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Review Article

Preparation and Recent Developments of Ti/SnO_2-Sb Electrodes

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Ti/SnO_2-Sb electrode, which is one of the dimensionally stable anode (DSA) electrodes, offers high specific conductivity, excellent electrocatalytic performance, and great chemical stability. For these reasons, Ti/SnO_2-Sb electrode has been extensively studied in the fields of wastewater treatment. This review covers essential research work about the advanced oxidation technology and related DSA electrodes. It gives an overview of preparation methods of SnO_2 electrodes, including sol-gel method, dip-coating method, electrodeposition method, chemical vapor deposition method, thermal decomposition method, magnetron sputtering method, and hydrothermal method. To extend service life and improve electrocatalytic efficiency, the review provides comprehensive details about the modification technologies of Ti/SnO_2-Sb electrode, such as doping modification, composite modification, and structural modification. In addition, the review discusses common problems in industrial applications of Ti/SnO_2-Sb electrode and highlights the promising outlook of Ti/SnO_2-Sb electrode.

1. Introduction

Nowadays, the treatment of wastewater has become a more challenging issue and a crucial factor for the sustainable development of modern industry, where essential measures are necessary to ensure clean environments. Most environmental pollutants can be successfully eliminated or converted to nontoxic materials by one or more processes, including biological method, physical method, and chemical methods. Regarding the different qualities of wastewater and the variety of organic pollutants in the wastewater, the wastewater treatment procedures would be complicated and different. Compared with biological method and physical method, the removal or alleviation of pollutants in water by a chemical method is easier to control and more effective, which has received an intensive interest for wastewater treatment.

Among the chemistry-based approaches, the advanced oxidation technique is the most common technique for removal of pollutants in wastewater. Advanced oxidation technology can decompose organic pollutants in wastewater into inorganics such as carbon dioxide and water through the hydroxyl radical OH^- generated in the oxidation process. Advanced oxidation technology has been widely used because of the following unique advantages: (1) the hydroxyl radical has a powerful oxidation ability; (2) the hydroxyl radical reaction is extensive and nonselective, so it can directly interact with various organic pollutants; (3) the advanced oxidation technology is relatively inexpensive, and the efficiency of organic pollutants degradation can be greatly improved by combining with other treatment technologies [1]. Among the advanced oxidation technologies as shown in Figure 1, electrocatalytic oxidation technology has attracted significant attention for wastewater treatment and is considered to be a potent tool for breaking up even the most resistant organic compounds.

Figure 2 presents the mechanism of electrocatalytic oxidation technology. In electrocatalytic oxidation

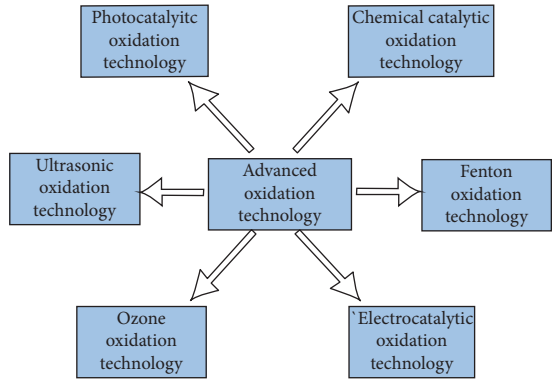

Figure 1: Classification of advanced oxidation technology.

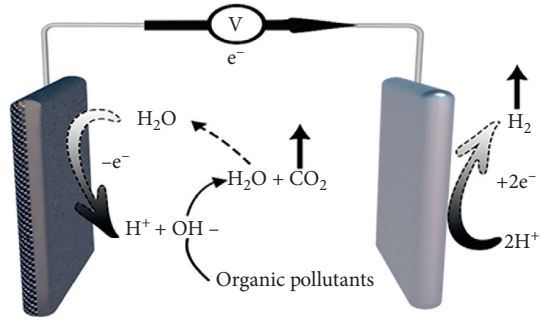

Figure 2: Schematic diagram of electrocatalytic oxidation technology.

technology, the electrocatalytic anode material plays a vital role. The electrocatalytic material will directly determine the oxidation process and the final oxidation products. Currently, the main electrocatalytic oxidation electrodes are noble metal electrodes, carbon electrodes, boron-doped diamond (BDD) electrodes, and metal oxide electrodes, of which metal oxide electrodes are also named as DSA electrodes. DSA electrodes have been widely used in various fields due to their advantages of stable size, long service life, high electrocatalytic performance, and low cost.

Table 1 compares four types of DSA electrodes, among which Ti/SnO_2-Sb electrode has a better degradation efficiency for organic pollutants, especially for some toxic organics that are difficult to be degraded, such as perfluorooctanoic acid, acid orange, *p*-chlorophenol, and other organic pollutions. Ti/SnO_2-Sb has a high oxygen evolution potential. The high oxygen evolution potential makes it possible to suppress the occurrence of oxygen evolution—the side reaction during oxidative degradation, thereby improving current efficiency and reducing reaction energy consumption. During the process of electrocatalysis, a large number of hydroxyl radicals can be generated, which significantly enhances the electrocatalytic efficiency of the Ti/SnO_2-Sb electrode. Besides, the doping of Bi, Sb, and other elements can greatly improve the conductivity and electrocatalytic ability of SnO_2 electrodes.

2. Synthesis of SnO_2-Sb Electrode

Currently, there are many preparation methods for Ti/SnO_2-Sb electrodes, including sol-gel method, electrodeposition method, thermal decomposition method, dip-coating method, hydrothermal method, sputtering method, ultrasonic atomization decomposition method, and self-assembly method. It is worth noting that different preparation methods will significantly affect the microstructure and performance of the Ti/SnO_2-Sb electrode. In this review, the fundamentals of each technology are briefly discussed in order to better understand their advantages and limitations for the applications in the removal and treatment of environmental pollutants in wastewater.

2.1. Sol-Gel Method. Sol-gel method mainly undergoes in few steps to generate the final metal oxide and those are hydrolysis, condensation, and drying process. The solution is obtained by dissolving tin-antimony inorganic salt in water or alcohol and then hydrolysis or alcoholysis produces sol particles. After the aggregation of sol particles, a gel is formed. By the pulling method or the coating method using the gel, the film on the titanium plate substrates can be prepared. The dried films are then heat-treated at different temperatures to obtain the electrodes. The tin-antimony oxide electrode prepared by the sol-gel method can cover the surface of the titanium substrate uniformly. However, it was found that when the concentration of the prepared sol is low,

Table 1: Comparison of different DSA electrodes.

