

RESEARCH ARTICLE

Synthesizing a Ternary $\text{TiO}_2/\text{g-C}_3\text{N}_4/\text{UiO-66}$ Photocatalyst From Waste Polyethylene Terephthalate Plastic to Treat Polystyrene Microplastics in Polluted Water

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ABSTRACT

A ternary $\text{TiO}_2/\text{g-C}_3\text{N}_4/\text{UiO-66}$ photocatalyst was synthesized using terephthalic acid recovered from waste polyethylene terephthalate bottles, providing a green strategy for both plastic upcycling and microplastics (MPs) degradation (TiO_2 = titanium dioxide; $\text{g-C}_3\text{N}_4$ = graphitic carbon nitride). Comprehensive characterizations confirmed the successful integration of TiO_2 , $\text{g-C}_3\text{N}_4$, and UiO-66 into a stable heterojunction with enhanced visible-light response and efficient charge transfer. Under visible-light irradiation, the composite achieved up to 90% degradation of polystyrene (PS) MPs within 30 h. Electrochemical impedance spectroscopy revealed its lowest charge-transfer resistance, correlating with superior separation of photogenerated carriers and strong reactive oxygen species (ROS) ($\cdot\text{O}_2^-$, $\cdot\text{OH}$) generation. These ROS actively fragmented PS chains into smaller oxidized intermediates, confirming the material's excellent photocatalytic potential for MPs removal and environmental purification.

1 | Introduction

The global escalation in plastic production and consumption has resulted in severe environmental contamination due to the persistence and poor degradability of polymeric materials [1, 2]. Among various plastics, polystyrene (PS) is particularly concerning because of its high stability, low recyclability, and potential to release toxic compounds upon degradation or incineration [3]. Once fragmented, PS forms microplastics (MPs)—particles smaller than 5 mm—that are now detected in soil, aquatic, and even atmospheric environments [4]. These MPs can be ingested by organisms, transferred through the food web, and potentially affected human health through oxidative stress, immune disruption, and developmental toxicity [4].

Conventional plastic waste management approaches, such as incineration and landfilling, are neither sustainable nor effective in mitigating long-term contamination [5]. Therefore, developing eco-friendly degradation technologies is essential. Among

various methods, photocatalytic degradation has emerged as a promising and green approach for converting persistent organic pollutants into harmless products like CO_2 and H_2O under mild conditions [6].

Titanium dioxide (TiO_2) is one of the most studied photocatalysts owing to its strong oxidation ability, low toxicity, and stability [7]. However, its wide bandgap (≈ 3.2 eV) restricts activity mainly to the UV region, which constitutes less than 5% of sunlight [8]. To enhance visible-light responsiveness, graphitic carbon nitride ($\text{g-C}_3\text{N}_4$) has been introduced as a complementary semiconductor due to its moderate bandgap (≈ 2.7 eV), good stability, and electronic tunability [9, 10]. The formation of $\text{TiO}_2/\text{g-C}_3\text{N}_4$ Z-scheme heterojunctions facilitates efficient charge transfer and reactive oxygen species (ROS) generation, thereby improving photocatalytic efficiency under visible light [11].

Recently, metal-organic frameworks (MOFs) such as UiO-66, a zirconium-based structure with terephthalic acid (TPA) linkers,

have attracted significant attention for environmental remediation due to their high porosity, tunable structure, and chemical stability [5]. Notably, UiO-66 can be synthesized from waste polyethylene terephthalate (PET), aligning with the concept of circular economy and sustainable material recovery [6]. Nevertheless, pristine MOFs exhibit limited photocatalytic activity because of poor charge transfer, motivating the design of MOF-based composites with semiconductors to achieve synergistic effects. Previous studies have demonstrated effective PS degradation using composites such as FeB/TiO₂ and TiO₂/CuPc under light irradiation [5, 6]. However, achieving efficient degradation of nanosized PS particles under visible light remains challenging.

In this study, a TiO₂@g-C₃N₄/UiO-66 photocatalytic composite was synthesized using recycled PET as a precursor. The composite was evaluated for its ability to degrade PS-MPs in aqueous systems under visible-light irradiation. The integration of TiO₂, g-C₃N₄, and UiO-66 is expected to enhance charge separation, broaden light absorption, and improve the photocatalytic degradation efficiency of PS MPs, offering a sustainable strategy for mitigating plastic pollution and promoting circular material utilization.

