

# Enhanced photocatalytic activity of $\text{Eu}_2\text{O}_3/\text{Ta}_2\text{O}_5$ mixed oxides on degradation of rhodamine B and 4-nitrophenol

Xia Yang<sup>a</sup>, Leilei Xu<sup>a</sup>, Xiaodan Yu<sup>a</sup>, Wei Li<sup>a</sup>, Kexin Li<sup>a</sup>, Mingxin Huo<sup>b,\*</sup>, Yihang Guo<sup>a,\*</sup>

<sup>a</sup> School of Chemistry, Northeast Normal University, Changchun 130024, PR China

<sup>b</sup> School of Urban and Environmental Sciences, Northeast Normal University, Changchun 130024, PR China

Received 19 October 2007; received in revised form 24 December 2007; accepted 4 January 2008

Available online 19 January 2008

## Abstract

Europium oxide/tantalum pentoxide ( $\text{Eu}_2\text{O}_3/\text{Ta}_2\text{O}_5$ ) mixed oxides with different  $\text{Eu}_2\text{O}_3$  dopings were prepared by a single-step sol–gel process via hydrolysis of tantalum pentachloride in the presence of europium nitrate. The products were in the amorphous and orthorhombic phase structures, respectively, based on the different calcination temperatures (200 and 500 °C). Composition, morphology, phase structure,  $\text{Eu}_2\text{O}_3$ -doping mode in the  $\text{Ta}_2\text{O}_5$  matrix and optical absorption property of the products were characterized by inductively coupled plasma atomic emission spectroscopy (ICP-AES), field emission scanning electron microscopy (FESEM), X-ray diffraction patterns (XRD), X-ray photoelectron spectroscopy (XPS) and UV–vis diffuse reflectance spectroscopy (UV–vis/DRS). The UV-light photocatalytic activity of the products was evaluated by degradation of aqueous rhodamine B (RB) and 4-nitrophenol (4-NP). The results showed that the photocatalytic activity of as-prepared  $\text{Eu}_2\text{O}_3/\text{Ta}_2\text{O}_5$  was higher than that of pure  $\text{Ta}_2\text{O}_5$  regardless of their phase structures. Among the tested samples,  $\text{Eu}_2\text{O}_3/\text{Ta}_2\text{O}_5$  with 0.49% Eu loading obtained with 200 °C exhibited the highest activity to degradation of the above two model molecules. The reasons of this enhanced photocatalytic activity were discussed.

© 2008 Elsevier B.V. All rights reserved.

**Keywords:** Europium oxide; Tantalum pentoxide; Sol–gel; Photocatalysis; Mixed oxides; RB; 4-NP

## 1. Introduction

Design and preparation of efficient heterogeneous photocatalytic materials still attracts much attention due to their potential activity for total destruction of organic compounds in polluted air and wastewater. Recently, photocatalytic activity for overall water decomposition and organic pollutant degradation on pure  $\text{Ta}_2\text{O}_5$ , nitrogen-doped  $\text{Ta}_2\text{O}_5$  or Ta-containing mixed oxides has been reported [1–9]. However, preparation and photocatalytic activity of rare-earth oxide-doped  $\text{Ta}_2\text{O}_5$  have seldom been reported so far.  $\text{Ta}_2\text{O}_5$  is a kind of semiconductor with wide band gap (4.0 eV) [10]. It has been reported that both amorphous and crystalline  $\text{Ta}_2\text{O}_5$  exhibited photocatalytic activity under UV-light irradiation [2]. In addition, rare-earth-doped anatase  $\text{TiO}_2$  showed enhanced UV-light photocatalytic activity compared with that of pure  $\text{TiO}_2$  [11–17]. The reason has been explained that rare-earth doping in the  $\text{TiO}_2$  matrix remarkably

changed the potential energy level of  $\text{TiO}_2$ , resulting in the creation of localized positive charge around Ti and/or formation of an oxygen vacancy. Therefore, the electron trapping efficiency was enhanced [17]. In addition, doping rare-earth oxide disturbed electronic structure of  $\text{TiO}_2$ , resulting in redshift of O 2p to Ti 3d charge transfer (CT) band. This decreased band gap energy played an important role in improving the photocatalytic activity of  $\text{TiO}_2$  [11,17,18].