DSA electrodes	Preparation method	Advantages and disadvantages	Application
Titanium-based manganese dioxide electrode	Thermal decomposition method, electrodeposition method	High-specific heat capacity, low cost, environmentally friendly	Electrocatalytic degradation of organic pollutants, anodizing methanol, electrowinning extraction of nonferrous metals, etc.
Titanium-based lead dioxide electrode	Electrodeposition method	Low cost, good corrosion resistance, good conductivity and simple preparation conditions, high brittleness, difficult machining, secondary pollution during application	Chemical power supply, chemical production, wastewater treatment, etc.
Titanium-based ruthenium-based, and iridium-based electrodes	Thermal decomposition method, electrodeposition method	Low chlorine evolution potential, good stability, short service life, low electrocatalytic ability	Chloralkali industry, wastewater treatment, electrowinning extraction of nonferrous metals, cathodic protection, etc.
Titanium-based antimony-doped tin dioxide electrode	Thermal decomposition method, electrodeposition method, vapor deposition method, etc.	Simple preparation conditions, low cost, high oxygen evolution potential, good electrochemical catalytic ability	Wastewater treatment, organic electrosynthesis, etc.

the spalling of the surface-active layer with poor binding strength would occur. Besides, the viscosity of the sol would also affect the performance of the electrode. At the same time, due to the inevitable volatilization of the solvent during the heat treatment process and the volumetric shrinkage caused by the temperature change, the formation of cracks in electrode would occur. As a result, the possible penetration of electrolyte into the electrodes with cracks and weak binding strength would cause corrosion of the titanium substrate and destroy the stability of the electrode material.

Sol-gel method is facile, economical, high-yield, and suitable for large-scale production of Ti/SnO_2-Sb electrode [2–5]. Duan et al. [2] fabricated an efficient Ti/Sb-SnO_2 electrode modified with nitrogen-doped graphene nanosheets (NGNSs) via a sol-gel method. Compared with a Ti/Sb-SnO_2 electrode, the NGNS-modified electrode possesses a smaller unite crystalline volume, smaller electrical resistivity, and lower charge-transfer resistance. The accelerated lifetime of Ti/Sb-SnO_2-NGNS electrode is prolonged significantly, which is 4.45 times as long as that of Ti/Sb-SnO_2 electrode. The results also indicated that introducing NGNS into the active coating can increase more reaction active sites to enhance the electrocatalytic efficiency. The electrochemical dye decolorization analysis demonstrates that Ti/Sb-SnO_2-NGNS presents efficient electrocatalytic performance for methylene blue and orange II decolorization. Zhong et al. [3] used $SnCl_4{\cdot}5H_2O$ and $SbCl_3$ as precursors to synthesize spherical SnO_2-Sb nanoparticles in ethanol solution by a sol-gel method and revealed the effects of calcination temperature and calcination time on the grain size, crystallinity, lattice parameters, and resistivity of the SnO_2-Sb nanoparticles. Zhou et al. [4] reported the preparation of Ti/SnO_2-Sb anode using sol-gel method and investigated the effects of applied current density, initial pH, and inorganic anions on the degradation kinetics.

2.2. Dip-Coating Method. Dip-coating technique process consists of several consecutive steps. First, the substrate should be immersed in the tin-antimony inorganic salt with a constant speed. Next, the substrate with thin layer of material is pulled up at a constant speed after remaining inside the solution for some time. Alternatively, the precursor solution can be spread by brushing over the pretreated substrate. The formed film then undergoes a series of heat treatment processes with the redox of the inorganic salt. As a result, a tin-antimony oxide film is obtained. The film properties and film thickness depend on several parameters, such as immersion time, withdrawal speed, number of dipping cycles, solution composition, concentration, and heating temperature [6, 7]. However, the preparation of thick films requires repeated steps of the dip-coating technique, which is time-consuming and labor-intensive. Besides, the limited solubility in the tin-antimony inorganic salt solution affects the application of the method.

Sun et al. [8] studied the electrochemical characteristics of Ti/SnO_2-Sb-Pd electrode prepared by the dip-coating method. Comparing the Ti/SnO_2-Sb electrode and Ti/SnO_2-Sb-Pt anode (as shown in Figure 3), it was found that the activity of tin oxide electrode doped with a small amount of Pd was greatly improved. These results suggested the greatly deteriorated electrochemical activity of both the deactivated undoped and Pd-doped Ti/SnO_2-Sb electrodes.

In addition to the traditional heat treatment in a muffle furnace, different heating sources have been employed for electrode performance improvements. Santos et al. [9] reported the optimization of Ti/SnO_2-Sb synthesis by CO_2 laser as the primary heating source for the first time. The laser-made Ti/SnO_2-Sb anode exhibited better electrocatalytic performance and improved the service life up to 5-fold as compared to the conventionally prepared anodes, envisaging its future applications in wastewater treatment.

2.3. Electrodeposition. Electrodeposition method, also known as electroplating, is an electric current driving deposition method. By controlling the composition of the electrolyte and the precipitation potential, the tin-antimony ions in the aqueous or nonaqueous solution are deposited on the titanium substrate by redox reactions. In general, the Ti/SnO_2-Sb

