

Article

Study on Photocatalytic Performance of Bi₂O₃-TiO₂/Powdered Activated Carbon Composite Catalyst for Malachite Green Degradation

Yajun Chen ^{1,*}, Man Cai ¹, Junfeng Li ¹ and Wenshuo Zhang ^{2,3}

¹ College of Environmental Science and Engineering, Yangzhou University, Yangzhou 225000, China; 17605274628@163.com (M.C.); 18118246107@163.com (J.L.)

² College of Pharmacy and Chemistry & Chemical Engineering, Taizhou University, Taizhou 225300, China; wenshuozhang@ustc.edu.cn

³ National Synchrotron Radiation Laboratory, University of Science and Technology of China, Hefei 230029, China

* Correspondence: 007822@yzu.edu.cn

Abstract: In this study, a Bi₂O₃-TiO₂/PAC ternary composite photocatalyst was successfully synthesized via a hydrothermal method, employing powdered activated carbon (PAC) as the support and using bismuth nitrate and tetrabutyl titanate as raw materials. The external morphology, microstructure, elemental composition, and optoelectronic properties of the catalyst were characterized by XRD, SEM, TEM, XPS, UV-Vis DRS, and BET analyses. The photocatalytic activity of the composite toward the degradation of malachite green (MG) was systematically evaluated under various conditions. The results revealed that the composite exhibited excellent photocatalytic activity, achieving a degradation efficiency of up to 99%. Apart from extremely acidic or alkaline conditions, MG removal efficiency increased with a rising solution pH. Moreover, the photocatalyst exhibited excellent adaptability and stability in the presence of coexisting inorganic anions and humic substances, indicating its broad potential for practical applications. Reactive-species-trapping experiments indicated that superoxide radicals ($\cdot\text{O}_2^-$) were the primary active species in the degradation process, with hydroxyl radicals ($\cdot\text{OH}$) and photogenerated holes (h^+) acting synergistically. Moreover, the catalyst maintained over 90% removal efficiency after five consecutive cycles, demonstrating its excellent stability and reusability. This work provides a promising strategy and theoretical foundation for the efficient photocatalytic treatment of MG-contaminated wastewater.

Keywords: photocatalysis; Bi₂O₃-TiO₂/PAC composite; malachite green



Academic Editors: Chiara Lo Porto and Vincenza Armenise

Received: 17 April 2025

Revised: 2 May 2025

Accepted: 8 May 2025

Published: 12 May 2025

Citation: Chen, Y.; Cai, M.; Li, J.; Zhang, W. Study on Photocatalytic Performance of Bi₂O₃-TiO₂/Powdered Activated Carbon Composite Catalyst for Malachite Green Degradation. *Water* **2025**, *17*, 1452. <https://doi.org/10.3390/w17101452>

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1. Introduction

Malachite green (MG) is a synthetic dye extensively employed in aquaculture, food processing, and the textile industry, owing to its inexpensive preparation, excellent antibacterial properties, and vibrant coloration [1,2]. However, MG has been recognized as a hazardous environmental pollutant, owing to its high toxicity, mutagenicity, and potential carcinogenicity. Once released into the environment, it may pose a serious risk to aquatic ecosystems and further endanger human health through bioaccumulation and prolonged exposure [3]. Moreover, some studies have shown that the toxicity of MG increases with extended exposure, elevated temperatures, and higher concentrations [4]. Therefore, it is essential to properly treat MG-containing wastewater prior to environmental discharge.

The remediation of industrial dye wastewater, exemplified by MG, remains a significant environmental concern.

The conventional methods for dye wastewater treatment, such as adsorption, filtration, chemical oxidation, and coagulation precipitation [5–7], can reduce the pollutant concentrations to a certain extent. However, these approaches are often hindered by inherent limitations, including low removal efficiency, high operational costs, and the generation of secondary pollution. These limitations make them inadequate for large-scale or practical applications. In contrast, photocatalytic degradation has been recognized as a viable solution because of its excellent efficiency and environmental compatibility for the removal of organic pollutants, particularly in water treatment [4,8,9]. Silva et al., 2021 synthesized $\text{Ag}_3\text{PO}_4/\text{SnO}_2$ composites via an in situ precipitation method and exhibited excellent photocatalytic activity toward Rhodamine B under visible light irradiation [10]. Among the various photocatalysts, titanium dioxide (TiO_2) has been one of the most extensively studied materials due to its relatively small band gap, high quantum yield, excellent chemical stability, low cost, and environmental friendliness [11]. Under UV irradiation, TiO_2 can be photoexcited to produce highly reactive oxidative species, including $\cdot\text{OH}$ and $\cdot\text{O}_2^-$, which are essential for the degradation of organic pollutants [12,13]. However, TiO_2 exhibits limited responsiveness to visible light, significantly constraining its photocatalytic efficiency under natural sunlight. Consequently, enhancing the visible light activity of TiO_2 has become a central focus in photocatalysis research. Bismuth oxide (Bi_2O_3), a novel photocatalyst with a narrower band gap (2.0–3.96 eV), demonstrates superior visible light absorption compared to TiO_2 [14,15]. Some studies have shown that the integration of Bi_2O_3 with TiO_2 can effectively reduce the overall band gap and boost light absorption. Furthermore, the heterojunction formed between Bi_2O_3 and TiO_2 can prolong the lifetime of photogenerated charge carriers, significantly improving their photocatalytic performance [16–18]. Silva successfully synthesized a $\text{Bi}_2\text{O}_3/\text{TiO}_2$ composite at scale and achieved the complete degradation of tetracycline antibiotics under visible light irradiation, highlighting the significant application potential of such composites [19]. Nevertheless, both TiO_2 and Bi_2O_3 are commonly used in powder form, which poses challenges such as poor dispersibility in aqueous media and difficulties in catalyst recovery and reuse. These practical issues hinder their direct application in continuous or large-scale water treatment processes.

To overcome these limitations, increasing attention has been directed toward the immobilization of photocatalysts on porous materials, such as activated carbon, zeolites, and silica gel. These materials typically possess large specific surface areas and well-developed porous structures, which can improve the dispersion and stability of photocatalysts [20–22]. Zhang et al., (2021) developed a $\text{Bi}_2\text{O}_3/\text{TiO}_2/\text{rGO}$ composite photocatalyst by employing reduced graphene oxide (rGO) as a two-dimensional support, achieving a degradation efficiency exceeding 90% for di-(2-ethylhexyl) phthalate [23]. Among the various porous supports, PAC is particularly notable for its extensive surface area and abundant surface functional groups. When combined with TiO_2 , PAC can not only extend the lifetime of photogenerated charge carriers and suppress electron–hole recombination, but also significantly enhance pollutant removal through the synergistic effects of adsorption and photocatalysis [24]. Additionally, anchoring nanoscale photocatalysts onto larger carriers like PAC enables more uniform dispersion in aqueous environments, simplifies post-treatment separation, and substantially improves catalyst recyclability and practical applicability in engineering large-scale wastewater treatment systems.

