

Article

Ti/SnO₂-Sb₂O_x-TiO₂ Electrodeposited from Methanesulfonate Electrolytes: Preparation, Properties, and Performance

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Abstract: In this study, Ti/SnO₂-Sb₂O_x-TiO₂ electrodes were produced using a sol-enhanced electrodeposition technique from methanesulfonate electrolytes. The surface microstructures of Ti/SnO₂-Sb₂O_x-TiO₂ were observed, and their phase constituents were determined. The surface features were analyzed by X-ray photoelectron spectroscopy. Linear sweep voltammetry and degradation tests were also conducted to determine the degradation performance. The results show that the addition of TiO₂ sol affects the microstructures of Ti/SnO₂-Sb₂O_x-TiO₂ electrodes, while a uniform coating surface can be obtained at a proper sol concentration in electrolytes. Adding TiO₂ sol also causes deep oxidation of Sb and generates more adsorbed oxygen on the electrode surface. The favorable surface features and the well-dispersed TiO₂ in the coatings of 10 mL/L TiO₂ modified Ti/SnO₂-Sb₂O_x-TiO₂ electrodes award them the best electrocatalytic performance, and their uniform coating surface prolongs the electrode service life.

Keywords: SnO₂-Sb₂O_x; sol-enhanced electrodeposition; organic degradation



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1. Introduction

The demand for effective effluents treatment technology has raised increasing research interests under growing public concern in environmental issues. The treatment of poorly biodegradable organics is essential in the effluents treatment industry. Advanced electrochemical oxidation technology (AEOT) is considered to be a promising method to deal with this issue, owing to its advantages in efficiency and flexibility [1–3]. Organic degradation reactions mostly occur on the anode surface, which plays a crucial role in the electrochemical treatment process. The primary anode materials applied in effluent treatment technology are precious metals (Pt, Au, etc.), carbon, boron-doped diamond (BDD), and metal oxide electrodes [4–6].

Titanium-supported antimony-tin oxide (Ti/SnO₂-Sb₂O_x) is a type of metal oxide electrode that can suppress the oxygen evolution reaction (OER) in order to allow for the effective degradation of organics by hydroxyl radicals. Originally, SnO₂-Sb₂O_x was utilized as a catalyst for organic conversion, whereas this mixed oxide was recently employed as an anode coating due to its adequate conductivity and chemical resistance. Moreover, SnO₂-Sb₂O_x can be efficiently and economically coated on a Ti substrate, making it into large areas for industrial applications. The inert Ti/SnO₂-Sb₂O_x electrodes, therefore, show unique advantages in effluent treatment technology due to their low preparation cost compared to precious metal and BDD, high OER potential, and solid electrocatalytic ability.

The conventional fabrication route for the Ti/SnO₂-Sb₂O_x electrode is thermal decomposition, in which, the precursor solution is transformed into SnO₂-Sb₂O_x after calcination [7]. However, the inferior stability and inadequate electrocatalytic ability limit the scale-up application of Ti/SnO₂-Sb₂O_x electrodes made from thermal decomposition. The electrodeposition

fabrication route was recently proposed to manufacture Ti/SnO₂-Sb₂O₃-TiO₂ electrodes. However, despite an enhanced stability, the electrodeposited Ti/SnO₂-Sb₂O₃-TiO₂ electrode suffers from a relatively low electrocatalytic performance.

Previous literature has investigated various modification methods for electrode performance enhancement. Venkatesha et al. [8] and Wang et al. [9] constructed a porous SnO₂-Sb₂O₃ coating based on anodized TiO₂ nanotubes, boosting the electrode electrocatalytic performance and long-term stability. Chen et al. [10] synthesized the dense spherical Ti/Sb-SnO₂ with a superior electrocatalytic performance using colloidal electrodeposition. Moreover, doping other elements and nanoparticles could effectively modify the electrodeposited Ti/SnO₂-Sb₂O₃ [11]. Liu et al. [12] and Qiao et al. [13] fabricated the Ti/SnO₂-Sb₂O₃-Ce electrodes for efficient electrocatalytic oxidation. Wu et al. [14] prepared a high-performance duplex-structured Ti/SnO₂-Sb₂O₃-CNT composite anode using the electrodeposition method. In our recent studies, a sol-enhanced electrodeposition method has been proposed to efficiently dope inert nanoparticles and achieve their well-dispersion into the coating [15–18], which can serve as a promising method for Ti/SnO₂-Sb₂O₃ modification.

This work explores the sol-enhanced electrodeposition route for the Ti/SnO₂-Sb₂O₃-TiO₂ electrodes in methanesulfonate electrolytes. Compared to the traditional oxidizing sulfuric sources, the methanesulfonate electrolyte better reveals the advantages of a low toxicity, excellent chemical stability, and outstanding biodegradability [19–21]. The influence of TiO₂ sol addition on the electrode's properties and performance is systematically investigated. We propose a novel modification procedure for Ti/SnO₂-Sb₂O₃, which will benefit the scale-up application for Ti/SnO₂-Sb₂O₃ in the effluent treatment industry. We also aim to thoroughly unveil the underlying mechanism and potential of its electrodeposition fabrication route.

2. Materials and Methods

2.1. Preparation of Ti/SnO₂-Sb₂O₃-TiO₂

The analytical grade chemicals were bought from Aladdin Reagent, Shanghai, China. A pure titanium (99.9%) plate was used in our work. The titanium substrate was pretreated as described in the procedures below: (1) grinding and polishing; (2) soaking into 15% wt sodium hydroxide (NaOH) solution for 2 h; and (3) soaking in 12% wt oxalic acid solution for 3 h. The TiO₂ sol was prepared as reported in the previous papers [22,23].

The proposed Ti/SnO₂-Sb₂O₃-TiO₂ electrodes were fabricated from the electrodeposition method. The electrodeposition fabrication route consists of the electrodeposition process and the following heat-treatment process.

