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基于微混合技术的可见光响应  $\text{TiO}_2$  光阳极  
的制备及其光电化学性能研究

Micromixing-assisted Fabrication and Photo-  
electrochemical Performance Investigation of  $\text{TiO}_2$   
Photoanodes with Visible Light Response

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**Micromixing-assisted Fabrication and Photo-  
electrochemical Performance Investigation of TiO<sub>2</sub>  
Photoanodes with Visible Light Response**



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## 摘要

基于  $\text{TiO}_2$  半导体特殊的光电化学性能，其有望成为绿色、环保的新型金属防腐技术中最为重要的光阳极材料。但是， $\text{TiO}_2$  存在可见光利用率低、光量子效率不高等限制其实际应用的问题。此外，由于传统混合方式在快速沉淀体系中难以有效调控均相化学过程，导致碱性液相环境制备有序  $\text{TiO}_2$  薄膜至今仍无法实现。基于上述考虑，本论文提出使用微混合设备辅助在碱性液相环境下制备出形貌可控的  $\text{TiO}_2$  薄膜光阳极，并通过沉淀剂对产物进行选择性的原位掺杂改性。通过数值模拟及实验测试深入研究了制备条件和掺杂对  $\text{TiO}_2$  薄膜光阳极光电化学性能的影响。本论文的主要研究内容和结果如下：

1、基于自行设计并制造的多孔分散微结构混合器，对液-液快速沉淀体系进行数值模拟及实验研究。结果表明，微混合器可在极短的时间内完成对液-液反应物的高效混合并有效抑制反应混合液中的均相化学反应，反应物流量、分散介质尺寸等对一次粒子的平均粒径及粒度分布有显著影响。

2、以微混合技术为背景，设计出一种在低温、碱性环境中高效合成有序锐钛矿纳米  $\text{TiO}_2$  薄膜的新工艺。实验发现，通过调节微观混合及液相沉积参数，产物的表面形貌可得到有效调控。当微观混合质量流率、沉积时间、温度分别为  $0.60 \text{ m/s}$ 、 $50 \text{ min}$ 、 $80 \text{ }^\circ\text{C}$  时，制得的  $\text{TiO}_2$  薄膜质量最佳。

3、通过调整 N 源浓度，制备出不同 N 掺杂物含量的  $\text{TiO}_2$  薄膜光阳极。结果表明，N 已进入  $\text{TiO}_2$  晶格并拓展了  $\text{TiO}_2$  的可见光响应区间， $\text{CO}(\text{NH}_2)_2$  浓度对薄膜生长效率和 N 掺杂能力有显著影响。N- $\text{TiO}_2$  光阳极具有优良的可见光诱导光电化学活性，在可见光照射下能对金属起到有效的光生阴极保护作用。

4、使用  $\text{Al}(\text{NO}_3)_3$  和  $\text{FeCl}_3$  分别作为 Al 源和 Fe 源，制备出 N/Al、N/Fe 共掺杂  $\text{TiO}_2$  薄膜光阳极。 $\text{Al}^{3+}$ 、 $\text{Fe}^{3+}$  进入  $\text{TiO}_2$  晶格并取代  $\text{Ti}^{4+}$  的位置，N/Al、N/Fe 共掺杂协同作用抑制了薄膜晶粒长大，提高了  $\text{TiO}_2$  光阳极的可见光响应活性和光量子效率，并抑制光生电子与价带中光生空穴的复合。

**关键词：**微混合技术； $\text{TiO}_2$  光阳极；掺杂改性；可见光响应；光电化学性能

## Abstract

TiO<sub>2</sub> semiconductor is expected as the most important photoanode material applied for the novel green and environmental friendly metal anticorrosion technology, because of its unique photoelectrochemical properties. Nevertheless, the inherent disadvantages of the TiO<sub>2</sub>, lies in their limited visible light utilization and quantum yield, have extremely restricted their practical applications. Moreover, the synthesis of well-ordered TiO<sub>2</sub> thin films from alkaline solution is still challenging due to the fact that the traditional mixing process possesses several disadvantages in controllable homogenous chemistry. Therefore, the work described in this paper is devoted to the preparation of TiO<sub>2</sub> thin film photoanodes from alkaline solutions with controllable morphology. In addition, several reactants are employed to doping anions and cations into the TiO<sub>2</sub> product. The effects of the operation conditions and doping modifications on the photoelectrochemical performance of the TiO<sub>2</sub> thin film photoanodes are investigated based on the numerical simulation and experimental measurements. The main research contents and results of this work were summarized as follows:

1. Based on the use of home-built micromixer, the liquid-liquid reaction system with fast precipitation process is investigated through numerical simulation and experimental measurements. It is found that the micromixer is advantageous for the ultrafast mixing at the microscale, and the homogeneous chemistry in the reactant solutions can be efficiently inhibited. The initial inlet velocity of the reactant solutions and the diameter of the microfiltration membrane have major impacts on the average size and distribution of the primary nano particles.

2. A novel micromixing-assisted route is designed to efficiently synthesize well-ordered anatase-TiO<sub>2</sub> thin films from alkaline solutions at low temperatures. The experimental results indicate that the surface morphology of the product can be controlled by carefully operating the parameters of micromixing and liquid phase deposition process. The quality of the of the surface morphology can be significantly

increased when the microxming mass flow rate, deposition time and preparation temperature is 0.60 m/s, 50 min and 80 °C, respectively.

3. The TiO<sub>2</sub> thin films photoanodes with various N contents are successfully synthesized through adjusting the concentration of the N-containing compounds. It is found that the N is well incorporated into the TiO<sub>2</sub> lattice and thus shifting the absorption zone of the TiO<sub>2</sub> to the visible domain. Intriguingly, we find that the concentration of CO(NH<sub>2</sub>)<sub>2</sub> plays a significant role in affecting the films growth rate and the N-doping capability. The as-prepared N-TiO<sub>2</sub> photoanodes exhibit excellent visible light-induced photoelectrochemical activities and can provide an effective photogenerated cathodic protection for the coupled metal electrode.

4. The TiO<sub>2</sub> thin films photoanodes co-doped with N/Al and N/Fe are successfully synthesized by using Al(NO<sub>3</sub>)<sub>3</sub> and FeCl<sub>3</sub> as Al and Fe source, respectively. The results indicate that the Ti<sup>4+</sup> in TiO<sub>2</sub> lattice is substituted by Al<sup>3+</sup> and Fe<sup>3+</sup> ions. The synergistic effect of N/Al and N/Fe co-doped has the benefit to restrain grain growth, as well as improving the visible light response activity of the TiO<sub>2</sub> photoanodes. Moreover, the recombination rate of photogenerated electron-hole is efficiently restricted due to the impregnation of Al and Fe.

**Keywords:** Micromixing technology; TiO<sub>2</sub> photoanodes; Doping modification; Visible light response; Photoelectrochemical performance

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