In this study, photocatalysts were immobilized onto PAC via a hydrothermal method, and the $\text{Bi}_2\text{O}_3\text{-TiO}_2/\text{PAC}$ composite photocatalysts were successfully synthesized. The composite was subsequently innovatively applied for the photocatalytic degradation of

MG in aqueous solution under visible light irradiation. The composites were systematically characterized in terms of their phase composition, microstructure, and optical properties. The photocatalytic degradation efficiency of MG was evaluated under various experimental conditions, including the catalyst dosage, the initial dye concentration, the solution pH, and the presence of coexisting ions. To further elucidate the photocatalytic mechanism, radical scavenging experiments were conducted to identify the dominant reactive oxygen species involved in the degradation process. Overall, this study aims to provide both theoretical understanding and practical implications for the efficient removal of persistent organic micropollutants from aqueous environments.

2. Materials and Methods

2.1. Chemicals

Tetrabutyl titanate ($\text{Ti}(\text{OBU})_4$, 98.0 wt%), bismuth nitrate pentahydrate ($\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$, 98.0 wt%), isopropanol (IPA), disodium ethylenediaminetetraacetate (EDTA), and benzoquinone (BQ) were purchased from Sigma-Aldrich (St. Louis, MO, USA). MG, glacial acetic acid (99.0 wt%), and absolute ethanol and PAC were obtained from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). All chemicals used in this study were of analytical reagent (AR) grade and used without further purification.

2.2. Synthesis of Bi_2O_3 - TiO_2 /PAC Composite Catalysts

The Bi_2O_3 - TiO_2 /PAC composites were synthesized via the hydrothermal method [4,20]. Solution A was prepared by slowly adding 10 mL of $\text{Ti}(\text{OBU})_4$ into 5 mL of glacial acetic acid, followed by mixing with 40 mL of absolute ethanol. The mixture was stirred at 350 rpm for 120 min under room temperature. Solution B was prepared by dissolving 1.391 g of $\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$ in 40 mL of deionized water, followed by ultrasonic treatment at 40 kHz for 30 min, and subsequent magnetic stirring at 350 rpm for 120 min. Solution B was then added dropwise to solution A under continuous stirring, followed by the gradual addition of 0.5 g of PAC. The combined solution was stirred for 5 h, and subsequently transferred to a Teflon-lined stainless steel autoclave for hydrothermal treatment at 150 °C for 12 h. The resulting product was dried at 130 °C, and subsequently calcined at 400 °C for 4 h to obtain the Bi_2O_3 - TiO_2 /PAC composite with a Bi/Ti molar ratio of 0.1 (denoted as 10%). Composites with Bi/Ti molar ratios of 0.06 (6%), 0.08 (8%), and 0.12 (12%) were synthesized using the same procedure.

2.3. Characterization

The morphology, structure, and chemical composition of the synthesized photocatalysts were characterized using the following techniques: X-ray diffraction (XRD, D8 Advance, Bruker, Billerica, MA, USA), X-ray photoelectron spectroscopy (XPS, ESCALAB 250Xi, Thermo Fisher Scientific, Waltham, MA, USA), scanning electron microscopy (SEM, S-4800II, Hitachi, Tokyo, Japan), Brunauer–Emmett–Teller surface area analysis (BET, ASAP 2460, Micromeritics, Norcross, GA, USA), and high-resolution transmission electron microscopy (HRTEM, Tecnai G2 F30, FEI, Hillsboro, OR, USA).

2.4. Photocatalytic Experiments

Photocatalytic activity was evaluated by the degradation of MG in aqueous solution. The experiments were carried out in a 50 mL quartz reactor containing 20 mL of MG solution at an initial concentration of 30 mg/L. A total of 5 mg of Bi_2O_3 - TiO_2 /PAC (10%) catalyst was added, and the suspension was stirred in the dark for 30 min to establish adsorption–desorption equilibrium. The photocatalytic reaction was subsequently initiated under irradiation from a 165 W xenon lamp. Samples were collected at predetermined time

spans, centrifuged at $6000 \times g$ rpm for 5 min, and then filtered through a $0.22 \mu\text{m}$ membrane to eliminate any suspended catalyst. The residual MG concentration in the supernatant was determined, and the degradation efficiency was calculated with the following equation:

$$\eta = \frac{C_0 - C_e}{C_0} \times 100\% \quad (1)$$

where C_0 is the initial concentration, and C_e is the equilibrium concentration after photocatalytic degradation. All experiments were conducted in triplicate, and the average values are reported as the ultimate results.

3. Results and Discussion

3.1. Characterization of $\text{Bi}_2\text{O}_3\text{-TiO}_2/\text{PAC}$ Composites

The structural properties of the $\text{Bi}_2\text{O}_3\text{-TiO}_2/\text{PAC}$ composite catalysts with varying Bi_2O_3 loadings were tested by XRD, and the corresponding patterns are presented in Figure 1. The synthesized TiO_2 exhibited distinct characteristic diffraction peaks at 2θ values of 25.3° , 38.2° , 48.1° , 55.1° , and 63.2° , which are characteristic of the anatase phase of TiO_2 [23]. Upon the incorporation of PAC and Bi_2O_3 , the noticeable broadening of diffraction peaks was observed, particularly at prominent planes such as (101) and (004). This peak broadening is likely attributable to structural modifications and the reduction in crystallinity induced by the addition of these components. Although Bi_2O_3 was introduced into the composite, no distinct diffraction peaks corresponding to crystalline Bi_2O_3 were detected. This observation is consistent with the previous studies [23] and can be attributed to low-level Bi_2O_3 loading and its high dispersion or amorphous state within the composite matrix. Moreover, the inherently amorphous nature of PAC contributes to broad, diffuse features or an elevated background signal in the XRD pattern, which may further obscure the weak diffraction signals of Bi_2O_3 , particularly at lower loadings [20]. Therefore, the presence and distribution of Bi_2O_3 in the composite should be further confirmed using complementary characterization techniques, such as XPS, SEM-EDS mapping, and HRTEM.

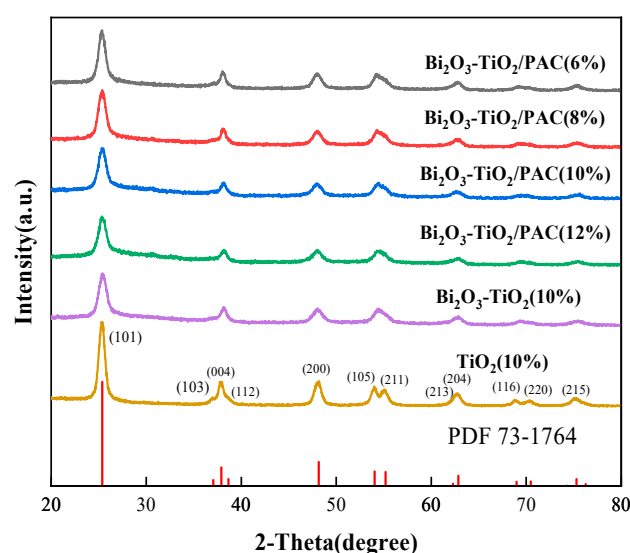


Figure 1. XRD spectra of synthesized $\text{Bi}_2\text{O}_3\text{-TiO}_2/\text{PAC}$.