2 | Results and Discussion

2.1 | Characteristics of Materials

The X-ray diffraction (XRD) patterns of individual components (g-C₃N₄, TiO₂, and UiO-66) and their composites are shown in Figure 1. The pristine g-C₃N₄ exhibits two characteristic diffraction peaks at $\approx 13.1^\circ$ and 27.5° , corresponding to the (100) and (002) planes, respectively, which are attributed to the in-plane structural packing and interlayer stacking of aromatic systems [12]. The TiO₂ sample showed prominent diffraction peaks at 25.3° , 37.8° , 48.0° , 53.9° , 55.1° , and 62.7° , indexed to the (101), (004), (200), (105), (211), and (204) planes of the anatase phase (JCPDS No. 21-1272) [13]. UiO-66 exhibited multiple low-angle peaks at 7.4° , 8.5° , and 25.8° , corresponding to the (111), (200), and (442) planes, confirming its highly crystalline framework typical of Zr-based MOFs [14].

In the composite materials, the hybrid samples TiO₂@g-C₃N₄/UiO-66 retained the characteristic peaks of all three components without the appearance of new peaks, indicating successful integration without phase transformation. This suggests good phase compatibility and structural stability during the synthesis. Preserving crystallinity is essential for the photocatalytic degradation of MPs in water, as it ensures efficient charge transfer and high surface area for adsorption and activation. g-C₃N₄ provides visible-light absorption, TiO₂ facilitates charge separation and ROS generation, and UiO-66 enhances adsorption and surface interaction with microplastic molecules [15, 16].

Fourier transform infrared (FT-IR) spectroscopy was utilized to investigate the functional groups present and to elucidate the structural evolution of the individual components and composite materials. As shown in Figure 2a, the FT-IR spectrum of pristine UiO-66 displays a distinct band at 1654 cm^{-1} , attributed to the C=O stretching vibration of the carboxylate group (-COO-) coordinated to Zr clusters. The band at 1582 cm^{-1} is assigned to the C=C stretching vibration of the conjugated benzene ring within the TPA linker (BDC), confirming the successful coordination of the organic ligand to the Zr⁴⁺ metal nodes in the framework [17]. The peak at 1387 cm^{-1} is assigned to O-C-O stretching, confirming the integrity of the organic linker, while the bands in the lower wavenumber region (739 and 653 cm^{-1}) are associated with Zr-O-Zr lattice vibrations within the metal cluster framework. The absence of significant changes in these bands in the composite materials indicates that the UiO-66 structure remains stable after hybridization. For TiO₂, the presence of broad bands centered around 3427 cm^{-1} is attributed to -OH stretching from adsorbed moisture or surface hydroxyl groups, which play a crucial role in surface reactivity and potential hydrogen bonding with other phases. The bands at 2920 and 2853 cm^{-1} correspond to Ti-OH bond vibrations, possibly originating from residual organics or surface-adsorbed hydrocarbons. Peaks at 825 and 653 cm^{-1} are typically assigned to Ti-O-Ti and Ti-O-C bending vibrations, reflecting the TiO₂ lattice [18]. The FT-IR spectrum of g-C₃N₄ exhibits multiple absorption bands in the range between 1200 and 1650 cm^{-1} , which are characteristic of C=N and C $\equiv$ N heterocyclic stretching vibrations from the heptazine rings.