In the electrodeposition process, the electrolyte consisted of 60 mL/L tin methanesulfonate (C₂H₆O₆S₂Sn, 50% wt in H₂O), 50 mL/L methane sulfonic acid (CH₄O₃S, 99%), antimony trichloride (SbCl₃, 99%), 2 g/L gelatin, and 2 g/L hydroquinone. The electrodes were prepared in an electrolytic cell as follows: the tin plate (40 mm × 40 mm) was used as the anode and the pretreated titanium substrate (20 mm × 30 mm × 0.1 mm) was used as the cathode. TiO₂ sol was prepared following the procedures in previous work [24–26]. A certain amount (5, 10, 15 mL/L) of as-prepared TiO₂ sol was slowly dropped into the stirring electrolytes. The electrodeposition was carried out at a current density of 30 mA/cm² for 20 min at 30 °C, and the electrolyte was stirred at 300 rpm. Then, in the heat treatment process, the samples were heat-treated in a muffle furnace at temperatures of 600 °C for 10 h to obtain the Ti/SnO₂-Sb₂O₃-TiO₂ electrodes. The thickness of electrodeposited SnO₂-Sb₂O₃ coating was ~26 μm, whereas TiO₂ sol addition hardly modified the coating thickness.

The Ti/SnO₂-Sb₂O₃ was also made by the traditional dip-coating method for comparison. Firstly, the pretreated titanium substrate was dipped into a mixture of 1.8 g of tin tetrachloride (SnCl₄), 2 mL of antimony oxide (Sb₂O₃), and 8 mL of isopropyl alcohol for 15 s, followed by a drying treatment at 100 °C for 10 min and then a heating treatment in a muffle furnace for 10 min at 600 °C. The drying and heating treatment process was repeated six times, and, finally, the titanium substrate was placed in a muffle furnace for ten hours at 600 °C to acquire the Ti/SnO₂-Sb₂O₃.

2.2. Structural and Elemental Characterization

X-ray diffraction (XRD-6000X, Shimadzu, Kyoto, Japan) identified the phase constituents at a step size of $0.1^\circ/\text{s}$. The surface morphology was observed by a scanning electron microscope (SEM, Phenom ProX, Eindhoven, The Netherlands) with an energy dispersive spectroscopy (EDS) detector, and the embedded simulation program completed the 3D surface imaging. The X-ray photoelectron spectroscopy (XPS, ESCALAB 250Xi, Thermo Fisher, Waltham, MA, USA) was utilized to analyze elemental details, during which, Al K α radiation at 1486.6 eV was used.

2.3. Electrochemical and Degradation Tests

The electrochemical performance of the electrode was tested by linear sweep voltammetry (LSV) using an electrochemical workstation (CH660E, Chenhua Instrument, Shanghai, China). A three-electrode system was used in the tests. Ti/SnO₂-Sb₂O₃-TiO₂ electrode (10 mm $\times$ 10 mm) was the working electrode. Platinum plate (20 mm $\times$ 20 mm) was the counter electrode, and saturated calomel electrode (SCE) was the reference electrode. A 0.25 mol/L Na₂SO₄ solution was used as the electrolyte solution. The scan range was from 0 to 4 V, at a scanning rate of 20 mV/s.

The benzoic acid degradation experiment was carried out in 100 mg/L benzoic acid solution at a volume of 200 mL. We used the Ti/SnO₂-Sb₂O₃-TiO₂ electrode (20 mm $\times$ 30 mm) as the anode and the same-sized titanium sheet as the cathode. A direct current power was employed to control the current density at 40 mA/cm², and the 300-rpm magnetic stirring was applied. The target solution sample was taken for detection after a certain degradation period. The degradation process was monitored using high-performance liquid chromatography (HPLC, Nexera, Shimadzu, Kyoto, Japan) equipped with a TC-C18 column. The mobile phase consisted of 90% water (0.1% trifluoroacetic acid) and 10% acetonitrile. The accelerated life was tested in 0.25 mol/L Na₂SO₄ at room temperature at 100 mA/cm². The prepared sample was the anode, and a titanium sheet was the cathode. The accelerated lifetime was calculated when the potential steeply raised and exceeded 5.0 V.

3. Results and Discussion

3.1. Structural Characterizations

Figure 1 presents the XRD profiles of Ti/SnO₂-Sb₂O₃ and Ti/SnO₂-Sb₂O₃-TiO₂ electrodes. The electrode prepared by the conventional dip-coating method reveals broad SnO₂ peaks, which indicates a large extent of the amorphous phase. In addition, the sharp Ti peaks imply the exposure of underneath titanium substrate. Such an observation is in good agreement with previous literature [7,27]. In contrast, distinct phase constituents are attained for these samples from the electrodeposition fabrication route. The crystallinity substantially increases, and several intense peaks of tetragonal rutile SnO₂ (JCPDS 99-0024) are discovered. Similar phenomena have also been reported in some earlier literature [7].

The incorporation of TiO₂ addition affects the phase composition significantly. When the TiO₂ addition was ≤ 10 mL/L, the SnO₂ peak intensity sharply decreases, which indicates a refinement for the prepared SnO₂ crystallites. As summarized in Table 1, the grain size of SnO₂ is estimated according to the Scherrer equation through full width at half maximum (FWHM) [28]. The results show that adding 10 mL/L sol leads to a significantly decreased grain size, whereas larger grains form when further increasing the TiO₂ sol concentration to 15 mL/L. During the electrodeposition process, the TiO₂ nanoparticles are in situ generated and then co-deposited to generate SnO₂-Sb₂O₃-TiO₂. These well-dispersed nanoparticles can serve as active sites for crystallite nucleation, thereby providing a driving force for nucleation and forming smaller SnO₂ crystallites [29]. However, the sharp SnO₂ peaks show an opposite phase variation at a 15 mL/L TiO₂ concentration. Under this condition, the generated TiO₂ nanoparticles tend to aggregate and decrease the active sites in the electrodeposition. It is noted that no TiO₂ diffraction peak is detected in the composite coatings due to the limited amount of embedded TiO₂ nanoparticles.