The specific surface area and pore structure of the composites with different Bi/Ti molar ratios were examined by N_2 adsorption–desorption isotherms, and the result is shown in Figure 2. According to the IUPAC classification, all the samples displayed type IV isotherms with distinct hysteresis loops (Figure 2a), characteristic of mesoporous structures. These mesopores are beneficial for promoting light scattering and enhancing photocatalytic

efficiency. The pore size distribution curves (Figure 2b) reveal that the composites possess a broad range of mesopores, and the well-developed porous architecture is favorable for the adsorption of MG molecules, thereby facilitating their subsequent photocatalytic degradation. As shown in Figure 2c, the composite with a Bi/Ti molar ratio of 10% exhibited the largest specific surface area and the smallest average pore size among all the samples (6%, 8%, 10%, and 12%), indicating its superior adsorption capacity. Furthermore, the UV-Vis DRS results (Figure 2d) demonstrated that the 10% Bi/Ti composite exhibited a more significant red shift in the absorption edge compared to that of pure TiO_2 , indicating an enhanced light-harvesting ability in the visible region. The estimated band gap energy of this composite was approximately 2.89 eV, which is lower than that of pristine anatase TiO_2 (typically 3.0–3.2 eV). This band gap narrowing confirms that the incorporation of Bi_2O_3 effectively extends the light response range of TiO_2 and improves its visible light photocatalytic activity. Based on its favorable surface area, pore structure, and optical properties, the Bi_2O_3 - TiO_2 /PAC composite with a 10% Bi/Ti molar ratio was selected for subsequent photocatalytic performance evaluations.

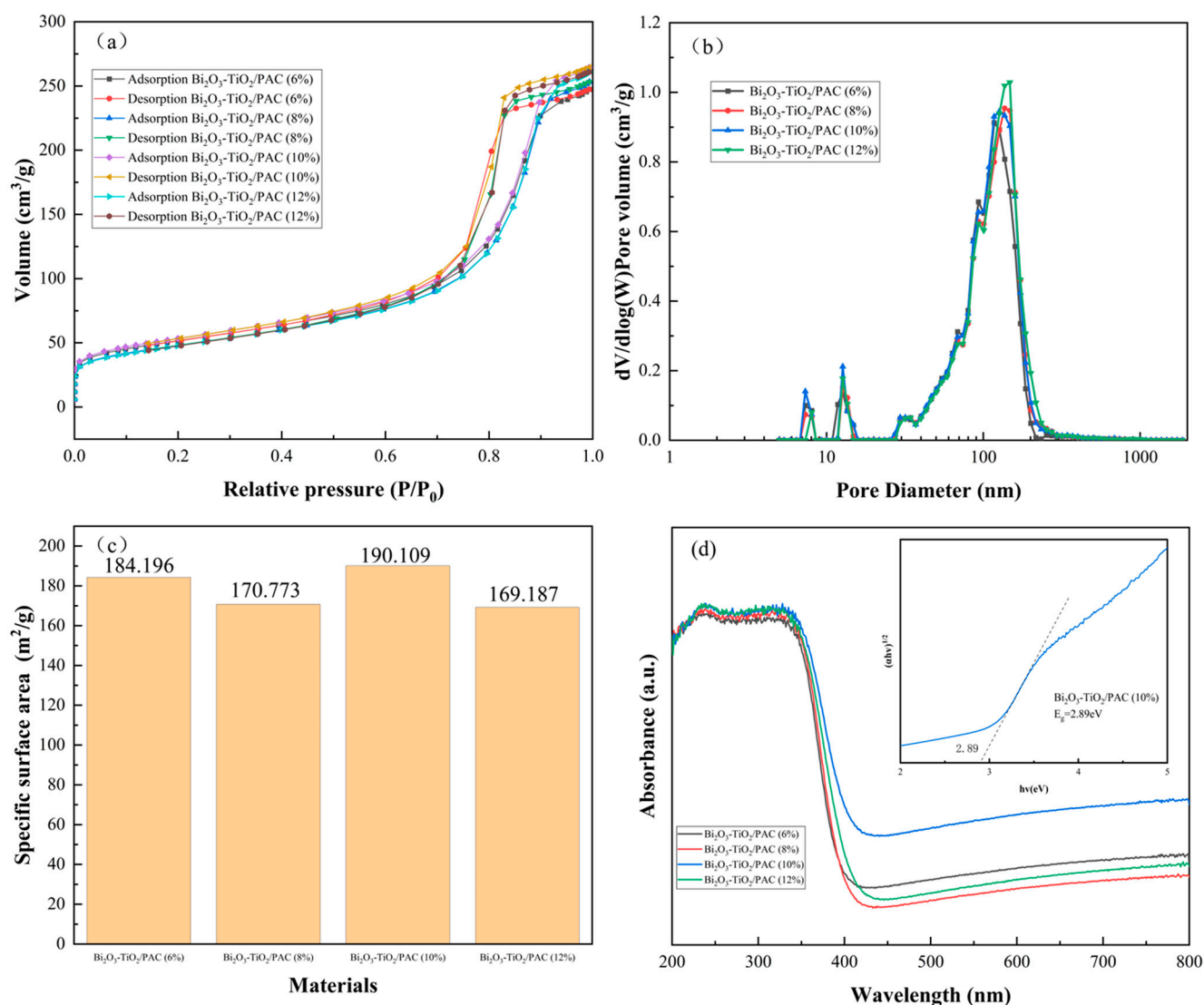


Figure 2. (a) N_2 - adsorption and desorption. (b) Pore diameter distribution. (c) Specific surface area. (d) UV-Vis-DRS.

The surface morphology of the composite was characterized using SEM and TEM, as shown in Figure 3. The composite catalyst was primarily composed of PAC, which

exhibited a layered morphology with distinct interlayer voids. Spherical or irregularly shaped TiO_2 and Bi_2O_3 nanoparticles with varying sizes were observed on the surface and within the interlayer regions of PAC. These particles were relatively uniformly distributed, with only minor aggregation present in some localized areas. SEM-EDS analysis (Figure 3b) confirmed the presence of Ti, Bi, C, and O elements in the composite, with minimal impurity signals. Furthermore, the elemental mapping images obtained from SEM (Figure 3c) clearly demonstrate the uniform distribution of TiO_2 and Bi_2O_3 throughout the PAC matrix, which is consistent with the SEM observations. The TEM image (Figure 3d) revealed that the PAC support exhibited an amorphous structure, with crystalline particles evenly anchored on its surface and embedded within the interlayer gaps. In the HRTEM image (Figure 3e), distinct lattice fringes were observed. The measured interplanar spacing of 0.351 nm corresponded to the (101) plane of anatase TiO_2 , while the spacing of 0.326 nm matched the (111) plane of Bi_2O_3 . These results further verify the successful formation and structural integrity of the Bi_2O_3 - TiO_2 /PAC composite.

