

# Research Progress on Electrochemical Oxidation of Aqueous Organic Pollutants Through Ti/SnO<sub>2</sub>-Sb Anodes

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## Abstract

The proliferation of persistent and toxic organic contaminants necessitates the development of efficient advanced oxidation processes (AOPs). Electrocatalytic oxidation, particularly using dimensionally stable anodes (DSA) like Ti/SnO<sub>2</sub>-Sb, presents a promising solution due to its high efficiency and environmental compatibility. The review details the primary preparation methods for Ti/SnO<sub>2</sub>-Sb electrodes, including thermal decomposition, electrodeposition, chemical vapor deposition, hydrothermal synthesis, and the widely used sol-gel method, analyzing their respective advantages and limitations. To address the electrode's key drawbacks of limited service life and moderate catalytic activity, various modification strategies are discussed, categorized into doping modification (using rare earth or precious metals) and structural modification (involving interlayers or 3D substrate architectures). These modifications significantly enhance electrode stability, oxygen evolution potential, and pollutant degradation efficiency. Furthermore, the successful application of modified Ti/SnO<sub>2</sub>-Sb electrodes in treating diverse wastewaters, such as dyeing, medical, and petrochemical effluents, is highlighted, demonstrating high removal rates for target pollutants and chemical oxygen demand (COD). This review underscores the potential of optimized Ti/SnO<sub>2</sub>-Sb electrodes as effective anodes for electrocatalytic water treatment and identifies ongoing research directions for improving their performance and durability.

## Keywords

Ti/SnO<sub>2</sub>-Sb, Electrocatalytic Oxidation, Electrode Modification, Organic Pollutants, Wastewater Treatment.

## 1. Introduction

Currently, with the expansion of urban areas and rapid industrialization, the presence of organic pollutants in water—such as dyes, antibiotics, and pesticides—poses a serious threat to both natural ecosystems and human health and safety. These pollutants exhibit high toxicity, persistent nature, and bioaccumulation properties [1, 2]. Traditional water treatment technologies, such as adsorption, biodegradation, sedimentation, extraction, membrane separation, coagulation, and ion exchange, struggle to effectively remove these pollutants [3-5]. Therefore, the development of novel, highly efficient treatment technologies is urgently needed. Advanced oxidation processes (AOPs) are capable of thoroughly organic pollutants degradation through oxidation into harmless CO<sub>2</sub> and H<sub>2</sub>O. These methods offer high treatment efficiency, which are not limited by refractory substances and have minimal impact on water quality. By utilizing strong oxidants such as O<sub>3</sub>, H<sub>2</sub>O<sub>2</sub>, •OH, etc., AOPs can oxidize various organic pollutants in a non-selective manner, without generating new contaminants [6]. Among them, electrocatalytic oxidation technology holds promising prospects due to its excellent environmental compatibility, ease operation, and outstanding degradation performance [7].

Based on the distinct pathways through which oxidation reactions occur, electrocatalytic oxidation methods can be categorized into direct oxidation and indirect oxidation. Pollutants can be oxidized via direct electron transfer at the electrode surface in the former pathway[8]. In contrast, indirect oxidation involves the initial generation of reactive oxygen species (ROS) at the anode surface, which subsequently participate in the pollutants oxidation reactions[9].

In the process of wastewater treatment, the selection of anode materials directly impacts both the degradation efficiency of organic pollutants and their service life, working as one of the key factors in electrocatalytic oxidation technology. Currently, electrocatalytic anodes are primarily categorized into noble metal electrodes, carbon-based electrodes, BDD electrodes, and metal oxide electrodes. Among these, metal oxide electrodes mainly include  $\text{PbO}_2$  electrodes and dimensionally stable anodes (DSA). DSA electrodes are widely utilized across various fields owing to their advantages such as dimensional stability, electrocatalytic stability, high degradation performance, and relatively low cost[10].

