A reusable chitosan/TiO$_2@$g-C$_3$N$_4$ nanocomposite membrane for photocatalytic removal of multiple toxic water pollutants under visible light

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**ABSTRACT**

Photocatalysis has been proved to be a promising approach in wastewater purification. However, it is hard to recycle powdery photocatalysts from wastewater in industry, but immobilizing them using larger materials can overcome this drawback. For that reason, TiO$_2@g$-C$_3$N$_4$ was embedded into chitosan to synthesize a highly reusable and visible-light-driven chitosan/TiO$_2@g$-C$_3$N$_4$ nanocomposite membrane (CTGM). CTGM showed enhanced photoactivity and the photocatalytic efficiencies of the toxic water pollutants methyl orange (M.O.), rhodamine B (Rh.B), chromium (VI) (Cr (VI)), 2,4-dichlorophenol (2,4-DCP) and atrazine (ATZ) were more than 90% under visible light at ambient conditions. Significantly, CTGM was easy to recycle and showed excellent reusability: there was no decrease in the photocatalytic decolorization efficiency of Rh.B throughout 10 cycles. A continuous-flow photocatalysis system was set up and 90% of Rh.B was effectively decolorized. A simple approach was developed to prepare a novel, effective and visible-light-driven membrane that was easy to reuse, and a feasible photocatalysis continuous-flow system was designed to be a reference for wastewater treatment in industry.

**Key words** | continuous-flow photocatalysis, immobilization, membrane, TiO$_2@g$-C$_3$N$_4$, wastewater purification

**HIGHLIGHTS**

- TiO$_2@g$-C$_3$N$_4$ was embedded into chitosan through a simple process.
- High-efficiency removal of multiple toxic water pollutants under visible light.
- CTGM is easy to recycle simply by washing with water.
- CTGM presents excellent reusability.
- A feasible photocatalysis continuous-flow system was designed.
INTRODUCTION

Wastewater pollution has become a worldwide concern (Ma et al. 2018). Many water pollutants are persistent and difficult to degrade in the environment, and it has been proven that heavy metals and organic pollutants are harmful to human health, leading to cancers, renal failure, angiocardiopathy, hypertension, etc. (Grandclement et al. 2017; Shahid et al. 2019), so suitable approaches to treating wastewater are urgently needed.

Semiconductor photocatalysis has been widely applied in wastewater purification because of its low energy requirements and mild reaction conditions (Zhang et al. 2018). Semiconductor photocatalysts can interact with light to generate active species for reduction or oxidation, thus effectively removing pollutants in wastewater (Kumar et al. 2019; Bellardita et al. 2020). Among the semiconductors, TiO2 has been widely applied to wastewater treatment due to its low cost, low toxicity and high chemical stability (Penboon et al. 2019; Ding et al. 2020). However, applications of pure TiO2 are restricted due to its poor use of visible light and the rapid recombination of photogenerated carriers (Xiu et al. 2020). As a metal-free organic polymeric semiconductor, g-C3N4 has attracted much attention in the field of photocatalysis due to its low cost, non-toxicity, high stability, visible-light response, attractive electronic properties and suitable band edge (Wang et al. 2018; Liu et al. 2020a). Constructing a heterojunction between g-C3N4 and TiO2 (TiO2@g-C3N4) has been proven to be one of the efficient methods for enhancing photogenerated carrier separation and responsiveness visible light (Xiao et al. 2018).