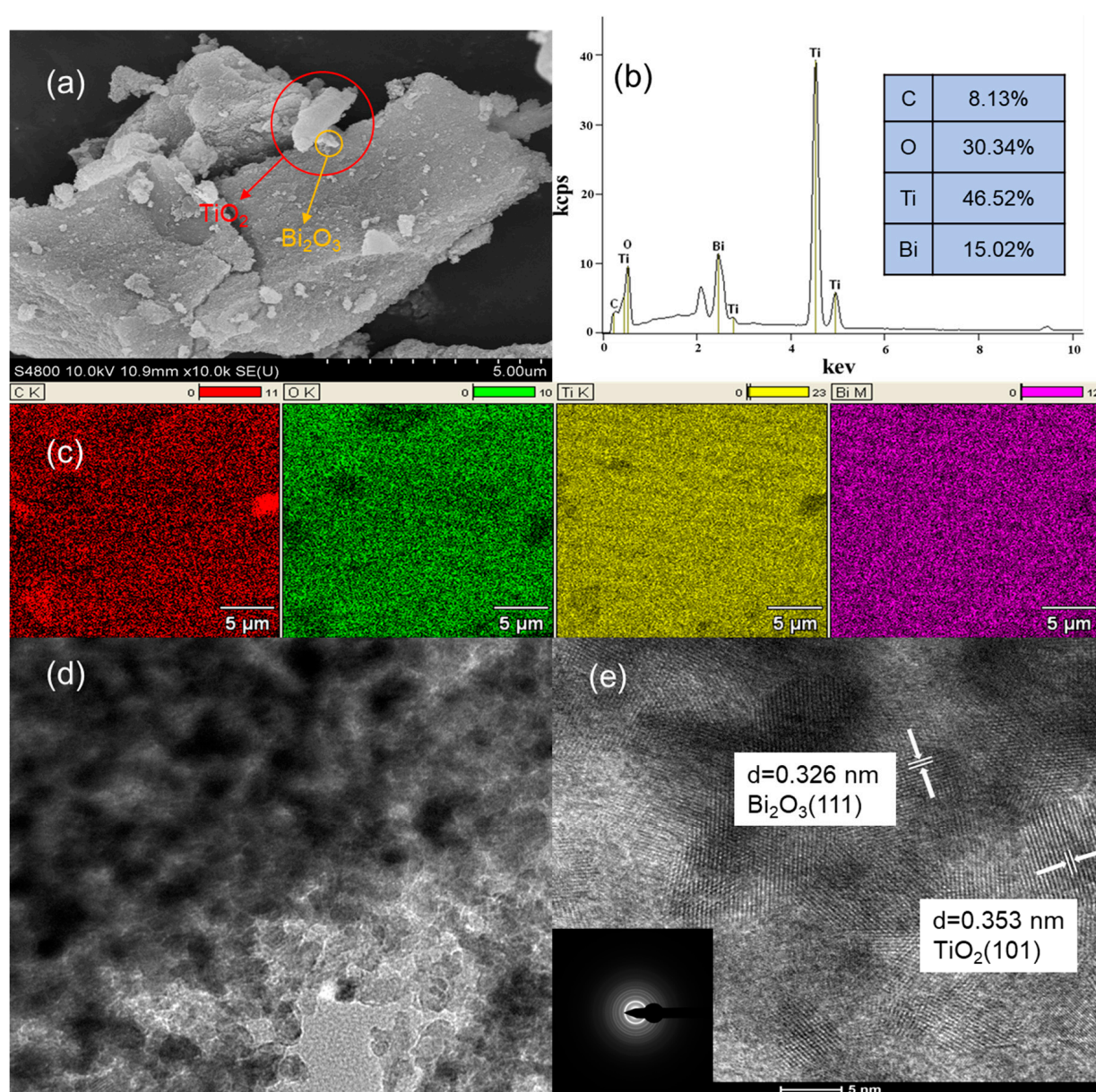


Figure 3. Surface morphology and composition analysis of Bi_2O_3 - TiO_2 /PAC(10%): (a) SEM image. (b) SEM-EDS spectra. (c) SEM-element mappings correspond to C, O, Ti and Bi. (d) TEM image. (e) HRTEM micrograph.

To explore the elemental composition and chemical states of the composite material, X-ray photoelectron spectroscopy (XPS) analysis was conducted, and the results are presented in Figure 4. The main elements detected in the composite were C, O, Ti, and Bi. As shown in Figure 4a, the two peaks at 164.8 eV and 159.8 eV correspond to Bi 4f_{5/2} and Bi 4f_{7/2}, respectively, indicating that bismuth is present in the +3 oxidation state as Bi₂O₃ [25]. Compared to the standard binding energies of Bi₂O₃ (163.9 eV and 158.8 eV), a slight positive shift was observed, suggesting electronic interactions between Bi₂O₃ and the other components in the composite during synthesis. Figure 4b shows two distinct Ti peaks at 464.8 eV and 458.9 eV, corresponding to Ti 2p_{1/2} and Ti 2p_{3/2}, respectively, indicating that titanium exists in the form of TiO₂ [26]. The C 1s spectrum (Figure 4c) can be deconvoluted into three peaks located at 288.4 eV (O–C=O), 285.5 eV (C–O–C), and 284.6 eV (C–C), confirming the presence of various carbon-containing functional groups [27]. The O 1s spectrum (Figure 4d) exhibits a peak at 530.2 eV, which is attributed to Ti–O bonds, while the peak at 531.5 eV is likely related to C=O, COOH, or O–H species [28,29].