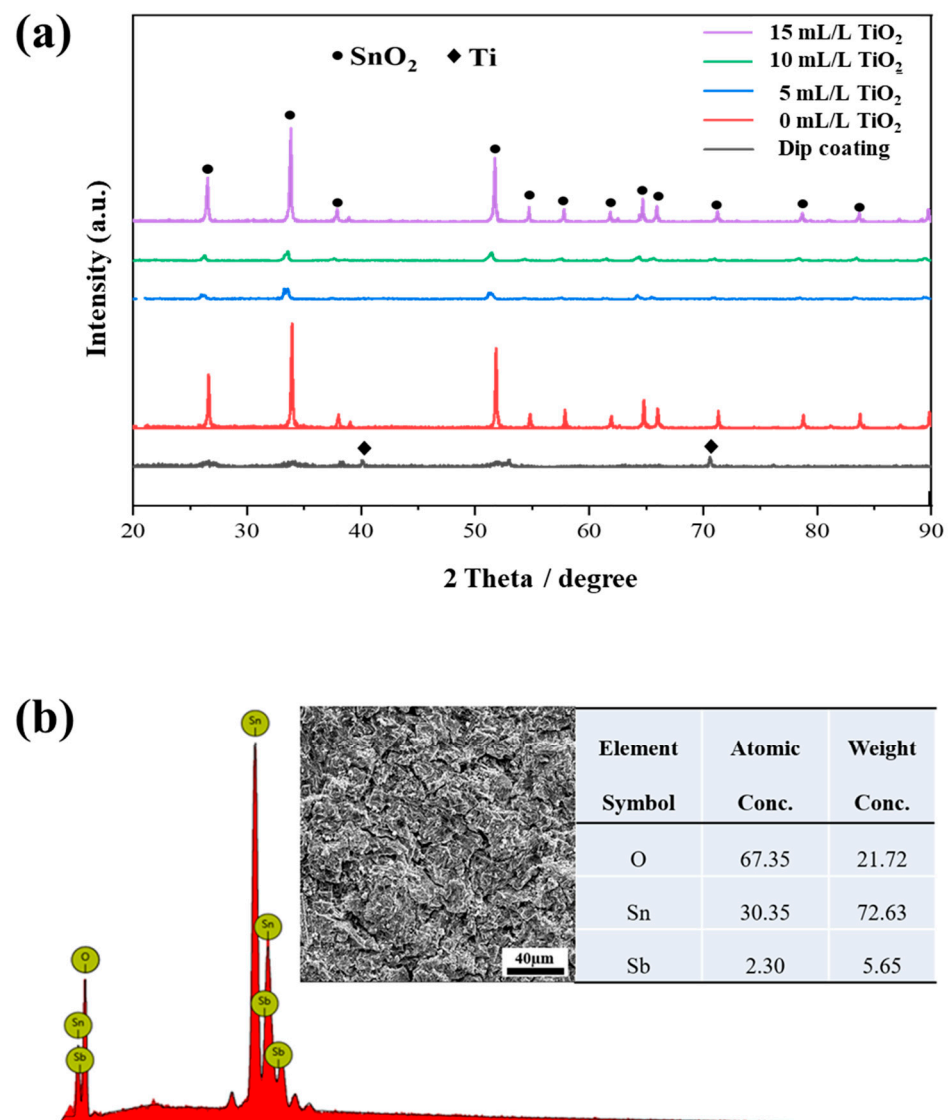


Figure 1. (a) XRD profiles recorded on the Ti/SnO₂-Sb₂O_x-TiO₂ with different concentrations of TiO₂ sol, and (b) the EDS results recorded on the electrodeposited Ti/SnO₂-Sb₂O_x.

Table 1. Estimated grain size of SnO₂-Sb₂O_x-TiO₂ prepared at different TiO₂ sol additions.

TiO ₂ Sol Addition	0 mL/L	10 mL/L	15 mL/L
Grain size (nm)	60	28	45

Figure 1b presents the elemental analysis recorded on the electrodeposited Ti/SnO₂-Sb₂O_x, proving the existence of antimony in the deposited coatings. The ratio of Sn/Sb is ~8 wt.% in SnO₂-Sb₂O_x, which is in good agreement with previous reports [7,30]. Such antimony content in the lattice provides good conductivity for the prepared electrode.

The surface microstructures of prepared Ti/SnO₂-Sb₂O_x-TiO₂ are presented in Figure 2. A typical mud-like coating surface is obtained using the conventional dip-coating method, as shown in Figure 2a. The cracks and holes in such coating surfaces could damage the electrode stability due to the oxide generation in the electrolysis process [7]. In the meantime, the electrodeposited Ti/SnO₂-Sb₂O_x-TiO₂ demonstrates a different surface morphology, showing a relatively compact coating surface with a much-improved uniformity. An increased addition of TiO₂ sol (≤ 10 mL/L) gives rise to a more compact surface, whereas an

addition of 15 mL/L TiO_2 sol causes a relatively non-uniform surface morphology due to the nanoparticle's aggregation.