However, the powdery TiO2@g-C3N4 is hard to separate from wastewater and reuse in industry, and could add potential threats to ecosystems and human health if released into the environment. Immobilization of TiO2@g-C3N4 using larger materials is a viable approach to overcoming the above drawbacks (Melinte et al. 2019). However, immobilization weakens the adsorption capacity of photocatalysis, leading to low reactivity (Yang et al. 2019). Thus, suitable supports should meet two requirements: (1) ease of separation from wastewater; (2) high adsorption potential for photocatalysts. As a support material, chitosan has attracted widespread interest because it is naturally abundant, non-toxic and biodegradable (Wang et al. 2018a; Kumar et al. 2020). Moreover, being a cationic polysaccharide with abundant hydroxyl and amino groups, chitosan has tremendous potential for removing various pollutants such as heavy metals, dyes, and pathogens, and these characteristics make it an ideal bio-adsorbent for wastewater treatment (Xiao et al. 2015b). Therefore, chitosan is an appropriate support to immobilize TiO2@g-C3N4.

This paper describes a simple and easy approach to immobilizing TiO2@g-C3N4 using chitosan and therefore preparing a highly reusable and visible-light-driven chitosan/TiO2@g-C3N4 nanocomposite membrane (CTGM). The membrane can be used for the efficient photocatalytic removal of different kinds of pollutants from wastewater, and shows excellent reusability without any decrease in photoactivity after 10 cycles. Compared to the slurry reaction system, continuous-flow chemistry is easy to scale up and has been extensively used for decades in the chemical industry.
(Yang et al. 2020). So a continuous-flow photocatalysis system was set up to decolorize rhodamine B (Rh.B), simulating wastewater treatment in industry. A simple and easy method was created to prepare visible-light-driven membranes with easy recovery and high reusability, and a feasible photocatalysis continuous-flow system was designed to provide a reference for scaling up water purification.

**EXPERIMENTAL METHODS**

**Chemicals**

Chitosan (biological reagent) was purchased from Sinopharm Chemical Reagent Co., Ltd (China). Titanium dioxide (TiO₂, Degussa P25, 80% anatase, 20% rutile, 50 m²·g⁻¹) was purchased from Shanghai King Chemical Co., Ltd (China). Urea (≥99.0%) was purchased from Xilong Scientifin Co., Ltd (China). Sodium hydroxide (NaOH, ≥96%), phosphoric acid (H₃PO₄, ≥85.0%), sulfuric acid (H₂SO₄, 98%), methyl orange (M.O.) (≥99.7%) and Rh.B (≥99.7%) were purchased from Beijing Chemical Works (China). Potassium dichromate (K₂Cr₂O₇, ≥99.95%) was purchased from Tianjin Guangfu Fine Chemical Research Institute. Atrazine (ATZ; 97%) was purchased from Shanghai Yuanye Bio-Technology Co., Ltd (China). 2,4-dichlorophenol (2,4-DCP; ≥99.5%) was purchased from Macklin (China). Acetic acid (≥99.5%) was purchased from Damao Chemical Reagent Factory. Methanol (≥99.9%) and acetonitrile (≥99.9%) were purchased from Thermo Fisher Scientific-CN.

**Preparation of the CTGM**

**Preparation of CTGM in different chitosan/TiO₂@g-C₃N₄ weight ratios**

The synthesis method of TiO₂@g-C₃N₄ was the same as in our previous work (Xiao et al. 2018). CTGM was made as follows: 20 mg chitosan was dissolved in 5 mL acetic acid (2.5% v/v); 25 mg TiO₂@g-C₃N₄ was added to the chitosan solution (chitosan/TiO₂@g-C₃N₄ = 4/5) and homogeneously dispersed by ultrasonication for 1 min. 1 mL of the mixed slurry was completely spread on the glass substrates (d = 4 cm) to dry at room temperature, after which the dried membranes were immersed in NaOH (0.25 M) for 1 hour. In the process, the positive charges of the acetic acid were neutralized to form a stable network among the chitosan polymeric chains (Bergonzi et al. 2019). Finally, the membranes were washed to neutral with deionized water and then dried at room temperature. The membranes obtained were labeled 4/5-CTGM. Membranes with different chitosan/TiO₂@g-C₃N₄ weight ratios of 2/1, 4/3 and 1/1 were prepared following the same process and labeled as 2/1-CTGM, 4/3-CTGM and 1/1-CTGM respectively.