Currently, DSA electrodes can be categorized into types such as titanium-based manganese dioxide electrodes, titanium-based lead dioxide electrodes, titanium-based ruthenium-series electrodes, titanium-based iridium-series electrodes, and titanium-based antimony-doped tin oxide electrodes[11]. Among them,  $\text{Ti/SnO}_2\text{-Sb}$  electrodes have been widely studied due to its advantages of easy preparation, low cost, and high oxygen evolution potential. However, the relatively short lifespan and moderate catalytic activity limit their large-scale applications[12]. Therefore, researchers often adopt methods such as doping modification during the preparation of  $\text{Ti/SnO}_2\text{-Sb}$  electrodes to improve the service life of  $\text{SnO}_2\text{-Sb}$  electrodes [10].

This article reviews the latest research progress in preparation methods, modification, applications in organic pollutant degradation and wastewaters treatment of  $\text{Ti/SnO}_2\text{-Sb}$  electrodes.

## 2. Preparation Method of $\text{Ti/SnO}_2\text{-Sb}$ Electrode

At present, the main preparation methods of  $\text{Ti/SnO}_2\text{-Sb}$  electrodes are thermal decomposition, electrochemical deposition, chemical vapor deposition (CVD), hydrothermal and sol-gel methods[13-17].

### 2.1. Thermal Decomposition Method

Thermal decomposition is a traditional process commonly used for large-scale production of  $\text{Ti/SnO}_2\text{-Sb}$  electrodes. This method typically involves dissolving various metal salts in aqueous or organic solvents, uniformly coating the resulting solution onto the electrode substrate surface, followed by drying and high-temperature decomposition treatments. The process culminates in high-temperature sintering to form metal oxide-coated electrodes through the repeated cycles of coating, drying, and decomposition. This method features straightforward operation, mature technology, and low requirements for equipment and tools, resulting in relatively low equipment costs. However, it demands strict control over thermal treatment temperatures, which directly impact the electrocatalytic performance of the coated electrodes. The excessively high temperatures cause volatile metal elements to evaporate during the conversion of salts to oxides, reducing metal content; simultaneously. Besides, the elevated temperatures accelerate the growth of metal oxide grains, resulting in excessively large grain sizes. This weakens the bond between the catalysts coating and the substrate, causing the coating to peel off easily and thereby reducing the life of the electrode. Conversely, the metal salts cannot fully oxidize if the heat treatment temperature is insufficient, similarly affecting the electrochemical performance of the electrode in practical applications and making it difficult to achieve the desired results. Man et al. prepared a Ce and Sb co-doped  $\text{SnO}_2$  nanoflower electrode via the hydrothermal method and thermal decomposition technology. It

was found that when treating simulated wastewater containing 20 mg/L of methylene blue (MB), this electrode achieved nearly 100% decolorization efficiency and 91.45% mineralization efficiency within 90 min[16].

## 2.2. Electrodeposition

Electrodeposition, also known as electroplating, is a technique for Ti/SnO<sub>2</sub>-Sb electrodes preparation. This method involves immersing the electrode substrate in an electrolyte containing the desired metal ions. Under the effect of an applied electric field, the metal ions undergo oxidation or reduction reactions through electron transfer, thereby depositing onto the substrate surface as metal or metal oxide. This process typically occurs at ambient temperature and pressure under mild conditions, yielding electrode materials with high crystallinity, strong substrate bonding, and excellent uniformity. In preparing tin-based electrodes, electrodeposition is commonly employed to form tin-antimony oxide interlayers, though it often requires alternating with thermal decomposition, resulting in a relatively complex procedure.

## 2.3. Chemical Vapor Deposition

CVD is a process for fabricating Ti/SnO<sub>2</sub>-Sb electrodes. This method vaporizes the targeted elements or compounds into gaseous phase, which are then transported via an inert carrier gas to the reaction chamber, where they undergo adsorption and gas-phase reactions on the heated electrode substrate surface, ultimately depositing to form a solid oxide coating. The prepared electrodes exhibit uniform coating thickness, rapid deposition rates, and excellent adaptability to substrate geometries. Consequently, it is widely employed for high-purity and high-density thin-film materials synthesis. However, this method faces limitations when fabricating tin-based electrodes such as Ti/SnO<sub>2</sub>-Sb, due to the high cost of required vapor-phase precursors.

