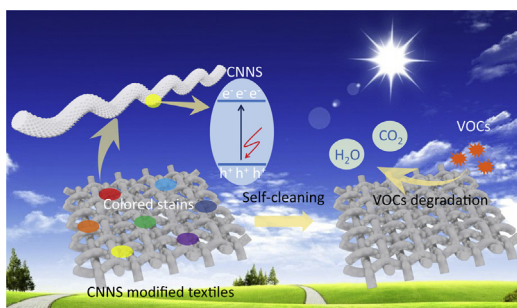


# Facile surface modification of textiles with photocatalytic carbon nitride nanosheets and the excellent performance for self-cleaning and degradation of gaseous formaldehyde

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## GRAPHICAL ABSTRACT



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## ABSTRACT

Great advances in photocatalysis have been made by developing various efficient photocatalysts, but investigation on practical applications of photocatalysis is relatively backward. Herein we report a facile surface modification approach to functionalize textiles with excellent ability for photocatalytic self-cleaning and degradation of indoor volatile organic pollutants. Graphitic carbon nitride nanosheets (CNNS) in colloidal suspension were directly sprayed onto the surface of cellulose fibers in textiles, and the powerful hydrogen bonding action between surface hydroxyl groups of cellulose and plentiful hydroxyl and amino groups of exfoliated CNNS from alkali-treating realizes high stability of CNNS modified textiles. Due to ultrathin 2D thickness and high visible light transparency, the modification of CNNS would not affect the hand feeling of textiles and shield their original colors. The obtained textiles show superior photocatalytic self-cleaning performance to remove stains from various colored pollutants under solar light irradiation, including industrial organic dyes and juices. Meanwhile, gaseous formaldehyde also can be efficiently decomposed using Xe lamp or commercial LED lamp as light sources. This work realizes photocatalytic performance of textiles using a simple spraying method, and it has great potential application in textile self-cleaning, not only for surface stains but also for volatile organic compounds from textile release.

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## 1. Introduction

Semiconductor photocatalysis has stimulated extensive interest since Fujishima et al. firstly reported the photocatalytic  $H_2O$ -splitting behavior on a  $TiO_2$  electrode at 1972 [1]. Up to date,

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numerous photocatalysts have been developed and applied in different fields, such as photo-degradation of organic pollutants in water or air [2–4], photocatalytic H<sub>2</sub> generation from water [5,6], photo-reduction of CO<sub>2</sub> to solar fuels [7,8] and photocatalytic redox reactions for organic synthesis [9,10]. Photocatalysis currently confronts application bottleneck due to the low solar light utilization efficiency, high recombination ratio of photo-generated charges and difficult recovery of powdered photocatalysts as well [11–13].

Modification of photocatalyst on supports can effectively promote the recycle performance and provide more active surface by avoiding aggregation of catalyst particles [14–16]. Moreover, it will bring additional photocatalytic application, for example indoor air purification [17,18], UV protection [19,20], anti-corrosion of metal materials [21], anti-fouling of filter membranes [22,23] and surface self-cleaning of substrate [24–26]. For these applications, it is vital to tightly and uniformly coat photocatalyst onto support surface to enhance the stability of photocatalyst layer. And for some cases, photocatalyst layers should have high transparency to prevent shielding of the color or transparency of supports. In addition, the hand feeling after coating photocatalysts is of great importance for textiles. To meet the above demands, the coated photocatalyst layer should have a thin thickness [27,28]. Among various photocatalysts, colloidal 2D nanosheets derived from layered compounds have high aspect-ratio morphology with large lateral size and ultrathin thickness in molecular size [29,30]. Surface charges of nanosheets inherited from precursors also facilitate the formation of interaction between photocatalysts and supports [31]. As for photocatalytic activity of materials, 2D nanosheets have unique advantages, such as large surface area to provide high density of active sites and short distance for charge diffusion to surface [32]. For instance, Yang et al. reported liquid exfoliation method of bulk g-C<sub>3</sub>N<sub>4</sub> to obtain 2D nanosheets. The structural features of 2D nanosheets lead to a very good photocatalytic activity for hydrogen evolution under visible light [33]. Yu et al. developed liquid-phase exfoliation into monolayered BiOBr nanosheets for photocatalytic oxidation and reduction. Compared to bulk BiOBr, monolayered BiOBr shows enhanced specific surface area, allowing more active sites [34]. Quinn et al. acquired aqueous dispersions of exfoliated molybdenum disulfide for use in visible-light photocatalysis by a sonication procedure [35].

