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Synthesis of MoS$_2$/g-C$_3$N$_4$ as a Solar Light-Responsive Photocatalyst for Organic Degradation

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Abstract

Photocatalytic degradation of organic contaminants is an attractive chemical process owing to its potential for using solar light for environmental applications. In this study, a novel molybdenum disulfide (MoS$_2$) and graphitic carbon nitride (g-C$_3$N$_4$) composite photocatalyst was synthesized using a low temperature hydrothermal method. MoS$_2$ nanoparticles formed on g-C$_3$N$_4$ nanosheets in the composite and this MoS$_2$ incorporation greatly enhanced the photocatalytic activity of g-C$_3$N$_4$. The photocatalyst was tested for the degradation of methyl orange (MO) under simulated solar light. Composite 3.0wt% MoS$_2$/g-C$_3$N$_4$ showed the highest photocatalytic activity for MO decomposition. MoS$_2$ nanoparticles increased the interfacial charge transfer and thus prevented the recombination of photo-generated electron–hole pairs. The new MoS$_2$/g-C$_3$N$_4$ photocatalyst material also displayed good stability during the photo-reactions and the recycled catalyst showed little reduction in activity during repeated test runs. The novel MoS$_2$/g-C$_3$N$_4$ composite is therefore shown as a promising catalyst for photocatalytic degradation of organic pollutants using solar energy.

Key words: Decoloration, graphitic carbon nitride (g-C$_3$N$_4$), molybdenum disulfide (MoS$_2$), photocatalysis, methyl orange, organic degradation
1. Introduction

Much attention has been focused on the photocatalytic strategy for the removal of environmental contaminants, as solar energy is an inexhaustible and environmentally friendly energy resource. A large number of semiconductor materials, such as metal oxides and metal sulfides (e.g. TiO₂, WO₃, CdS, ZnS and ZnO), have been developed as active catalysts for photocatalysis of organic pollutants. Efforts have been made in recent years to use non-metal materials in the synthesis of new photocatalysts with improved reactivity and stability. Wang et al. reported a novel polymeric photocatalyst, graphitic carbon nitride (g-C₃N₄) that exhibits excellent photocatalytic hydrogen production using solar energy. The metal-free g-C₃N₄ photocatalyst possesses a good electronic property as well as a high thermal and chemical stability, making it a valuable material for photocatalytic applications. However, the photocatalytic efficiency of bare g-C₃N₄ is still limited due to the high rate of recombination of the photo-generated electron–hole pairs. For further improvement, co-catalyst materials, including noble metals, semiconductors and carbon allotropes, have been combined with g-C₃N₄ to fabricate new composite photocatalysts.

It is well known that loading precious metals, such as Pt and Au, effectively enhances the activity of photocatalysts. However, these metals are rare and expensive to apply. Molybdenum disulfide (MoS₂) is an emerging photocatalytic cocatalyst material that may be used as a substitute for noble metals in synthesizing photocatalysts. As an indirect-gap semiconductor, MoS₂ has a rather narrow band gap (1.29 eV), and its conduction band (CB) and valence band (VB) edge potentials (-0.1 and +2.0 eV) are more positive than most photosensitive semiconductors. The difference between the CB edge potentials of a semiconductor and MoS₂ allows electron transfer from the semiconductor to MoS₂, which would make the CB electrons more mobile and hence promote the separation of electron-hole pairs on the semiconductor. Metal oxides and metal sulfates show improved
photocatalytic activity when MoS$_2$ is added as a synergistic cocatalyst.$^{11,12}$ Recently, a g-C$_3$N$_4$-MoS$_2$ composite was synthesized by mixing g-C$_3$N$_4$ and MoS$_2$ together, which was found to be effective for photocatalytic H$_2$ generation.$^{10}$ In this study, we prepared the g-C$_3$N$_4$-MoS$_2$ composites using a facial low temperature hydrothermal method to deposit MoS$_2$ as nanoparticles on the g-C$_3$N$_4$ sheets. The resulting composite catalyst exhibited a high photocatalytic activity and good stability for the degradation of methyl orange under simulated solar light.

