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ORIGINAL PAPER

Optimization of $Ti/SnO_2–Sb_2O_5$ anode preparation for electrochemical oxidation of organic contaminants in water and wastewater

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Abstract The preparation of antimony-doped tin oxide anodes on a titanium substrate ($Ti/SnO_2–Sb_2O_5$ anodes) by dipping in a solution of tin chloride and antimony chloride and annealing at high temperatures was optimized for the potential applications of drinking water disinfection, wastewater effluent disinfection, and industrial waste stream treatment. The effectiveness of $Ti/SnO_2–Sb_2O_5$ anodes prepared under different conditions was evaluated by using hexanol as a probe molecule to measure the extent of oxidative reactions, and anode performance was monitored by cyclic voltammetry. A large factorial matrix consisting of tin chloride concentration × antimony chloride concentration × annealing temperature was first evaluated, and the optimum conditions were found to be 20% tin chloride and 1% antimony chloride in the dip solution and an annealing temperature of 500°C. Further investigation showed that the rate of withdrawal from the dip solution, the number of coatings of the dip solution, and the addition of oxygen during annealing did not significantly affect anode performance. Under optimum preparation conditions, $Ti/SnO_2–Sb_2O_5$ anodes showed no loss of performance over 1,280 cycles of cyclic voltammetry, suggesting that their performance can be sustained over long periods of use. The result of this research is a simple preparation method for effective and long-lived $Ti/SnO_2–Sb_2O_5$ anodes; this method could be easily adopted by a utility for pilot- or full-scale disinfection of water and wastewater and the treatment of industrial waste streams.

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1 Introduction

Numerous chemical and physical agents have been used for the disinfection of drinking water and wastewater effluents; however, all of the processes currently in use have disadvantages. Chlorine is the most commonly used disinfectant because of its low cost; however, it is not effective against *Giarda* and *Cryptosporidium* [1]. Furthermore, the formation of chlorinated byproducts [2, 3] and the possibility of tank ruptures and the release of toxic gas to the atmosphere [4] create significant safety concerns. Ozone and ultraviolet (uv) light are sometimes used as alternatives to chlorine, but these technologies are limited by high operation and maintenance costs [5]. As an alternative to these technologies, several electrochemical systems have recently been studied for water disinfection using electrodes composed of titanium [6], carbon cloth [7, 8], graphite [9], titanium nitride [10], platinum [11], platinum coated with niobium [12], titanium coated with ruthenium oxide (RuO_2) [13–16], and boron-doped diamond [17, 18]. Hydroxyl radical generated at the anode surface is likely the disinfecting agent in these systems [12, 19, 20].

Titanium anodes coated with antimony-doped tin oxide ($Ti/SnO_2–Sb_2O_5$) also generate hydroxyl radical [21, 22]. Recently, Loge et al. [23] investigated the efficacy of $Ti/SnO_2–Sb_2O_5$ anodes for wastewater effluent disinfection. They were able to achieve multi-log disinfection of coliform bacteria, and provided an economic analysis demonstrating that full-scale effluent disinfection with

Ti/SnO_2–Sb_2O_5 anodes may be more economical than uv disinfection. Their results suggest that Ti/SnO_2–Sb_2O_5 anodes have a high degree of potential for the full-scale disinfection of wastewater effluents. These anodes have also been used to treat COD in wastewater [24] and to treat landfill leachate resulting in decolorization, COD removal, and complete elimination of ammonia [25]. Furthermore, tin oxide and antimony-doped tin oxide anodes can also oxidize organic compounds such as phenol [26–28] *p*-methoxyphenol [29], pentachlorophenol [30], bisphenol-A [31], aniline [32], and acetominophen [33]. These results suggest that Ti/SnO_2–Sb_2O_5 anodes can destroy organic contaminants in water simultaneously with the inactivation of pathogens.