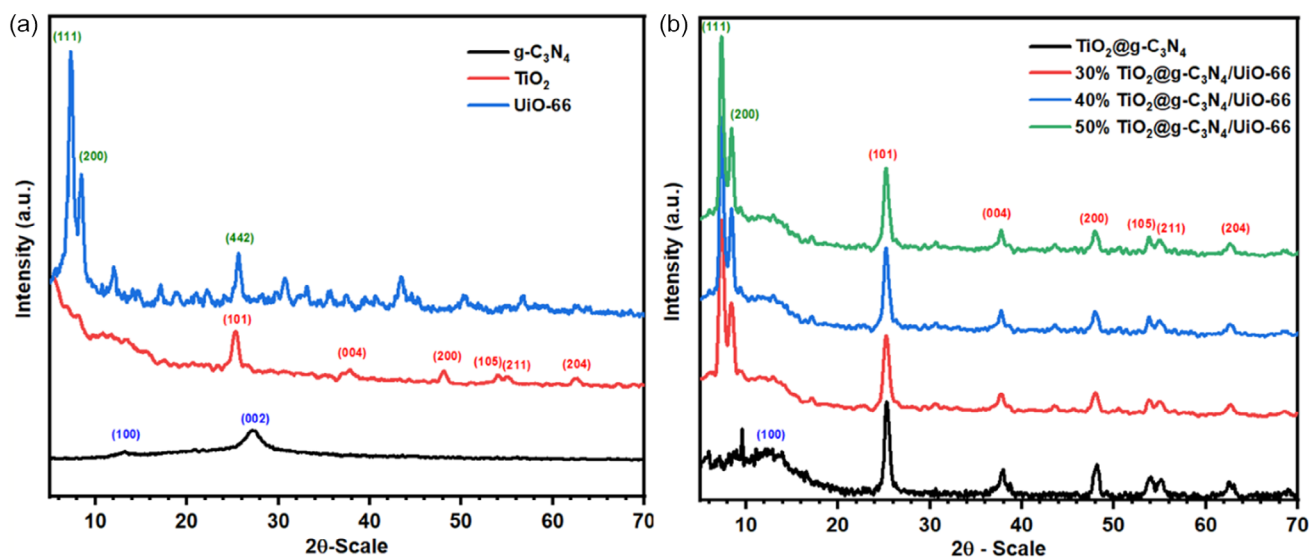


FIGURE 1 | XRD pattern of (a) g-C₃N₄, TiO₂, UiO-66 and (b) TiO₂@g-C₃N₄ and 30–50% TiO₂@g-C₃N₄/UiO-66 composites. XRD = X-ray diffraction.

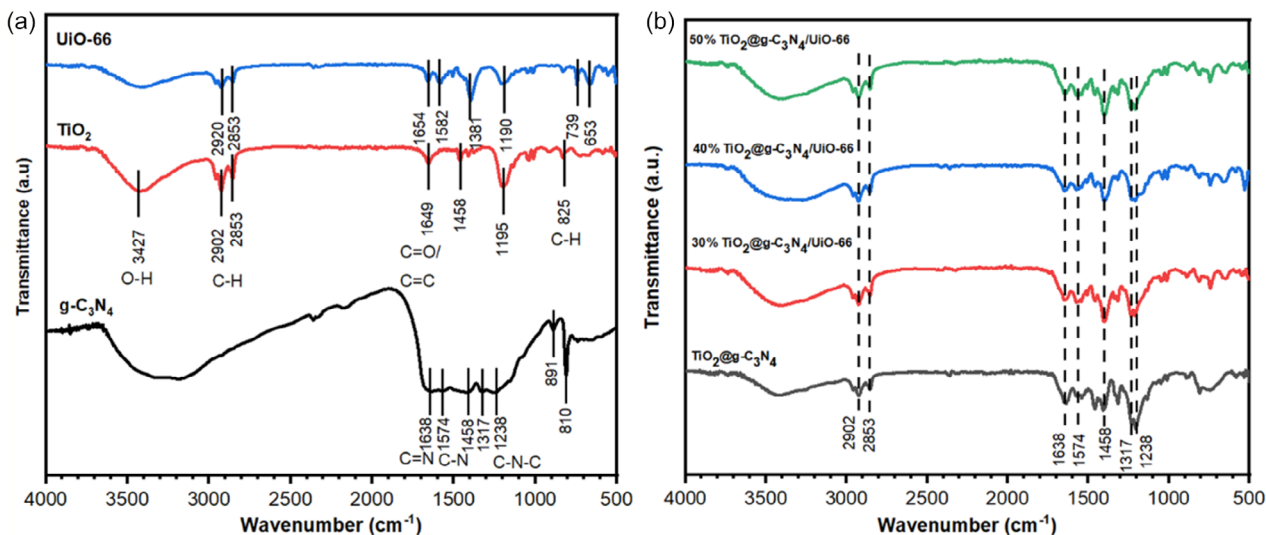


FIGURE 2 | FT-IR spectra of (a) $g\text{-C}_3\text{N}_4$, TiO_2 , and UiO-66, and (b) $\text{TiO}_2@g\text{-C}_3\text{N}_4$ and 30–50% $\text{TiO}_2@g\text{-C}_3\text{N}_4/\text{UiO-66}$ composites. FT-IR = Fourier transform infrared.

Specifically, bands at 1638, 1574, 1458, and 1317 cm^{-1} confirm the conjugated planar structure of polymeric $g\text{-C}_3\text{N}_4$ [19]. The sharp peak at 810 cm^{-1} corresponds to the breathing mode of triazine units, a signature feature of $g\text{-C}_3\text{N}_4$ [20]. The retention of these features in the composite spectra confirms the preservation of the $g\text{-C}_3\text{N}_4$ structure during synthesis.

