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RESEARCH ARTICLE

PHOTOCATALYTIC PERFORMANCE OF TB, N CO-DOPED TITANIA NANOCOMPOSITES

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ABSTRACT

In this study, the photocatalytic performance of Tb, N co-doped titania nanocomposites was evaluated by degradation of methylene blue. The effects of calcination temperature and N doping amount on the catalytic activity were analyzed. In addition, the photocatalytic reaction kinetics of the nanocomposites was also discussed. The results showed that Tb-N-TiO_2 exhibites the highest photocatalytic performance when the calcination temperature is 600 °C and the optimal doping amount of N is 6 %. Tb-N-TiO_2, N-TiO_2 and Tb-TiO_2were fitted to the pseudo-first-order reaction kinetics, and the reaction rate constants were 0.03768 h^{-1}, 0.56742 h^{-1} and 0.06289 h^{-1}, respectively.

INTRODUCTION

With the development of industry, dye wastewater pollution has become one of the most serious environmental problems in the world. As a common dye, methylene blue is a kind of aromatic heterocyclic compound. The carcinogenic and teratogenic characteristics of methylene blue wastewater have seriously endangered human health. Moreover, the dye contaminants could consume a large amount of dissolved oxygen, which affects the normal growth of aquatic organisms and leads to water deterioration. Therefore, dye wastewater processing technology has received much attention. Since Fujishima and Honda discovered the phenomenon of decomposing water to hydrogen by illuminating TiO_2 electrode in 1972, photocatalysis has become a new hot spot in the field of catalysis research(6). The principle of photocatalytic process is that after a semiconductor photocatalyst is excited by light, photons with a certain energy are absorbed by ground-state electrons, resulting in electron transitions to generate photo-generated electron-hole pairs. They migrate to the surface of the catalyst and form a highly active oxidation and reduction active potential. Thereafter, a series of photochemical reactions have been occurred, and strong oxidizing substances such as hydroxyl radicals are generated, which can not only eliminate a large number of pathogenic microorganisms, but also effectively degrade chemical pollutants. TiO_2 is widely used in photocatalysis due to its chemical stability, photocorrosion resistance, non-toxicity and low cost.

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However, there are still some defects that limit the practical application of TiO_2, which is not responsive to visible light and has low quantum efficiency. To solve these problems, the modification of TiO_2 has attracted a lot of attention. The purpose of modification includes broadening the photoresponse range, promoting effective separation of photogenerated charges, inhibiting carrier recombination to improve photon quantum efficiency, and improving the stability of photocatalysts. Element doping is one of the important modification methods. Many reports have focused on single-element doping of TiO_2. However, recent studies have shown that co-doping of TiO_2 with two or more elements can achieve better photocatalytic performance than single-element. In this paper, using terbium and nitrogen co-doped TiO_2composites as catalysts, the effects of calcined temperature and nitrogen doping amount on the degradation of methylene blue were investigated. In addition, the effectiveness of single-element doped and double-element doped samples was compared, and the photocatalytic reaction kinetics were analyzed.

MATERIALS AND METHOD

Methylene blue (≥99.7% purity) was purchased by the company of Shanghai macklin biochemical technology. The chemical structure of methylene blue is shown in Fig.1. All of the reagents were of analytical grade and used without any further purification. Tb, S co-doped TiO 2 nanocomposites were fabricated by sol-gel method via a facile step. The photocatalytic performance was evaluated by degradation of methylene blue in aqueous solution. Firstly, 100 mL of 10 mg/L methylene blue was placed in a beaker, then 3 drops of hydrogen peroxide solution were added, followed by an

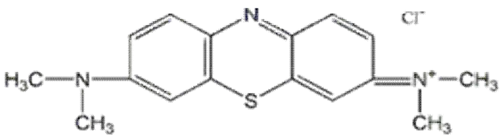

Fig.1. Chemical structure of methylene blue

appropriate amount of catalyst. Priorto the photocatalytic reaction, stirring in the dark for 0.5 h. Subsequently, turning on the lamp to start the photocatalytic reaction. 5 ml of the suspension was taken and centrifuged every 0.5 h, and measure the absorbance of supernatantat 664 nm by a HITACHI U-2910 spectrophotometer.

RESULTS AND DISCUSSION

Fig.2 shows the effect of calcined temperature of Tb, N co-doped composites on photocatalytic performance. When the calcination temperature was in the range of 300-600°C, an increment of degradation rate was observed with the increase of calcination temperature. This was attributed to the combined effects of the crystallinity, crystallite size and specific surface area of the nanocomposites. The effect of nitrogen doping content on the photocatalytic performance is shown in Fig.3. When the nitrogen doping content was 6%, the composite exhibited the highest photocatalytic performance. Therefore, Tb-N-TiO_2 nanocomposite with 6% nitrogen doping content which calcined at 600°C was selected for subsequent experiments.

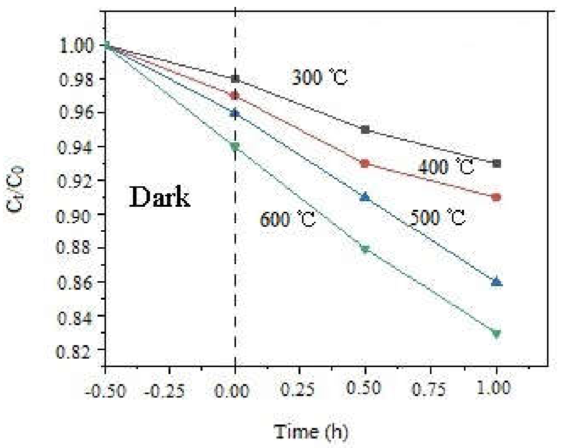

Fig.2. Photocatalytic activity of Tb-N-TiO_2 (nitrogen doping content is 6%)calcined at different temperatures.

