductivity, low cost, simple synthesis process and excellent structure adjustability. Therefore, it is very conducive to accurately and controllably adjust their electrochemical properties through the doping/dedoping process [15, 16]. However, conductive polymers will undergo severe volume expansion and contraction during repeated charge and discharge, which easily leads to mechanical structure degradation and active material shedding. As a result, their cycling stability is poor, capacity decays fast, and their working potential window is relatively narrow, which objectively limits the overall energy density of the device. To overcome these shortcomings, researchers usually composite conductive polymers with highly stable carbon materials (such as activated carbon, carbon nanotubes, graphene, etc.), and use the carbon skeleton to provide mechanical support and efficient conductive network while significantly improving rate performance and cycle life, so as to maintain higher specific capacitance.

4.3. MXene and novel two-dimensional materials

MXene and its derivatives have become a new generation of two-dimensional materials that are very active and attract much attention in the field of supercapacitors due to their clear layered structure, excellent conductivity, abundant surface functional groups and outstanding mechanical flexibility. More importantly, such materials mainly store electricity through the surface Faradaic pseudocapacitance mechanism, so high energy density can be obtained without traditional electric double layer capacitance. Their two-dimensional layered structure itself is conducive to rapid ion intercalation and migration, and also naturally solves the volume expansion problem during charge and discharge. When MXene is composited with photosensitive materials or conductive polymers, the MXene-based electrode can make full use of photogenerated carriers to promote charge transfer and redox reactions under light, so the specific capacitance and rate performance are greatly improved [5]. At the same time, thanks to its inherent flexibility, MXene can be used to construct bendable and stretchable flexible supercapacitors that maintain stable electrochemical output directly on carbon cloth, nickel foam or self-supporting films under bending or twisting conditions [15]. Furthermore, reasonable bionic structure design (such as layered "brick-mortar" configuration) and favorable composite with other two-dimensional materials (SnS_2, graphene) can effectively inhibit sheet stacking, thereby improving mechanical strength and cycling stability at the same time [17]. Therefore, it can be concluded that MXene and novel two-dimensional materials provide an ideal electrode platform for developing high-energy, high-power, flexible and wearable supercapacitors by virtue of their conductivity, pseudocapacitance behavior, structure adjustability and mechanical adaptability.

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4.4. Transition metal oxides

Metal oxide materials used in supercapacitors mainly serve as pseudocapacitive electrodes, which store charges through fast and reversible Faradaic redox reactions on the electrode surface. Therefore, they can greatly improve specific capacitance and energy density, and well make up for the shortage of low energy storage capacity of traditional carbon materials relying on electric double layer. Among many metal oxides, tin dioxide (SnO_2) has become a research hotspot of supercapacitor electrode materials due to its excellent physical and chemical properties [12, 5]. Specifically, tin dioxide has prominent advantages such as high theoretical specific capacitance, sufficient raw material sources, low cost, environmental friendliness and non-toxicity, which is highly consistent with the development direction of green energy materials. More importantly, its wide working potential window is conducive to improving the energy density of the device, and its low electrochemical potential helps promote fast and efficient surface Faradaic redox reactions, so the pseudocapacitance contribution is remarkable [3]. The SnO_2 nanostructure is highly adjustable, and can be conveniently and controllably prepared into various morphologies such as nanoparticles, nanowires and nanosheets to increase electrochemical active sites. However, bulk tin dioxide has poor conductivity and is prone to large volume changes during cycling. Therefore, in practical applications, it is often composited with highly conductive carbon materials (such as carbon nanotubes, graphene, carbon nanofibers, etc.) to construct a stable conductive network and design a reasonable volume buffer structure. Furthermore, by systematically regulating the nanomorphology of metal oxides and optimizing the electrode-electrolyte interface, the exposure rate of active sites can be effectively improved, the ion diffusion path can be shortened, and the charge storage performance can be maximized.