## 2.4. Hydrothermal Method

The hydrothermal method is a technique for electrode preparation that utilizes a sealed high-pressure reactor with water or other solvents as the reaction medium, creating a high-temperature and high-pressure environment through heating. The specific procedure involves placing the electrode substrate into insoluble metal salts aqueous solution (or organic solvents), and heating the sealed system to high-temperature and high-pressure conditions. Under these conditions, the insoluble substances gradually dissolve and undergo multiphase chemical reactions, forming a supersaturated solution. Eventually, the material precipitates and crystallizes on the substrate surface, producing a uniform oxide coating to obtain the desired electrode. This method is particularly suitable for synthesizing well-crystallized thin-film materials under relatively mild conditions.

## 2.5. Sol-gel Method

The sol-gel methods are the most commonly used techniques for Ti/SnO<sub>2</sub>-Sb electrodes preparation. These methods firstly involve pretreating the titanium substrate with processes such as oxalic acid etching to enhance adhesion. Subsequently, precursor salts containing tin (SnCl<sub>2</sub>·2H<sub>2</sub>O) and antimony (SbCl<sub>3</sub>) are dissolved in ethanol solvents, with the addition of complexing agents to form a uniform and stable sol. The sol is deposited onto the substrate surface via dip-coating or spin-coating, dried at low temperature to form a gel, and then sintered in a muffle furnace at 500-650 °C to crystallize the coating. This coating and sintering process is typically repeated multiple times to form a dense, uniform oxide film. The core of this method lies in refining SnO<sub>2</sub> grains, enhancing conductivity, and boosting electrocatalytic activity through antimony doping (with an optimized molar ratio of approximately Sn: Sb = 9: 1). The sol-gel methods offer advantages such as precise molecular-level composition control, excellent coating uniformity, and relatively simple equipment. However, they suffer from

drawbacks including a lengthy process cycle, potential substrate oxidation during high-temperature sintering, and limitations on single-coating thickness.

### 3. Modification Methods of Ti/SnO<sub>2</sub>-Sb Electrode

The relatively short service life of Ti/SnO<sub>2</sub>-Sb electrodes limits their application. In recent years, researchers have conducted extensive studies on how to improve the lifespan of Ti/SnO<sub>2</sub>-Sb electrodes, focusing primarily on enhancing the bonding strength between the catalytic layer and the substrate. A series of electrode modification techniques have been developed, which can be mainly categorized into two types: doping modification and structural modification[18].

#### 3.1. Doping Modification

The primary role of introducing foreign elements is to refine grain size, enhance crystallinity, reduce charge transfer resistance, and contribute to the formation of a crack-free surface[18]. The dopant elements can be categorized into two groups: rare earth metals and precious metals.

##### 3.1.1. Doping of Rare Earth Element

Rare earth elements (such as Ce, La, and Yb) have both unique electrochemical and physicochemical structures[19]. Doping these elements into the crystal lattices enhance the electrocatalytic performance and extend electrode lifetimes[20, 21]. Hong et al. investigated the fabrication of La- and Eu-doped Ti/SnO<sub>2</sub>-Sb electrodes and their electrochemical degradation of methylene blue (MB)[22]. The study found that the La-modified Ti/SnO<sub>2</sub>-Sb electrode exhibited excellent electrochemical performance, with an oxygen evolution potential (OEP) of 1.84 V, which is 0.15 V higher than that of the undoped electrode. The La-doped electrode also demonstrated superior wastewater treatment capability, achieving a 45% increase in degradation efficiency of 50 ppm MB within 25 min compared to the Ti/SnO<sub>2</sub>-Sb-Eu electrode. Zheng et al. successfully fabricated a Ti/Sm-Mn-Sb-SnO<sub>2</sub> electrode through co-doping with a Sm-Mn composite interlayer for the electrochemical oxidation degradation of polystyrene nanoplastics (PS NPs) in water[23]. The study demonstrated that the Ti/Sm-Mn-Sb-SnO<sub>2</sub> anode exhibited the optimal PS NP removal efficiency of 58.75% and the longest accelerated electrode lifetime of 825 h. The doping of composite interlayer elements resulted in a more uniform and dense crack structure on the anode surface, while also formed a more complete crystalline structure, effectively increasing the active sites and specific surface area for the electrochemical process.