Results to date obtained in bench scale investigations suggest that Ti/SnO_2–Sb_2O_5 anodes may provide an effective and economical alternative to other treatment systems for contaminant destruction and water and wastewater effluent disinfection. These anodes appear to have the greatest potential for full-scale application in water treatment and wastewater effluent systems that use single chamber systems, such as plug-flow (PF) or continuously stirred tank reactors. Antimony-doped tin oxide anodes have a significant advantage over other electrode systems for such single chamber applications because of the minimal back reactions that occur at the cathode [34]. However, scale-up and long-term use of Ti/SnO_2–Sb_2O_5 anode systems require that the electrodes be prepared using conditions that provide the most effective and efficient performance. Antimony-doped tin oxide anodes have previously been prepared on titanium substrates by spray pyrolysis [35], sol-gel application [36], chemical vapor deposition [37], and manual application of tin–antimony solutions using a paintbrush [38]; however, Loge et al. [23] reported that mechanical dip-coating of titanium rods in a tin–antimony solution was a simple and cost-effective preparation method. Because Ti/SnO_2–Sb_2O_5 anodes are not currently commercially available, utilities would need an effective method to prepare the anodes for pilot- or full-scale use. The purpose of this research was to determine the most effective conditions for the preparation of such dip-coated Ti/SnO_2–Sb_2O_5 anodes for the electrochemical disinfection of drinking water and wastewater effluents and the treatment of industrial waste streams.

2 Experimental

2.1 Materials

Hexanol, carbon tetrachloride, chloroform, hexachloroethane, hexane, sodium bicarbonate, sodium sulfate, sodium perchlorate, isopropanol, tin chloride pentahydrate, antimony chloride, and tin sulfate were purchased from Sigma-Aldrich. Hydrochloric acid and sulfuric acid were obtained from J.T. Baker. Titanium rods (2 mm diameter) were purchased from Goodfellow Metals.

2.2 Development of preparation methods for Ti/SnO_2–Sb_2O_5 anodes

The initial conditions for investigating the preparation of Ti/SnO_2–Sb_2O_5 anodes were based on the procedures of Lipp and Pletcher [38] and Loge et al. [23]. Titanium rods were prepared by bead blasting, sonication in isopropanol, boiling in 11.5 M HCl, rinsing in distilled water, and air drying. Layers of tin–antimony were then applied by dipping the rods in isopropanol containing tin chloride and antimony chloride, drying at 100°C for 10 min, and annealing in a muffle furnace for 20 min.

Because Ti/SnO_2–Sb_2O_5 anodes are not commercially available, a matrix of conditions was employed to evaluate the most effective procedure for preparing the anodes. The variables investigated included tin chloride concentration in the dipping solution (5–20 mg L^{-1}), antimony chloride concentration in the dipping solution (0.1–10 g L^{-1}), annealing temperatures (400–600°C), and oxygen flow rates during annealing (0–8 L min^{-1}). Anodes were dried for 20 min between each dip. After the preparation of a group of eight anodes was complete, the anodes were placed in a holder fitted to a 200 mL batch reactor matched with stainless steel cathodes. The anodes and cathodes were connected to a Tenma 72-4045A DC power source with constant current of 200 mA. The current was monitored, and current falling below 200 mA signified the end of the anode service life. Anode service lifetimes are listed in Table 1.

2.3 Reactor systems

Two reactor systems were used to investigate the oxidation effectiveness of Ti/SnO_2–Sb_2O_5 anodes. A single chamber reactor was used to assess the effectiveness of the anodes under the practical constraints of a typical effluent disinfection system; e.g., a PF reactor in which both the anodes and the cathodes are configured in a single reactor. In addition, a dual chamber reactor was used for cyclic voltammetry. The dual chamber reactor consisted of two 200 mL beakers joined by a fitted glass tube fit together by a junction holding an o-ring and a bracket that held the junction in place. A Nafion membrane was placed in the junction to separate reactions at the anode and cathode. The anodes and cathodes were connected to a cyclic voltammeter. All reactions were conducted in triplicate, and control systems were evaluated in the same reactors without the application of current.