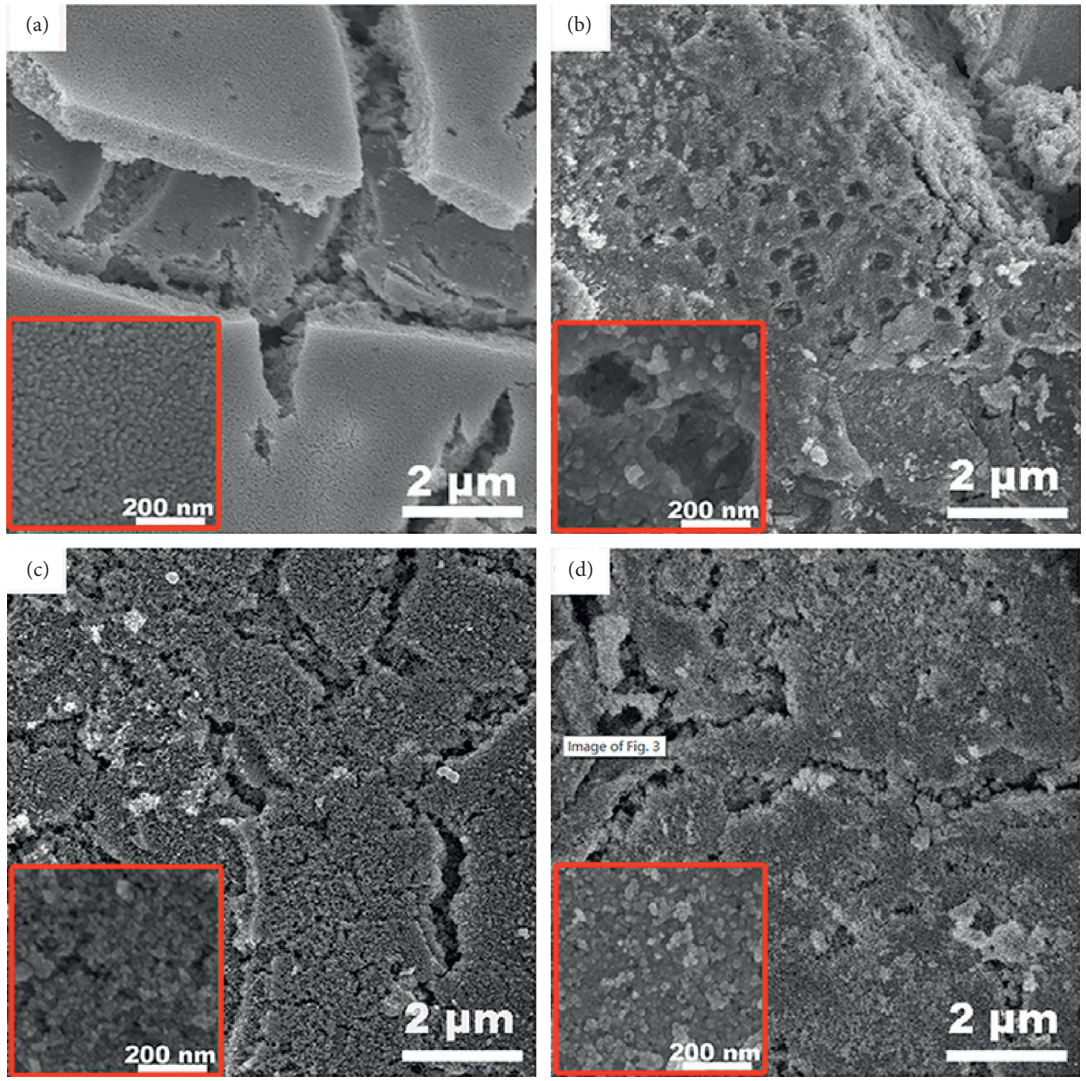

Figure 3: SEM images of (a) freshly prepared undoped Ti/SnO_2-Sb electrode, (b) deactivated undoped Ti/SnO_2-Sb electrode, (c) freshly prepared Pd-doped Ti/SnO_2-Sb electrode, and (d) deactivated Pd-doped Ti/SnO_2-Sb electrode; the insets show the enlargement of local area [8].

electrode prepared by electrodeposition has excellent binding strength and uniform thickness without cracks. Besides, the electrochemical deposition process is relatively simple and easy to control. Electrodeposition gives a precise control of the structure and performance of the coating.

The electrodeposition generally starts with the cathode electrodeposition step in electrolytic solutions, followed by the calcination step to form mixed metal oxides [10–12]. Duan et al. [12] fabricated different Ti/SnO_2-Sb electrodes using pulsed electrodeposition in a colloidal medium. Compared with an electrode prepared in an aqueous solution, these electrodes had a rougher and more compact surface with smaller grain sizes. The electrodes prepared in a colloidal medium displayed high oxygen evolution potentials, low charge exchange resistances, significantly enhanced lifetimes, and superior electrocatalytic activities. These electrodes also performed better for the adsorption and decolorization of methyl orange. Sun et al. [13] provided an eco-friendly, economical, and facile method for fabricating Ti/SnO_2-Sb electrode with high performance via electrodeposition using a deep eutectic solvent, which showed better electrochemical activity and longer service life than the conventional Ti/SnO_2-Sb electrode electrodeposited using aqueous solvent.

Furthermore, some recent research directly fabricated SnO_2-Sb anode by pulse reverse electrodeposition technique, without the traditional calcination procedures [14, 15]. Wu et al. [14] developed a unique SnO_2-Sb anode on TiO_2 nanotubes by a pulse reverse electrodeposition method, which presented a high oxygen evolution potential and a strong electrochemical oxidation ability. Compared with the SnO_2-Sb electrode prepared by traditional sol-gel method, the SnO_2 electrode prepared by the pulse electrodeposition method had a higher crystallinity and lower oxygen vacancy concentration. Besides, the SnO_2 electrode exhibited total organic carbon removal rates and mineralization efficiency, which are 2.6 times and 3.3 times of that for the traditional SnO_2 electrode, respectively.

2.4. Chemical Vapor Deposition. Chemical vapor deposition has been widely used in the development of various thin film materials with high purity and density. During chemical vapor deposition, the constituents of a vapor phase, often diluted with an inert carrier gas, are introduced into a reaction chamber and adsorbed on a heated substrate surface, which results in a solid coating via a chemical reaction. During the process, the substrate temperature is critical and can influence the occurrence of different reactions. Besides, several gradients or variable coatings can also be obtained by changing the gas phase composition.

Yao et al. [16] fabricated the Ti/SnO_2 anodes by chemical vapor deposition using a gas phase mixture of $SnCl_4$ and H_2O as a precursor at 550°C. It was found that the new electrodes had compact microstructure, high overpotential for oxygen evolution, and superior activity for pollutant oxidation. The surface quality of the electrode prepared by the chemical vapor deposition method could be well controlled, but the relatively expensive vapor precursor limits the chemical vapor deposition method in the application of Ti/SnO_2-Sb synthesis. To overcome the cost and throughput limitations of high vacuum techniques of the chemical vapor deposition method, Scott et al. [17] developed SnO_2-Sb thin films via mist chemical vapor deposition, a nonvacuum solution-based technique that involves the gas-assisted transport of ultrasonically generated aerosols from simple aqueous Sn and Sb precursors. The electrical properties of the resulting films were similar to those achieved using molecular beam epitaxy and other more sophisticated high vacuum techniques.

2.5. Self-Assembly Method. The self-assembly technique is the spontaneous association and organization of numerous individual units, such as molecules, nanomaterials, micrometers, or larger-scale components into coherent and well-defined structures without external intervention. Self-assembly involves diffusion followed by the association of molecules through noncovalent interactions, such as hydrogen bonding, ionic bonding, hydrophobic interactions, and van der Waals interactions. These interactions, although weak, are capable of forming the higher ordered structures.

Liu et al. [18] synthesized self-assembled 3D flower-like SnO_2 architectures. Fan et al. [19] fabricated SnO_2 mesoporous films through a sol-gel-based self-assembly process using Pluronic P-123 as a structure-directing agent. The self-assembled 3D flowerlike or mesoporous SnO_2 architectures possessed excellent electrical conductive performance and high specific surface area; thus, the SnO_2 electrodes would present remarkable electrochemical performance. Li et al. [20] prepared a SnO_2/TiO_2-NTs electrode with a 2D macropore structure using a self-assembly method with liquid crystal flexible membrane. It was found that this electrode showed good photocatalytic performance and electrocatalytic performance. Therefore, it exhibited high-efficiency photoelectric synergistic oxidation performance when degrading organic pollution. Fan et al. [21] fabricated a new SnO_2-Sb electrode with a high specific surface area and excellent electrocatalytic oxidation performance by evaporation-induced self-assembly. The results showed that the SnO_2-Sb electrode had nano-scale particles, high crystallinity, and ordered porous structures. The porous structures provided more in situ active sites and promoted the adsorption of organic pollutants. Therefore, the mesoporous SnO_2-Sb electrodes possessed better electrochemical activity than traditional tin-antimony oxide electrode and higher kinetic constant, and its initial mineralization current efficiency could also be greatly improved. Wang et al. [22] synthesized SnO_2-Sb nano-thin film electrodes by template-assisted self-assembly, which presented good electrical conductivity, and had extremely high crystallinity with an ordered mesoporous structure.