2. Experimental

2.1 Synthesis of g-C$_3$N$_4$ from thiourea

Ten grams of thiourea powder was put into an alumina crucible with a cover that was placed in a tube furnace. The chemical was heated to 550 °C at a heating rate of 10 °C /min and kept at this temperature for 2 h in air. The air exhaust released during the thermal treatment was absorbed by a dilute NaOH solution (0.05 M). The resulting yellow powder collected was g-C$_3$N$_4$.\textsuperscript{13}

2.2 Synthesis of g-C$_3$N$_4$ supported MoS$_2$ nanoparticles

Half a gram prepared g-C$_3$N$_4$ was dispersed with sonication in 100 mL DI water with ammonium molybdate ((NH$_4$)$_6$Mo$_7$O$_{24}$$\cdot$4H$_2$O) and Na$_2$S•4H$_2$O. The mixture was stirred for 2 h to obtain a homogeneous solution. Ten mL 35% HCl solution was added and the solution was heated to 90 °C. After 30 min, 1 g NH$_2$OH•HCl was added and the solution was kept at this temperature for another 4 h to grow MoS$_2$ on g-C$_3$N$_4$.\textsuperscript{14} The composite, g-C$_3$N$_4$-MoS$_2$, was washed thoroughly with DI water and then dried in an oven at 60°C for 12 h.

2.3 Photocatalytic degradation of methyl orange
Methyl orange (MO), one of important classes of commercial dyes, was selected as a model chemical compound for photocatalytic experiments. Given the short life of its excited-state and its high stability under visible and near UV irradiation, MO has often been used as a model dye for testing photo-redox reactions that result in color reduction. For a typical MO decomposition test, 50 mg of the photocatalyst powder was first dispersed in 50 mL of water in a quartz photo-tube by sonication, followed by the addition of MO to an initial concentration of 20 mg/L. Before exposure to light, the suspension was stirred in the dark for 10 h to allow adsorption of MO by the catalyst. Photocatalysis of MO was performed in an XPA-7 photo-reactor (Xujiang Electromechanical Plant, Nanjing, China) with a 500 W xenon lamp for the simulated solar (SS) light. During the photocatalytic test, 3 mL of the solution was sampled from the photo-tube at regular time intervals to measure the MO concentration. Each sample was filtered and its absorbance (ABS) was measured by a UV-visible spectrophotometer (UV-vis lambda 25, Perkin Elmer) at 463 nm to determine the MO concentration in the solution.

2.4 Material characterization

The crystal forms of the photocatalysts were analyzed from their X-ray diffraction (XRD) patterns using a Bruker D8 Advance X-ray powder diffractometer. The morphology of the catalysts was examined by transmission electron microscopy (TEM) (Philips Tecnai G220 S-TWIN, Amsterdam, the Netherlands). Fourier transform infrared spectroscopy (FT-IR) (Perkin Elmer, FT-IR Spectrophoeometer Spectrum One B) was used to characterize the functional groups of different catalysts. The chemical states of MoS₂ nanoparticles were detected by X-ray photo-emission spectroscopy (XPS) (PHI 5600 Multi-Technique XPS System, Physical Electronics). The UV-vis diffusive reflectance spectra (DRS) of the catalyst powders were recorded by a spectrophotometer (Hitachi U-3010). The surface area and pore structure of catalyst powder samples was determined by a Beckman Coulter
SA3100 surface area analyzer from the nitrogen adsorption-desorption isotherm at liquid nitrogen temperature (77 K).