5. Preparation methods of tin dioxide materials

As a typical transition metal oxide, tin dioxide has always received great attention in the field of supercapacitor electrode materials due to its high theoretical specific capacitance, abundant redox active sites and excellent thermal stability. However, it goes without saying that its poor conductivity and severe volume expansion during charge and discharge limit its practical application. Therefore, the academic community has developed various structural design and composite strategies to improve its electrochemical performance. At present, the preparation methods of tin dioxide-based materials can be naturally and hierarchically summarized into three directions: First, directly synthesize nanostructured SnO_2 (nanosheets, nanofibers) by solvothermal or hydrothermal method, and increase the specific surface area and shorten the ion diffusion path through morphology engineering [4, 15]. Second, composite SnO_2 with highly conductive carbon materials (reduced graphene oxide, carbon nanotubes) or conductive polymers (polypyrrole) to form binary or ternary composite materials with synergistic effects, so as to perfectly solve the two major problems of poor conductivity and unstable structure of single oxide [10, 15, 18]. Third, adopt electrostatic self-assembly or in-situ polymerization strategy to uniformly and controllably load SnO_2 nanoparticles on the carbon skeleton to construct a three-dimensional porous aerogel or core-shell heterostructure, which can not only buffer volume changes, but also facilitate charge transport [10, 4, 18]. From the perspective of specific processes, hydrothermal/solvothermal method is the most commonly used and mature method for preparing SnO_2 nanosheets and nanofibers due to its simple operation and easy morphology control [15, 18]. Electrospinning combined with calcination can produce hollow or porous SnO_2 nanofibers, which greatly increases the contact area between active materials and electrolyte [4]. The colloidal electrostatic self-assembly method more

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ingeniously uses the interaction between oppositely charged colloidal particles to uniformly composite SnO_2 with graphene, and then obtains lightweight porous aerogel through hydrothermal reduction and freeze-drying [18]. As a good complement, the chemical oxidative polymerization method can uniformly coat polypyrrole on the SnO_2 surface to form a clear core-shell structure, so the obtained electrode material has high cycling stability and excellent flexibility [15]. More importantly, various methods connect and complement each other from morphology regulation, component composite to structure integration, which systematically and solidly taps the potential of tin dioxide-based materials in high-performance supercapacitors. The specific preparation methods and key parameters are shown in Table 1.

Table 1. Types and preparation methods of SnO_2 materials

Material Type	Main Raw Materials	Preparation Method	References
SnO_2/carbon nanotube composite material	Tin source; carbon-based skeleton; auxiliary reagent	Wet chemical method	[10]
SnO_2-based nanoparticles and doping systems	Tin source; doping component (copper source); precipitant	Coprecipitation method	[19]
SnO_2/metal oxide heterostructure	Tin source; doping component (iron source); polymer template; solvent	Sol-gel electrospinning + calcination	[4]
SnO_2/carbon cloth and conductive polymer composite material	Tin source; carbon-based skeleton; complexing agent; conductive polymer monomer; oxidant	Solvothermal method + chemical oxidative polymerization	[15]
SnO_2/graphene aerogel composite material	Tin source; carbon-based skeleton (graphene precursor)	Electrostatic self-assembly + hydrothermal reduction + freeze-drying	[18]

6. Performance study of tin dioxide materials for supercapacitors

Since tin dioxide-based electrode materials show great diversity in specific capacitance, conductivity, cycling stability and other aspects, their application scenarios, advantages and disadvantages are also very clear.

Tin dioxide/carbon composite materials have outstanding performance in power density and cycle life. Loading tin dioxide nanoparticles on conductive skeletons such as vertically aligned carbon nanotubes or reduced graphene oxide helps construct a three-dimensional conductive network, resulting in extremely low charge transfer resistance and effectively buffered volume expansion during charge and discharge. Therefore, it is very suitable for supercapacitors requiring fast charge-discharge and large current output [10, 15]. However, such materials generally have the problem of limited tin dioxide loading, so the overall specific capacitance still has room for improvement, and the interfacial bonding strength between the carbon skeleton and tin dioxide still needs to be optimized [4]. In terms of energy density, tin dioxide/metal oxide heterostructures show unique advantages. Compared with single tin dioxide, rationally designed core-shell or hollow heterostructures can make full use of the synergistic energy storage of two-component redox reactions, thus obtaining specific capacitance and energy density far exceeding those of single materials [3, 20]. However, it must be objectively pointed out that the preparation process of such materials is relatively complex (electrospinning combined with high-temperature calcination), and the lattice matching and interfacial compatibility between different metal oxides need careful

