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Carbon coated titanium electrode for the electrolytic decoloration of methylene blue

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ABSTRACT

Electrolysis is widely adopted in water treatment. The major challenge of electrolysis technology focuses on developing better electrode materials. In this study, we reported that the hydrothermal coating of titanium electrode with carbon (C-Ti electrode) could significantly improve the decoloration performance of Ti electrode. The decoloration kinetics of C-Ti electrode was much faster than that of naked Ti electrode at the same current density. The presence of chloride electrolytes, e.g. NaCl and KCl, was crucial for the efficient electrolysis. The optimized pH for C-Ti electrode was pH 7. Beyond methylene blue, C-Ti electrode showed high decoloration performance for other dyes, such as congo red, acidic blue 92 and remazol brilliant blue R. Mechanistically, the electrolytic oxidation of MB was mainly via indirect pathway, where Cl^- was oxidized into ClO^- for the oxidation of other substances. Moreover, the performance of C-Ti electrode largely remained in the recycling evaluations. The implication to the applications of C-Ti electrode in water treatment is discussed.

1. Introduction

Water pollution has been become the serious environmental problem nowadays, which hinders the development of human society [1,2]. Many technologies are involved in water treatment, such as electrolysis, active sludge method, advanced oxidation process and adsorption [3,4]. In particular, electrolysis is widely adopted in water treatment, because electrolysis is of relative low cost and general applicability [5–8]. Electrolysis can remediate antibiotics, heterocyclic compounds, heavy metals, and so on. The major efforts in the electrolytic remediation should be dedicated to develop high-performance electrode materials [9,10].

Recently, graphene is considered as promising material for electrode amendment, due to its extraordinary electrochemical properties [11–13]. Graphene amended electrodes have been used for the detection and remediation of pollutants with high performance [14–18]. However, graphene modification of electrode suffers several limits. Firstly, graphene modification was heterogeneous process, which makes the electrode surface heterogeneous. Secondly, the reproducibility of graphene modified electrode is poor, because the aggregation and stacking of graphene sheets are random. Thirdly, graphene is expensive, which largely increases the price of electrode. Despite these drawbacks, the experience of graphene modification of electrode suggests that carbon coating would definitely improve the performance of electrode. Therefore, new homogenous carbon modification method is demanded for electrode amendment.

Herein, we reported that the hydrothermal coating of carbon on Ti electrode (C-Ti electrode) could significantly improve the decoloration performance of naked Ti electrode. The decoloration performance was compared between C-Ti electrode and Ti electrode at the same current density. The visible absorption spectra of methylene blue (MB) solution during the electrolysis were recorded. The influences of electrolytes and pH values were investigated. Beyond MB, the electrolysis of other dyes, such as congo red (CR), acidic blue 92 (AB92) and remazol brilliant blue R (RBBR), was also measured. The mechanism of the electrolytic oxidation was preliminarily concerned. Moreover, the performance of C-Ti electrode in the recycling evaluations was determined. The implication to the applications of C-Ti electrode in water treatment is discussed.

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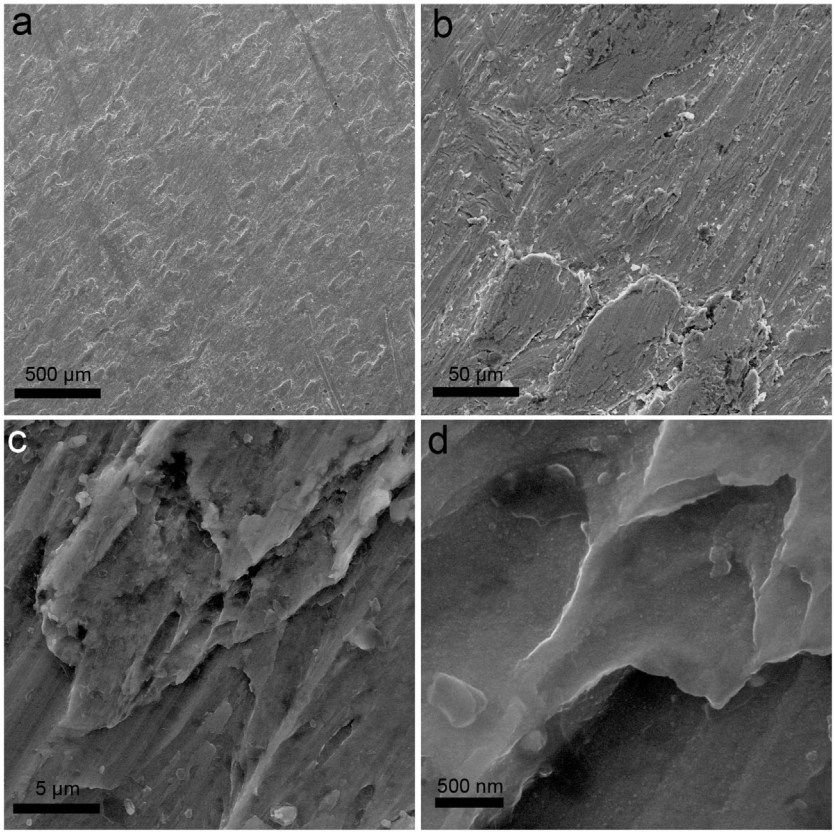