2.6. Ultrasonic Spray Pyrolysis Method. The liquid in the form of thin film when allowed to flow on a vibrating surface (frequency >20 kHz) breaks up into fine droplets. This phenomenon is known as ultrasonic atomization. During ultrasonic atomization, the metal salt solution is sprayed onto the pretreated titanium substrate. After dry and heat treatment, an oxide film on the substrate could be obtained. For the ultrasonic spray pyrolysis technology, the required operation equipment is relatively inexpensive, and the decomposition conditions are easy to control. Besides, the microstructure of the film surface can be optimized by the formation of ultrafine liquid droplets via ultrasonic atomization, thereby improving the performance of the film.

Until now, many literatures have reported SnO_2 film prepared via ultrasonic spray pyrolysis [23–27]. Chen et al. [25] fabricated Ti/SnO_2-Sb electrode with different nickel concentrations by a spray pyrolysis technique. The nickel and antimony co-doped Ti/SnO_2 anodes showed different structures and onset potential for oxygen evolution. SnO_2 thin films grew in preferential orientation along the (101) plane as the nickel concentration increases. The onset potential of oxygen increased due to the introduction of nickel doping. These results are significant in developing advanced SnO_2-based electrodes with a high oxidation potential to treat a broad kind of organic pollutant. Sánchez-García et al. [26] used tin dichloride ethanol solution as the raw material to quickly prepare tin dioxide film by an ultrasonic spray pyrolysis method. The results showed that the film prepared by this method was uniform and dense, whose surface could be completely oxidized. Besides, the film thickness and surface uniformity could also be well controlled. Yao [27] deposited the Sb doping SnO_2 film on the titanium substrate by ultrasonic spray pyrolysis at 600°C. The results showed that the crystal size of SnO_2-Sb increased with the increasing doping content. When the doping level of Sb was about 3%, the Ti/SnO_2-Sb electrode possessed the lowest resistivity and the best electrocatalytic activity.

2.7. Hydrothermal Method. Hydrothermal synthesis refers to heterogeneous reactions in a sealed and heated solution above ambient temperature and pressure. Hydrothermal synthesis facilitates the solubility and chemical reactions of the hardly soluble metal salt precursors in hot water (or organic solvent) under high pressure, followed by

supersaturation and crystallization. Generally, the pretreated titanium substrate is usually put into an aqueous solution under high-temperature and high-pressure conditions, and a tin-antimony oxide film is directly formed on the titanium substrate after the hydrothermal reaction.

Xu et al. [28] embedded SnO_2-Sb into TiO_2 nanotubes by hydrothermal synthesis to form a three-dimensional electrode (TiO_2-NTs/Sb-SnO_2). Compared with the Ti/SnO_2-Sb_2O_5 electrode prepared by conventional methods, the TiO_2-NTs/SnO_2-Sb electrode prepared exhibited a higher density and longer service life. An et al. [29] developed SnO_2-Sb film coated with sol layer by a hydrothermal method and spin coating method with the formation of solution-based transparent conductive oxides. The results showed that the SnO_2-Sb film with five sol layers had excellent resistivity and light transmittance. Besides, the increase in the density and thickness of the sol coating layer could improve the performance of the film. However, the increase in carrier concentration led to a reduction of mobility. Xu and Lian [30] designed a novel SnO_2-Sb nanorod anode with Ti sheet as a substrate (Ti/SnO_2-Sb-NRs) through the hydrothermal process. The results indicated that Ti/SnO_2-Sb-NRs anode possessed SnO_2-Sb-NRs which were about 70 nm in width, as well as a better wettability, a lower charge-transfer resistance, a larger current at constant potential, and a longer lifetime than the conventional Ti/SnO_2-Sb anode prepared by pulse electrodeposition. Yang et al. [31] successfully fabricated Ti/SnO_2-Sb electrode with a nano-scaled sphere-stacking structure was using a solvothermal synthesis approach, which possessed superior electrochemical properties to an electrode prepared using dip-coating methods.

2.8. Magnetron Sputtering Method. Magnetron sputtering is a versatile method to create very dense films with good adhesion. Magnetron sputtering is a plasma-based coating method that generates a magnetically confined plasma near the surface of a target. Then, positively charged energetic ions from the plasma collide with the negatively charged target material, and atoms from the target are ejected or sputtered, which then deposit on a substrate. The magnetron sputtering method has been widely employed in the preparation of various alloys and compounds. Montero et al. [32] deposited SnO_2-Sb thin films from metal targets onto glass substrates by direct current magnetron sputtering at room temperature. The results showed that the deposited films exhibited amorphous or nanocrystalline structures. Besides, the optical and electrical properties of the film mainly depended on the deposition parameters. Yan et al. [33] obtained Ti/SnO_2-Sb anodes with a microrod structure prepared by magnetron sputtering. The SnO_2 coating on the above Ti substrate by magnetron sputtering with postannealing was comprised of microrod and had longer service time than the traditional Ti/SnO_2 anode.

3. Research Progress of Ti/SnO_2-Sb Electrode

Ti/SnO_2-Sb electrode has various advantages, but it is worth noting that the relatively short service life and unstable electrocatalytic efficiency limit the application of titanium-based tin-antimony oxide electrode. In recent years, many studies aim to improve both the service life and electrocatalytic performance of tin-antimony oxide electrodes. For instance, it was found that increasing the bonding strength between the surface-active layer and the substrate could prevent the corrosion failure inside the electrodes. Therefore, it is of great importance to develop appropriate modification techniques for electrodes. Currently, the modification technology includes doping modification, structural modification, and composite modification.

3.1. Doping Modification. Doping different elements into Ti/SnO_2-Sb electrodes can effectively promote electrocatalytic oxidation performance, electrode life, and surface morphology of electrodes. The doping elements for the modification of Ti/SnO_2-Sb electrode can be included as follows:

(1) Doping of rare Earth metals, such as Nd, Eu, and Dy [34–36]. Zhu et al. [35] investigated Ti/SnO_2-Sb electrodes doped with different rare Earth elements (Ce, Dy, La, and Eu), which were prepared by the thermal decomposition method at 550°C (as shown in Figure 4). The results demonstrated that the electrocatalytic degradation performances of Ti/SnO_2-Sb electrodes were improved to different levels by doping different rare Earth ions. According to the improvement effects, the electrodes doped with four rare Earth elements rank in the order: Ti/SnO_2-Sb-La electrode > Ti/SnO_2-Sb-Eu electrode > Ti/SnO_2-Sb-Dy electrode > Ti/SnO_2-Sb-Ce electrode. It worth noting that Ti/SnO_2-Sb-La electrode has a higher oxygen evolution potential, better electrocatalytic activity, and longer electrode life. Li et al. [36] prepared a Pd-modified Ti/SnO_2-Sb anode via thermal decomposition. It was found that Pd doping could effectively change the surface morphology and lattice parameters of the metal oxide electrode. Pd can also facilitate the entry of more Sb into SnO_2 crystals and promote the reduction of Sb. At the same time, with Pd doping, the lifetime of the electrode was increased more than 40 times, demonstrating the good potential for application in industry.