Motivated by the above work, here we prepared  $\text{Eu}_2\text{O}_3$ -doped  $\text{Ta}_2\text{O}_5$  mixed oxides via a one-step sol–gel method. The procedure mainly included three steps: (1) hydrolysis of  $\text{TaCl}_5$  in the presence of  $\text{Eu}(\text{NO}_3)_3$ ; (2) aging the resulting sol of (Eu, Ta)-O<sub>2</sub> until formation of the gel; (3) calcination of the gel particulate at 200 and 500 °C, respectively, to produce  $\text{Eu}_2\text{O}_3/\text{Ta}_2\text{O}_5$  mixed oxides. Composition, morphology, phase structure, optical absorption property of the products and  $\text{Eu}_2\text{O}_3$ -doping mode in the  $\text{Ta}_2\text{O}_5$  matrix were characterized by ICP-AES, FESEM, XRD, UV–vis/DRS and XPS. The UV-light photocatalytic activity of as-prepared mixed oxides was evaluated by degradation of aqueous RB and 4-NP. Under the same conditions, the activity of pure  $\text{Ta}_2\text{O}_5$  was tested for comparison.

\* Corresponding authors. Tel.: +86 431 85098705; fax: +86 431 85098705.  
E-mail address: [guoyh@nenu.edu.cn](mailto:guoyh@nenu.edu.cn) (Y. Guo).

## 2. Experimental

### 2.1. Catalyst preparation

TaCl<sub>5</sub> (3 mmol) was dissolved in 172 mmol of ethanol with stirring for 30 min. Another solution containing 69 mmol of ethanol, 111 mmol of distilled water and Eu(NO<sub>3</sub>)<sub>3</sub>·6H<sub>2</sub>O in the required stoichiometry was slowly added into the above TaCl<sub>5</sub>/EtOH solution. The mixture was stirred for 90 min until the transparent sol was obtained. The sol was aged at 45 °C for 48 h and then heated at 80 °C for 3 h. The obtained gel was calcined at 200 and 500 °C for 4 h, respectively, at a rate of 2 °C min<sup>-1</sup>, and then washed with double distilled water for three times. The final products were obtained after being dried at 45 °C in a vacuum for 12 h. The products were represented by *x*-ET-*t*, where *x* referred to weight percentage of Eu (%), E to Eu<sub>2</sub>O<sub>3</sub>, T to Ta<sub>2</sub>O<sub>5</sub> and *t* calcination temperature (°C).

### 2.2. Catalyst characterization

Dopings of Eu in the products were determined by a Leeman Prodigy ICP-AES, and they were 0.32%, 0.49%, 0.68% and 0.93%, respectively, for the products calcined at 200 °C, and 0.52% for the product calcined at 500 °C. UV–vis/DRS of the products were recorded on a Cary 500 UV–vis-NIR spectrophotometer. XRD patterns of the products were recorded on a Rigaku D/max-3c X-ray diffractometer, and the particle sizes were estimated from the diffraction peak broadening with the Scherrer formula,  $D = 0.89\lambda/\beta\cos\theta$ , where *D* refers to the crystal size,  $\lambda$  the wavelength of X-ray radiation (0.154 nm for Cu K $\alpha$  radiation),  $\beta$  the full width at half maximum and  $\theta$  is the diffraction angle. The FESEM micrographs of the products were performed on a XL-30 ESEM FEG scanning electron microscopy. XPS were performed on a VG-ADES 400 instrument with Mg K $\alpha$ -ADES source at a residual gas pressure of below 10<sup>-8</sup> Pa. TOC results were carried out using a Shimadzu TOC-500 Total Organic Carbon analysis.