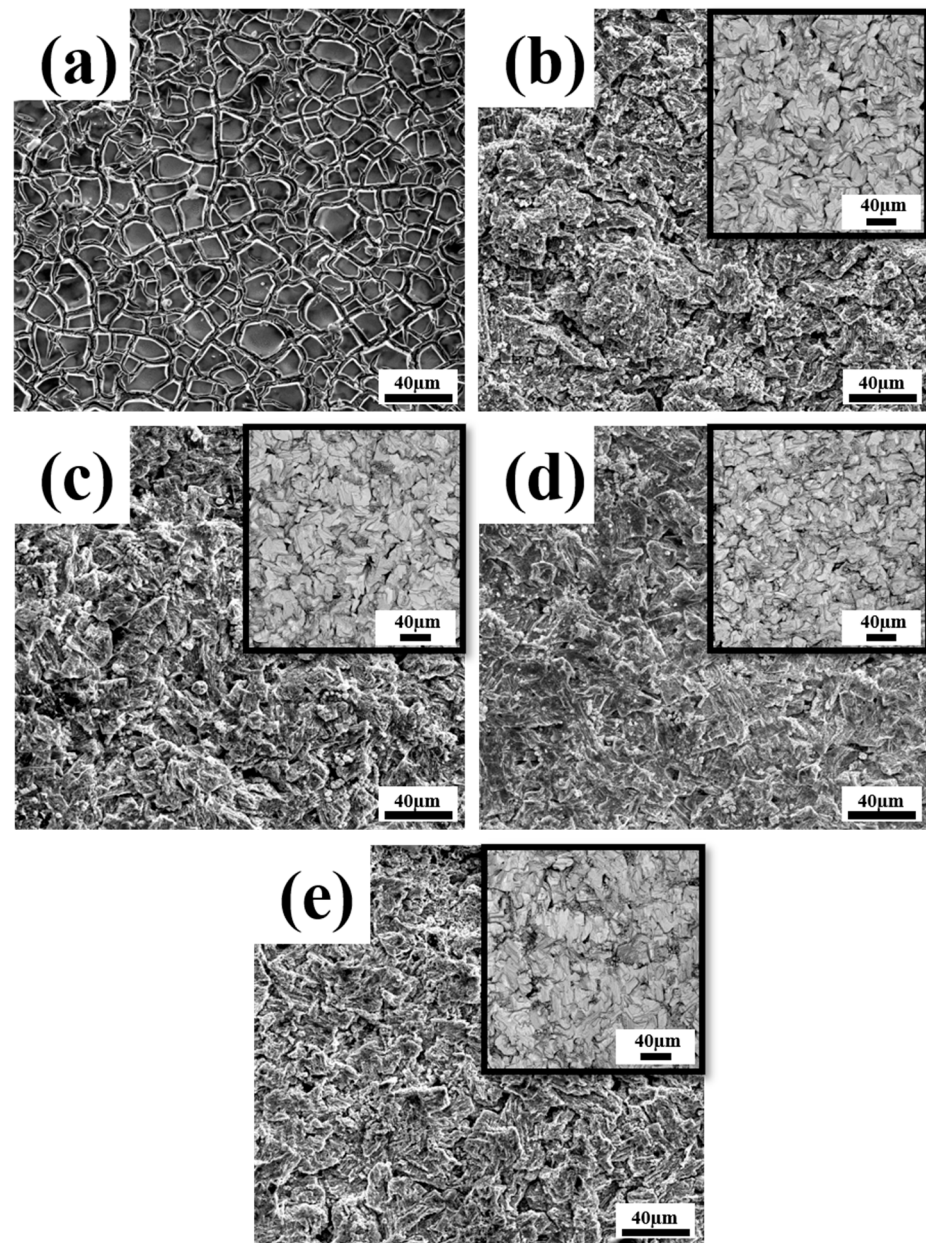


Figure 2. The surface morphologies of (a) $\text{Ti/SnO}_2\text{-Sb}_2\text{O}_x$ made by traditional dip-coating, and $\text{Ti/SnO}_2\text{-Sb}_2\text{O}_x\text{-TiO}_2$ electrodeposited with different additions of TiO_2 sol: (b) 0 mL/L (c) 5 mL/L, (d) 10 mL/L, and (e) 15 mL/L (insets are the corresponding Sn-Sb-TiO_2 coatings before the heat treatment process).

A similar morphologic variation can be seen for the Sn-Sb-TiO_2 layers before the heat-treatment process, as shown in the inset images. The TiO_2 addition refines the surface morphology of Sn-Sb-TiO_2 at a TiO_2 sol concentration ≤ 10 mL/L, whereas an excessive sol concentration caused relatively non-uniform morphology. The surface structures of $\text{Ti/SnO}_2\text{-Sb}_2\text{O}_x\text{-TiO}_2$ electrodes are highly associated with the early electrodeposited Sn-Sb-TiO_2 layers, with some features remaining after the oxidation reactions during heat treatment.

The surface features were further characterized by a 3D imaging technique, as shown in Figure 3. These images show the surface roughness and uniformity for the electrodeposited samples. The proper addition of TiO_2 sol brings a more uniform and compact surface, depicted in Figure 3b,c. The observation corresponds with our earlier XRD and

SEM findings, which proves that the 10 mL/L TiO_2 sol addition refines the surface with a decreased crystallite size and leads to a relatively open surface morphology with an improved uniformity. Nevertheless, the 15 mL/L TiO_2 addition causes a non-uniform coating surface, depicted in Figure 3d.

