



# Preparation of a coated Ti anode for producing acidic electrolyzed oxidizing water



Wenwei Tang<sup>a,\*,1</sup>, Yanfei Li<sup>b,1</sup>, Weihao Li<sup>c</sup>, Xuejin Chen<sup>b,\*\*</sup>, Xiping Zeng<sup>d,\*\*\*</sup>

<sup>a</sup> Department of Chemistry, Shanghai Key Laboratory of Chemical Assessment and Sustainability, Tongji University, 1239 Siping Road, Shanghai, 200092, China

<sup>b</sup> School of Medicine, Shanghai Jiao Tong University, South Chongqing Road, Shanghai, 200025, China

<sup>c</sup> Handan Municipal Centre for Disease Control and Prevention, Handan, 056000, China

<sup>d</sup> School of Life Science and Technology, Tongji University, Shanghai, 200092, China

## ARTICLE INFO

### Article history:

Received 11 May 2015

Received in revised form

5 September 2015

Accepted 25 October 2015

Available online 30 October 2015

### Keywords:

Acidic electrolyzed oxidizing water

RuO<sub>2</sub>–SnO<sub>2</sub>–TiO<sub>2</sub>

Electrochemical properties

Disinfection effect

## ABSTRACT

Coated Ti anodes have been prepared for producing acidic electrolyzed oxidizing water (AEOW). To improve the performance of anodes, the influence factors (sintering temperature, heat treatment time and dip-coating times) were systematically explored. The results show that the optimum sterilizing effect of AEOW is obtained in the case of sintering at 450 °C for 1 h with as many dip-coatings as possible. The sintering temperature is a critical factor. A lower temperature leads to better reaction selectivity, whereas a higher temperature results in a longer lifetime. A short heat treatment time leads to inadequate coating oxidation, a long heat treatment time prolongs the lifetime but results in high SEC. Increasing the number of dip-coatings could enhance the coating-substrate adhesion, thus extending the anode lifetime. The electrode prepared by optimization could replace the platinized Ti anode to produce AEOW. AEOW could be used as an effective disinfectant for eliminating *Escherichia coli* and *Staphylococcus aureus*. The sterilization ability is based on the combined effect of active chlorine concentration (ACC), low pH and high oxidation–reduction potential (ORP).

© 2015 Elsevier Ltd. All rights reserved.

## 1. Introduction

In recent years, large amounts of infectious diseases and drug-resistant bacteria have emerged due to globalization, deteriorating environment and anti-biotic abuse, and are striking people frequently by sudden and severe epidemic diseases. Therefore, we must implement highly effective disinfection systems in the food industry to guarantee food safety, prevent infectious diseases and create a healthy living environment.

Acidic electrolyzed oxidizing water (AEOW) has a high oxidation–reduction potential (ORP) of over 1100 mV, a low pH of 2–3, and an active chlorine concentration (ACC) of 30–100 mg/L and is characterized by its high efficiency, broad spectrum antimicrobial activity, low cost and high-level security (Udompijitkul, Daeschel, &

Zhao, 2007). AEOW is currently applied in various fields (Arealos-Sánchez et al., 2013; Wang et al., 2014; Zhang et al., 2011). It can effectively kill common bacteria, such as *Salmonella*, *Escherichia coli*, *Vibrio parahaemolyticus*, and *Listeria monocytogenes* in food sterilization (Ding, Rahman, Purev, & Deog-Hwan, 2010; Issa-Zacharia, Kamitani, Miwa, Muhimbula, & Iwasaki, 2011; Xie, Sun, Pan, & Zhao, 2012), reduce the rate of bacterial infection and control the probability of food decay in food storage and preservation (Audenaert et al., 2012; Jadeja, Hung, & Bosilevac, 2013), and improve livestock's immunity and antiviral ability in farm disinfecting (Fasenko, Christopher, & McMullen, 2009; McCarthy & Burkhardt, 2012).

However, the application of AEOW is still limited due to its high production cost. To solve this problem, it is necessary to develop an inexpensive and effective process to prepare qualified electrode to replace the traditional expensive Pt-plated Ti electrode. Recently, a low-cost RuIrTi-coated Ti electrode was developed to produce AEOW with similar performance as that of Pt-plated Ti electrode (Cheng, Zhang, & Cui, 2007). Zeng et al. (Zeng, Chen, Wu, Wang, & Soowohn, 2007) studied the effect of IrO<sub>2</sub> loading on RuO<sub>2</sub>–TiO<sub>2</sub> anodes. These studies improved the performance of electrodes and

\* Corresponding author.

