



# Novel adsorption and photocatalytic oxidation for removal of gaseous toluene by V-doped TiO<sub>2</sub>/PU under visible light



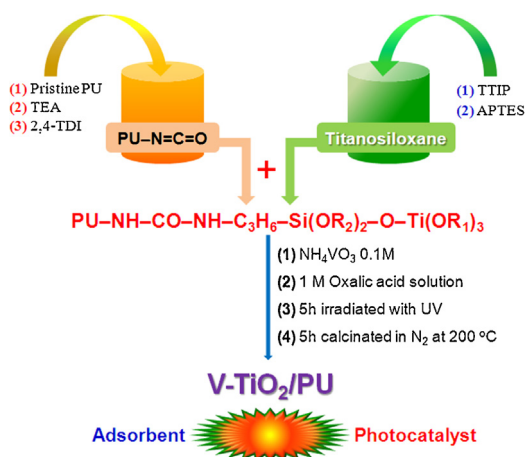
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## HIGHLIGHTS

- V doping was used to enhance the photocatalytic activity of TiO<sub>2</sub> photocatalyst.
- The enhanced TiO<sub>2</sub> was fixed on porous polyurethane (PU) by chemical bonds.
- The PU was used as a substrate to increase the adsorption ability of the photocatalyst.
- V-TiO<sub>2</sub>/PU adsorbed and photocatalytically oxidized toluene gas under visible light.
- The V/TiO<sub>2</sub> ratio of 6wt% was optimal for enhancing the photocatalytic activity.

## GRAPHICAL ABSTRACT



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## ABSTRACT

In this study, V was used as a dopant to defect into the TiO<sub>2</sub> lattice, leading to formation of Ti<sup>3+</sup> and V<sup>4+</sup> in the lattice. The presence of Ti<sup>3+</sup> and V<sup>4+</sup> introduced into the TiO<sub>2</sub> lattice increased the electron–hole pair generation capacity and electron–hole pair separation efficiency of the TiO<sub>2</sub>, leading to enhancement of the photocatalytic activity of the photocatalyst. Porous polyurethane (PU) was used to immobilize the V-doped TiO<sub>2</sub> by creating chemical bonds. The use of porous substrate contributed to the increased adsorption ability of the enhanced photocatalyst, as well as expanded its application for the removal of toluene from aerosols. Under dark conditions, the V-TiO<sub>2</sub>/PU only exhibited adsorption ability for toluene treatment in aerosol. Under visible light conditions, the V-TiO<sub>2</sub>/PU exhibited high photocatalytic oxidation ability for the removal of toluene in aerosol. The photocatalytic oxidation ability was found to depend on the V to TiO<sub>2</sub> ratio. The optimal V content in V/TiO<sub>2</sub> for enhancing the photocatalytic activity of TiO<sub>2</sub> was determined to be 6 wt%. Even under visible light irradiation, the 6% V-TiO<sub>2</sub>/PU sample could photocatalytically remove 80% of the toluene in 200-ppmV inlet gas, while 89.3% of the removed amount was mineralized into CO<sub>2</sub> and H<sub>2</sub>O.

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## 1. Introduction

Ambient emissions of volatile organic compounds (VOCs) have become ubiquitous and serious environmental burdens [1]. Piumetti et al. reported that VOCs emissions contribute to the formation of tropospheric ozone, photochemical smog and stratospheric ozone depletion [2]. Many VOCs, such as benzene and chloroform, are regulated to be toxic and carcinogenic to humans [3,4]. Rezaee et al. reported that the inhalation of toluene leads to nervous system complications, such as reduction in thinking, memory and muscular abilities, as well as some level of loss in both hearing and color vision [5]. Therefore, it is necessary to remove VOCs from the environment to improve the environmental quality, as well as to eliminate their adverse effects on human health.

Traditional methods, including adsorption, incineration, wet scrubbing, membrane separation and bioreaction, have widely been applied to the removal of VOCs [6–8]. However, these methods are associated with several disadvantages, such as the generation of secondary pollution, high costs, high-energy consumption and difficulty operating, particularly when low concentrations of VOCs need to be removed [6]. For example, the use of adsorptive materials to adsorb VOCs only succeeds in transferring the contaminants to another phase, rather than eliminating them. Therefore, numerous studies have been conducted to investigate effective techniques, which are both economically feasible and actually remove the VOCs, rather than merely transferring them from one phase to another [9–11]. Among such methods, photocatalytic oxidation processes using  $\text{TiO}_2$  and other semiconductors as photocatalysts have been extensively investigated [10–12].  $\text{TiO}_2$  is considered to be the most suitable photocatalyst because of its high photocatalytic activity, chemical stability, non-toxicity, relative inexpensiveness and availability [11]. Applications of  $\text{TiO}_2$  for the removal of VOCs have received growing interests in recent years [11,13–15]. The  $\text{TiO}_2$  photocatalyst exhibited high removal efficiency of toluene and ethyl benzene under UV irradiation [13].  $\text{TiO}_2$  has a wide band gap energy (3.2 eV), thus it can only be activated by UV light, causing limited application of the photocatalyst for VOCs removal in the visible light region [10,11,16]. Therefore, numerous studies have been conducted to enhance the photocatalytic activity of pure  $\text{TiO}_2$  to allow use of the photocatalyst even under visible light, which requires less energy consumption than the production of light in the UV region [16]. The reported studies adopted methods including doping metals or non-metals such as V, Cr, Fe, N, C and S into the  $\text{TiO}_2$  lattice, and modifying or sensitizing the  $\text{TiO}_2$  surface with noble metals such as Pt, Pd and Au [16–22]. A few studies have used V as the doping agent to enhance the photocatalytic activity of  $\text{TiO}_2$ . For example, V-doped  $\text{TiO}_2$  was found to exhibit high photocatalytic efficiency, even in the visible region [16,23]. However, explanations regarding the mechanism of how V-doping affects the photocatalytic efficiency are still controversial. Several discrepancies also exist among the different studies concerning the oxidation states and the optimal V-doping content in the  $\text{TiO}_2$  lattice. In addition, it is difficult to separate or regenerate the V-doped  $\text{TiO}_2$  photocatalyst powder from experimental systems after use.