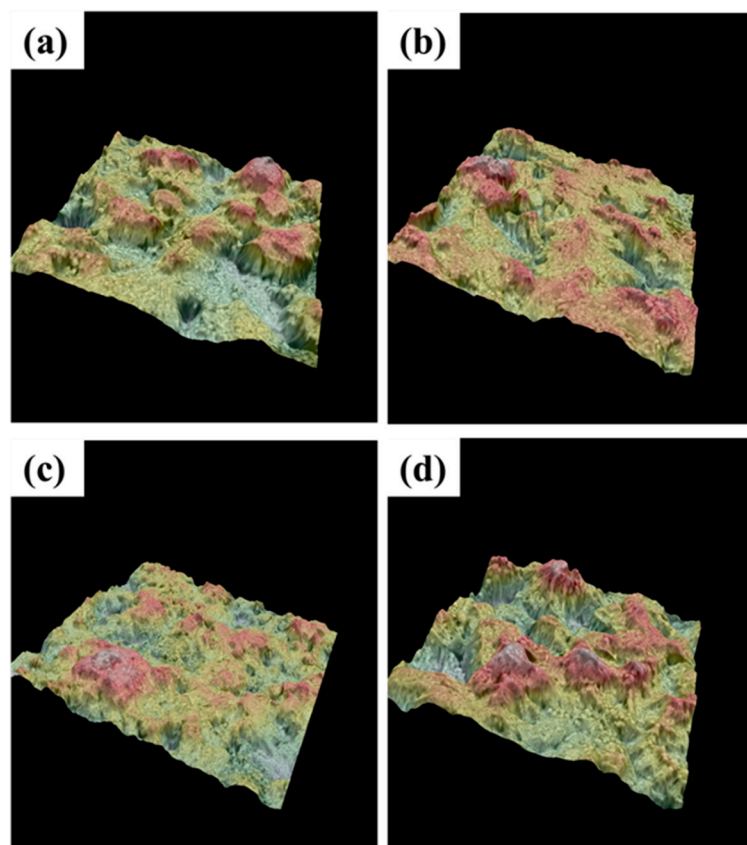


Figure 3. The 3D-imaging surface (depicted with heat map) for the prepared $\text{Ti}/\text{SnO}_2\text{-Sb}_2\text{O}_x\text{-TiO}_2$ samples electrodeposited with different additions of TiO_2 sol: (a) 0 mL/L, (b) 5 mL/L, (c) 10 mL/L, (d) 15 mL/L.

3.2. Elemental and LSV Analysis

The elemental analysis characterizes the electrodeposited samples, as shown in Figure 4. Figure 4a shows the overall spectra of the electrodeposited $\text{Ti}/\text{SnO}_2\text{-Sb}_2\text{O}_x$ and $\text{Ti}/\text{SnO}_2\text{-Sb}_2\text{O}_x\text{-TiO}_2$ samples, where prominent peaks of Sb and Sn are detected. The presence of Ti peaks in Figure 4b proves the successful incorporation of TiO_2 into the prepared $\text{SnO}_2\text{-Sb}_2\text{O}_x\text{-TiO}_2$ coatings, whereas the $\text{SnO}_2\text{-Sb}_2\text{O}_x$ sample shows no Ti peak. The weak Ti peaks correlate with the limited content of TiO_2 existing on the electrode surface. During the sol-enhanced electrodeposition, the TiO_2 nanoparticles are in situ generated in electrolytes. The attached organic chains avoid the agglomeration of nanoparticles, which are then co-deposited in the coating with excellent dispersion.

Figure 4c,d compare the spectra of Sb and O on the electrodeposited $\text{Ti}/\text{SnO}_2\text{-Sb}_2\text{O}_x$ and $\text{Ti}/\text{SnO}_2\text{-Sb}_2\text{O}_x\text{-TiO}_2$. The results elaborate in detail on the state of antimony oxide and absorbed oxygen with and without TiO_2 sol addition. Both Sb^{3+} and Sb^{5+} exist in the prepared coatings, indicating that there are two oxidation states for antimony after the heat treatment process. In addition, the detected oxygen can be categorized as O_{lat} (i.e., lattice oxygen species) and O_{ads} (hydroxyl oxygen species), which matches with previous studies [13,31].