(2) Doping of precious metals, such as Pt, Ru, and Ir [37, 38]. Berenguer et al. [38] compared Pt- and Ru-doped Ti/SnO_2-Sb anodes to conventional Ti/RuO_2 and Ti/Co_3O_4 anodes. It was found that the Ti/SnO_2-Pt anode exhibits the best electroactivity, fastest kinetics, and highest current efficiency among the studied anodes but poor electrochemical stability. The introduction of small amounts of Ru (3.25–9.75 at.%) brings about a slight loss of the electrocatalytic performance, but it causes a remarkable increase in the stability of the electrode.

(3) Doping with iron group metals and other metals, such as Fe, Co, and Ni [37, 39]. Yang et al. [37] studied six elements of Fe, Ni, Co, Ru, Ce, and Pd doped into the optimized SnO_2-Sb electrode, which aimed to reveal the most effective dopants and the

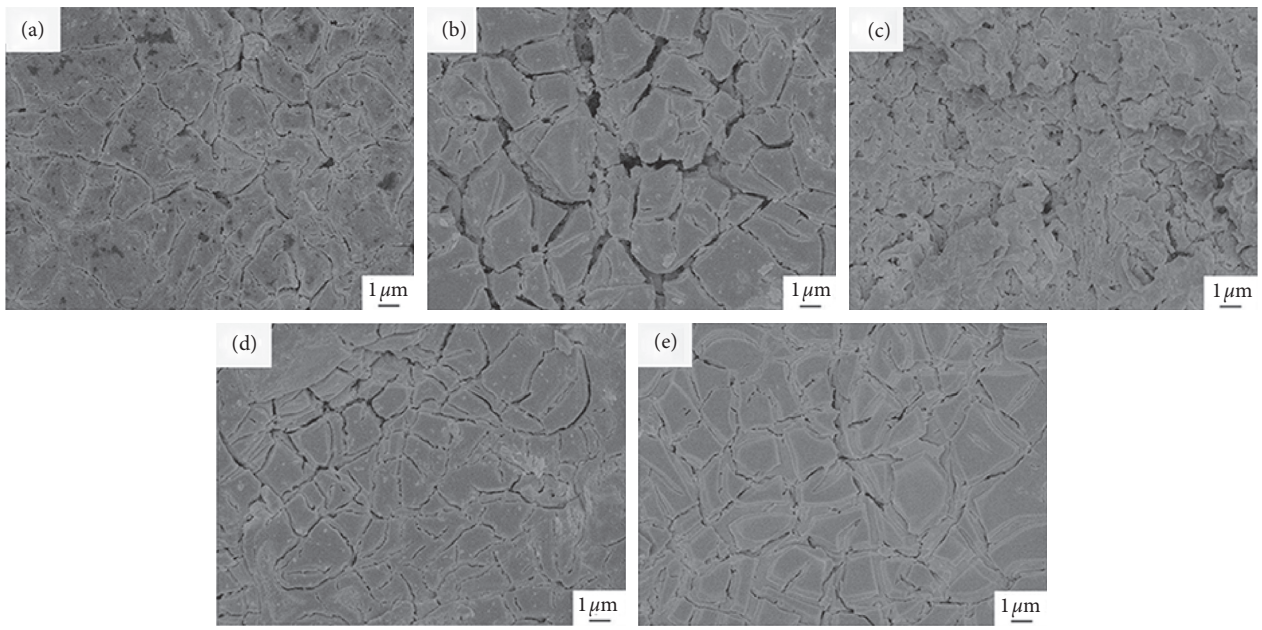

Figure 4: SEM images of Ti/SnO_2-Sb electrodes undoped and doped with different rare earths: (a) undoped; (b) Ce; (c) Dy; (d) La; (e) Eu [35].

best combinations to improve the electrocatalytic activity of SnO_2 electrode to pollutants. The results showed that the optimal doping amount of Sb in the Sb-SnO_2 electrode was 5–10%. The doping of Ni and Fe enhanced the electrocatalytic activity of the electrode. Notably, the doping of Ni significantly increased the degradation efficiency of phenol and the removal rates of total organic carbon. However, the doping of precious metals, such as Pd, Ru, and Co, led to a significant reduction in the oxygen evolution potential of the electrode, resulting in low efficiency of the electrode when degrading pollutants. Liang et al. [39] investigated the effect of Mo with different molar ratios on the characterization of Ti/SnO_2-Sb-Mo electrodes prepared by the sol-gel method (as shown in Figure 5). The results showed that the electrode at the Mo content of 1 at.% provided optimal catalytic activity for phenol degradation and the longest lifetime. The Ti/SnO_2-Sb-Mo electrode coating with 7 at.% Mo presented the highest oxygen evolution overpotential, indicating the diverse effects for different Mo molar ratio doping.

3.2. Compound Modification. Compound modification methods mainly include metal-metal carbide compound, metal nitride compound, nanocarbon material compound, and organic polymer compound [10, 40–44]. Since the electrode prepared by the electrodeposition method has good binding strength and the morphology and thickness of the deposited layer can be controlled by adjusting the parameters, the nanomaterial is mainly doped into the coating by means of electrodeposition. Duan et al. [10] prepared Ti/SnO_2-Sb electrode modified with TiN nanoparticles by a pulse electrodeposition method. The prepared Ti/Sb-SnO_2-TiN electrode had a dense film structure and a small unit cell volume (as shown in Figure 6). Compared to traditional Ti/SnO_2-Sb electrode, Ti/Sb-SnO_2-TiN electrode had higher decolorization efficiency and kinetic rate constant (as shown in Figure 7). Zhang et al. [41] prepared a new type of CNT-modified Ti/SnO_2-Sb electrode by pulse electrodeposition. Compared with the electrode without CNT modification, the electrode modified with CNT had a larger specific surface area and smaller microcrystalline particles. Besides, the electrode modified with CNT had higher oxygen evolution potential, kinetic rate constant, chemical oxygen demand, total organic carbon removal rate, and mineralization current efficiency. Pahlevani et al. [42] synthesized Ti/SnO_2-Sb electrode modified by graphene oxide using dip-coating technique and thermochemical decomposition, with the formation of uniform SnO_2 nanoparticles ranging 15–26 nm and presence of reduced graphene oxide. The graphene oxide modified anode led to a higher oxygen evolution overpotential and less energy being consumed for competitive reactions, resulting in high electrocatalytic activity for the fabricated anode.