**Preparation of 4/5-CTGM of different thicknesses**

Following the same process, a mixed slurry with the chitosan/TiO₂@g-C₃N₄ weight ratios of 4/5 was prepared. 0.5 mL, 0.75 mL, 1.0 mL, 1.5 mL of the mixed slurry were each spread on glass substrates, after which the preparation process was same as described in ‘Preparation of CTGM in different chitosan/TiO₂@g-C₃N₄ weight ratios’. The resulting 4/5-CTGM of different thicknesses were labeled as 0.50–4/5-CTGM, 0.75–4/5-CTGM, 1.00–4/5-CTGM, 1.50–4/5-CTGM. Pure chitosan membranes labeled as CS were prepared following the steps of 0.75–4/5-CTGM without TiO₂@g-C₃N₄. 25 mg TiO₂@g-C₃N₄ was directly dispersed into 5 mL deionized water by ultrasonication, and 0.75 mL of the slurry was spread on the glass substrates to obtain pure TiO₂@g-C₃N₄ membranes.

**Characterization methods**

The surface morphology and chemical components of the materials were analyzed using a scanning electron microscope (SEM, JSM-7610FPlus, Jeol, Japan). The specific surface area and pore structure of materials were analyzed using the Brunauer–Emmett–Teller method (BET, ASAP 2460 3.01, Micromeritics, USA). The hydrophilicity of materials was analyzed using a contact angle goniometer (CA, JC2000D, Shanghai Zhongchen Corporation, China). The functionalization of materials was analyzed using Fourier transform infrared spectroscopy (FTIR, 6700, Nicolet, USA). The crystal phases of materials were analyzed using an X-ray diffractometer (XRD, Ultima IV, Rigaku, Japan). The UV–vis diffuse reflection spectra (DRS) of materials were obtained using a UV–vis spectrometer (UV-3600, Shimadzu, Japan).

**Photocatalytic studies**

All photocatalytic experiments were carried out in a batch photoreactor (Figure S1, Supplementary Information) at ambient conditions. The membrane was placed below the aqueous solution. With a cold water bath, the reactor (d = 4.5 cm, h = 5.5 cm) was covered with a quartz plate under
a 100 W LED lamp (400 nm ≤ λ ≤ 800 nm). At each sampling time, a certain amount of the reaction solution was removed for analysis. For comparison, photolysis experiments were carried out at the same conditions without photocatalyst and absorption experiments were carried out in the dark. The absorbances of M.O. and Rh.B were measured at 464 nm (OD 464) and 554 nm (OD 554), respectively, on a UV-vis spectrophotometer (N4, Jingke, China). The concentration of Cr(VI) was measured at 540 nm using the diphenylcarbazide colorimetric method. The concentrations of 2,4-DCP and ATZ were analyzed using high performance liquid chromatography (HPLC, 1260 Infinity II, Agilent, USA). The above reactions were performed in triplicate to obtain reproducible results and the average values were reported with their standard deviation.

To prove the reusability of 0.75-4/5-CTGM, a recycling experiment was carried out. After each cycle of photocatalytic decolorization of Rh.B, 0.75-4/5-CTGM was fully washed using deionized water and then dried at 60 °C for the next reaction.

The Langmuir-Hinshelwood pseudo-first-order model is widely applied to analyze the photocatalytic kinetics for the transformation of various pollutants in water at a dilute concentration (Jiang et al. 2016), which can be expressed as the following equation (Equation (1)):

\[ -\ln \left( \frac{C_t}{C_0} \right) = k_{app} \times t \]  

where \( C_0 \) (mg/L) and \( C_t \) (mg/L) are the concentrations of pollutants at the beginning and time \( t \) respectively, \( k_{app} \) (min\(^{-1}\)) is the apparent first-order rate constant, and \( t \) (min) is the irradiation time.