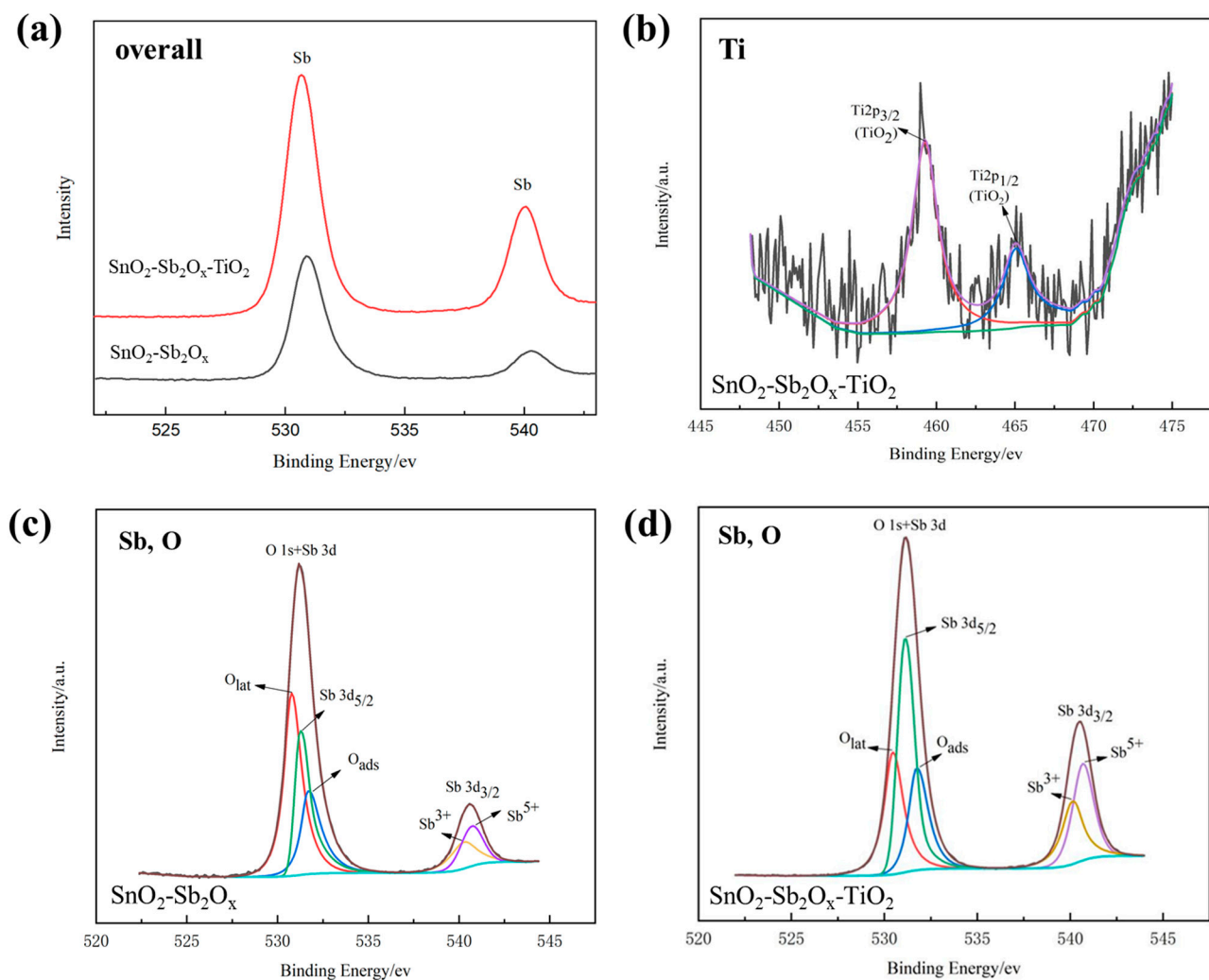


Figure 4. XPS spectra of Ti/SnO₂-Sb₂O_x and Ti/SnO₂-Sb₂O_x-TiO₂ (10 mL/L TiO₂ sol addition): (a) the overall spectra, (b) Ti spectra for Ti/SnO₂-Sb₂O_x-TiO₂, (c) Sb, O spectra for Ti/SnO₂-Sb₂O_x, and (d) Sb, O spectra for Ti/SnO₂-Sb₂O_x-TiO₂.

Table 2 summarizes the comparison between the Ti/SnO₂-Sb₂O_x and Ti/SnO₂-Sb₂O_x-TiO₂ in XPS analysis. The listed atom ratios of O_{ads}/O_{lat} and Sb⁵⁺/Sb³⁺ provide a qualitative judgment for the element variation. The atom ratio of O_{ads}/O_{lat} is higher in Ti/SnO₂-Sb₂O_x-TiO₂. It is noted that adsorbed oxygen is a potent oxidizing agent. In the meantime, the Sb 3d_{3/2} is split into two oxidation states, Sb⁵⁺ and Sb³⁺, respectively. The higher ratio of Sb⁵⁺/Sb³⁺ implies that deeper surface oxidation occurs in the heat treatment process. The higher valence of Sb generally offers more excessive electrons and functions as dominant donors for the SnO₂-Sb semiconductor, improving the conductivity of the prepared electrode [32]. The natural donors of oxygen vacancies are inhibited simultaneously, and the lattice oxygen is reduced. To conclude, the results imply that the addition of TiO₂ sol can help to generate favorable surface features of the chemical composition and oxygen state for the improved electrocatalytic performance.

Table 2. Calculated atom ratios for Ti/SnO₂-Sb₂O_x and Ti/SnO₂-Sb₂O_x-TiO₂.

Samples	Atom Ratio	
	Sb ⁵⁺ :Sb ³⁺	O _{ads} :O _{lat}
0 mL TiO ₂ sol sample	1.13	0.49
10 mL TiO ₂ sol sample	1.34	0.78

Figure 5 presents the linear sweep voltammetry recorded on the Ti/SnO₂-Sb₂O₃ and Ti/SnO₂-Sb₂O₃-TiO₂. The oxygen evolution reaction (OER) potential decreases in the order below: 10 mL/L TiO₂ modified sample (2.34 V) > 5 mL/L TiO₂ modified sample (2.27 V) > 15 mL/L TiO₂ modified sample (2.19 V) > 0 mL/L TiO₂ modified sample (2.15 V). The picture indicates that adding a certain TiO₂ sol increases the electrode's OER potential. When the amount of TiO₂ sol increases to 15 mL/L, the OER potential shows a downward tendency, resulting from the nanoparticle aggregation in the coating [33]. In general, the effective organic degradation process favors a high OER potential, which avoids the OER (i.e., the side-reaction) in the electrolysis process.

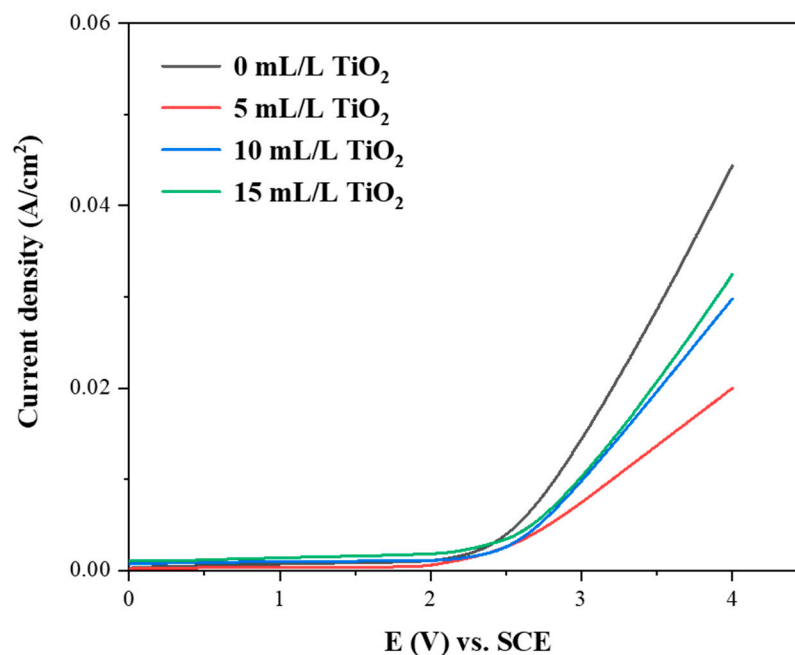


Figure 5. LSV results of electrodeposited Ti/SnO₂-Sb₂O₃ and Ti/SnO₂-Sb₂O₃-TiO₂.