### 2.3. Photocatalytic tests

The photoreactor was designed with an internal light source (50 W high pressure mercury lamp with main emission wavelength 313 nm) surrounded by a quartz jacket, where the suspension included the solid catalyst (0.20 g) and an aqueous RB (25 mg L<sup>-1</sup>, 100 mL) or 4-NP (50 mg L<sup>-1</sup>, 100 mL) completely surrounded by the light source. The suspension was ultrasonicated for 10 min and stirred in the dark for 30 min to obtain a good dispersion and establish adsorption–desorption equilibrium between the organic molecules and the catalyst surface. The acidity of the suspension was neutral. The temperature of the suspension was maintained at 30 ± 2 °C by circulation water through an external cooling coil, and the system was open to air. Decreases in the concentrations of dyes were analyzed by a Cary 500 UV–vis-NIR spectrophotometer at  $\lambda = 554$  nm. At given intervals of illumination, some reaction solution was taken out, and then centrifuged and filtrated. Finally, the filtrates were analyzed. The concentrations of the 4-NP in suspensions

were monitored by a Shimadzu LC-8A high pressure liquid chromatography (HPLC) equipped with a UV detector ( $\lambda = 210$  nm). Before HPLC determination, the withdrawn suspensions were centrifuged and filtered with the microporous membrane.

## 3. Results and discussion

### 3.1. Catalyst preparation and characterization

Hydrolysis of TaCl<sub>5</sub> in the presence of Eu<sup>3+</sup> resulted in the sol of (Eu, Ta)-O<sub>2</sub> species, which was formed with a large number of surface hydroxyl groups due to incomplete condensation. After aging the sol at 45 °C for 48 h, homogeneous gel was formed. After dryness, the gel at 80 °C for 3 h, the gel particulate was yielded. Calcination of the particulate at 200 and 500 °C, respectively, Eu-containing Ta<sub>2</sub>O<sub>5</sub> with amorphous and orthorhombic phase were produced. The morphology, phase structure, Eu-doping mode in Ta<sub>2</sub>O<sub>5</sub> matrix and optical absorption property of the products were characterized, and the results were described below.

FESEM images (Fig. 1) reveal that both 0.49-ET-200 and 0.52-ET-500 exhibit spherical shape with average particle size of 40 and 25 nm, respectively. In addition, aggregation among the particles occurred, particularly for the 0.52-ET-500 sample. Decrease of the particle size for the 0.52-ET-500 compared with the 0.49-ET-200 was due to the shrinkage of the particles during the process of calcination at relatively high temperature.

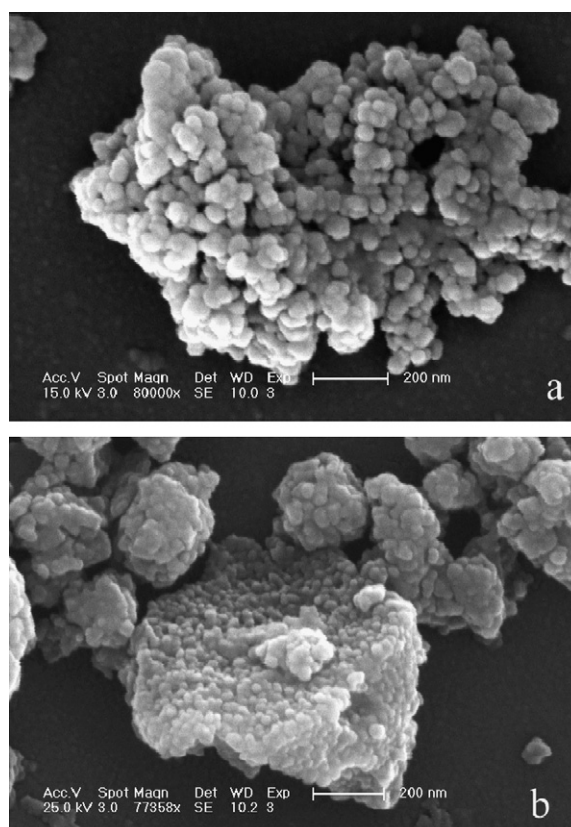


Fig. 1. FESEM images of: (a) 0.49-ET-200 and (b) 0.52-ET-500.

Download English Version:

<https://daneshyari.com/en/article/596730>

Download Persian Version:

<https://daneshyari.com/article/596730>

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