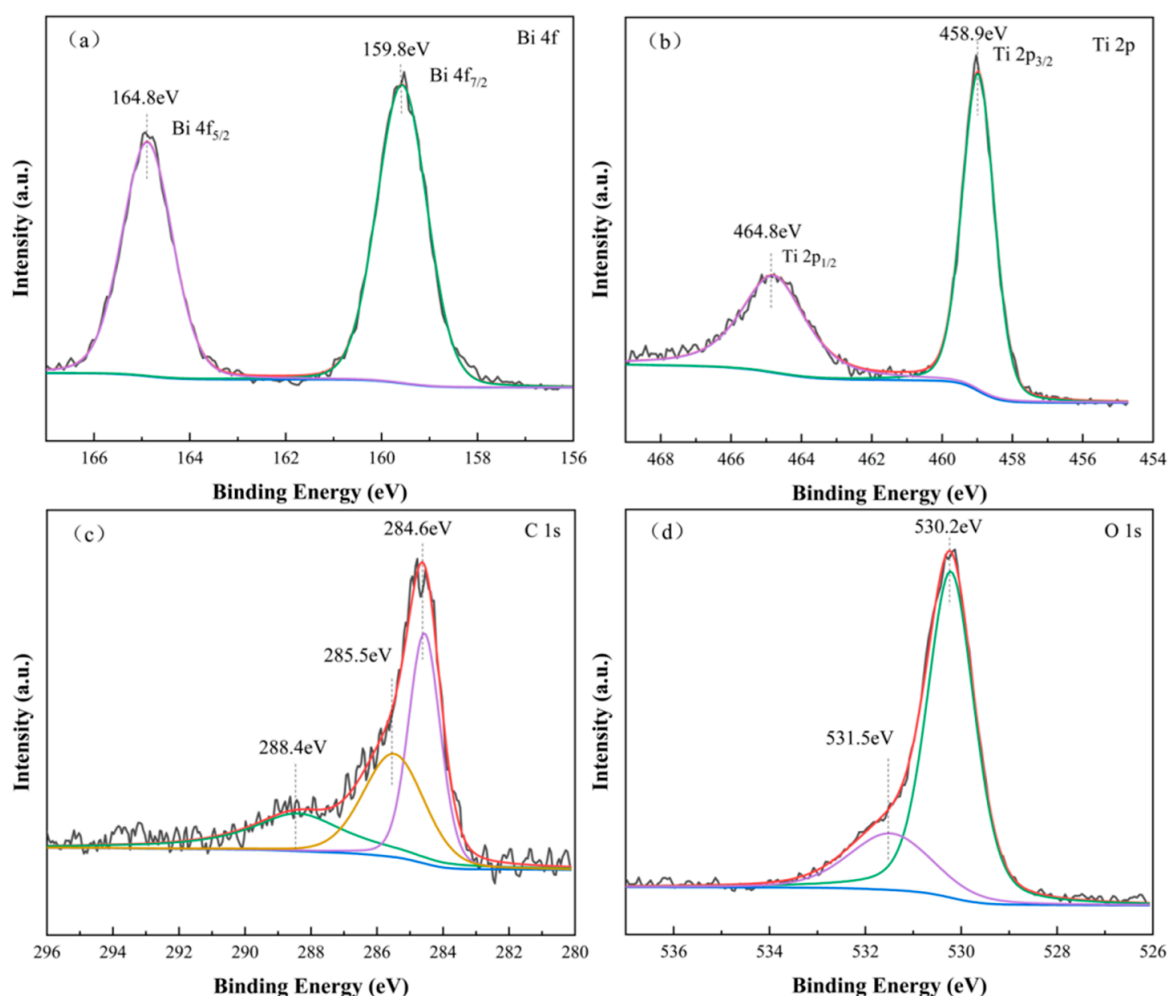


Figure 4. XPS spectra of Bi₂O₃-TiO₂/PAC (10%): (a) Bi 4f. (b) Ti 2p. (c) C 1s. (d) O 1s.

3.2. Photocatalytic Degradation Performances

3.2.1. Effect of Bi₂O₃-TiO₂/PAC Dosage

The effect of catalyst dosage on degradation efficiency is illustrated in Figure 5. As the dosage increased from 0.25 g/L to 0.5 g/L, the removal efficiency of MG rose from 97.5% to 99.5%. When the dosage was further increased to 0.75 g/L, MG was almost completely removed during the dark adsorption stage. When the catalyst dosage was insufficient, the number of available active sites on the catalyst surface was limited, and

incident photons could not be fully utilized, resulting in the reduced generation of electron–hole pairs [30], which in turn weakened the catalyst’s degradation capacity. As the dosage increased, more active sites became available, allowing for photons from the light source to fully interact with the catalyst surface, thereby enhancing the production of reactive charge carriers. At the same time, the increased amount of catalyst facilitated greater adsorption of MG molecules, leading to the more complete photocatalytic degradation process and a significant improvement in MG removal efficiency. Considering both practical effectiveness and economic feasibility, a dosage of 0.25 g/L was selected for use in the subsequent experiments.

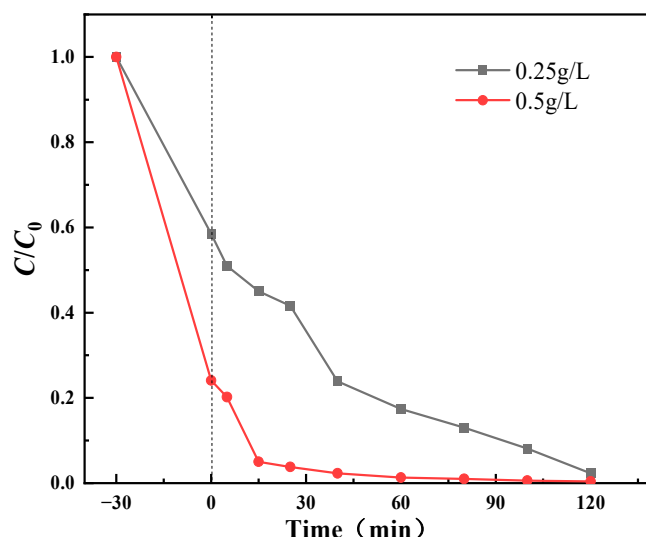


Figure 5. Effect of catalyst dosage on degradation rate (experimental conditions: [MG] = 20 mg/L; pH = 7).

In comparison with other photocatalysts used for the removal of contaminants from aqueous solutions, Bi₂O₃-TiO₂/PAC exhibits an excellent performance (Table 1), which can be attributed to its large specific surface area and abundant porosity. These structural features facilitate adsorption, mass transfer, and electron transfer, thereby promoting the generation of reactive species and enhancing the degradation of target pollutants.

Table 1. Comparison of degradation efficiencies between Bi₂O₃-TiO₂/PAC and other photocatalysts for pollutants removal from aqueous solutions.

Materials	Structure	Surface Area (Before–After/m ² /g)	Bandgap Energy (Before–After/eV)	Pollutant	Degradation Efficiency
Bi ₂ O ₃ -TiO ₂ /PAC	Mesopore	84.56–190.1	3.2–2.89	MG	99%
Cu-TiO ₂	Micropores	43–46	3.08–2.78	OC	75% [31]
Mn-TiO ₂	Mesopore	3.20–2.21	3.20–2.21	TPOME	60.2% [32]
ZnO-TiO ₂	Mesopore	50.05–107.98	3.26–2.76	TC	92% [32]
TiO ₂ -SiO ₂	Mesopore	217–256	3.22–3.22	TC	96% [33]
BiVO ₄ -TiO ₂	Mesopore	60.6–95.3	3.2–3.03	FA	97% [34]
TiO ₂ -WO ₃	Mesopore	95–117	3.0–2.6	MB	90% [35]

3.2.2. Effect of Initial MG Concentration

The effect of the initial MG concentration on photocatalytic degradation activity is shown in Figure 6. As the concentration increased from 10 mg/L to 35 mg/L, the final removal efficiency remained relatively high, ranging from 98% to 99%; however, the required reaction time increased significantly. At 10 mg/L, MG was almost completely

degraded during the early stages of the photocatalytic reaction. The degradation of MG at 20 mg/L and 30 mg/L showed minimal variation, with complete degradation achieved within 120 min. When the initial MG concentration reached 35 mg/L, the degradation efficiency after 120 min was approximately 90%, and the reaction rate plateaued with further irradiation. This suggests that at higher concentrations, the photocatalytic capacity of the catalyst approached saturation. At this stage, a large number of MG molecules may have occupied the available active sites on the catalyst surface, thereby hindering effective interaction between the photons and the catalyst. This limitation reduces the generation and separation efficiency of photogenerated electron–hole pairs, ultimately lowering the degradation rate [30]. Although satisfactory degradation was still observed at higher concentrations, the reaction time was significantly prolonged, and the degradation kinetics slowed. Therefore, in practical applications, the catalyst dosage and reaction duration should be optimized based on pollutant concentration to enhance photocatalytic efficiency and achieve an optimal removal performance.