3. Results and Discussion

3.1 Characterization of the photocatalysts

The commonly used precursors for g-C₃N₄ are nitrogen rich compounds with a pre-bonded triple or double C–N core structure, such as cyanamide and dicyandiamide, which are unstable, highly explosive and toxic. In this study, a low cost, non-toxic chemical, thiourea, was used as the raw material to produce g-C₃N₄. The process is simple and easy to perform and can be completed in 2 h at 550 °C in the air atmosphere. MoS₂ nanoparticles were formed and deposited on g-C₃N₄ by the reduction of molybdate with NH₂OH·HCl at 90 °C. The reaction mechanisms for this process can be described as follows:

\[
\text{(NH}_4\text{)}_6\text{Mo}_7\text{O}_{24} + \text{Na}_2\text{S} \xrightarrow{\text{HCl}} \text{(NH}_4\text{)}_2\text{MoS}_4 + \text{NH}_4\text{OH} + \text{HCl} \rightarrow \text{MoS}_2
\]

The composite sample was first characterized by XPS to verify its chemical composition. As shown in Fig. S1a, the Mo3d spectrum exhibits two peaks at 229.4 eV (Mo3d₅/₂) and 232.4 eV (Mo3d₃/₂), indicating a +4 oxidation state for Mo in the composite. The peak at 163.2 eV in Fig. S1b can be attributed to S2p of S²⁻. These binding energies are all close to the reported values for MoS₂.¹⁰,¹⁵-¹⁷

Fig. 1 presents the XRD patterns of the g-C₃N₄ and g-C₃N₄-MoS₂ samples. The g-C₃N₄ spectra show a strong peak at 27.4° corresponding to the tight inter-layer stacking distance (0.325 nm) of the aromatic planes in g-C₃N₄. The other pronounced peak at 13.11° corresponds to an in-plane structural repeating motif with a repeated distance of 0.675 nm. Pure MoS₂ nanoparticles were prepared using the same method as described for the
composite, without the addition of g-C3N4. As shown in Fig. 1, the XRD profile for pure MoS2 displayed low and broad diffraction peaks. The pattern of broad peaks was likely caused by the combination of small-size particles, disorder in crystallinity and strains in the crystallites. The XRD pattern for g-C3N4-MoS2 is nearly the same as that for g-C3N4, due to the small percentage of MoS2 and its low diffraction intensity. The FT-IR spectra also show little difference between pure g-C3N4 and g-C3N4-MoS2 (Fig. S2).

The optical absorption property of g-C3N4-MoS2 composites with various MoS2 contents was analyzed. According to the DRS in Fig. 2, pure g-C3N4 displays absorption from UV through the visible range up to 460 nm, which can be ascribed to the band gap of g-C3N4 (2.7 eV). In the UV (≤400 nm) region, the composites’ absorption abilities follow the order g-C3N4-0.03MoS2 > g-C3N4-0.05MoS2 > g-C3N4-0.01MoS2 > g-C3N4. Generally, in the low energy visible region, the DRS intensity strengthened with an increase of the MoS2 content in the g-C3N4-MoS2 composite. The differences in optical adsorption agree well with the colors of the catalyst powders, presented in Fig. 2, which range from yellow to brown. The DRS features suggest that MoS2 doping can enhance the response of the catalyst composite towards solar light.

The TEM image of g-C3N4 in Fig. 3a shows a clear nanosheet structure similar to graphene. The nanosheets are very thin and transparent to the electron beam. The TEM image of pure MoS2 in Fig. 3b shows aggregated nanoparticles in a wide size distribution. At a higher magnification in Fig. 3c, a relatively weak crystallinity and strain of the MoS2 in the crystallites can be observed. The characteristic crystal lattice of MoS2 (0.62 nm) can be identified at the edge of the nanoparticles. The morphology and microstructure of the g-C3N4-
MoS2 composite are shown in detail in the TEM image in Fig. 3d. MoS2 nanoparticles on the g-C3N4 surface can be seen at the edge of the composite, and the MoS2 deposition does not appear to disrupt the g-C3N4 nanosheets.

Fig. 3.