Therefore, in the present study, porous polyurethane (PU) was used as a substrate to immobilize the V-doped  $\text{TiO}_2$  photocatalyst in order to overcome the disadvantages faced by powder photocatalyst. This study operated under the hypothesis that the immobilization of V-doped  $\text{TiO}_2$  onto porous PU (V doped  $\text{TiO}_2/\text{PU}$ ) could also lead to an increase in the adsorption ability of the synthesized photocatalyst for the photocatalytic oxidation removal of toluene in aerosol under visible light irradiation. Based on the obtained physicochemical characteristics of the synthesized V-doped  $\text{TiO}_2/\text{PU}$  and its photocatalytic efficiency for the removal of toluene, the suitable mechanism of the V-doping effects were also

investigated, and the V-doping content was optimized for enhancing the photocatalytic activity of  $\text{TiO}_2$ .

## 2. Materials and methods

### 2.1. Material preparation

In the first step, pristine PU was activated by a mixed solution of toluene, toluene-2,4-diisocyanate (2,4-TDI) and anhydrous triethylamine (TEA) to synthesize isocyanated PU, which is PU containing isocyanate groups (NCO) on its surface [24]. In the second step, titanium isopropoxide (TTIP) was reacted with  $\gamma$ -aminopropyl triethoxysilane (APTES) to synthesize amino titanasiloxane, which contains Si—O—Ti bonds and an amine group ( $\text{NH}_2$ ) [24]. In the third step, the amino titanasiloxane was immobilized on the PU surface, based on the urea bond formed from the reaction between the isocyanate groups of the isocyanated PU and the amino group ( $\text{NH}_2$ ) of the amino titanasiloxane [24]. Finally, ammonium metavanadate ( $\text{NH}_4\text{VO}_3$ ) was used as a precursor of the V-dopant to achieve synthesis of V- $\text{TiO}_2/\text{PU}$ . The volume of 0.1 M  $\text{NH}_4\text{VO}_3$  solution added was controlled in order to synthesize 0, 2, 4, 6, 8 and 10% V- $\text{TiO}_2/\text{PU}$  materials, which the ratios of V/ $\text{TiO}_2$  were 0, 2, 4, 6, 8 and 10% wt, respectively. The material preparation processes are schematically described in Fig. 1, and the detailed synthesis procedure is outlined in the following sections.

### 2.2. Experiments for removal of toluene

#### 2.2.1. Experimental apparatus

Fig. 2 shows a continuous experimental apparatus for removal of toluene in aerosol using V- $\text{TiO}_2/\text{PU}$  materials. The experimental apparatus is composed of three main parts including gas generator, reaction chamber and analyzer systems. The gas generator system included a toluene gas source, a synthetic air tank, a humidifier, a humidity controller, flow rate meters and a gas mixer. A cylinder of 1000 ppm toluene in nitrogen gas was used as the toluene source tank. Synthetic air, containing 20.5% vol  $\text{O}_2$  and 79.5% vol  $\text{N}_2$ , was used for dilution of the toluene to obtain a flow rate of 200 mL/min with 200 ppm toluene in the gas mixture. The humidifier and humidity controller were employed to ensure that the relative humidity (RH) of the gas mixture remained constant at 50% (RH). The reaction chamber was a dark cover cask (25 × 50 × 50 cm)

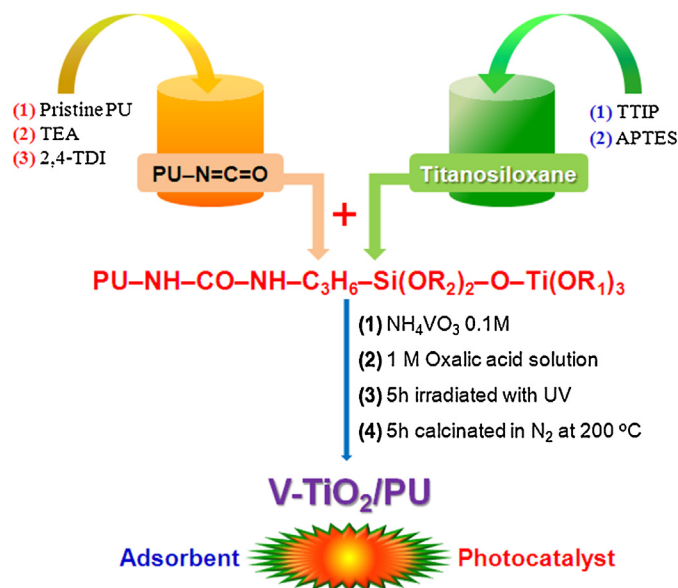


Fig. 1. Material preparation processes.

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