\*\* Corresponding author.

\*\*\* Corresponding author.

E-mail addresses: [tangww@tongji.edu.cn](mailto:tangww@tongji.edu.cn) (W. Tang), [chenxuejin@shsmu.edu.cn](mailto:chenxuejin@shsmu.edu.cn) (X. Chen), [zengxp@tongji.edu.cn](mailto:zengxp@tongji.edu.cn) (X. Zeng).

<sup>1</sup> These authors contributed equally to this work.

were helpful to the development of RuO<sub>2</sub>–SnO<sub>2</sub>–TiO<sub>2</sub> electrodes used in AEW generation. As reported (Cui, Feng, & Liu, 2009; Depauli & Trasatti, 1995; Smith & Okinaka, 1983), introducing Sn into coated Ti electrodes can refine the coating grains and improve the coating-substrate adhesion and stability of the active elements. In addition, the partial replacement of precious metals such as Ru and Ir by Sn, can significantly reduce the cost of anodes. A series of nano-scale RuO<sub>2</sub>–SnO<sub>2</sub>–TiO<sub>2</sub>-coated anodes, determined as an optimum formula of Ru<sub>25</sub>Sn<sub>7</sub>Ti<sub>68</sub> (Zeng, Zhao, Tang, & Zheng, 2011b; Zeng et al., 2012) have been prepared in our laboratory by using the sol–gel method with n-butyl titanate as the raw material and using the modified Pechini sol–gel method with RuCl<sub>3</sub>·3H<sub>2</sub>O and SnCl<sub>4</sub>·5H<sub>2</sub>O as the raw materials.

To achieve improved electrochemical performance (low chlorine evolution, low oxygen evolution potential, low specific energy consumption and long lifetime) and develop a less expensive nanoscale RuO<sub>2</sub>–SnO<sub>2</sub>–TiO<sub>2</sub>-coated anode, further studies on the preparation conditions of coated Ti anodes, including the sintering temperature, heat treatment time and number of dip-coatings, have been conducted based on the optimum formula in this paper. The process parameters were confirmed by means of chlorine evolution potential test, cyclic voltammetry test, accelerated life and the performance of AEW produced by the coated Ti anode. The disinfection capacity, and effect of AEW on *Escherichia coli* and *Staphylococcus aureus* (*E. coli* and *S. aureus*) was examined by suspension quantitative germicidal test. Furthermore, the membrane permeability of *S. aureus* was investigated by fluorescence microscopy.

## 2. Materials and methods

### 2.1. Preparation of the coated Ti anode

#### 2.1.1. Pretreatment of the Ti substrate

The electrode was prepared with a 20 × 20 × 1 mm titanium plate (Shanxi Ti Company, China) as the substrate. The polished titanium plates were degreased in detergent (0.1%, v/v) and acetone (5%, v/v) for 15 min with ultrasound, respectively. Then they were immersed in sulfuric acid/nitric acid (1:1, v/v) for 2 h. After that, the plates were etched with boiling oxalic acid (10%, w/w) for 2 h. Finally, the pretreated Ti plate was stored in ethyl alcohol after cleaning in distilled water (Song, Zhan, & He, 2010; Yang, Zou, Huo, Cai, & Xiao, 2009).

#### 2.1.2. Preparation of the coating materials

A TiO<sub>2</sub> sol was prepared as follows: 10 mL tetra-n-butyl titanate (TBTA) was dissolved in 5 mL isopropanol, and 3 mL acetylacetone was added as inhibitor. After vigorous stirring for 0.5 h, the solution was marked as solution A. A mixture of 2 mL distilled water, 2.5 mL isopropanol and a small amount of acetic acid was denoted as solution B. And solution C was composed of PEG-4000 (5%, w/w) and 2.5 mL isopropanol. Solution B was added dropwise into solution C under stirring, then solution A was also added dropwise. TiO<sub>2</sub> sol was obtained by stirring the mixture for 0.5 h and aging overnight.