##### 3.1.2. Doping of Precious Metals

Precious metals (such as Ru, Ir and Pt) typically play unique role in electrode modification, which can serve as both catalytic activity promoter and structural stabilizer, due to their unique electronic structures and catalytic properties[24-26]. Guo et al. prepared Ti/SnO<sub>2</sub>-Sb-Ru electrodes with varying Ru contents via the sol-gel methods[12]. results indicate that the Ti/SnO<sub>2</sub>-Sb-Ru(6) electrode exhibits outstanding electrocatalytic performance. Electrochemical degradation experiments demonstrated that under an initial pH of 4, the Ti/SnO<sub>2</sub>-Sb-Ru(6) electrode achieved a decolorization efficiency of 98.08% and COD removal efficiency of 76.98% at initial MB concentration of 300 mg/L. Moreover, the Ti/SnO<sub>2</sub>-Sb-Ru(6) electrode demonstrated a service life exceeding 1211 days, significantly surpassing conventional electrodes. Its degradation efficiency remained stable at approximately 93% throughout 10 repeated experiments. Wu et al. prepared Ti/SnO<sub>2</sub>-Sb-Ir electrodes doped with low iridium content and employed them for the oxygen evolution reaction in acidic water electrolysis[27]. The results indicate that the electrode surface is relatively compact. Iridium dioxide doping significantly enhanced the electrocatalytic activity and stability of the Ti/SnO<sub>2</sub>-Sb electrode during the oxygen evolution reaction.

### 3.2. Structural Modification

The core concept of structural modification lies in enhancing coating adhesion, increasing active surface area, and optimizing electron transport pathways, thereby synergistically improving stability and catalytic activity[10]. Structural modification primarily encompasses interlayer structural modification and Ti-based bulk structural modification.

#### 3.2.1. Interlayer Structural Modification

The intermediate layer introduction can increase the bonding force between the SnO<sub>2</sub>-Sb catalytic layer and the Ti substrate, prolonging the electrode's lifetime. Tang et al. successfully fabricated Ti/TiN/SnO<sub>2</sub>-Sb electrodes using arc spraying for the TiN intermediate layer and electrodeposition[28]. Under conditions of 30 mA/cm<sup>2</sup> current density, 2 cm electrodes spacs, pH 7, and 180 min duration, the hydroxychloroquine (HCQ) removal efficiency reached 85.5%. Moreover, the accelerated lifespan of the Ti/TiN/SnO<sub>2</sub>-Sb electrode reached 309 min, demonstrating a 4.4-fold increase in durability compared to the Ti/SnO<sub>2</sub>-Sb electrode.

#### 3.2.2. Ti-based Bulk Structural Modification

Ti-based bulk structural modification means a 3D surface structure formation through the anodization of the substrate. This 3D architecture can enhance the bonding force between the SnO<sub>2</sub>-Sb catalytic layer and the Ti substrate, thereby enhance the electrode's stability. Wang et al. successfully fabricated a Ti/SnO<sub>2</sub>-Sb-HFs electrode with a 3D hierarchical flower-like structure via a hydrothermal method[29]. They found that this novel electrode exhibits a larger electrochemical active surface area, lower charge transfer resistance, and higher oxygen evolution potential compared to the conventional Ti/SnO<sub>2</sub>-Sb electrode. After 5 hours of degradation, the removal efficiencies of Acid Red 73 (AR 73) and COD reached 98.7% and 83.9%, respectively. Moreover, its accelerated service life was 9.3 times longer than that of the traditional Ti/SnO<sub>2</sub>-Sb electrode.