Table 1 Factorial optimization of tin oxide anode preparation

Tin chloride/mass/volume	Annealing temperature/°C	Service life/h
Antimony chloride = 0.1% (mass/volume)		
5	500	18
5	550	16
5	600	1
10	500	84
10	550	66
10	600	0.9
20	500	120
20	550	88
20	600	1.0
Antimony chloride = 0.5% (mass/volume)		
5	500	6
5	550	47
5	600	<0.1
10	500	119
10	550	110
10	600	<0.1
20	400	<0.1
20	550	124
20	600	2
Antimony chloride = 1.0% (mass/volume)		
5	500	108
5	550	114
5	600	6.5
10	500	98
10	550	114
10	600	27
20	500	>180
20	550	174
20	600	22
Antimony chloride = 2.0% (mass/volume)		
5	500	34
5	550	57
5	600	24
10	500	55
10	550	96
10	600	8
20	450	8
20	500	112
20	550	144
20	600	24

2.4 Evaluation of oxidation by Ti/SnO_2–Sb_2O_5 anodes

1-Hexanol was chosen for its high reactivity with OH• and other oxidants ($k_{OH\bullet} = 6.0 \times 10^9$ M^{-1} s^{-1} [39]), and its low potential for reactivity with reductants ($k_{e^-} = < 10^6 M^{-1} s^{-1}$ [39]) [40]. Oxidation experiments were conducted using a solution of 1 M sodium sulfate and 1 mM 1-hexanol. 1-Hexanol was analyzed on a Hewlett-Packard 5890 gas chromatograph using a DB-5 capillary column with flame ionization detection. The injector port and detector port temperatures were 240 and 350°C, respectively. The initial oven temperature was 70°C, the program rate was 20°C min^{-1}, and final temperature was 210°C.

2.5 Effect of number of SnO_2–Sb_2O_5 layers

Antimony-doped tin oxide anodes were prepared with 8, 16, and 24 coatings of the dipping solution using the optimum tin chloride concentration, antimony chloride concentration, drying time, and oxygen flow rate during annealing. Each of the sets of anodes was then evaluated for effectiveness by monitoring hexanol oxidation.

2.6 Effect of dip withdrawal rate

The effect of the rate at which the titanium rods were withdrawn from the tin chloride–antimony chloride solution was investigated using a pulley system with a variable speed motor. Five different rates were used: 0.03, 0.1, 0.3, 0.7, and 1.7 cm s^{-1}. The performance of the anodes was evaluated by cyclic voltammetry and the activity was evaluated by measuring the oxidative loss of hexanol in dual chamber reactors.

2.7 Anode service life

A Ti/SnO_2–Sb_2O_5 anode prepared using the optimal conditions was evaluated by continuous cyclic voltammetry. The cycles were repeated continuously for 1,280 cycles.

2.8 Cyclic voltammetry

Antimony-doped tin oxide anodes prepared under various conditions were characterized by cyclic voltammetry using an EG&G Model 173 potentiostat and EG&G Model 175 universal programmer. The reactions were conducted in a two-compartment cell; the anode being evaluated and a calomel reference electrode were suspended in one cell, and a platinum wire cathode was suspended in the other cell. The electrolyte in the cells consisted of an aqueous solution of 0.5 M sodium sulfate and 10 mM potassium ferricyanide. Voltammagrams were conducted at scan rates of 5, 10, 20, and 50 mV s^{-1}, with the exception of the repeated voltammagrams for assessing anode longevity, which were conducted using a scan rate of 20 mV s^{-1}.

2.9 Statistical analysis

The most common data obtained in this study featured concentrations of a constituent as a function of time affected by a given variable (e.g., the rate of oxidation of hexanol affected by different anode preparation methods). These data were analyzed using completely randomized design analysis of variance with one way treatment structure and repeated measures using a 95% confidence interval.

3 Results and discussion

3.1 Optimization of Ti/SnO_2–Sb_2O_5 anode preparation

A three-level factorial design was used to determine the optimal conditions for Ti/SnO_2–Sb_2O_5 anode preparation. The factors evaluated included tin chloride concentration × antimony chloride concentration × annealing temperature. The most effective results from the optimization are listed in Table 1. Anode service lifetimes varied significantly based on the preparation conditions. For all doses of tin chloride and antimony chloride, annealing temperatures <500°C resulted in short service lives for the anodes (i.e., <0.5 h). Furthermore, annealing at 600°C also resulted in short service lifetimes of <0.1–24 h. Depending on the tin chloride and antimony chloride concentrations in the dipping solution, 500 or 550°C was the optimum annealing temperature. The most effective condition for Ti/SnO_2–Sb_2O_5 anode preparation was 20% tin chloride, 1% antimony chloride, and an annealing temperature of 500°C; this formulation is slightly different from the 2% antimony chloride reported in a previous study [23]. The optimized conditions provided a service life that exceeded the 180 h of testing.