The specific surface area and pore volume of the catalyst samples were measured using nitrogen adsorption (Fig. S3). The g-C3N4-MoS2 composites have a larger surface area than g-C3N4 (17.5 m²/g), but there is little difference between the composites with different MoS2 loading percentages. The N2 adsorption-desorption isotherms in Fig. S3a indicate that g-C3N4-0.03MoS2 has a greater N2 adsorption capability than pure g-C3N4. The diameter of the main pores increased with the MoS2 content, from 18 nm for g-C3N4 to 30 nm for g-C3N4-0.03MoS2 (Fig. S3b), which is likely to be the cause of the composite’s increased surface area.

3.2 Photocatalytic decomposition of methyl orange

The photocatalytic activity of the different catalysts was tested by the decoloring or decomposition of 20 mg/L MO under simulated solar light. Prior to the photo-tests, the changes in MO concentration caused by the adsorption of the catalyst materials were determined in the dark. The adsorption capability of g-C3N4 clearly increased with MoS2 deposition (Fig. S4). This is attributable to the increases in the surface area and pore size of the composites (Fig. S3). After adsorption, the equilibrium MO concentrations were used as the initial concentrations for the subsequent photocatalysis tests (Fig. 4). Composite g-C3N4-MoS2 exhibited a much greater photocatalytic activity than bare g-C3N4 for MO destruction. The photocatalytic efficiency for MO decoloration under SS light follows the order g-C3N4-0.03MoS2 > g-C3N4-0.05MoS2 > g-C3N4-0.01MoS2 > g-C3N4. Integration of 1% MoS2 into g-C3N4 greatly increased its photocatalytic activity. Increase of the MoS2 doping ratio to 3% further increased the activity of the photocatalyst. However, when the MoS2 ratio increased to
5%, the photo-activity of the catalyst decreased according to the MO degradation tests. The half-time ($T_{1/2}$) of MO decoloring by the photocatalysts can be determined from the MO reduction curves. Under SS light, the $T_{1/2}$ of MO for bare g-C3N4 was 1253 min, whereas the $T_{1/2}$ for g-C3N4-0.03MoS2 was only 213 min.

**Fig. 4.**

An unsuccessful attempt was made to further increase the activity of the photocatalyst by annealing the g-C3N4-0.03MoS2 composite at different temperatures. The photo-activity of the catalyst actually decreased after the annealing treatment; even at the maximum annealing temperature of 400 °C (g-C3N4 becomes unstable at 400 °C or higher). Detailed experiment and results are reported in the Supporting Information (Fig. S5).

The stability of the g-C3N4-0.03MoS2 composite as a photocatalyst was evaluated by repeating the MO decoloration tests. As shown in Fig. S6, the adsorption capacity of g-C3N4-0.03MoS2 decreased after the first use, but little further decrease was observed in the following test runs. The photo-activity of the catalyst was stable, as indicated by the repeated MO decoloring tests (Fig. 5). No significant deactivation was found for the recycled photocatalyst after four runs.

**Fig. 5.**

### 3.3 Photocatalytic MO degradation mechanisms by g-C3N4-MoS2

The mechanisms of photocatalytic MO degradation by the g-C3N4-MoS2 composite are illustrated in Fig. 6. MO molecules are readily adsorbed via conjugation by the s-triazine rings (C$_3$N$_3$) of g-C3N4, which is beneficial to the subsequent MO decomposition. Under SS light, irradiation of photosensitive g-C3N4 leads to electron excitation and electron–hole pair formation. The CB and VB edge potentials of polymeric g-C3N4 are reported at -1.13 and +1.57 eV. In comparison, the CB and VB edge potentials of MoS2 are more positive, at -0.1
and +2.0 eV. The difference between the CB edge potentials of g-C₃N₄ and MoS₂ allows electron transfer from the CB of g-C₃N₄ to the CB of MoS₂, which is favorable to the separation of electron-hole pairs on g-C₃N₄.⁸,¹⁰ Therefore, MoS₂ nanoparticles on the g-C₃N₄ sheets apparently act as electron collectors, similarly to noble metals, conducting photo-generated electrons from the main photocatalyst and thus preventing the electrons from recombining with the holes. Moreover, the presence of MoS₂ could provide more active sites for MO adsorption and degradation. The electrons would eventually be accepted by dissolved oxygen in the aqueous phase to facilitate the electron–hole separation. The holes can directly oxidize MO molecules adsorbed on the catalyst surface, resulting in color reduction. Moreover, the holes react with water (or hydroxyl) to form hydroxyl free radicals (·OH), which are a strong oxidant for MO decomposition.²,¹⁸ The MO degradation in water results in decoloration of the solution and formation of CO₂ and intermediate products, such as phenolic compounds and carboxylic acids. The major reaction steps during the photocatalytic process may be described by the following equations (1–6):