Li et al. [43] investigated the electrocatalytic degradation of aniline by Ti/SnO_2-Sb, Ti/Sb-SnO_2/Pb_3O_4, and Ti/Sb-SnO_2/PbO_2 anodes in different electrolytes. In 5 wt‰ Na_2SO_4 electrolyte, aniline could be degraded relatively faster on the Ti/Sb-SnO_2 anode. While in 5 wt‰ NaCl electrolyte, Ti/Sb-SnO_2/Pb_3O_4 would be more beneficial for aniline degradation. Besides, the Ti/Sb-SnO_2/PbO_2 could exhibit a better capability to enhance biodegradability in both electrolytes. Jin et al. [44] deposited SnO_2-Sb and α-PbO_2 onto the surface of a titanium substrate, followed by the fabrication of β-PbO_2 doped with Fe element and

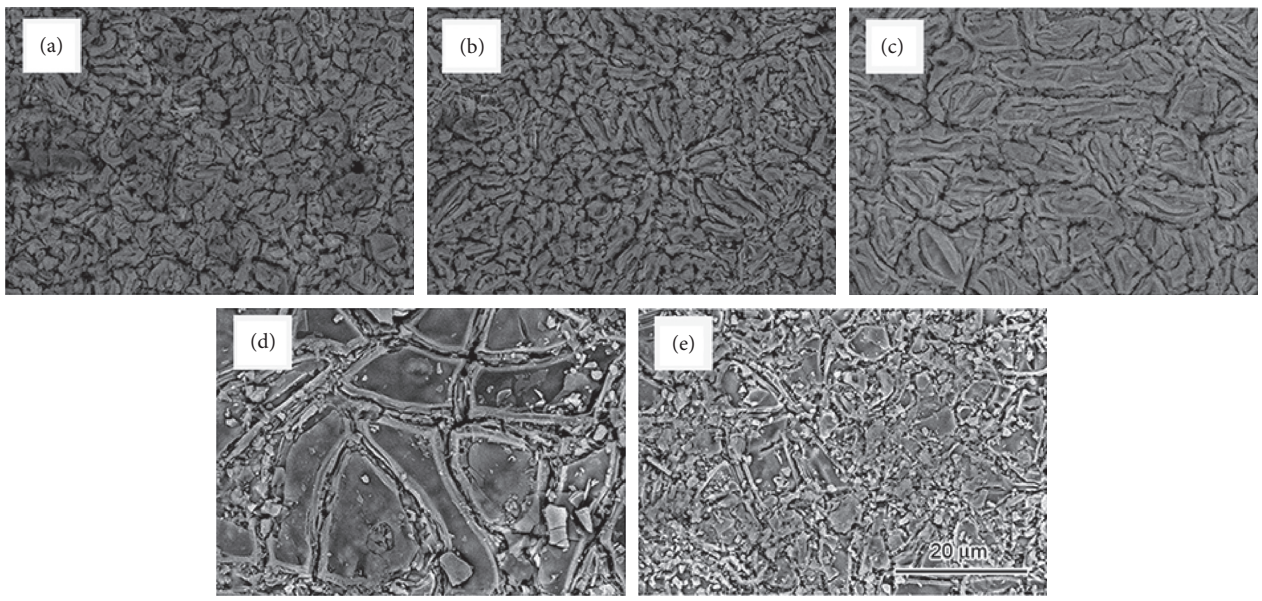

Figure 5: SEM images of Ti/SnO_2-Sb-Mo electrodes of different Mo doping molar ratios: (a) 0.0 at.%; (b) 1.0 at.%; (c) 3.0 at.%; (d) 5.0 at.%; (e) 7.0 at.% [39].

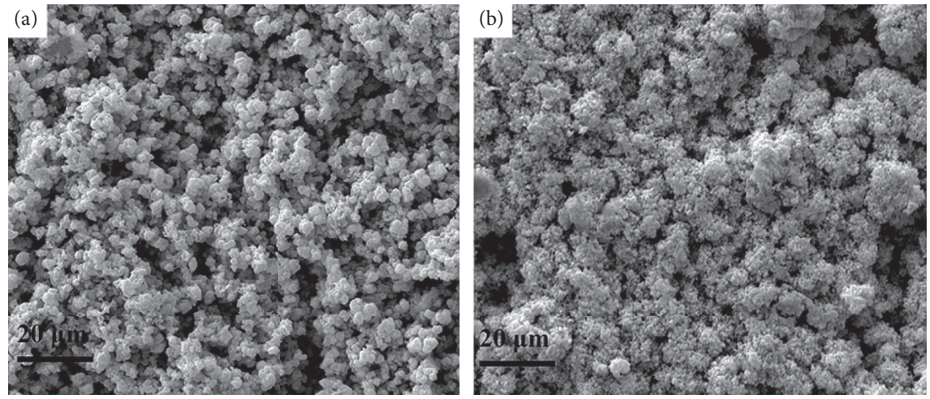

Figure 6: SEM images of (a) Ti/Sb-SnO_2 electrode and (b) Ti/Sb-SnO_2-TiN electrode [10].

polytetrafluoroethylene (PTFE) thereon. The optimized superhydrophobic electrode modified with PTFE exhibited lower charge-transfer resistance and good oxidative ability towards organics. Due to the collaborative contribution of α-PbO_2 and PTFE, the stability of the Ti/SnO_2-Sb/α-PbO_2/Fe-β-PbO_2-PTFE electrode was found to be significantly improved. Such superhydrophobic Ti/SnO_2-Sb/a-PbO_2/Fe-b-PbO_2-PTFE electrodes could effectively degrade organic pollutants under low voltages, which is of great significance for reducing energy consumption.

3.3. Structural Modification. The structural modification methods for Ti/Sb-SnO_2 electrodes mainly include intermediate layer structural modification and active layer structural modification. The intermediate layer structure modification method consists of the addition of the intermediate layer and the modification of the titanium-based structure. The addition of the intermediate layer can increase the binding strength between the active layer and the substrate, prevent the substrate from being passivated after corrosion, extend the service life, and improve the stability of the electrode during electrocatalytic degradation. Santos et al. [45] used 4 or 16 repetitive alternating Sn and Sb electrodeposition methods on the titanium foil with Pt as the intermediate layer, and finally, Ti/Pt/SnO_2-Sb_2O_4 electrode was obtained after high-temperature calcination. Compared with traditional Ti/SnO_2-Sb_2O_4 electrode, SnO_2-Sb_2O_4 electrode with Pt as the middle layer had a higher service life, electrochemical stability, and mechanical stability. Furthermore, it was found that the Ti/Pt/SnO_2-Sb_2O_4 electrode had high removal rates of chemical oxygen demand to chloroauric acid, diclofenac, and ibuprofen. Bi et al. [46] prepared Ti/SnO_2-Sb-La electrodes by the sol-gel method with two kinds of Ce-Mn and Fe-Mn composite intermediate layers, a Mn intermediate layer, or no intermediate layer (as shown in Figure 8). The results indicated the lifetime of the electrode with the intermediate layer