In Figure 2b, the FT-IR spectra of the composite materials ($\text{TiO}_2@g\text{-C}_3\text{N}_4/\text{UiO-66}$) show the coexistence of the major functional group vibrations from each component, suggesting that the hybridization process did not compromise the individual structures. Notably, slight shifts and broadening in the peaks (e.g., C $\equiv$ N and C=N stretching) in the composites compared to pristine $g\text{-C}_3\text{N}_4$ suggest the occurrence of interfacial interactions among $g\text{-C}_3\text{N}_4$, TiO_2 , and UiO-66. These shifts may arise from hydrogen bonding between $g\text{-C}_3\text{N}_4$ and surface hydroxyl groups on TiO_2 or coordinative interactions with the metal clusters in UiO-66. Such

interactions are essential as they can facilitate interfacial charge transfer, reduce recombination of photogenerated electron–hole pairs, and enhance photocatalytic efficiency [21].

The scanning electron microscopy (SEM) images provide clear insights into the morphological characteristics of the synthesized materials. As shown in Figure 3a, UiO-66 exhibits well-defined polyhedral particles with relatively uniform distribution, which is characteristic of its crystalline MOF structure [14]. In contrast, $g\text{-C}_3\text{N}_4$ (Figure 3b) displays an aggregated sheet-like morphology with a rough surface, consistent with its intrinsic layered structure [22]. Upon the incorporation of TiO_2 , the $\text{TiO}_2@g\text{-C}_3\text{N}_4$ composite (Figure 3c) reveals agglomerated nanoparticles distributed over the $g\text{-C}_3\text{N}_4$ matrix, indicating the successful anchoring of TiO_2 onto the support [23].

More notably, the SEM images of the 40% $\text{TiO}_2@g\text{-C}_3\text{N}_4/\text{UiO-66}$ composite (Figure 3d–f) clearly demonstrate the coexistence of

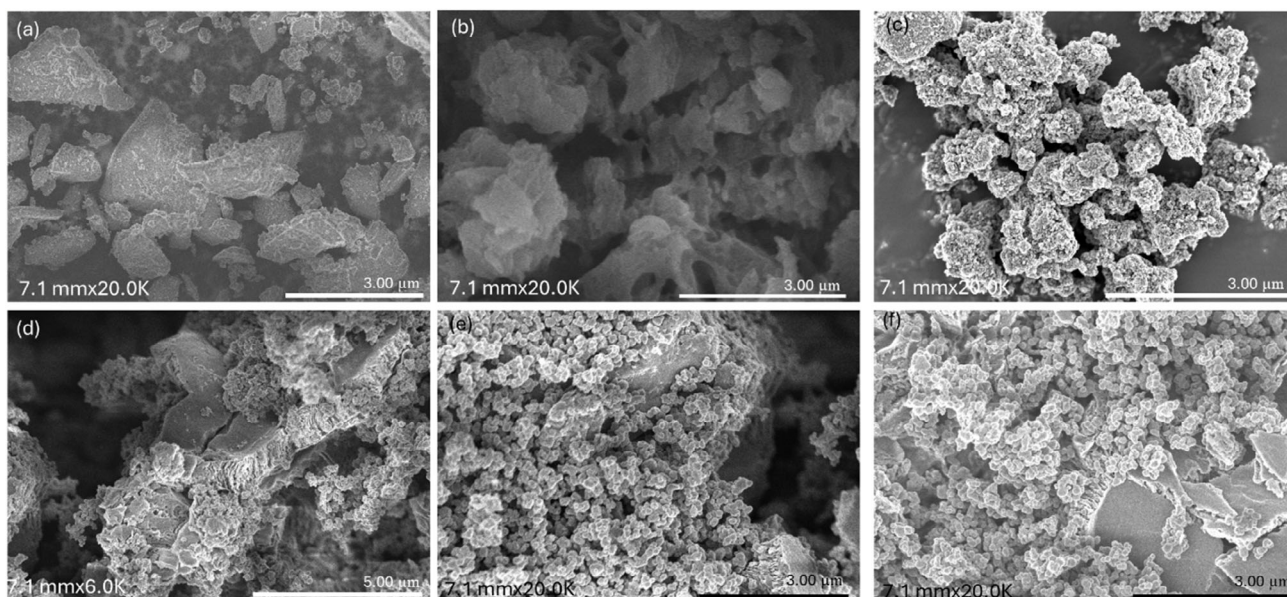


FIGURE 3 | SEM images of (a) UiO-66, (b) $g\text{-C}_3\text{N}_4$, (c) $\text{TiO}_2@g\text{-C}_3\text{N}_4$, and (d–f) 40% $\text{TiO}_2@g\text{-C}_3\text{N}_4/\text{UiO-66}$. SEM = Scanning electron microscopy.