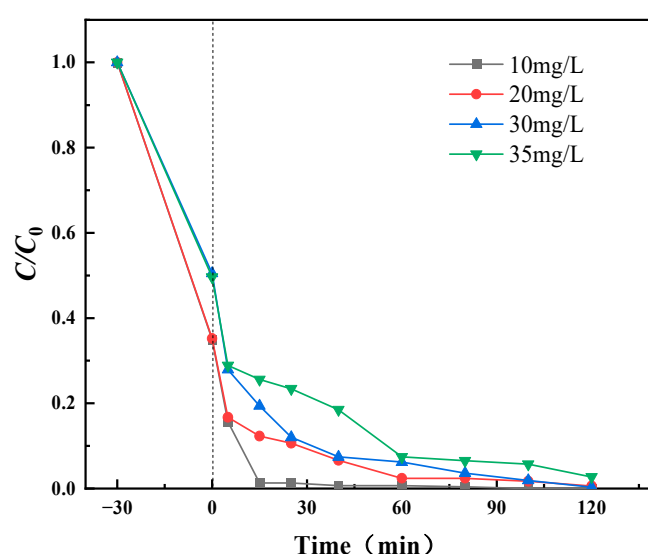


Figure 6. Effect of initial MG concentration on MG degradation (experimental conditions: [catalyst] = 0.25 g/L; pH = 7).

3.2.3. Effect of Solution pH

To investigate the impact of solution pH on the photocatalytic degradation efficiency, the pH was adjusted over a range from 3.39 to 9.13 using 0.1 mmol/L HCl and NaOH, while maintaining a catalyst dosage of 0.25 g/L and an initial MG concentration of 30 mg/L. The experimental results presented in Figure 7 show that under acidic conditions (pH 3–4), the degradation efficiency was relatively low (88%). As the pH increased, the degradation rate gradually improved, achieving a maximum of 98.91% at pH 9.13.

This phenomenon indicates that an acidic environment suppresses the photocatalytic degradation efficiency of MG. The pH of the solution not only influences the generation of reactive species—thereby directly affecting the efficiency of photocatalysis—but also alters the surface charge distribution of both the catalyst and the MG molecules, which in turn impacts their adsorption behavior on the catalyst surface [13]. According to the relevant literature, photoinduced electrons (e^-) can react with dissolved oxygen to form superoxide radicals ($\cdot O_2^-$) (Equation (2)) [23]. The generated $\cdot O_2^-$ may further react with protons (H^+) and electrons to produce hydroxyl radicals ($\cdot OH$) and hydroxide ions (OH^-) (Equation (3)) [36,37]. Under acidic conditions, a high number of H^+ ions tend to scavenge $\cdot O_2^-$ radicals, thereby decreasing their availability in the system. As demonstrated in the radical scavenging experiments discussed later, $\cdot O_2^-$ is primarily responsible for the pho-

photocatalytic degradation of MG by the $\text{Bi}_2\text{O}_3\text{-TiO}_2/\text{PAC}$ composites. Thus, acidic conditions hinder MG degradation primarily by depleting the key reactive species $\cdot\text{O}_2^-$. Although acidic environments may also promote $\cdot\text{OH}$ generation via Equation (3), the contribution of $\cdot\text{OH}$ to MG degradation is significantly smaller than that of $\cdot\text{O}_2^-$, a conclusion supported by the results of the scavenging experiments. Conversely, in highly alkaline environments, excessive OH^- ions may consume photoinduced holes (h^+) (Equation (4)), thereby inhibiting the formation of $\cdot\text{OH}$ and suppressing the overall photocatalytic reaction [38]. The catalyst exhibited robust photocatalytic activity across a wide pH range, with only a slight deterioration in performance under acidic conditions. Notably, the MG removal efficiency remained above 88% under all the tested pH values, demonstrating the excellent stability and adaptability of the catalyst.

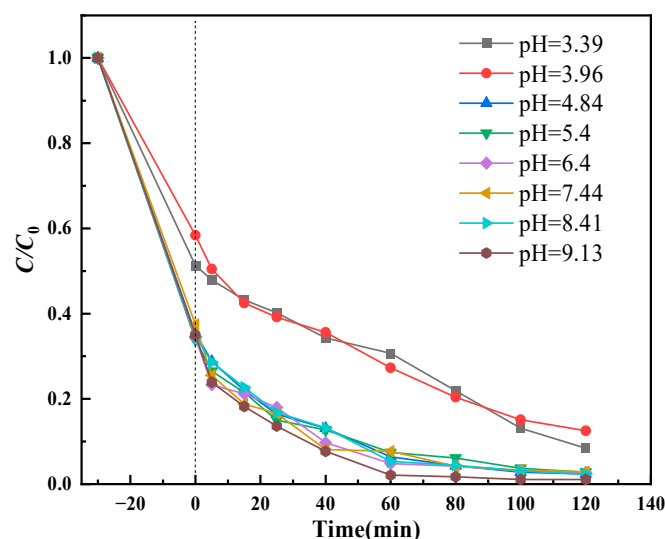
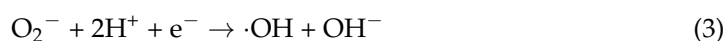


Figure 7. Effect of solution pH on MG degradation (experimental conditions: [catalyst] = 0.25 g/L, [MG] = 30 mg/L; pH = 7).

3.2.4. Effect of Coexisting Anions

Various inorganic anions are commonly present in natural water bodies, and their presence can influence the photocatalyst's efficiency in removing target pollutants through multiple mechanisms. On the one hand, these anions may compete with the target pollutants for adsorption on the active sites of the catalyst surface or consume reactive species generated during the photocatalytic process, thereby decreasing catalytic efficiency. On the other hand, certain anions can be transformed into reactive radicals with some oxidative potential or induce local electrostatic fields near the catalyst surface, facilitating the separation of photogenerated electron-hole pairs and enhancing the degradation reaction [39]. Therefore, the presence of common anions in environmental waters can exert both inhibitory and promoting effects on photocatalytic degradation.