3.2 Effect of oxygen flow rate during anode annealing

The effect of the oxygen flow rate on the oxidation effectiveness of the Ti/SnO_2–Sb_2O_5 anodes, using hexanol as a probe molecule for oxidant detection, is shown in Fig. 1. Statistical analyses confirmed that the oxidant generation rates for each of the two air flow rates used in the annealing process were not significantly different. These results indicate that there was no significant difference in the effect of air flow rates on Ti/SnO_2–Sb_2O_5 anode performance, and suggest that the ambient oxygen concentration in the muffle furnace (i.e., 21%) is sufficient to promote the oxidation of tin (II) to tin (IV) oxide. Therefore, a supplemental oxygen supply would not be needed at a decentralized location, such as at a utility public water facility, to prepare Ti/SnO_2–Sb_2O_5 anodes with optimal performance characteristics.

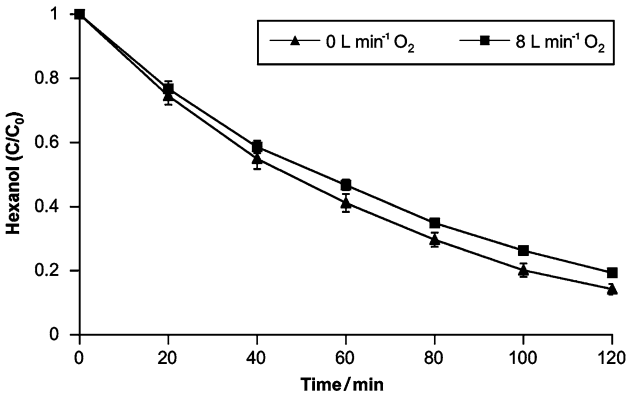

Fig. 1 Relative oxidant activity of Ti/SnO_2–Sb_2O_5 anodes, measured by loss of the oxidant probe hexanol, with and without oxygen flow during annealing. Experimental conditions: single chamber reactor containing 1 M sodium sulfate and 1 mM hexanol, with 200 mA applied current

3.3 Effect of number of tin–antimony coatings on anode performance

The effect of the number of tin chloride–antimony chloride coatings on oxidant activity was evaluated using anodes prepared under the optimum preparation conditions of 20% tin chloride, 1% antimony chloride, and an annealing temperature of 500°C. Antimony-doped tin oxide anodes prepared with 8, 16, and 24 layers of tin chloride–antimony chloride coatings were evaluated for oxidative activity using hexanol as the oxidant probe. The effect of the number of coatings on oxidant generation is shown in Fig. 2. These data suggest that the

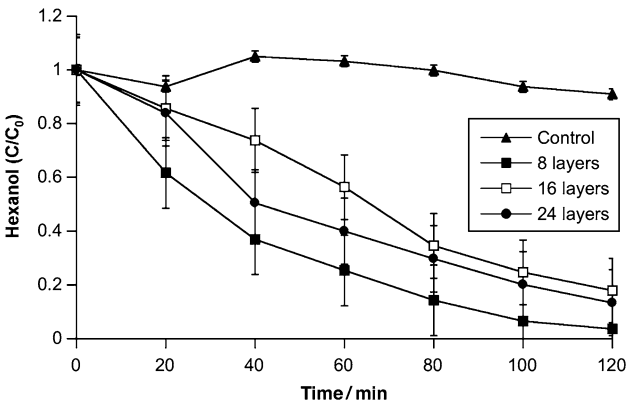

Fig. 2 Relative oxidant activity of Ti/SnO_2–Sb_2O_5 anodes, measured by loss of the oxidant probe hexanol, for anodes with 8, 16, and 24 layers of tin oxide coating. Experimental conditions: single chamber reactor containing 1 M sodium sulfate and 1 mM hexanol, with 200 mA applied current (experiments) or no current (control)

number of tin–antimony coatings had no effect on oxidant activity. Statistical analysis confirmed that the oxidant generation rates for each of the three numbers of coatings were not significantly different. The results shown in Fig. 2 were also confirmed by evaluating cyclic voltammograms on Ti/SnO_2–Sb_2O_5 anodes prepared with the three different levels of coatings (Fig. 3a–c). These three essentially identical voltammograms confirm that the increased numbers of tin–antimony coatings had minimal effect on the characteristics of the Ti/SnO_2–Sb_2O_5 anodes.