**Setup of the continuous-flow photocatalysis system**

A certain amount of the mixed slurry with the chitosan/TiO\(_2\)@g-C\(_3\)N\(_4\) weight ratios of 4/5 was coated onto the inner wall of a borosilicate glass tube (d = 0.6 cm, l = 10 cm), which was then used as the photoreactor. As shown in Figure S2 ( Supplementary Information), the continuous-flow photocatalysis system consists of the photoreactor placed under a 300 W xenon lamp (CEL-HXFS00, Beijing China Education Au-light Co., Ltd), a pump and a storage tank containing the Rh.B solution (V = 10 mL). In the continuous-flow photocatalysis system, the flow velocity, irradiation intensity and concentration of Rh.B are changed to different values for exploring their influence on the photocatalytic decolorization of Rh.B.

**RESULTS AND DISCUSSION**

**Characterization**

**Surface morphology**

The SEM images in Figure 1 show the surface morphology of the CS and the 0.75-4/5-CTGM. Figure 1(a) shows the smooth and dense surface morphology of the CS, and Figure 1(b) reveals its layer thickness is 2.5 μm. Figure 1(c) shows a rough surface morphology and the rough, porous and fluffy microstructure of 0.75-4/5-CTGM due to the addition of TiO\(_2\)@g-C\(_3\)N\(_4\). As shown in Figure 1(d), the thickness of the 0.75-4/5-CTGM is 21 μm. The porous and fluffy microstructure of 0.75-4/5-CTGM can increase its specific surface area as shown in Table 1, and a larger specific surface area can improve adsorption and provide more reactive sites (Cao et al. 2016; Zhang et al. 2021). The SEM image and the energy dispersive X-ray analysis (EDX) mapping of Ti, C, N and O of 0.75-4/5-CTGM are shown in Figure 1(e) and 1(f). As shown in Figure 1(f), the elemental mapping of Ti is uniformly dispersed in the membrane, demonstrating that TiO\(_2\)@g-C\(_3\)N\(_4\) was evenly embedded in the 0.75-4/5-CTGM.

**Water contact angle**

Figure 2 shows the water contact angles (WCA) of the CS and the 0.75-4/5-CTGM. The CS presents the WCA of 93.16° > 90° in Figure 2(a), indicating its hydrophobicity. 0.75-4/5-CTGM has a WCA of 17.50° < 90° in Figure 2(b), indicating its good hydrophilicity. As a hydrophilic material containing many functional groups such as hydroxyl and amine (Venkatesh et al. 2020), TiO\(_2\)@g-C\(_3\)N\(_4\) increases 0.75-4/5-CTGM’s hydrophilicity. The large surface roughness of 0.75-4/5-CTGM is also good for hydrophilicity (Duan et al. 2020), which leads to better anti-pollution properties (Liu et al. 2020b).

**FTIR spectra**

The FTIR spectra of CS, 0.75-4/5-CTGM and TiO\(_2\)@g-C\(_3\)N\(_4\) are shown in Figure 3(a). For the 0.75-4/5-CTGM FTIR spectra, strong broad bands extending from 3,300 to 3,500 cm\(^{-1}\) (OH and -NH stretches), broad bands extending from 2,867 to 2,939 cm\(^{-1}\) (CH stretching vibrations) and 1,060 cm\(^{-1}\) (C-O stretching) indicate the presence of chitosan (Li et al. 2019; Zheng et al. 2020). Moreover, the peaks...
at 1,253–1,620 cm\(^{-1}\) (typical vibrational modes of C-N and C = N) and 806 cm\(^{-1}\) (tri-s-triazine stretching vibration) both appear in the FTIR spectra of 0.75–4/5-CTGM and prove the existence of g-C\(_3\)N\(_4\) (Du et al. 2020). The peak of Ti-O-Ti does not appear in the FTIR spectra of