3.3. The Degradation Performance of Ti/SnO₂-Sb₂O₃-TiO₂ Electrodes

The electrocatalytic performance of the prepared electrode was studied by degradation tests, as demonstrated in Figure 6. Benzoic acid (BA) is a typical organic pollutant used as a target in degradation tests. The results show that adding TiO₂ sol influences the electrocatalytic ability. The degradation performance decreases following the order below: 10 mL/L TiO₂ modified sample > 5 mL/L TiO₂ modified sample > 15 mL/L TiO₂ modified sample > 0 mL/L TiO₂ modified sample. After the 10 h electrolysis, 98% BA is electrochemically combusted by the 10 mL/L TiO₂ modified SnO₂-Sb electrodes, whereas only 79% BA is degraded on the non-doped sample in the same duration.

Figure 6b shows the kinetic fitting curve of benzoic acid concentration with degradation time. The logarithm of benzoic acid concentration and degradation time showed a good linear relationship during the degradation process, which proved that the degradation process of benzoic acid follows the primary reaction kinetic model:

$$\ln \left(\frac{C_0}{C_t} \right) = kt \quad (1)$$

where C_0 represents the initial concentration of benzoic acid, C_t represents the concentration of benzoic acid at a specific moment, and k is the reaction rate constant. Table 3 lists the calculated k value for the degradation process on different electrodes. The larger k value of the 10 mL/L TiO₂ modified sample represents the electrode's faster degradation rate of benzoic acid.

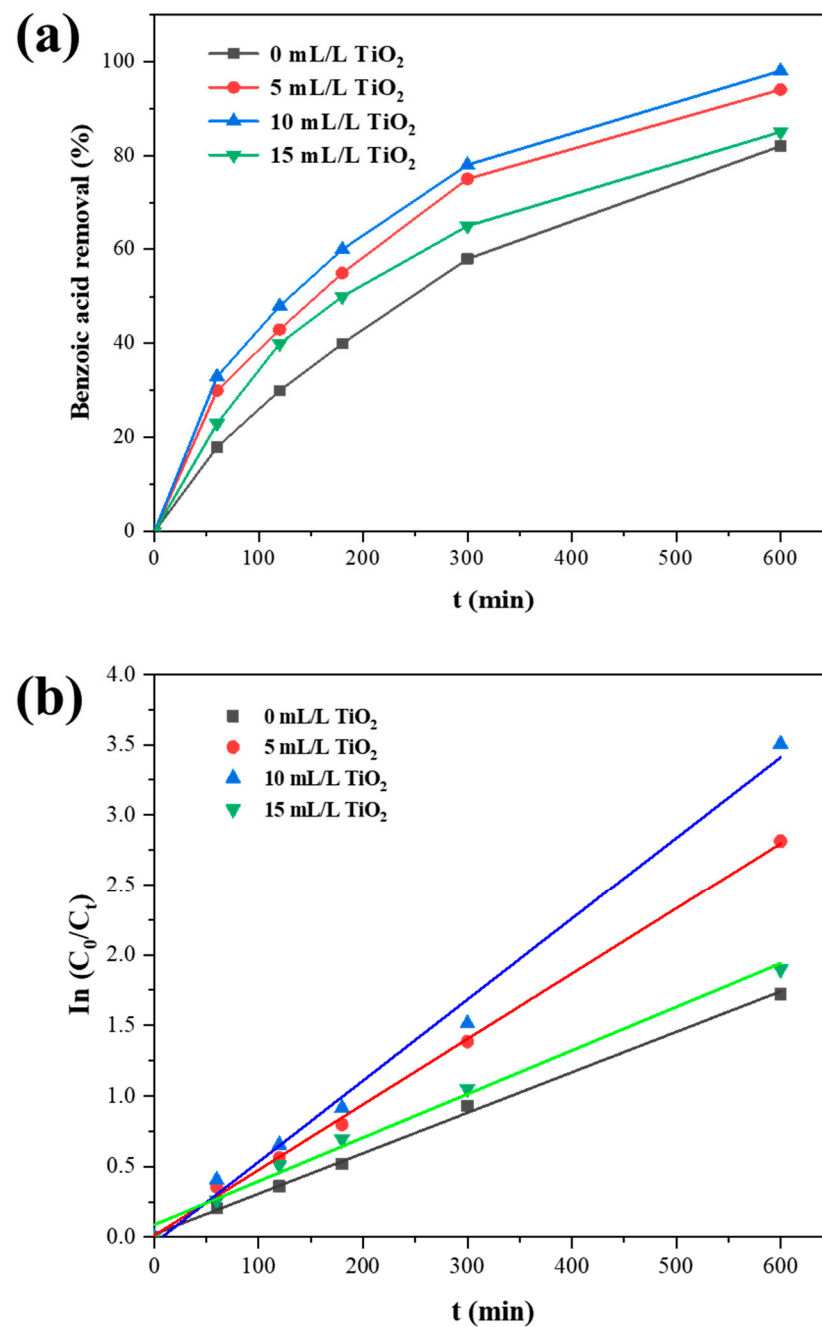


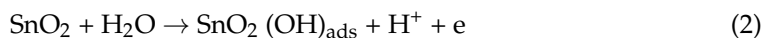
Figure 6. (a) The benzoic concentration after a certain duration of degradation on different electrodes, (b) the fitted curve for the BA degradation process, where C_t is the BA concentration and C_0 is the initial BA concentration.

Table 3. Degradation ability of Ti/SnO₂-Sb₂O₃-TiO₂ electrodes with different concentrations of TiO₂ sol in electrolytes, where k is kinetics coefficients.

	0 mL/L TiO ₂	5 mL/L TiO ₂	10 mL/L TiO ₂	15 mL/L TiO ₂
K (min ⁻¹)	2.87×10^{-3}	4.64×10^{-3}	5.75×10^{-3}	3.09×10^{-3}

The results prove that the proper addition of TiO₂ sol enhances the electrocatalytic performance of Ti/SnO₂-Sb₂O₃-TiO₂ electrodes. In general, the electrochemical combustion of organics is achieved by the hydroxyl radicals—a robust oxidizing agent that breaks the

chemical bonds and finally turns the organic into H_2O and CO_2 . The organic degradation pathway is proposed as follows in Equations (2) and (3) [13,34]:



where R stands for the organic and $(OH)_{ads}$ stands for the physisorbed hydroxyl radicals. The higher content of Sb^{5+} increases the electrode conductivity and helps to generate surface adsorbed oxygen species. Besides, the uniform electrode surface with numerous peaks and valleys could provide many active sites for degradation reactions for the 5 and 10 mL/L TiO_2 modified electrodes. The well-dispersed TiO_2 nanoparticles also offer additional reaction sites, further improving the degradation ability for the TiO_2 sol-enhanced electrodes. As for the 10 mL/L TiO_2 modified $Ti/SnO_2-Sb_2O_x-TiO_2$ electrode, both the superior surface feature and well-dispersed TiO_2 award it the best electrocatalytic performance in the examined samples.