3.4 Effect of dip withdrawal rate

The effect of anode withdrawal rate from the tin–antimony dip solution was evaluated by measuring oxidant activity using hexanol as an oxidant probe and by comparing cyclic voltammograms. The oxidation of hexanol in dual chamber reactors for dip withdrawal rates of 0.03, 0.1, 0.3, 0.7, and 1.7 cm s^{-1} are shown in Fig. 4a–e. These data suggest that dip withdrawal rate had no effect on the potential of Ti/SnO_2–Sb_2O_5 anodes to generate oxidants. Statistical analyses confirmed that the oxidant generation rates for each of the five dip withdrawal rates were not significantly different. The influence of dip rate was also investigated using cyclic voltammetry. The voltammograms were essentially identical, which in concert with the data of Fig. 4, indicates that dip solution withdrawal rate did not have a significant influence on anode characteristics and the relative degree of oxidant production. The initial hypothesis related to the influence of dip withdrawal rate was that a slower dip rate might provide more uniform coating of the anodes. However, dip withdrawal rate had no effect on Ti/SnO_2–Sb_2O_5 anode performance, which will simplify their preparation.

3.5 Anode longevity

Varying results have been reported for the service life of Ti/SnO_2–Sb_2O_5 anodes. Correa-Lozano et al. [41] reported a service life of less than 12 h for Ti/SnO_2–Sb_2O_5 anodes at a current density 100 mA cm^{-2} when used for the electrolysis of aqueous acidic solutions. The deactivation of these electrodes has been proposed to be due to the formation of a nonconductive tin hydroxide in the outer layer of the anode [42, 43]. Chen et al. [44] reported that Ti/SnO_2–Sb_2O_5 anodes underwent some irreversible damage within seconds, as measured by ability to treat chemical oxygen demand, although they were still able to treat COD by 75% in subsequent usage. In contrast, Lipp and Pletcher [38] found that Ti/SnO_2–Sb_2O_5 anodes showed no change in function over 48 days, and scanning electron microscopy showed no change in the surface appearance of the anodes. In addition, Loge et al. [23] reported a service lifetime of greater than 30 days for Ti/SnO_2–Sb_2O_5 anodes.

A long service life of Ti/SnO_2–Sb_2O_5 anodes is critical for these electrochemical systems to be cost-effective.

