



g-C₃N₄/UiO-66 nanohybrids with enhanced photocatalytic activities for the oxidation of dye under visible light irradiation



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ABSTRACT

In this study, g-C₃N₄ nanosheets coated UiO-66 nanohybrids were fabricated via a simple solvothermal method. The physicochemical parameters, structural and electrochemical properties of g-C₃N₄/UiO-66 nanohybrids (CNUO-x) were investigated. The nanohybrids were systematically characterized by XRD, FT-IR, TGA, TEM, SEM, EDS elemental mappings, XPS, N₂ adsorption-desorption isotherms, PL and DRS. It was found that the addition of moderate g-C₃N₄ could enhance the separation and migration rate of photo-induced charges, consequently leading to the increase of photocatalytic efficiency. The CNUO-1 nanohybrid exhibited better photocatalytic activity for the oxidation of Rh B under visible light irradiation, and displayed about 6.0 and 3.3 times faster than that of g-C₃N₄ and UiO-66, respectively. Meanwhile, the nanohybrid showed excellent stability and reusability in four cyclic experiments. Finally, the increased photocatalytic reaction mechanism was also proposed. ·O₂⁻, h⁺ and ·OH were further found to be the main active contributors.

1. Introduction

Nowadays, more and more pollutants have attracted the attention of researchers [1–15]. Photocatalysis technologies have tempted huge concern, because they can efficiently convert pollutants to H₂O and CO₂ [16–25]. Many conventional semiconductors metal oxide, such as zinc oxide, titanium dioxide et al., have been used as photocatalysts to remove environmental pollutants in the past several years [18–25]. However, two fundamental issues limit those further applications in many fields. On one hand, photo-induced electron-hole pairs easily rapid recombination, resulting in the decrease of photocatalytic efficiency; the other hand, visible light response is poor, and the electron-hole pairs are stimulated by ultraviolet light. In order to solve these problems, some attempts have been tried [26–28]. However, to synthesize new eco-friendly and efficient photocatalysts still are a tremendous challenge.

Recently, metal organic frameworks (MOFs), emerging as new porous crystal materials, are made up of inorganic secondary building units and organic linkers, which have attracted tremendous attention in many fields, such as photocatalysis, catalysis, molecular sensing,

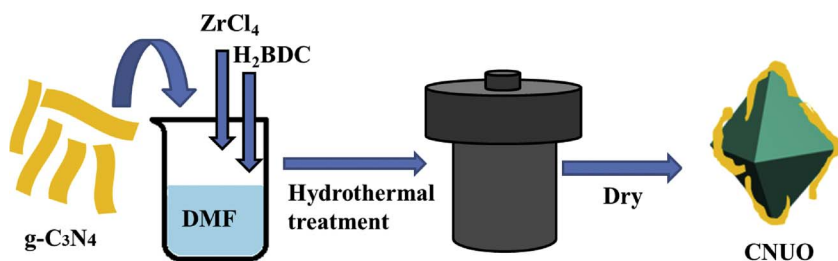
separation, gas storage and adsorption et al. [29–39]. So far, MOFs based photocatalysts have been used in the degradation of environmental pollutant, hydrogen evolution, and carbon dioxide reduction [40–44].

Based on previous studies, MOFs heterojunction photocatalysts have been more and more concerned, which can solve low light harvesting efficiency and rapid recombination of photo-induced electron-hole pairs, thereby increasing the photocatalytic performance [45–47]. Recently, it is found that delocalized π–π conjugated structures can generate a fast separation rate of photo-generated charge in electron transfer courses. Among these π–π conjugated structures, carbon nitride (C₃N₄) and reduced graphene oxide have been extensive studied as photocatalysts due to easy synthesis, low cost, high environmental stability, and strong visible light response [48,49]. Importantly, the recombination of C₃N₄ and semiconductor can enhance the transfer efficiency of photo-induced carriers, facilitating the separation of photo-induced carriers, which increase the photocatalytic activity [50–54].

UiO-66, a zirconium-based MOF with photochemical properties, possesses accessible micropore/mesopore diameters and large specific

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Scheme 1. Schematic illustration for the synthesis of the g-C₃N₄/UiO-66 nanocomposite.

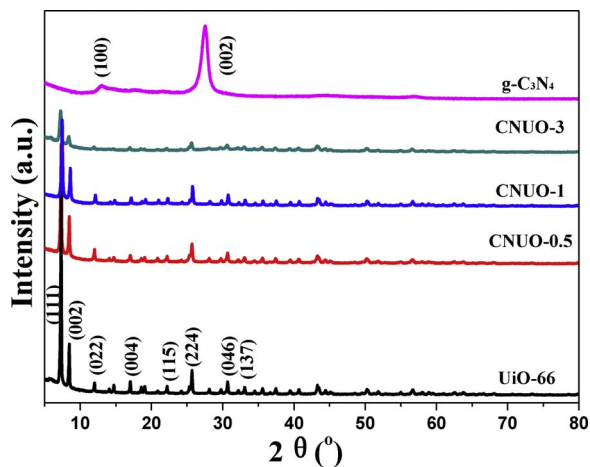


Fig. 1. XRD patterns of UiO-66, g-C₃N₄ and CNUO nanocomposites.