Herein, we developed a facile strategy to fabricate 2D graphitic carbon nitride nanosheet (CNNS) modified textiles by directly spraying colloidal suspension of CNNS. The CNNS obtained from an alkali exfoliation has plentiful hydroxyl and amino groups [36,37] on surface that are favorable for forming hydrogen bonds between CNNS and cellulose based textiles. The modification of CNNS does not affect the hand feeling and color of textile substrates. These obtained CNNS modified textiles have photocatalytic activity for degradation of colored organic pollutants, consequently resulting in an excellent photocatalytic self-cleaning property for textiles. We also investigated its performance for photocatalytic removal of gaseous formaldehyde under Xe lamp or indoor LED irradiation.

## 2. Experimental

### 2.1. Synthesis of g-C<sub>3</sub>N<sub>4</sub>

The bulk g-C<sub>3</sub>N<sub>4</sub> was synthesized by thermal poly-condensation of melamine, which was reported in our previous literatures [38]. 10 g of melamine was put into a semi-closed alumina crucible with a cover and heated at 520 °C for 4 h in a muffle furnace with a ramping rate of 10 °C min<sup>-1</sup>.

### 2.2. Exfoliation of g-C<sub>3</sub>N<sub>4</sub> into CNNS

Colloidal suspension of CNNS was obtained by a simple alkali treatment at a reflux condition [36]. Typically, 1 g g-C<sub>3</sub>N<sub>4</sub> was dispersed into 50 mL NaOH aqueous solution (0.2 mol L<sup>-1</sup>) and then heated at 100 °C for 12 h. After collecting the sediment by removal of the clear supernatant, it was re-dispersed into 50 mL deionized water. Centrifuging the mixture at 12,000 rpm for 5 min to remove precipitation, we obtained CNNS colloidal suspension. In addition, the solid residue could be further dispersed into water, and finally all of solids were dispersed to form CNNS suspension by repeated water washing.

### 2.3. Modification of CNNS on textiles

Colloidal suspension of CNNS was added into an atomizer, and then sprayed it onto the surface of various cellulose based textiles, including a cotton T-shirt. After drying these CNNS modified textiles by natural sunlight or in an oven at 60 °C, the CNNS which was not tightly coated on surface was removed by repeated water washing. Finally, drying at 60 °C for 12 h, we obtained CNNS modified textiles.

The loading percentage of CNNS on textile was calculated by the following equation:

$$\text{CNNS\%} = \frac{m_0 - m'}{m_0} \times 100\% \quad (1)$$

where  $m_0$  and  $m'$  are the masses of textile before and after CNNS loading.

### 2.4. Characterizations

Crystal structures of samples were characterized by X-ray diffraction (XRD) technology on a DX-2700 diffractometer (Dandong Haoyuan Instrument Co. Ltd, China) using Cu K $\alpha$  radiation. Fourier transform infrared (FTIR) spectra were recorded with an FTIR spectrometer (Nicolet Avatar 370, USA) in the range of 4000–500 cm<sup>-1</sup>. UV–vis absorption spectra and diffuse-reflectance spectra (DRS) were obtained using a UV–vis spectrophotometer (Shimadzu UV-2450, Japan) equipped with an integrating sphere assembly. The morphology of materials was observed on a scanning electron microscope (SEM, FEI Quanta 250 FEG).

### 2.5. Photocatalytic tests

#### 2.5.1. Photocatalytic degradation of organic dyes in aqueous solution

The photocatalytic activity of CNNS modified textile was firstly estimated by removal of Rhodamine B (RhB) in aqueous solution under visible light irradiation using a 300 W Xe lamp (HSX-F300, Beijing NBet) with a 420 nm cutoff filter as the light source. The photocatalytic reaction was carried out in a 150 mL quartz reactor with a circulating water system to maintain a constant temperature. In each experiment, the CNNS modified textile was immersed into 100 mL aqueous solution of RhB (1 mg L<sup>-1</sup>), and the distance between light and surface of the solution was set at about 15 cm. In order to make sure the adsorption-desorption equilibrium of solution, the reactor was stirred in the dark for 10 min before illumination. During the photoreaction process, the solution was continuously stirred with a dynamoelectric stirrer and approximately 3 mL of the mixture was collected every 5 min. The concentration was determined by measuring the maximum absorbance at 554 nm for RhB using a Shimadzu UV-2450 spectrophotometer.

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