This study systematically investigated the effects of several common anions found in natural water bodies (Cl^- , HCO_3^- , NO_3^- , and SO_4^{2-}) on the photocatalytic degradation efficiency of MG. As shown in Figure 8, the MG degradation rate in the control system (without background anions) reached 98.6%. After the introduction of Cl^- , HCO_3^- , NO_3^- , and SO_4^{2-} , the degradation rates decreased to 91.1%, 98.4%, 94.7%, and 86.7%, respectively. Except for HCO_3^- , the other three anions inhibited the photocatalytic degradation to

varying extents. Notably, at the initial stage of the reaction, the system containing HCO_3^- demonstrated a faster degradation rate than that of the control, which may be attributed to the pH increase induced by HCO_3^- addition. This higher pH likely facilitated the generation of superoxide radicals ($\cdot\text{O}_2^-$), thereby enhancing the oxidative capacity of the photocatalytic process. Although the previous studies have reported that Cl^- , HCO_3^- , NO_3^- , and SO_4^{2-} can react with photogenerated holes (h^+) or hydroxyl radicals ($\cdot\text{OH}$) to produce less oxidative species ($\cdot\text{Cl}$, $\cdot\text{CO}_3^-$, $\cdot\text{NO}_3$, and $\cdot\text{SO}_4^-$) (Equations (5)–(8)) [20], which could theoretically suppress pollutant degradation, the experimental findings here suggest that HCO_3^- can actually enhance the degradation process under specific conditions. This effect may be closely related to its pH-regulating role and its influence on the generation pathways of reactive oxygen species. Furthermore, HCO_3^- may also react with $\cdot\text{OH}$ to form $\cdot\text{CO}_3^-$, a radical with comparatively less oxidative potential (Equation (9)) [28]. In summary, although the presence of background anions introduces some interference in the photocatalytic degradation of MG, the extent of inhibition is relatively minor. These results demonstrate that the $\text{Bi}_2\text{O}_3\text{-TiO}_2/\text{PAC}$ composite catalyst exhibits strong environmental adaptability and holds considerable promise for practical water treatment applications.

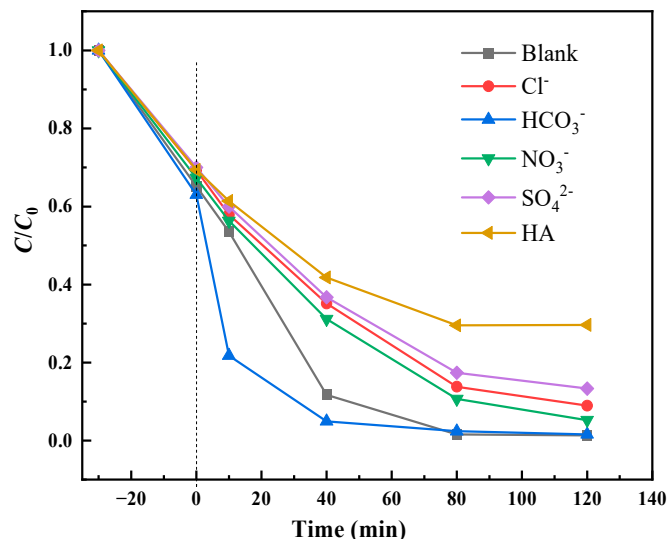
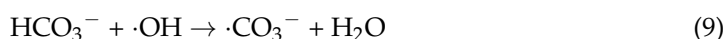


Figure 8. Effect of coexisting anions on MG degradation (experimental conditions: [catalyst] = 0.25 g/L; [MG] = 30 mg/L; [anions] = 10 mM and pH = 7).

In addition to inorganic anions, natural organic matter (NOM) is also widely present in natural water bodies, with humic acid (HA) being one of its major components. The previous studies have shown that the presence of NOM can significantly influence photocatalytic degradation processes [22]. In this study, HA was selected as a representative NOM to investigate its impact on the photocatalytic degradation performance of MG. The experimental results revealed that the final degradation rate of MG dropped significantly to 70.4% after the addition of HA compared to 98.6% in the blank system without background substances, indicating a pronounced inhibitory effect. This suppression can primarily be at-

tributed to the competitive consumption of reactive species between HA and MG molecules during the photocatalytic process, which reduces the availability of these species for MG degradation. Moreover, HA itself may undergo transformation during the photocatalytic reaction, producing complex intermediate products that can also scavenge reactive radicals such as $\cdot\text{OH}$ and $\cdot\text{O}_2^-$, thereby further diminishing the overall photocatalytic efficiency [22]. These findings highlight that the presence of natural organic matter can exert a significant negative impact on photocatalytic degradation. In real water treatment applications, NOM may substantially impact the catalyst performance, and thus, its interference should be carefully considered and appropriately addressed in the design and implementation of photocatalytic treatment systems.

3.2.5. Stability and Reusability of Prepared Catalysts

The stability of a photocatalyst is a key parameter in evaluating its feasibility for practical engineering applications, and it is commonly assessed by its recyclability. To investigate the reusability of the $\text{Bi}_2\text{O}_3\text{-TiO}_2/\text{PAC}$ (10%) composite, five consecutive photocatalytic degradation experiments were conducted under identical conditions. After each cycle, the catalyst was recovered by centrifugation, thoroughly washed multiple times with deionized water and ethanol, and subsequently dried at 30°C for reuse. As illustrated in Figure 9, the MG degradation efficiency of the composite catalyst showed only a slight decrease from 98.8% in the first cycle to 90.0% after the fifth cycle, indicating excellent stability and reusability. Although a minor decline in photocatalytic activity was observed, the overall efficiency remained at a high level, suggesting that the catalyst possessed strong resistance to deactivation during repeated use. This stability may be attributed to robust interfacial interactions between TiO_2 and PAC, which help maintain the structural integrity and catalytic performance of the composite over multiple cycles. Overall, these results demonstrate the $\text{Bi}_2\text{O}_3\text{-TiO}_2/\text{PAC}$ system's strong potential for long-term and sustainable application in real-world wastewater treatment.

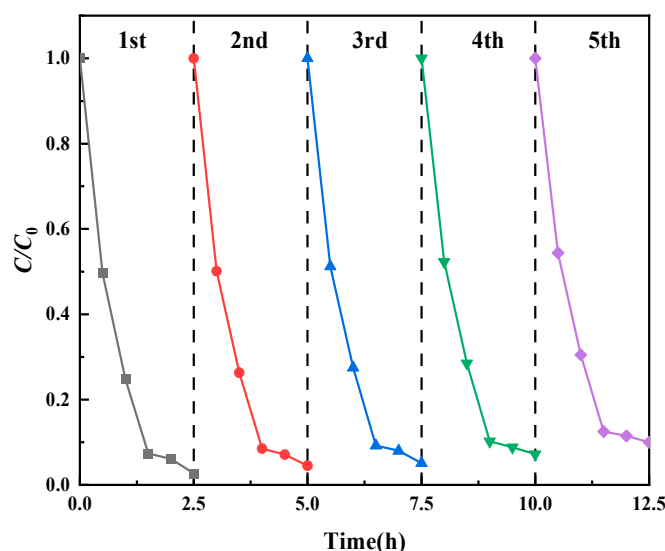


Figure 9. Stability test of MG degradation (experimental conditions: [catalyst] = 0.25 g/L; [MG] = 30 mg/L and pH = 7).