RuO<sub>2</sub> and SnO<sub>2</sub> sols were prepared by a modified Pechini sol–gel method. The precursor solution was prepared by dissolving citric acid (CA) in ethyl alcohol. RuCl<sub>3</sub>·3H<sub>2</sub>O and SnCl<sub>4</sub>·5H<sub>2</sub>O were added to the precursor solution in a CA: Ru (Sn) molar ratio of 3:1.

The RuO<sub>2</sub> and SnO<sub>2</sub> sols were added to the TiO<sub>2</sub> sol to improve the stability and to form a homogeneous sol.

#### 2.1.3. Depositing of the Ti substrate

A Ti sheet was dipped vertically into the previously formed sol solution and elevated at a slow speed, then dried under an infrared lamp for 15 min. After the 5 dip-coatings, the sheet was annealed at

450 °C for 10 min in a muffle furnace to oxidize the coating layer completely. By repeating this process, a series of Ti substrates with different oxide coatings was obtained after placing the substrates in the muffler for 1 h at 450 °C.

### 2.2. Characterization of the coated Ti anode

#### 2.2.1. Chlorine evolution potential test

All electrochemical tests were conducted with an AUTOLAB model PGSTAT30.

A three-electrode testing system featuring a Luggin capillary with a salt bridge filled with a saturated KCl solution was constructed. A saturated calomel electrode served as the reference electrode, the prepared electrode as the anode and a Pt electrode as the cathode. Electrochemical tests were conducted in an H-type electrobath with a saturated NaCl solution. The fast linear scanning method was employed over a potential range of 0–1.5 V at a scanning speed of 50 mV s<sup>-1</sup>.

#### 2.2.2. Oxygen evolution potential test

Using a saturated calomel electrode as the reference electrode, a double salt bridge was constructed using a saturated KCl solution and a 0.5 mol L<sup>-1</sup> K<sub>2</sub>SO<sub>4</sub> solution. The salt bridge featured a Luggin capillary, and a three-electrode testing system was built as previously described. The H-type electrobath was filled with a 0.5 mol L<sup>-1</sup> H<sub>2</sub>SO<sub>4</sub> solution. The testing parameters were identical to those used in the chlorine evolution potential test.

#### 2.2.3. Cyclic voltammetry test

The tests were conducted in a saturated NaCl solution and a 0.5 mol L<sup>-1</sup> H<sub>2</sub>SO<sub>4</sub> solution, respectively, over a potential range of –0.25 to –1.3 V, at scanning speeds of 10, 20, 50, 100 and 200 mv s<sup>-1</sup>.

#### 2.2.4. Accelerated lifetime test

Using a 1.0 mol L<sup>-1</sup> H<sub>2</sub>SO<sub>4</sub> solution as the testing solution, a pure Ti plate as cathode, and the prepared electrode as anode, the electrolysis was conducted under a constant current of 2 A cm<sup>2</sup> at 40 °C, and the inter-plate distance was set at 1.5 cm. The accelerated lifetime of the electrode was then recorded when a 10 V elevation over the initial bath voltage was reached.

### 2.3. Generation and specific energy consumption (SEC) of AEW

An 8.0 g L<sup>-1</sup> NaCl solution was added to the self-made static cell (500 mL, made by polytetrafluoroethylene), then was electrolyzed at 8.0 V. The prepared electrodes worked as anode and cathode with a spacing of 5 mm. The ORP, pH and SEC of the AEW were tested and determined.

SEC is defined as the energy consumption of an electrolysis system that produces a unit mass of total oxidative substance (measured by ACC), calculated as follows:

$$SEC(\text{kW}\cdot\text{h}/\text{kg}) = \frac{UIt}{cV} \times 1000 \quad (1)$$

*U* – Average voltage of electrolysis (V); *I* – Average current of electrolysis (A); *t* – Electrolysis time (h); *c* – Concentration of the total oxidative substance (mg L<sup>-1</sup>); *V* – Volume of electrolyte (L).

### 2.4. Measurement of ACC (or TOS)

The iodometric method was used to measure TOS or ACC as follows: 10.00 mL disinfectant was added to a 150.00 mL conical beaker, and then 10.00 mL KI solution (10%) and 10.00 mL H<sub>2</sub>SO<sub>4</sub>

Download English Version:

<https://daneshyari.com/en/article/6401080>

Download Persian Version:

<https://daneshyari.com/article/6401080>

[Daneshyari.com](https://daneshyari.com)