## 4. Application of Ti/SnO<sub>2</sub>-Sb Electrodes in Organic Wastewater Treatment

### 4.1. Dye Wastewater

Organic dyes are difficult to eliminate from wastewater because of their aromatic chemical structure and non-biodegradable nature. Moreover, these compounds are toxic and carcinogenic, posing significant threats to human health[30]. Cao et al. fabricated a Ti/SnO<sub>2</sub>-Sb-Cu electrode via the sol-gel methods[31]. The study demonstrated that under the conditions of pH 4, current density of 20 mA/cm<sup>2</sup>, and an initial concentration of 50 mg/L, the optimal degradation efficiencies of MB and COD removal reached 99% and 79.5%, respectively.

### 4.2. Medical Wastewater

The discharged amount of medical wastewater increase continually due to the rapid development of the healthcare industry, frequent occurrence and wide spread of various diseases[32]. Medical wastewaters are complex in composition and highly biotoxic, posing severe challenges to both the natural environments and public health safety. The typical pollutants include antibiotics, nonsteroidal anti-inflammatory drugs (NSAIDs), antibiotic resistance genes (ARGs), and disinfection byproducts (DBPs)[33]. These substances have low biodegrade, and carry potential pathogenic and carcinogenic risks, posing long-term threats to nature ecosystems and human health[34]. Lan et al. prepared nitrogen-doped carbon quantum dot (N-CQDs)-modified Ti/SnO<sub>2</sub> anodes (Ti/SnO<sub>2</sub>-N-CQDs) via electrodeposition for tetracycline (TC) degradation[35]. The study revealed that the Ti/SnO<sub>2</sub>-N-CQDs-10 electrode achieved a 98.8% tetracycline removal efficiency within 60 min.

### 4.3. Petrochemical Wastewater

Harmful dissolved organic compounds, such as aromatic compounds, long-chain hydrocarbons, surfactants, and heterocyclic compounds, are often coexisted in petrochemical wastewaters[36]. There are inefficient solidification, biological treatment, and adsorption processes for the compounds because of their varying physicochemical properties[37]. Zhao et al. prepared Ti/SnO<sub>2</sub>-Sb electrodes via the sol-gel methods and applied them to the electrocatalytic treatment of actual petrochemical reverse osmosis concentrate[38]. The results revealed that at a current density of 20 mA/cm<sup>2</sup>, the electrode achieved a COD removal efficiency of 94.5% and a DOC removal efficiency of 43.4%.

## 5. Conclusion

Ti/SnO<sub>2</sub>-Sb electrodes have emerged as a highly promising and cost-effective anode material for the electrocatalytic degradation of refractory organic pollutants in water. This review has outlined the fundamental preparation techniques, highlighting the sol-gel methods as prevalent approach due to the compositional control and uniformity, despite challenges like long processing times. The core limitation of electrode deactivation has been actively addressed through targeted modification strategies. Doping with elements such as La, Ce, Ru, or Ir refines the electrode's microstructure, enhances conductivity, and improves catalytic activity and stability. Concurrently, structural modifications, including the introduction of interlayers (e.g., TiN) or the creation of 3D hierarchical substrates, effectively strengthen the coating-substrate adhesion and increase the electrochemically active surface area, thereby extending the electrode's operational lifespan significantly. Moreover, in treating complex wastewater streams containing dyes, pharmaceuticals, and petrochemical effluents, the modified Ti/SnO<sub>2</sub>-Sb electrode demonstrates remarkable practical efficacy, achieving high degradation rates and mineralization efficiency. In summary, advancing the material design and engineering innovation of Ti/SnO<sub>2</sub>-Sb electrodes holds significant potential for promoting highly efficient and sustainable electrochemical wastewater treatment.

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