3.3. Insight into the Photocatalytic Mechanism

During the photocatalytic process, the primary reactive species involved include $\text{OH}^\cdot$, h^+ , and $\cdot\text{O}_2^-$. To further elucidate the dominant reaction mechanism of the $\text{Bi}_2\text{O}_3\text{-TiO}_2/\text{PAC}$ composite in the photocatalytic degradation of MG, a series of reactive-species-trapping experiments was conducted. IPA, EDTA, and BQ were used as specific scavengers for $\cdot\text{OH}$,

h^+ , and $\cdot O_2^-$, respectively [20,39,40], to investigate the contribution of each reactive species to the photocatalytic process. As shown in Figure 10, in the control system without any scavengers, the MG removal efficiency reached 99%. Upon the addition of IPA, EDTA, and BQ, the degradation efficiency decreased to 89%, 77%, and 63%, respectively, indicating that all these three reactive species played important roles in the photocatalytic degradation process. Among them, the most significant inhibition was observed with the addition of BQ, suggesting that $\cdot O_2^-$ was the dominant species in the degradation of MG. The impact of EDTA also revealed a substantial role of h^+ , while $\cdot OH$ had a comparatively weaker effect. In summary, the Bi_2O_3 - TiO_2 /PAC composite photocatalytic system achieves efficient degradation of MG primarily through an oxidative mechanism dominated by $\cdot O_2^-$, with h^+ and $\cdot OH$ acting synergistically.

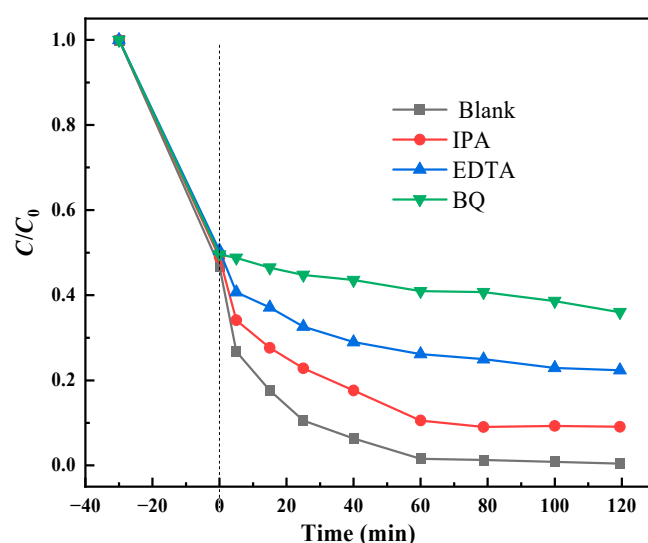


Figure 10. Effect of different scavengers on MG degradation (experimental conditions: [catalyst] = 0.25 g/L; [MG] = 30 mg/L and pH = 7; [IPA] = 10 mM; [EDTA] = 2 mM; [BQ] = 2 mM).

Figure 11 illustrates the proposed photocatalytic mechanism for MG degradation over the Bi_2O_3 - TiO_2 /PAC composite. In the schematic, the gray region represents the layered structure of PAC, which, owing to its large specific surface area and well-developed porosity, provides an ideal platform for the uniform dispersion and stabilization of TiO_2 and Bi_2O_3 nanoparticles, forming a well-integrated ternary composite. At the onset of the reaction, PAC efficiently adsorbs MG molecules, enriching the pollutant concentration at the catalyst surface, and thereby facilitating subsequent photocatalytic degradation. Its porous architecture also offers abundant active sites to support the reaction. Upon exposure to sufficient light energy, the electrons in the valence band (VB) of TiO_2 are excited to the conduction band (CB), generating electron–hole pairs (e^-/h^+) [41–43]. Due to the favorable band alignment between TiO_2 and Bi_2O_3 , photogenerated electrons rapidly transfer from TiO_2 to Bi_2O_3 , effectively suppressing e^-/h^+ recombination, extending the lifetime of reactive species [44] and enhancing the overall photocatalytic efficiency. The migrated electrons in the CB react with dissolved oxygen (O_2) to form superoxide radicals ($\cdot O_2^-$), while the holes in the VB oxidize water molecules or surface hydroxyl groups to produce hydroxyl radicals ($\cdot OH$). Additionally, some holes (h^+) can directly oxidize MG molecules [41,42]. These reactive species— $\cdot O_2^-$, $\cdot OH$, and h^+ —work synergistically to break down MG. Their combined oxidative activity is particularly effective for degrading dye pollutants that are highly sensitive to radical-mediated reactions.

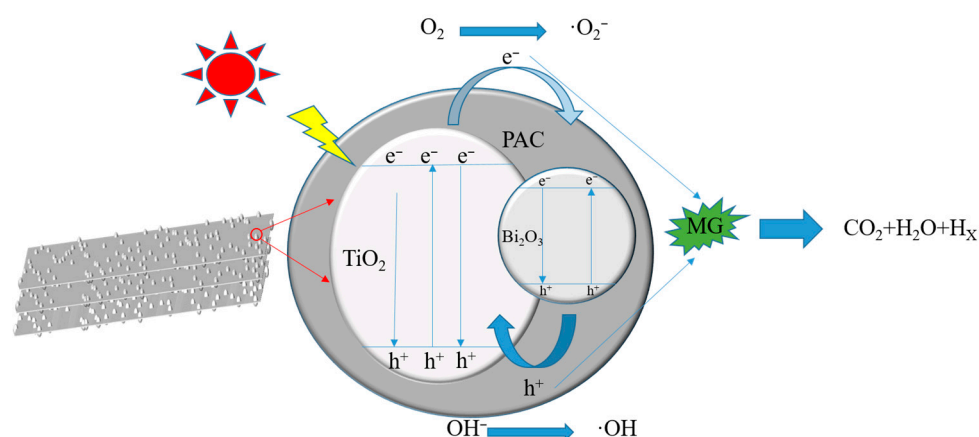


Figure 11. Proposed photocatalytic mechanism of MG/Bi₂O₃-TiO₂/PAC system.

4. Conclusions

In this study, Bi₂O₃-TiO₂/PAC was successfully synthesized and applied for the treatment of MG solution. Ti and Bi were uniformly distributed on the surface and within the interlayer structure of PAC as TiO₂ and Bi₂O₃, respectively. The incorporation of Bi₂O₃ enhanced light absorption in the UV-visible region, significantly improving the photocatalytic degradation efficiency of MG. The use of PAC as a support material facilitated the dispersion of metal oxides and enhanced the structural stability of the catalyst. Active-species-trapping experiments revealed that $\cdot\text{O}_2^-$ played a dominant role in the photocatalytic process, with h^+ and $\cdot\text{OH}$ also contributing. The catalyst demonstrated excellent environmental adaptability, maintaining high activity in the presence of common inorganic anions and across a wide pH range. The cyclic stability tests showed that the catalyst retained over 90% MG removal efficiency after five consecutive cycles, indicating its excellent stability, reusability, and strong potential for practical applications in wastewater treatment.

Author Contributions: Conceptualization, Y.C.; methodology, Y.C. and W.Z.; software, M.C. and J.L.; validation, Y.C.; formal analysis, M.C. and J.L.; investigation, Y.C.; resources, Y.C.; data curation, W.Z.; writing—original draft preparation, Y.C.; writing—review and editing, W.Z.; visualization, Y.C.; supervision, W.Z. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Data Availability Statement: The original contributions presented in this study are included in the article. Further inquiries can be directed to the corresponding author.

Conflicts of Interest: The authors declare no conflicts of interest. The funders had no role in the design of this study; in the collection, analyses, or interpretation of data; in the writing of the manuscript; or in the decision to publish the results.

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