both UiO-66 and $\text{TiO}_2\text{@g-C}_3\text{N}_4$ phases. The nanoparticles are homogeneously dispersed and anchored on the surface of UiO-66, forming a porous and interconnected structure. Such a hierarchical architecture not only preserves the intrinsic crystalline features of UiO-66 but also ensures the effective dispersion of $\text{TiO}_2\text{@g-C}_3\text{N}_4$ within the MOF framework. This synergistic morphology is expected to provide a larger surface area, enhanced interfacial contact, and abundant active sites, thereby facilitating charge separation and improving photocatalytic performance (Figure 4).

X-ray photoelectron spectroscopy (XPS) analysis of the $\text{TiO}_2\text{@g-C}_3\text{N}_4/\text{UiO-66}$ composite clearly reveals the coexistence of C, N, O, Zr, and Ti, confirming the successful integration of $\text{g-C}_3\text{N}_4$, TiO_2 , and UiO-66 into a single material (Figure 4) [24]. The high-resolution spectra provide insight into the electronic structure and chemical states of the elements. A notable feature is observed in the C 1s spectrum, where the main peak appears at 283.5 eV, showing a negative shift of ≈ -1.3 eV relative to the typical $\text{C}\equiv\text{C}$ bond at 284.8 eV, which suggests an increased electron density on carbon atoms within the composite [25]. In contrast, the Zr $3d_{5/2}$ peak is shifted to 182.7 eV, representing a positive shift of +0.5 eV compared to pristine UiO-66, indicating that Zr centers become more electron deficient [26]. The binding energies of N 1s (400.1 eV) and Ti $2p_{3/2}$ (458.5 eV) remain almost unchanged, implying preservation of the triazine framework of $\text{g-C}_3\text{N}_4$ and the Ti^{4+} oxidation state in TiO_2 [20, 27]. Additionally, the O 1s peak at 531.8 eV corresponds to surface hydroxyl or chemisorbed oxygen species, which are known to provide active sites for photocatalytic reactions [28]. Taken together, these XPS findings confirm strong interfacial electronic interactions and indicate electron transfer from UiO-66 (Zr nodes) to $\text{g-C}_3\text{N}_4$, which facilitates charge separation and rationalizes the enhanced photocatalytic activity of the $\text{TiO}_2\text{@g-C}_3\text{N}_4/\text{UiO-66}$ composite.

The nitrogen adsorption–desorption isotherms of $\text{TiO}_2\text{@g-C}_3\text{N}_4$, UiO-66, and the $\text{TiO}_2\text{@g-C}_3\text{N}_4/\text{UiO-66}$ composite at 77 K are shown in Figure 5. As shown in Figure 5a, both $\text{TiO}_2\text{@g-C}_3\text{N}_4$ and the $\text{TiO}_2\text{@g-C}_3\text{N}_4/\text{UiO-66}$ composite exhibited type IV adsorption–desorption isotherms as mesopores, whereas UiO-66 belonged to type I as micropores, according to the IUPAC classification. The UiO-66 possesses the highest specific surface area ($1017.27 \text{ m}^2/\text{g}$) and the largest average pore volume ($0.314 \text{ cm}^3/\text{g}$). In contrast, $\text{TiO}_2\text{@g-C}_3\text{N}_4$ showed the lowest surface area ($53.45 \text{ m}^2/\text{g}$) and pore volume ($0.127 \text{ cm}^3/\text{g}$). Upon integration of the photocatalytic $\text{TiO}_2\text{@g-C}_3\text{N}_4$ with the MOF UiO-66, the resulting $\text{TiO}_2\text{@g-C}_3\text{N}_4/\text{UiO-66}$ composite achieved a surface area of $305.87 \text{ m}^2/\text{g}$ and a pore volume of $0.211 \text{ cm}^3/\text{g}$. Evidently, after coupling with $\text{TiO}_2\text{@g-C}_3\text{N}_4$, both the surface area and the average pore volume decreased, which may be attributed to the $\text{TiO}_2\text{@g-C}_3\text{N}_4$ nanoparticles dispersing and adhering to the surface or filling the pores of the MOF framework. Brunauer–Emmett–Teller (BET) analysis also indicated that the pore size of the $\text{TiO}_2\text{@g-C}_3\text{N}_4/\text{UiO-66}$ composite (3.985 nm) did not deviate substantially from that of pristine UiO-66 (4.671 nm), suggesting that the deposition of $\text{TiO}_2\text{@g-C}_3\text{N}_4$ nanoparticles on the UiO-66 surface provided additional active sites for adsorption and catalytic reactions.