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regulation, so their large-scale application is still limited to some extent [4, 21]. Tin dioxide/conductive polymer composite materials have excellent application prospects in the field of flexible energy storage: coating conductive polymers such as polypyrrole on the surface of tin dioxide nanosheets by chemical oxidative polymerization not only improves the overall conductivity of the electrode, but also endows the carbon cloth substrate with excellent flexibility and bendability, so it is very suitable for wearable electronic devices. However, there are indeed non-negligible problems: conductive polymers are prone to swelling and degradation during long-term cycling, resulting in capacitance attenuation, and the balance between polymer layer thickness and ion diffusion rate still needs to be optimized [15]. Finally, element-doped tin oxide materials have a clear and attractive application direction in multifunctionality: doping copper ions into the tin dioxide lattice by coprecipitation method can reasonably adjust its optical band gap and greatly enhance surface antibacterial activity, so it is an ideal choice for combining energy storage and antibacterial functions [19]. However, it must be recognized that simple element doping has limited direct improvement on electrochemical energy storage performance. To achieve high energy density, it still needs to be reasonably coupled with other composite means. In summary, different types of tin dioxide-based electrode materials have their own advantages and disadvantages. To promote their practical application in high-performance supercapacitors, future research should focus on solving key problems such as volume expansion inhibition, interfacial stability improvement, multi-component synergy and flexible device integration.

Table 2. Performance, advantages and disadvantages of SnO_2 materials

Material Type	Specific Capacitance	Cycling Stability	Energy Density	Power Density	References
SnO_2/carbon nanotube composite material	133-262 F g^{-1}	90–95% 1000–2000 cycles	0.18–0.51 kW/kg	20–31 Wh/kg	[18]
SnO_2-based nanoparticles and doping systems	100–974 F g^{-1} for doping systems (depending on dopant)	Poor for pure SnO_2, improved to 83–98% by doping	13–34 Wh kg^{-1}	0.23–0.95 kW kg^{-1}	[3] [21] [20]
SnO_2/metal oxide heterostructure	400–562 F g^{-1}	>90% retention rate (over 3000 cycles)	0.65–6.5 kW kg^{-1}	37–50 Wh kg^{-1}	[4]
SnO_2/carbon cloth and conductive polymer composite material	175–494 mF cm^{-2}	90.5% (10000 cycles)	0.12–0.46 mWh cm^{-3}	1.9–38 mW cm^{-3}	[15]
SnO_2/graphene aerogel composite material	310–541 F g^{-1}	93%–97.3% (10,000 cycles)	30–160 Wh kg^{-1}	8.3 kW kg^{-1}	[22] [23]

7. Conclusion

Tin dioxide (SnO_2) materials have received extensive and lasting attention in the field of supercapacitors due to their high theoretical capacitance, abundant redox active sites, low cost and environmental friendliness. However, it goes without saying that its poor intrinsic conductivity and severe volume expansion during charge and discharge both limit its practical application. Therefore, in recent years, the academic community has developed various clear optimization strategies with respective mechanism advantages: compositing with carbon materials to construct a conductive network, so as to improve charge transfer efficiency and enhance structural stability; introducing more active sites and buffering volume changes through heterostructure engineering; shortening ion diffusion paths by morphology regulation; endowing flexibility and excellent interfacial properties

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with conductive polymer coating; expanding multifunctionality such as antibacterial properties through element doping. The synergistic effect of various strategies has greatly improved the specific capacitance, cycle life and energy density of tin dioxide-based electrodes, which has shown excellent application prospects in flexible solid-state devices. However, it must be objectively seen that the actual capacitance of current tin dioxide-based materials is still far lower than the theoretical value, the volumetric energy density still has room for improvement, and the structural fatigue problem in long-term cycling has not been completely solved. Therefore, the reasonable direction in the future is to carry out multi-component synergistic design, defect engineering regulation, micro-nano structure integration and all-solid-state device development, and systematically promote the practical application of tin dioxide-based materials in high-performance energy storage systems.

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