<table>
<thead>
<tr>
<th>Photocatalyst</th>
<th>Surface area (m(^2)/g)</th>
<th>Pore volume (cm(^3)/g)</th>
<th>Average pore size (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CS</td>
<td>27.20</td>
<td>0.072</td>
<td>10.65</td>
</tr>
<tr>
<td>TiO(_2)@g-C(_3)N(_4)</td>
<td>44.27</td>
<td>0.057</td>
<td>5.12</td>
</tr>
<tr>
<td>0.75–4/5-CTGM</td>
<td>59.04</td>
<td>0.150</td>
<td>10.16</td>
</tr>
</tbody>
</table>
0.75–4/5-CTGM, probably due to the low content of TiO$_2$. According to the above results, the as-synthesized 0.75–4/5-CTGM consists of TiO$_2$@g-C$_3$N$_4$ and chitosan.

**XRD patterns**

The XRD patterns of CS, g-C$_3$N$_4$, TiO$_2$, TiO$_2$@g-C$_3$N$_4$ and 0.75–4/5-CTGM are shown in Figure 3(b). For 0.75–4/5-CTGM, the characteristic diffraction peaks of TiO$_2$ are well matched to either the anatase or the rutile phase because both of the phases exist in the commercial TiO$_2$ (P25) (Giannakopoulou et al. 2017); and the minor peak around 13.2° is well matched to (100) crystal faces of g-C$_3$N$_4$ (Li et al. 2021). An amorphous diffraction peak around 20.2° indicates the presence of chitosan in 0.75–4/5-CTGM (Farhadian et al. 2019). The above analysis results indicate that TiO$_2$@g-C$_3$N$_4$ was successfully immobilized by chitosan and the crystal structures of TiO$_2$@g-C$_3$N$_4$ remained unchanged during the preparation process.

**UV-vis DRS spectra**

As shown in Figure 4, there is a bathochromic shift in 0.75–4/5-CTGM compared to TiO$_2$@g-C$_3$N$_4$. TiO$_2$@g-C$_3$N$_4$ exhibits a strong absorption band showing a steep edge with the extrapolated onset of 422 nm and a weak absorption band starting from the onset to the end around 705 nm. By contrast, 0.75–4/5-CTGM exhibits a strong absorption band showing a steep edge with the extrapolated onset of 440 nm and an enhanced absorption band starting from the onset to the end at around 730 nm. So 0.75–4/5-CTGM performs higher absorption of visible light.

**Photocatalytic performance**

As shown in Figure 5(a), the photocatalytic activities of CTGM are improved with the decrease of the chitosan/TiO$_2$@g-C$_3$N$_4$ weight ratio. The decolorization efficiency of M.O. reaches 96.35% at 120 min under visible light when the chitosan/TiO$_2$@g-C$_3$N$_4$ weight ratio reaches 4/5. It leads to poor film formation of the mixed slurry when the chitosan/TiO$_2$@g-C$_3$N$_4$ weight ratio is below 4/5. As shown in Figure 5(b), the thickness of the 4/5-CTGM influences the photocatalytic activities. For the 0.50–4/5-CTGM, 0.75–4/5-CTGM, 1.00–4/5-CTGM and 1.50–4/5-CTGM, the decolorization efficiencies of M.O. are 83.16%, 93.53%, 96.35% and 96.00% respectively at 120 min under visible light, with no appreciable gaps among the 0.75–4/5-CTGM, 1.00–4/5-CTGM and 1.50–4/5-CTGM. Therefore, 0.75–4/5-CTGM was chosen for the following experiments, considering the cost and activity. As shown in Figure 5(c), M.O. was hardly decolorized without photocatalyst under visible light, indicating that there was no self-decolorization.
of M.O. under this reaction condition. 93.53% of M.O. was decolorized on 0.75–4/5-CTGM, while only 53.76% and 68.93% of M.O. was decolorized over CS and TiO2@g-C3N4 under visible light. The photoactivity of 0.75–4/5-CTGM was obviously improved because of the synergistic effect between adsorption of chitosan on anionic dyes and photocatalysis of TiO2@g-C3N4 (Zainal et al. 2013). While in the dark, the decolorization efficiency of M.O. on the 0.75–4/5-CTGM was much lower, indicating the visible-light-driven photocatalytic activity. The kinetic curve of decolorization of M.O. on the 0.75–4/5-CTGM under visible light shows a good linear correlation between ln(Cf/C0) and t (R² = 0.998), which indicates the experiment data fit well with the pseudo-first-order kinetic model and the kapp is 0.0238 min⁻¹ (Figure S3, Supplementary Information).