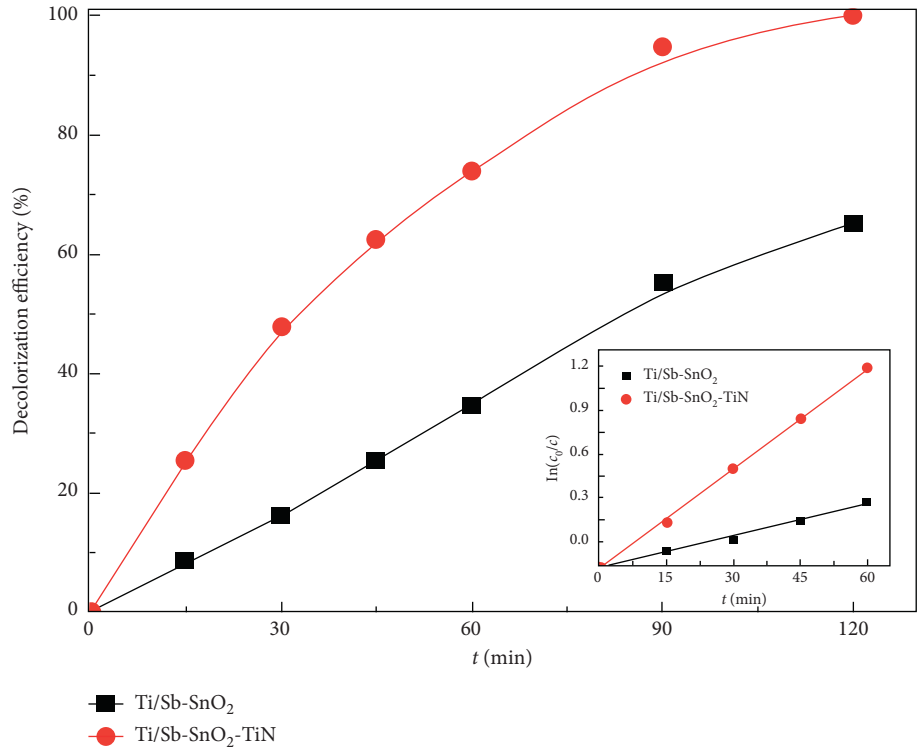

Figure 7: Variations in decolorization efficiency with time. The inset is the pseudo-first-order of methylene blue decolorization on Ti/Sb-SnO_2 and Ti/Sb-SnO_2-TiN electrodes [10].

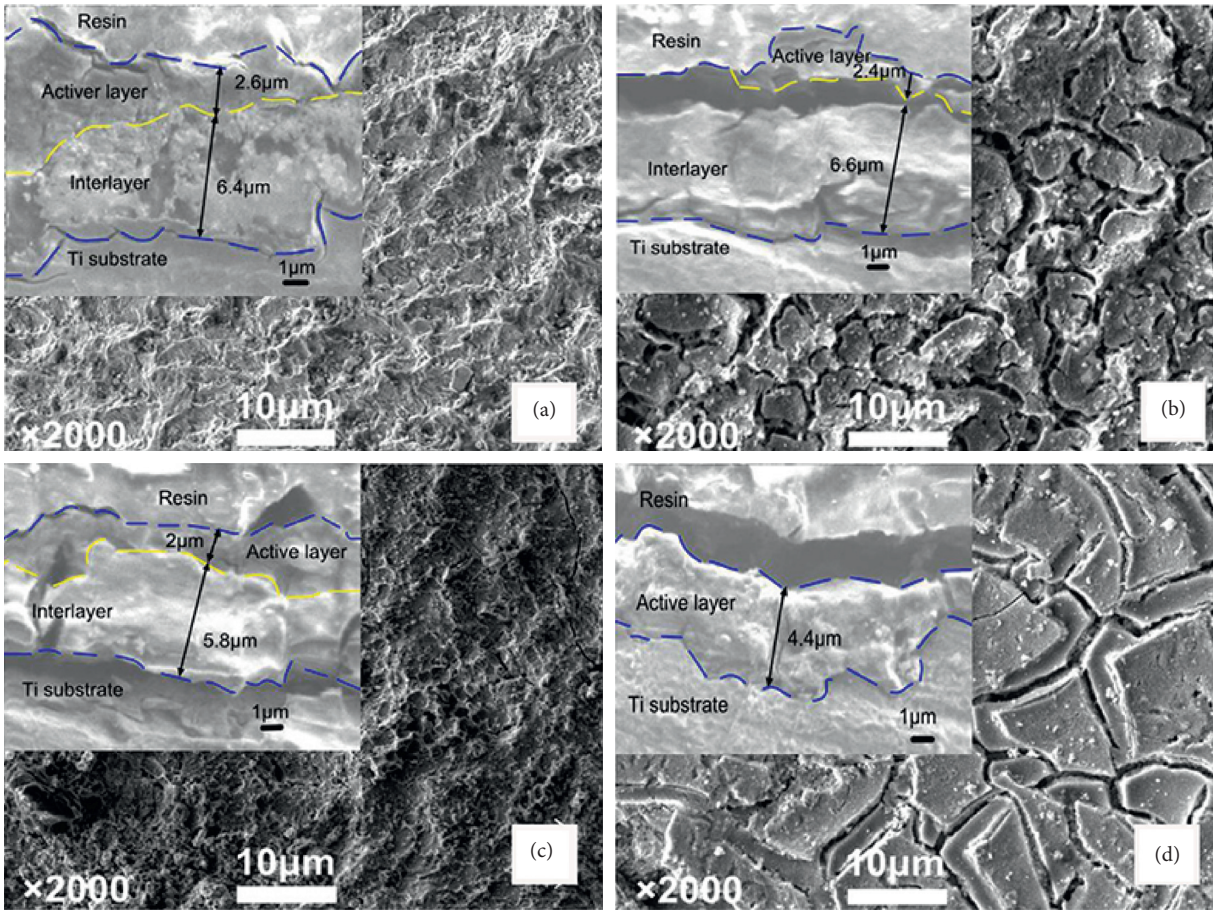

Figure 8: SEM images of different electrodes: (a) Ti/Ce-Mn/SnO_2-Sb-La; (b) Ti/Fe-Mn/SnO_2-Sb-La; (c) Ti/. Mn/SnO_2-Sb-La; (d) Ti/SnO_2-Sb-La [46].

