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# Enhanced visible-light photocatalytic performance of a monolithic tungsten oxide/graphene oxide aerogel for nitric oxide oxidation



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### ABSTRACT

Photocatalysis is considered a promising technique for removal of pollutants from indoor air. However, the low selectivity and limited recyclability of photocatalysts in powder form currently limit their practical application. In this work, we reported the successful preparation of a monolithic tungsten oxide (WO<sub>3</sub>)/graphene oxide (GO) aerogel photocatalyst through a cost-effective freeze-drying method. GO not only acts as a macroscopic support, but also increases the catalyst surface area from 46 to 57 m<sup>2</sup>/g, enhances the light absorption in the visible-light region, and raises the separation efficiency of photogenerated electron-hole pairs. The Obtained WO<sub>3</sub>/GO aerogel exhibited an outstanding visible-light photocatalytic degradation rate of nitric oxide of 51%, which was 3.3 times that of pristine WO<sub>3</sub> powder. In addition, the aerogel displayed excellent selectivity, with a generation fraction of toxic nitrogen dioxide of as low as 0.5%. This work presents a facile synthesis route to fabricate a monolithic WO<sub>3</sub>/GO aerogel photocatalyst with great promise for air purification.

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# 1. Introduction

With the widespread use of fossil fuels and the rapid development of modern industry, gas pollution is becoming a serious issue that we face today. Nitrogen oxides [consisting of mainly nitric oxide (NO) and nitrogen dioxide (NO<sub>2</sub>)] are common air pollutants that can lead to not only acid rain, tropospheric ozone depletion, and the greenhouse effect, but also respiratory disease and skin allergies [1–6]. Therefore, various techniques have been developed for NO removal, including physical adsorption [7–9], biofiltration [10,11], and thermal catalytic reduction [12,13]. However, the above-mentioned methods are usually complex, expensive, and unsuitable to remove low concentrations of NO (ppb level). Recently, photocatalysis has been considered as a promising technique to achieve low-concentration NO removal. Various photocatalysts such as metals and metal oxides (Bi, TiO<sub>2</sub>, BiOX: X = Cl, Br, I, or CO<sub>3</sub>,) [14–20], perovskite-type oxides (ATiO<sub>3</sub>, where A = Ba, Sr or Pb) [21–23], and metal-free compounds (C<sub>3</sub>N<sub>4</sub>) with high NO photodegradation activity have been reported [24,25]. However, most reported photocatalysts generate a high fraction of NO<sub>2</sub>, which is more poisonous than NO, leading to secondary pollution. Therefore, the development of photocatalysts with high selectivity for NO removal is still a great challenge.

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Tungsten oxide (WO<sub>3</sub>) is a typical n-type semiconductor that has attracted great attention because of its nontoxicity, stable physicochemical properties, and good resistance to photocorrosion [26–34]. Importantly, both theory and experiments have proved that WO<sub>3</sub> possesses a relatively low valence-band edge, which endows it with strong oxidation ability and makes it a promising candidate for NO removal [35,36]. However, the inherent deficiencies of WO<sub>3</sub>, including its low specific surface area, wide bandgap, and high recombination rate of photogenerated electron-hole pairs, severely lower its photocatalytic performance. In addition, most of the reported WO<sub>3</sub> photocatalysts are in powder form, so they have a strong tendency to agglomerate and are difficult to recycle.

To overcome these typical limitations of photocatalysts, a three-dimensional (3D) macroscopic graphene aerogel and its derivatives (reduced graphene oxide (RGO) aerogel and graphene oxide (GO) aerogel) with large surface area, low density, and high porosity have been developed. For example, Li and co-workers synthesized BiOBr/RGO, TiO<sub>2</sub>/RGO, and Cu<sub>2</sub>O/RGO nanocomposite aerogels via a freeze-drying method [37-40]. These nanocomposite aerogels exhibited high activity for the degradation of pollutants and were easily separated from the aqueous reaction systems to allow easy recycling, opening a new avenue for the development of integrated photocatalysts. In these systems, the graphene aerogel acts as not only the substrate but also has a positive effect on photocatalytic performance because it increases light absorption and slows the recombination of electron-hole pairs [41-44]. Moreover, the monolithic structure of photocatalyst aerogels makes them easy to recycle.