surface area [55–61]. Importantly, UiO-66 displays also good structural stability in waste water treatment. So far, g-C₃N₄/UiO-66 composite was applied in H₂ production and CO₂ reduction [62,63]. Up to present, few reports on g-C₃N₄/UiO-66 photocatalyst for waste water treatment are reported [64]. Here, g-C₃N₄/UiO-66 nanohybrids have been successfully fabricated via a simple solvothermal method. The photocatalysts are characterized by XRD, FT-IR, TGA, TEM, HRTEM, SEM, EDS elemental mappings, XPS, N₂ adsorption-desorption isotherms, PL and DRS. The new g-C₃N₄/UiO-66 nanohybrid is used as photocatalyst for the oxidation of RhB under visible light irradiation. The stability and reusability of g-C₃N₄/UiO-66 nanohybrids are evaluated. The results show that g-C₃N₄/UiO-66 can effectively enhance photocatalytic performance of RhB. A possible photocatalytic mechanism has been proposed in detail.

2. Experimental

2.1. Synthesis of the materials

2.1.1. g-C₃N₄

The g-C₃N₄ was synthesized by calcined with melamine as precursor [29,65]. In a typical synthesis, a certain amount of melamine was put into a ceramic crucible with a lid, and then placed in a muffle furnace and heated at 550 °C at a heating rate of 2 °C min⁻¹ for 4 h. The resulting yellowish powder was gathered for further use.

2.1.2. UiO-66

UiO-66 was obtained by a traditional solvothermal method according to the previous reported method [66]. Typically, Zirconium tetrachloride (0.053 g, 0.227 mmol) and terephthalic acid (0.034 g, 0.227 mmol) were dissolved in N, N-2-dimethylformamide (5 mL), and then the obtained mixture was transferred to a Teflon-lined stainless steel autoclave (50 mL). The autoclave was sealed and heated in an oven at 120 °C for 24 h. After cooling to room temperature, the sample

was centrifuged and purified with ethanol for several times to make sure the completely remove of the occluded DMF molecules. The white UiO-66 power was dried at 80 °C for 12 h to obtain the final products.

2.1.3. g-C₃N₄/UiO-66 nanocomposites

A series of g-C₃N₄/UiO-66 nanocomposites with different mass ratios of g-C₃N₄ vs UiO-66 were synthesized by a facile one-pot solvothermal reaction. Firstly, a certain amount of g-C₃N₄ powder was dispersed into 10 mL DMF under ultrasonication for 60 min. Then a mixture of 0.053 g ZrCl₄ and 0.034 g terephthalic acid were dissolved in 5 mL N, N-2-dimethylformamide at room temperature by ultrasonic until the solution become homogeneous. The g-C₃N₄ solution was added into the above solution with continuously ultrasonic for another 1 h. Then the mixture was subjected to solvothermal conditions in a Teflon-lined stainless steel autoclave at 120 °C for 24 h. And subsequent steps of the above suspension were the same as that of UiO-66. The final products were named as CNUO-x, where the x represents the weight content of g-C₃N₄.

2.2. Characterizations

The textural properties of the samples were determined by nitrogen physisorption at liquid nitrogen temperature (−196 °C) using a Quantachrome autosorb-iQ-2MP apparatus. The specific surface area was calculated by using the BET (Brunauer-Emmett-Teller) model. XRD experiments were carried out on a Bruker D8 Advance X-ray diffractometer by a monochromatic detector equipped with Cu K α radiation, operating at 40 kV and 40 mA, and a scan rate of 5° min⁻¹. The morphology of the samples was obtained from a FEI Tecnai G2 F20 transmission electron microscope (TEM) and a Hitachi FESEM-4800 scanning electronic microscopy (SEM). TG analysis was conducted on a Perkin Elmer STA8000 thermal analyzer. X-ray photoelectron spectroscopic (XPS) (Thermo ESCALAB 250XI) analysis was performed to examine the surface property and composition of the sample. The binding energies (Eb) were calibrated internally by the carbon deposit C1 s binding energy at 284.6 eV. The photoluminescence (PL) spectra were conducted on a Hitachi F-7000 fluorescence spectrophotometer at an excitation wavelength of 320 nm. The Fourier transform infrared (FT-IR) spectroscopy was conducted on Nicolet iS10. UV–vis diffuse reflectance spectra (UV-vis DRS) were recorded by a UV-2600 UV–vis spectrometer with a wavelength range of 200–800 nm.

2.3. Evaluation of photocatalytic activity

The photocatalytic degradation of RhB aqueous solution was conducted over CNUO nanocomposites under visible light irradiation. A 500W Xe lamp (PLS-SXE 500, CEALIGHT Beijing) with a 420 nm cutoff filter (420 nm < λ < 760 nm) was used as the light source (light intensity 50 mW cm⁻²). Typically, 20 mg of photocatalyst sample and 50 mL RhB aqueous solution (10 ppm) were mixed into the photoreactor. In the dark condition, the solution should magnetically stir for 60 min to achieve the adsorption-desorption equilibrium. At scheduled time intervals, a certain amount of sample solution was taken from the reactor and separated by centrifugation. The concentration of RhB remained in the supernatant solution was calibrated at its

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