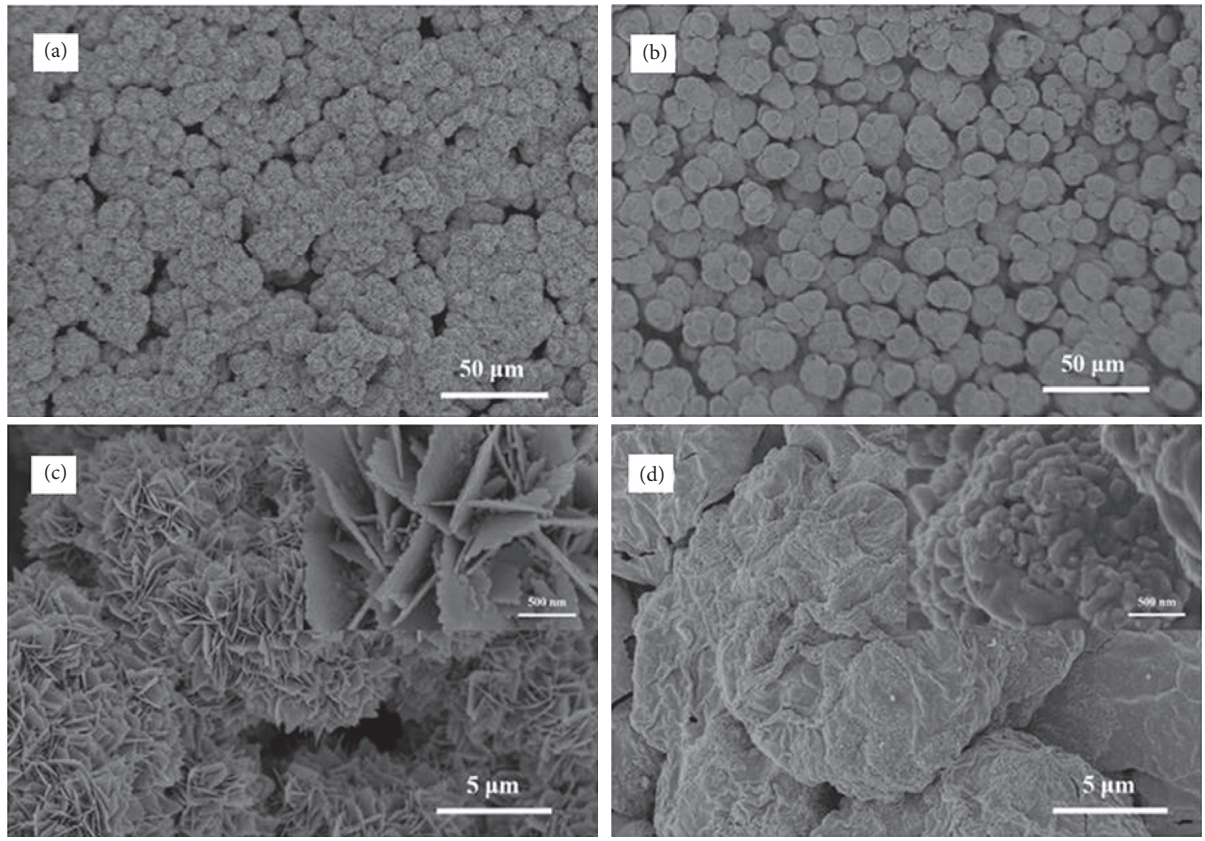

Figure 9: SEM images of the surface morphologies of (a, c) Ti/SnO_2-Sb-HFs electrode and (b, d) Ti/SnO_2-Sb electrode [50].

increased. Especially, the electrochemical performance of the electrode can be significantly improved by adding a Ce-Mn composite intermediate layer with high oxygen evolution potential. Moreover, such addition also enhances the capacity of the electrode for electrocatalytic oxidation degradation of phenol to some degrees, showing high removal rate of phenol and the maximum removal rate of chemical oxygen demand.

The primary way to modify the titanium-based structure is to obtain a highly ordered array of titanium dioxide nanotubes (TiO_2-NTs) by anodizing the titanium plate surface. TiO_2-NTs as a tubular template can increase the contact area between the surface-active layer and the matrix and the specific surface area of the active layer to provide more active sites for the catalytic layer. Wang et al. [47] prepared Sb-doped SnO_2 coatings on different TiO_2-NTs substrates by electrodeposition with different anodic oxidation times and voltages of the titanium substrate. The results indicated that TiO_2-NTs could significantly improve the electrocatalytic efficiency of the electrodes. Meanwhile, the pore size and length of TiO_2-NTs played a crucial role in the degradation efficiency of TiO_2-NTs/SnO_2-Sb electrode. When the pore size of TiO_2-NTs was 85 nm and the length was 5 μm, the electrode had the best degradation efficiency on target pollutants.

The active layer structure modification method is actually to prepare Ti/SnO_2-Sb electrode with special active layer structure to improve the electrocatalytic ability of the electrode. Asim et al. [48] developed a simple template removal method to prepare two different types of layered porous SnO_2-Sb electrodes (honeycomb Ti/Sb-SnO_2 electrodes and mesh Ti/Sb-SnO_2 electrodes). In terms of microstructures, both the two types of electrodes presented extremely high porosity and were arranged layer by layer in multiple directions. The high specific surface area results in more electrochemically active sites on the electrode surface. Compared with traditional SnO_2-Sb electrodes, these two types of porous electrodes exhibited better electrochemical activity and higher reaction rates for electrocatalytically degrading organic pollutants. Besides, the presence of porous channels could significantly promote adsorption and diffusion of pollutant molecules. Moir et al. [49] prepared a 3D SnO_2-Sb nanoelectrode with a disordered macropore structure by the template method. The results showed that the 3D macropore structure improved the specific surface area of the active layer significantly, so the electrode possessed an efficient electrocatalytic performance. Recently, Wang et al. [50] achieved a novel Ti/SnO_2-Sb electrode with a 3D hierarchical flower-like structure (HFs), which was firstly prepared using a hydrothermal method. Compared the traditional Ti/SnO_2-Sb electrode, this novel Ti/SnO_2-Sb-HFs electrode has a larger electrochemical active surface area, a lower charge-transfer resistance, and a higher oxygen evolution potential (as shown in Figure 9). Especially, the stability of the electrode is improved most obviously.

4. Conclusion

Electrocatalytic oxidation technology has attracted much interest in the field of water treatment technology. Based on this technology, it is of great importance to choose proper electrode materials. Among them, Ti/Sb-SnO_2 electrode has been widely used in various fields due to its good conductivity and stability. However, the shortages including low catalytic performance and poor stability limited the industrial applications of the electrode. To expand the application of the Ti/Sb-SnO_2 electrode, the following problems are urgent to be solved:

(1) The traditional Ti/Sb-SnO_2 electrode has relatively poor stability and short service life. Although modification technology can improve its service life to a certain extent, the service life is still shorter than other DSA electrodes.

(2) The electrode synthesized by electrodeposition has good binding strength and long service life, but generally with low catalytic efficiency, which affects the efficiency of degrading organic pollutants in wastewater. In addition, when the electrode is prepared by the electrodeposition with the aqueous solution system, the stress in coating caused by hydrogen inclusions would produce blisters or cracks.

(3) Most of the modification techniques for Ti/SnO_2-Sb electrodes have their own limitations. Therefore, developing Ti/Sb-SnO_2 electrodes with high catalytic efficiency and good binding strength still needs to be investigated.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Authors' Contributions

Fangcheng Cao and Jingyi Tan contributed equally to this paper.

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