In this work, we report the fabrication of a monolithic  $WO_3/GO$  aerogel photocatalyst via a cost-effective freeze-drying process. GO acts as a scaffold to support  $WO_3$  and also enhances the photocatalytic NO degradation activity of  $WO_3$ . Moreover, the obtained  $WO_3/GO$  aerogel exhibits excellent selectivity, with  $NO_2$  generation fraction as low as 0.5%. Our results demonstrate that  $WO_3/GO$  aerogels show great potential for use in indoor air purification.

#### 2. Experimental

#### 2.1. Materials

All chemical reagents were purchased from Chengdu Kelong Co. Ltd. in analytical grade and used without any further purification treatment.

#### 2.2. Synthesis of WO<sub>3</sub> and GO

Hexagonal WO<sub>3</sub> powder was prepared via a hydrothermal route according to our previous work [45,46]. In a typical procedure, ammonium metatungstate (248 mg) and acetic acid (2 mL, 25 vol %) were placed in a 25-mL Teflon-lined stainless-steel autoclave. The autoclave was heated at 180 °C for 48 h and then cooled to room temperature. The solid residue was washed with deionized water and then dried at 70 °C to obtain WO<sub>3</sub> powder.

GO was synthesized through a modified Hummers method [40]. The final GO solution was diluted to a concentration of 4 mg/mL for use in further experiments.

## 2.3. Synthesis of monolithic WO<sub>3</sub>/GO aerogel

The prepared WO<sub>3</sub> powder (360 mg) was dispersed in deionized water (90 mL) by ultrasonication at 300 W and 40 kHz for 1 h. GO solution (10 mL, 4 mg/mL) was added dropwise to the above suspension. The mixture was stirred for 24 h to achieve a uniform suspension and then frozen at -80 °C for 2 h. After freeze-drying for 48 h, 90 wt% WO<sub>3</sub>/GO aerogel was obtained. For comparison, WO<sub>3</sub>/GO aerogels with different contents of WO<sub>3</sub> (70 and 80 wt%) were prepared via a similar synthesis process, and are denoted as 70 wt% WO<sub>3</sub>/GO and 80 wt% WO<sub>3</sub>/GO, respectively. Unless otherwise stated, the WO<sub>3</sub>/GO aerogel referred to is the 90 wt% WO<sub>3</sub>/GO sample.

#### 2.4. Characterization

Powder X-ray diffraction (XRD) was performed with a PANalytical X'pert diffractometer operated at 40 kV and 40 mA using Cu  $K_{\alpha}$  radiation. Transmission electron microscopy (TEM) and high-resolution transmission electron microscopy (HRTEM) images were recorded on an FEI Tecnai G2 20 microscope operated at 200 kV. Ultraviolet-visible (UV-Vis) diffuse reflection spectra (DRS) were measured on a Shimadzu UV-2600 spectrophotometer. Fourier transform infrared (FTIR) spectra were collected on a Nicolet 6700 spectrometer. X-ray photoelectron spectroscopy (XPS) was performed on a Thermo ESCALAB 250Xi spectrometer with Al  $K_{\alpha}$  radiation at 1486.6 eV. Brunauer-Emmett-Teller (BET) surface area measurements were performed using the nitrogen adsorption-desorption technique on a Quantachrome ASAP 2020 HD88 surface analyzer. Photoluminescence spectra (PL) were measured using Hitachi F-7000 with the light source of MVL-210 (Mejiro Genossen Inc.).

#### 2.5. Photocatalytic measurements

Photocatalytic removal of NO at ppb level was measured using a custom-made testing system [19]. In this experiment, photocatalyst (400 mg) was split equally between two petri dishes that were used in the activity testing. The flow rates of air and NO were 2.0 and 12.0 mL/min, respectively. The removal ratio ( $\eta$ ) of NO was calculated by Eq. (1):

$$\eta(\%) = (1 - C/C_0) \times 100\%, \tag{1}$$

where *C* is the concentration of NO during the reaction and  $C_0$  is the initial concentration of NO. In addition, the generation ratio ( $\omega$ ) of NO<sub>2</sub> during the photocatalytic reaction was calculated by:

$$\omega(\%) = [C(NO_2) - C_0(NO_2)]/C_0(NO_x) \times 100\%, \quad (2)$$
  
where  $C(NO_x)$  is the total concentration of nitrogen oxides (NO<sub>2</sub>  
and NO;  $C(NO_x) = C(NO) + C(NO_2)$  and  $C_0(NO_2)$  is the initial  
concentration of NO<sub>2</sub> before the photocatalytic reaction.

#### 3. Results and discussion

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