Except for M.O., the decolorization of Rh.B and the removal of several kinds of typical aqueous pollutants, including Cr(VI), 2,4-DCP and ATZ, on the 0.75–4/5-CTGM under visible light were studied. As shown in Figure 6, the photocatalytic efficiencies of all the pollutants were over 90% without any strong oxidizer on the 0.75–4/5-CTGM under visible light, and all the kinetics data of photocatalytic reactions of Rh.B, Cr(VI), 2,4-DCP and ATZ fit well with the pseudo-first-order correlation (Figure S3).

Reusability study

The recycling performance of 0.75–4/5-CTGM is shown in Figure 7. As shown in Figure 7(a), there was no decrease in the decolorization efficiency of Rh.B throughout 10 cycles and the decolorization efficiency was at least 93.83%. Additionally, 0.75–4/5-CTGM kept its initial appearance throughout 10 cycles (as shown in Figure 7(c)), while most of TiO2@g-C3N4 was washed away after just one use (as shown in Figure 7(e)). Compared to immobilized photocatalysts in other studies, as shown in Table 2, 0.75–4/5-CTGM as synthesized presents high photoactivity and excellent reusability for decolorizing Rh.B under visible light. Moreover, the recovery process of 0.75-CTGM from wastewater was easy, thus demonstrating that the membrane has great practical value.
Continuous-flow photocatalysis

Using the optimized reaction conditions, the continuous-flow photocatalysis system shown in Figure S2 was set up to simulate the general process of industrial wastewater purification. The influences of irradiation intensity, flow velocity and concentration for the decolorization of Rh.B were studied and the results are shown in Figure 8. A positive dependence of the photocatalytic activity on the incident light intensity, which has been generally observed for photocatalytic processes catalyzed by both semiconductors and metal nanoparticles, is considered to be an
experimental signature of light-induced reactions mediated by photogenerated charge carriers (Kasap et al. 2016; Aslam et al. 2017; Huang et al. 2017). Figure 8(a) shows that the photocatalytic performance for Rh.B decolorization shows a positive dependence on the incident light intensity as expected (V = 4.5 L/min, C = 5 ppm). It is proved that chitosan does not prevent TiO$_2$@g-C$_3$N$_4$ from responding to light and can even have a good synergistic effect on visible light absorption, as shown in Figure 4. In Figure 8(b), the decolorization efficiencies of Rh.B are shown under different flow velocities (I = 100 mW/cm$^2$, C = 5 ppm). The photocatalytic decolorization efficiency