The comparison of photocatalytic performance of Tb-N-TiO_2, Tb-TiO_2 and N-TiO_2 within 3h is displayed in Fig.4. It can be clearly seen that Tb-N-TiO_2 exhibited better photocatalytic performance than Tb-TiO_2 and N-TiO_2. The Langmuir-Hinshelwood kinetic model was used to study the kinetics of photocatalytic reactions. The ordinate is $-\ln(C_t/C_0)$, the abscissa is the reaction time t, and the fitting analysis was performed. The slope is the reaction rate constant. The result is displayed in Fig.5. and Table 1. It can be seen from Table 1 that the reaction rate constant of N-TiO_2, Tb-N-TiO_2 and Tb-TiO_2 as catalysts were 0.03768 h^{-1}, 0.56742 h^{-1} and 0.06289 h^{-1}, respectively. The corresponding correlation coefficient were 0.87794, 0.84960, 0.95560, demonstrating that they are all in line with the first-order reaction model.

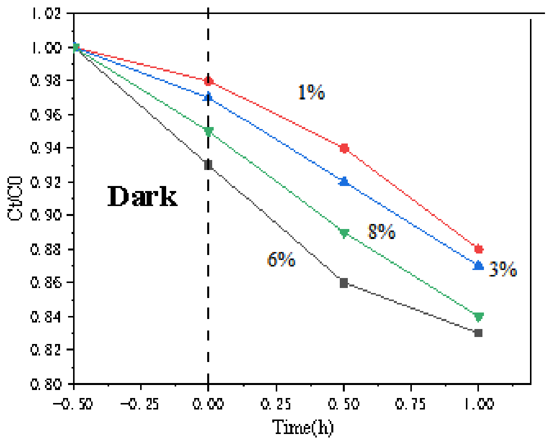

Fig. 3. Photocatalytic activity of Tb-N-TiO2 with different nitrogen doping content.

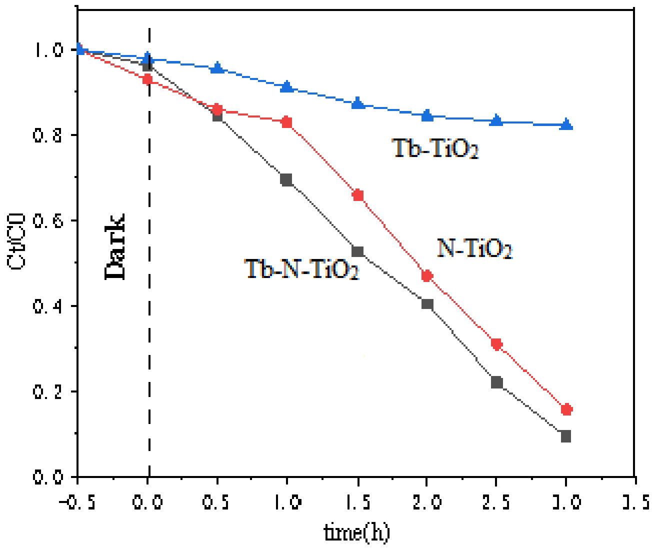

Fig.4. Photocatalytic performance of Tb-N-TiO_2, Tb-TiO_2 and N-TiO_2 within 3h

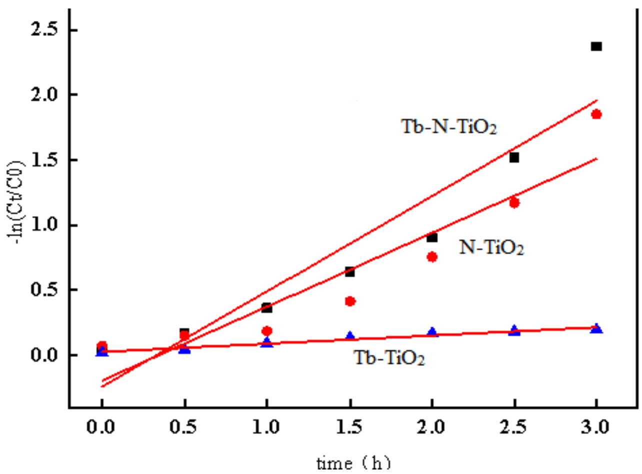

Fig.5.Fitting curve of pseudo-first-order kinetics.

Table1. Parameters of pseudo-first-order kinetics

Sample	N-TiO_2	Tb-N-TiO_2	Tb-TiO_2
Reaction rate constant (h^{-1})	0.56742	0.73140	0.06289
Correlation coefficient (R^2)	0.84960	0.87794	0.95560

Conclusions

Tb-N-TiO_2 with nitrogen doping content of 6% which calcined at 600 °C showed better photocatalytic performance than Tb-TiO_2 and N-TiO_2. The synergistic effect of terbium and nitrogen effectively make an enhancement of photocatalytic performance. Tb-N-TiO_2 nanocomposites exhibited excellent photocatalytic activity, suggesting that it is a promising candidate in practical application of dye wastewater treatment.

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