Accelerated life tests were also conducted to investigate the electrode performance, as shown in Figure 7, comprehensively. The dip-coated $Ti/SnO_2-Sb_2O_x$ electrode was tested for comparison, which shows an inferior long-term stability due to its open surface structure being vulnerable to corrosion attack. During the degradation process, the corrosion attack results in the generation of titanium oxide, a non-conductive inert material that inhibits the degradation process and causes coating defoliation.

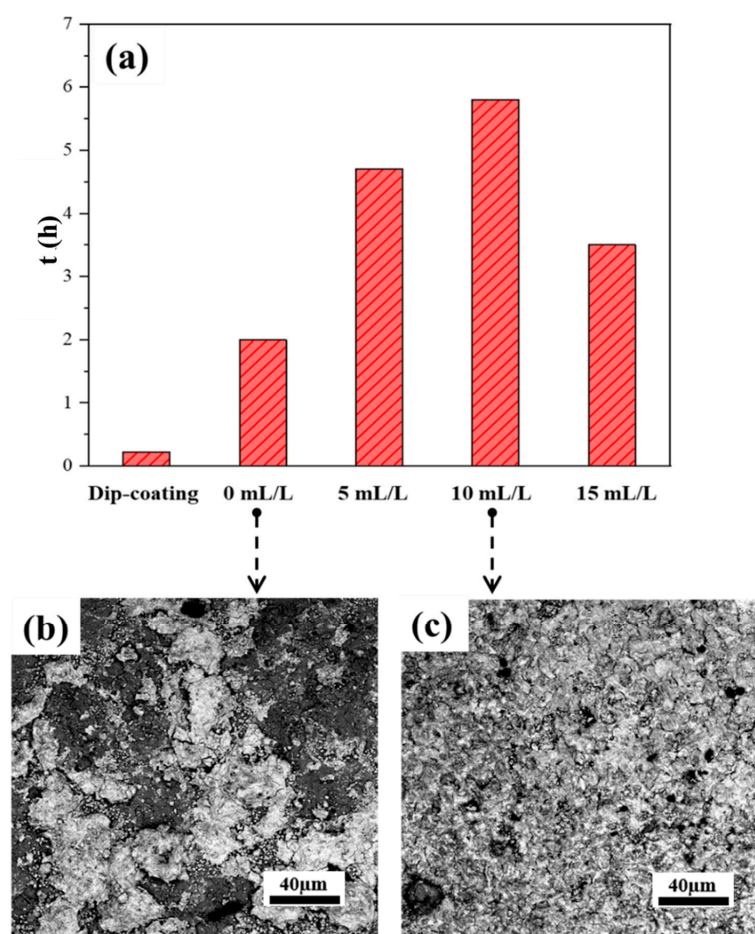


Figure 7. The accelerated service life results, (a) the tested accelerated service life for samples prepared by dip-coating and sol-enhanced electrodeposition with different TiO_2 sol concentrations, (b) surface morphology of electrodeposited $Ti/SnO_2-Sb_2O_x$ after 2 h test, and (c) surface morphology of $Ti/SnO_2-Sb_2O_x-TiO_2$ (10 mL/L TiO_2 sol doped) after 2 h test.

The electrodeposited Ti/SnO₂-Sb₂O₃-TiO₂ shows a much-improved stability in the accelerated life tests. A relatively compact coating fully covers underneath the Ti substrate for these samples. An increasing TiO₂ addition gradually prolongs the accelerated life at a concentration less than 10 mL/L. Figure 7b,c demonstrate the surface morphology of the undoped sample and 10 mL/L TiO₂ sol modified samples after 2 h accelerated life tests. The undoped SnO₂-Sb₂O₃ coating is almost peeled off from the substrate, whereas the electrolysis process only slightly changes the surface morphology of the Ti/SnO₂-Sb₂O₃-TiO₂ electrode.

The electrode stability is highly correlated to the different morphologic features. The proper addition of TiO₂ sol leads to a uniform surface with fewer cracks and holes, thereby improving the electrode stability. Moreover, well-dispersed nanoparticles in the coating can prevent corrosion penetration and enhance the corrosion resistance for electrodes. In contrast, excessive TiO₂ addition harms the coating uniformity due to the nanoparticle aggregation, negatively affecting the electrode stability.

4. Conclusions

This study develops the Ti/SnO₂-Sb₂O₃-TiO₂ electrodes by applying sol-enhanced electrodeposition from methanesulfonate electrolytes. The results show that adding TiO₂ sol affects the surface morphologies of Ti/SnO₂-Sb₂O₃-TiO₂ electrodes. A uniform coating surface can be obtained at a suitable sol addition. Furthermore, the addition of TiO₂ sol leads to deep oxidation of Sb and helps to generate more adsorbed oxygen. The organic degradation performance decreases in the following order: 10 mL/L TiO₂ sol modified sample > 5 mL/L TiO₂ modified sample > 15 mL/L TiO₂ modified sample > 0 mL/L TiO₂ modified sample. The favored surface feature and well-dispersed TiO₂ in the 10 mL/L TiO₂ modified Ti/SnO₂-Sb₂O₃-TiO₂ electrode results in the best electrocatalytic performance in the examined samples, and its uniform coating surface with fewer cracks and holes prolong the electrode service life.

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