\[
g\text{-C}_3\text{N}_4 + \text{hv} \rightarrow g\text{-C}_3\text{N}_4 (e^- + h^+) \tag{1}
\]

\[
g\text{-C}_3\text{N}_4 (e^-) + \text{MoS}_2 \rightarrow g\text{-C}_3\text{N}_4 + \text{MoS}_2 (e^-) \tag{2}
\]

\[
\text{MoS}_2 (e^-) + 1/2\text{O}_2 \rightarrow \text{MoS}_2 + \text{O}_2^- \tag{3}
\]

\[
g\text{-C}_3\text{N}_4 (h^+) + \text{MO} \rightarrow \text{CO}_2 + \text{H}_2\text{O} + \text{other products} \tag{4}
\]

\[
g\text{-C}_3\text{N}_4 (h^+) + \text{OH}^- \rightarrow g\text{-C}_3\text{N}_4^+ + \cdot\text{OH} \tag{5}
\]

\[
\cdot\text{OH} + \text{MO} \rightarrow \text{CO}_2 + \text{H}_2\text{O} + \text{other products} \tag{6}
\]
4. Conclusions

Novel g-C₃N₄-MoS₂ composite photocatalysts were prepared using a facile, low temperature hydrothermal method, with MoS₂ formed as nanoparticles on the g-C₃N₄ nanosheets. The composite catalysts exhibited much greater photocatalytic activity than pure g-C₃N₄ for the decoloration and degradation of methyl orange under simulated solar light, and the 3.0wt% MoS₂/g-C₃N₄ composite had the greatest activity. An annealing process failed to further increase the activity of the photocatalysts. The enhanced photocatalytic activity can be attributed to the synergetic function of MoS₂ in the composite catalyst. The presence of MoS₂ nanoparticles apparently increased the interfacial charge transfer and thus reduced electron–hole recombination. The photocatalyst also displayed good stability during the photo-reactions and no obvious deactivation was found for the recycled catalyst after four test runs. The MoS₂/g-C₃N₄ composite is thus a promising photocatalyst with a high reactivity and stability for photocatalytic degradation of organic pollutants using solar energy.

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References


19.
Figure Captures

Fig. 1. XRD patterns of pure MoS$_2$, pure g-C$_3$N$_4$, and the g-C$_3$N$_4$-MoS$_2$ composite.

Fig. 2. UV-vis diffusive reflectance spectra of g-C$_3$N$_4$ and g-C$_3$N$_4$-MoS$_2$ composites.

Fig. 3. (a) TEM image of pure g-C$_3$N$_4$, (b) TEM images of pure MoS$_2$, (c) HR-TEM images of pure MoS$_2$ and (d) TEM image of g-C$_3$N$_4$-MoS$_2$ composite.

Fig. 4. Degradation of MO under simulated solar light by pure g-C$_3$N$_4$, and the g-C$_3$N$_4$-MoS$_2$ composites.

Fig. 5. Photocatalytic MO degradation by the recycled photocatalysts in repeated test runs.