Fig. 1. SEM images of C-Ti electrode at ×100 (a), ×1000 (b), ×10000 (c) and ×40000 (d) magnifications.

2. Materials and methods

2.1. Materials

Graphite, glucose and MB were obtained from Sinopharm Chemical Reagent Co. Ltd., China. Congo red and acidic blue 92 were provided by J&K Scientific Ltd., China. Remazol brilliant blue R was obtained from Sigma Co., USA. Flat Ti pieces (2 cm × 7 cm) were purchased from local market. The rest were of analytical grade. All reagents were used without purification.

2.2. Preparation of C-Ti electrode

Ti electrode was firstly corroded with acetic acid. Briefly, flat Ti pieces (2 cm × 7 cm) were immerged into acetic acid. The solution was heated to 80 °C for 3 h. Then, the Ti pieces were washed with deionized water to remove the remnant acetic acid. The Ti pieces were dried under vacuum to give naked Ti electrode.

Naked Ti electrode was placed in a Teflon tube and added with 80 mL of glucose solution (1000 mg/80 mL). The mixture was sonicated for 30 min and then hydrothermally treated at 120 °C for 6 h to reach the carbonization of glucose. After cooling to room temperature, the C-Ti electrode was washed with deionized water and dried under vacuum.

C-Ti electrode was characterized by scanning electron microscopy (SEM, Quanta 200FEG, FEI, Netherland) and X-ray photoelectron spectroscopy (XPS, Kratos, UK). The chronoamperometry (CA) of C-Ti and naked Ti electrode was measured on electrochemical workstation (CHI-604E, Shanghai Chenhua Instrument Co., Ltd.). Briefly, Ag/AgCl electrode was set as the reference electrode. Pt electrode was used as the counter electrode. The electrolyte was 0.2 M NaOH. The potential was set at 0.5 V and the current was recorded within 1 min.

2.3. Decoloration of MB

In the decoloration experiments, C-Ti electrode was set as the anode and the naked Ti electrode was set as the cathode. The distance of the two electrodes was 1.5 cm. At 298 K, 100 mL of MB (pH 7, 50 mg/L) was added and electrolyzed at the current density of 75 mA/cm^2 in the presence of NaCl (0.09 mol/L). At designed intervals, the solution was collected to measure the absorbance at 664 nm. The absorbance spectra of solutions were recorded. The electrolysis of MB by naked Ti electrode was performed following the same protocol by changing the anode to naked Ti electrode. The cycle voltammetry (CV) and electrochemical polarization curves of both electrodes were performed on electrochemical workstation with Ag/AgCl electrode as counter electrode. The decoloration of MB by C-Ti electrode was performed without electricity to evaluate the contribution of adsorption of MB on carbon.

2.4. Influence of electrolyte and pH

To investigate the influence of electrolyte, the electrolysis was performed in the presence of NaCl, KCl, or Na_2SO_4 (0.09 mol/L) and in the absence of electrolyte following aforementioned protocol. To investigate the influence of pH value, the electrolysis was performed at pH 1–11.

2.5. Decoloration of various dyes

The electrolytic decoloration of AB92, CR and RBBR was performed following the same protocol. The dye concentrations were 50 mg/L. The distance of electrodes was 1.5 cm. The pH was adjusted to 7. The concentration of NaCl was 0.09 mol/L. The current density was set as 75 mA/cm^2.