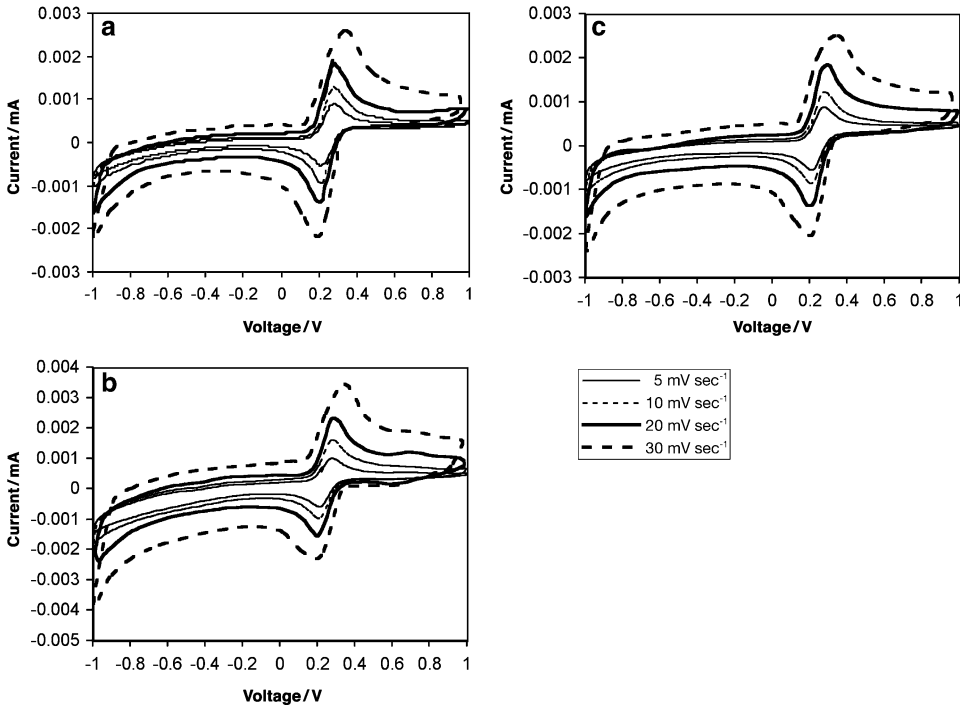

Fig. 3 Cyclic voltammograms for Ti/SnO_2–Sb_2O_5 anodes with variable numbers of coating layers; **a**, 8 coatings; **b**, 16 coatings; **c**, 24 coatings

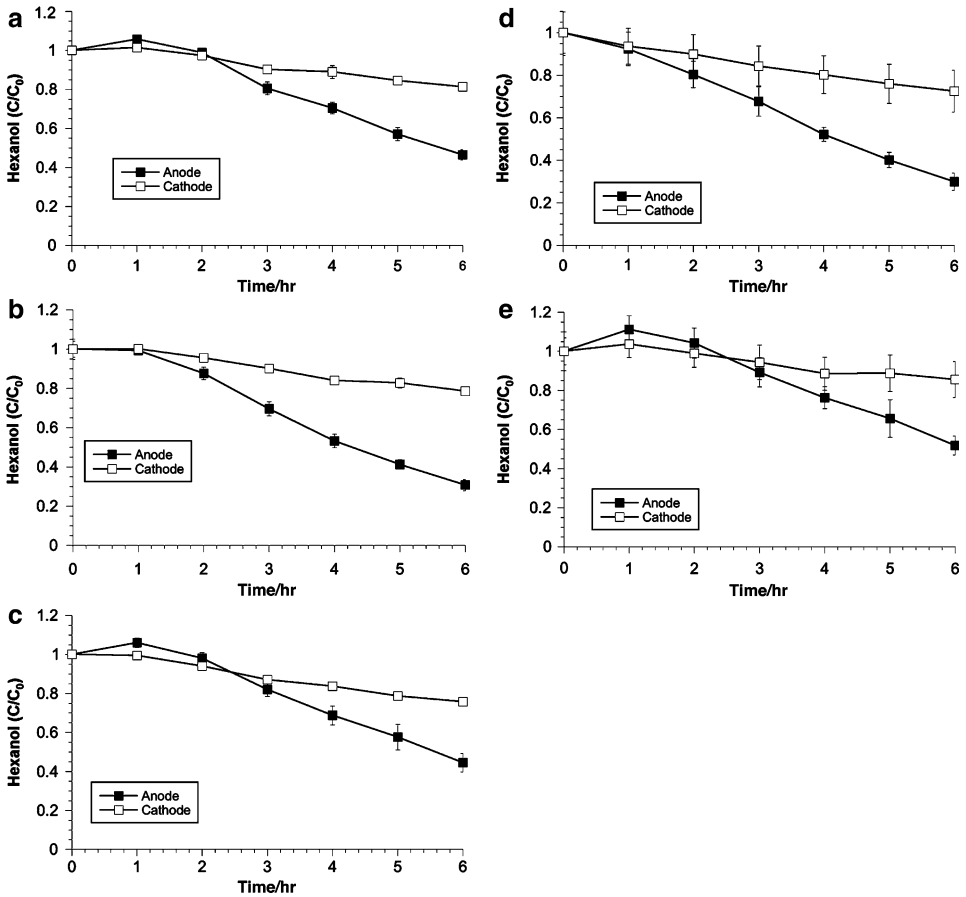

Fig. 4 Relative oxidant activity quantified by loss of hexanol in the anode cell and the cathode cell for Ti/SnO_2–Sb_2O_5 anodes withdrawn from the tin chloride–antimony chloride–isopropanol solution at different rates; **a**, 0.03 cm s^{-1}; **b**, 0.1 cm s^{-1}; **c**, 0.3 cm s^{-1}; **d**, 0.7 cm s^{-1}; **e**, 1.7 cm s^{-1}. Experimental conditions: dual chamber reactor containing 1 M sodium sulfate and 1 mM hexanol, with 200 mA applied current