<table>
<thead>
<tr>
<th>Photocatalyst</th>
<th>Light source and intensity</th>
<th>Dosage (g/g Rh.B)</th>
<th>k (min$^{-1}$)</th>
<th>Cycles</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>g-C$_3$N$_4$ immobilized by carboxymethyl cellulose and β-cyclodextrin</td>
<td>300 W Xenon lamp, λ $\geq$420 nm, I = 280 mW/cm$^2$</td>
<td>2,000</td>
<td>40.4 $\times$ 10$^{-3}$</td>
<td>5</td>
<td>Qi et al. (2019)</td>
</tr>
<tr>
<td>TiO$_2$ immobilized by polyethylene glycol</td>
<td>UV black light lamp, λ $\leq$365 nm, I = 12.8 W/m$^2$</td>
<td>500</td>
<td>0.93 $\times$ 10$^{-3}$</td>
<td>5</td>
<td>Hu et al. (2017)</td>
</tr>
<tr>
<td>TiO$_2$@g-C$_3$N$_4$ immobilized by Fe$_3$O$_4$@SiO$_2$ magnetic core</td>
<td>Sunlight</td>
<td>50</td>
<td>20.0 $\times$ 10$^{-3}$</td>
<td>3</td>
<td>Narzary et al. (2020)</td>
</tr>
<tr>
<td>TiO$_2$@g-C$_3$N$_4$ immobilized by chitosan</td>
<td>100 W LED lamp, 400 nm $\leq$ λ $\leq$ 800 nm, I = 20 mW/cm$^2$</td>
<td>150</td>
<td>23.8 $\times$ 10$^{-3}$</td>
<td>10</td>
<td>This work</td>
</tr>
</tbody>
</table>

Figure 8 | The influences of (a) irradiation intensity, (b) flow velocity and (c) concentration for the photocatalytic decolorization of Rh.B in the photocatalysis continuous-flow system.
of Rh.B was gradually improved with the flow velocity increasing to 4.5 L/min, but decreased at the flow velocity of 6 L/min. In the experiment, many small and dense bubbles were produced in the photoreactor when the flow velocity increased to 6 L/min. It was observed that there were many bubbles if the flow velocity was excessively high and they might have limited the mass transfer between the substrate and the 0.75–4/5-CTGM coated onto the inner wall of the borosilicate glass tube and the absorption of light on 0.75–4/5-CTGM. The widespread occurrence of trace pollutants in water has become a serious threat to environmental safety and human health (Munoz et al. 2021). As shown in Figure 8(c), different concentrations of Rh.B were decolorized in the photocatalysis continuous-flow system (I = 100 mW/cm², V = 4.5 L/min). At the low concentration of 5 ppm, 90% of Rh.B was decolorized, which demonstrates that trace pollutants in water can be effectively removed in the photocatalysis continuous-flow system. This is a feasible photocatalysis continuous-flow system that serves as a reference for wastewater treatment in industry.

**CONCLUSIONS**

In summary, TiO2@g-C3N4 was embedded in chitosan to prepare a highly reusable and visible-light-driven membrane using an easy process. The rate of chitosan and TiO2@g-C3N4 and the slurry dosage were optimized to obtain 0.75–4/5-CTGM. 0.75–4/5-CTGM has a rough, porous and fluffy surface morphology and a larger specific surface area. TiO2@g-C3N4 is evenly dispersed on the surface with unchanged crystal structures. 0.75–4/5-CTGM performs effective photocatalysis due to the synergistic effect between the absorption of chitosan and the photocatalysis of TiO2@g-C3N4. In addition, 0.75–4/5-CTGM absorbs visible light more efficiently because of the obvious red shift compared to TiO2@g-C3N4. Based on the above characters, the photocatalytic efficiencies of 0.75–4/5-CTGM for M.O., Rh.B, Cr (VI), 2,4-DCP and ATZ are all over 90%. The high hydrophilicity enhances the anti-pollution capacity of 0.75–4/5-CTGM, and so improves its reusability. In the recycling experiment, 0.75–4/5-CTGM was easy to recycle, simply needing to be washed in water and there was no decrease in photocatalytic activity after 10 cycles. A photocatalysis continuous-flow system was also set up to effectively decolorize Rh.B under visible light, simulating wastewater treatment in industry. Consequently, this work provides some novel and deep insights into immobilizing photocatalysis for purifying wastewater.

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**DATA AVAILABILITY STATEMENT**

All relevant data are included in the paper or its Supplementary Information.

**REFERENCES**


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