Fig. 6. Schematics of the mechanisms of photocatalytic MO degradation by the g-C$_3$N$_4$-MoS$_2$ composite under SS light.
Fig. 1. XRD patterns of pure MoS$_2$, pure g-C$_3$N$_4$ and the g-C$_3$N$_4$-MoS$_2$ composite
Fig. 2. UV-vis diffusive reflectance spectra of g-C₃N₄ and g-C₃N₄-MoS₂ composites.
Fig. 3. (a) TEM image of pure g-C₃N₄, (b) TEM images of pure MoS₂, (c) HR-TEM images of pure MoS₂ and (d) TEM image of g-C₃N₄-MoS₂ composite.
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Supporting Information

Synthesis of MoS$_2$/g-C$_3$N$_4$ as a Solar Light-Responsive Photocatalyst for Organic Degradation

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Fig. S1. XPS spectra of MoS$_2$ nanoparticles deposited on g-C$_3$N$_4$: (a) Mo (3d) spectrum and (b) S (2p) spectrum.

Mo$_{3d_{5/2}}$ Mo$_{3d_{3/2}}$ S$_{2p}$
Fig. S2. FT-IR profiles of pure g-C₃N₄ and the g-C₃N₄-MoS₂ composite.

The FT-IR spectrum of pure g-C₃N₄ clearly shows several peaks at the frequency characteristic of the vibrational modes related to the chemical bonding between carbon and nitrogen. The adsorption peak at 810 cm⁻¹ corresponds to the breathing mode of the triazine units. Several strong bands in the 1240-1645 cm⁻¹ region can be attributed to the stretching modes of C-N heterocyclics (e.g. 1321 cm⁻¹ C-N stretching and 1641 C=N cm⁻¹ stretching). The broad absorption band at ~3200 cm⁻¹ can be assigned to the stretching modes of secondary and primary amines and their intermolecular hydrogen-bonding interactions. No major differences can be seen between the pure g-C₃N₄ and the g-C₃N₄-MoS₂ composite spectra. Therefore, MoS₂ doping did not change the functional groups of g-C₃N₄ and no new chemical bonds were generated during this process. However, the intensity of the peaks (1100-1750 cm⁻¹) for the g-C₃N₄-MoS₂ composite is relatively lower than those of pure g-C₃N₄. This was likely caused by the layer of MoS₂ on the g-C₃N₄ surface, which decreased the transmittance of infrared rays from g-C₃N₄.
Fig. S3. (a) N₂ adsorption-desorption isotherms at 77 K and (b) pore size distributions of g-C₃N₄ and g-C₃N₄-MoS₂.
Fig. S4. Change in MO concentration after adsorption by the different catalyst materials (g-C₃N₄ and its composites with different amounts of MoS₂)
Thermal treatment of MoS$_2$/g-C$_3$N$_4$

The g-C$_3$N$_4$-0.03MoS$_2$ composite was placed in a crucible that was placed in a tube furnace. An N$_2$ flow (50 mL/min) was applied for 2 h to remove air from the furnace. The temperature was increased at a rate of 5 °C/min to the required temperature (200, 300 or 400 °C) and then maintained at that temperature for 2 h. A temperature higher than 400 °C will break the structure of g-C$_3$N$_4$, so this was set as the upper temperature limit. The furnace was cooled down naturally to room temperature to complete the annealing process.

The materials obtained were characterized by XRD and the results are shown in Fig. S5a. No major differences can be seen between the different samples. The catalysts were tested for photocatalytic MO degradation under the same conditions. Fig. S5b shows that their adsorption capacity decreased after the annealing treatment. In addition, the photoactivity of the thermally treated catalyst did not show any increase for MO degradation (Fig. S5c). The annealing process therefore did not enhance the photocatalytic activity of g-C$_3$N$_4$-0.03MoS$_2$. 

![Graphs showing XRD and Photocatalytic activity results](image)
Fig. S5 (a) XRD patterns of g-C$_3$N$_4$-0.03MoS$_2$ annealed at different temperatures in the N$_2$ atmosphere, (b) the adsorption capability of the different catalyst materials and (c) photocatalytic degradation of MO by the different photocatalysts.

Fig. S6. The adsorption capacity of the recycled catalyst materials