Anodes studied for the opitmization matrix (Table 1) were tested for only 180 h; therefore, an anode prepared using the optimum conditions was evaluated by repeating cyclic voltammetry through 1,280 cycles. The voltammograms were graphed sequentially on 26 separate figures, all of which were nearly identical; the first and last 50 cycles are shown in Fig. 5a–b. These results confirm a long service life for Ti/SnO_2–Sb_2O_5 anodes prepared using the optimum conditions described in this study. The wide-ranging results reported in previous studies, many of which are in contrast to the results of this investigation, indicate that the preparation method for Ti/SnO_2–Sb_2O_5 anodes is critical for effective performance and longevity. The results of this study demonstrate that Ti/SnO_2–Sb_2O_5 anode longevity can be increased significantly using the conditions reported in this paper. Antimony-doped tin oxide anodes are not commercially available; therefore, they would need to be prepared by a utility or other entity in a pilot or demonstration disinfection study. The results of this research would streamline Ti/SnO_2–Sb_2O_5 anode preparation by minimizing an emphasis on some parameters (e.g., number of coatings, dip withdrawal rate, oxygen flow rate) while focusing on the composition of the dip solution and the annealing temperatures.

4 Conclusions

The preparation of Ti/SnO_2–Sb_2O_5 anodes was investigated and the performace of anodes prepared by different methods was evaluated in single- and dual-chamber reactors and by cyclic voltammetry. The preparation of Ti/SnO_2–Sb_2O_5 anodes was optimized by varying the tin chloride and antimony chloride concentrations in the dip solution, the rate at which the titanium rods were withdrawn from the dip solution, the number of coatings of the dip solution, the annealing temperature, and the oxygen flow rate during annealing. The results show that the concentrations of tin chloride and antimony chloride in the dipping solution, in conjunction with the annealing temperature, have a significant effect on Ti/SnO_2–Sb_2O_5 anode performance. The optimum condition for the preparation of Ti/SnO_2–Sb_2O_5 anodes consisted of 20% (mass/volume) tin chloride and 1% (mass/volume) antimony chloride in the dip solution and an annealing temperature of 500°C. However, varying the number of tin chloride–antimony chloride coatings between 8 and 24 had minimal effect. Similarly, the rate of withdrawal from the dipping solution and the oxygen flow rate during annealing had no effect on anode performance. Under optimum preparation

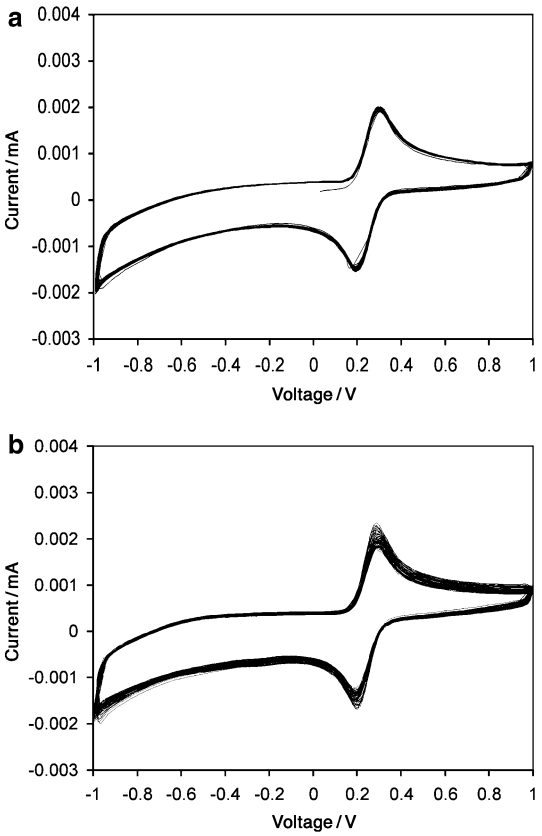

Fig. 5 Cyclic voltammograms for a Ti/SnO_2–Sb_2O_5 anode prepared using optimum conditions and subjected to repeating cyclic voltammetry at 20 mV s^{-1} for 1,280 cycles; **a**, the first 50 cycles; **b**, the last 50 cycles

conditions, Ti/SnO_2–Sb_2O_5 anodes showed no loss of performance over 1,280 cycles of cylcic voltammetry, suggesting that their performance can be sustained over long periods of use.

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