Photoluminescence (PL) and UV–Vis diffuse reflectance spectroscopy (DRS) analyses were conducted to investigate the optical properties and photoinduced charge separation efficiency of the individual components (UiO-66, TiO_2 , $\text{g-C}_3\text{N}_4$) and the composites $\text{g-C}_3\text{N}_4\text{@TiO}_2/\text{UiO-66}$ with varying loadings (Figure 6). The pristine TiO_2 and $\text{g-C}_3\text{N}_4$ display strong PL emission peaks in the ranges between 390–450 nm and 430–520 nm, respectively. These high PL intensities suggest a high recombination rate of photogenerated electron–hole (e^-/h^+) pairs, which is typically

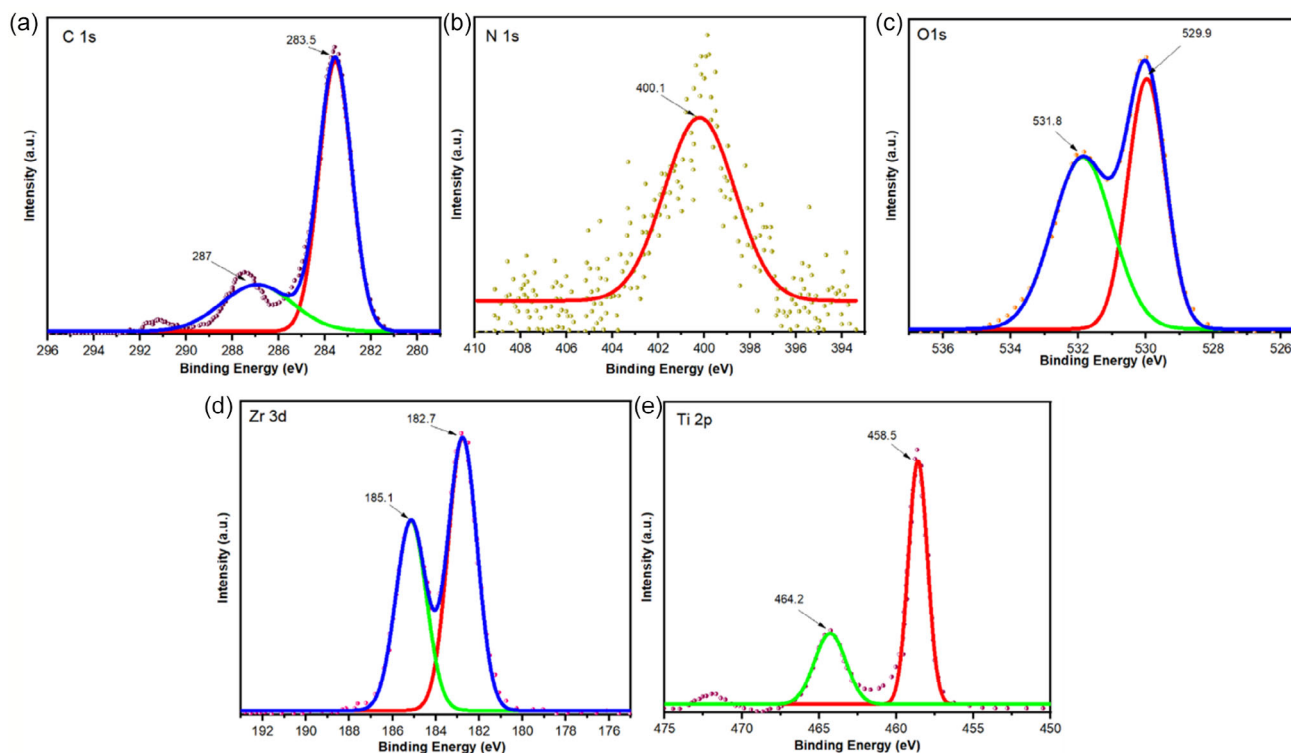


FIGURE 4 | XPS spectra of the $\text{TiO}_2\text{@g-C}_3\text{N}_4/\text{UiO-66}$ composite: (a) C 1s, (b) N 1s, (c) O 1s, (d) Zr 3d, and (e) Ti 2p.

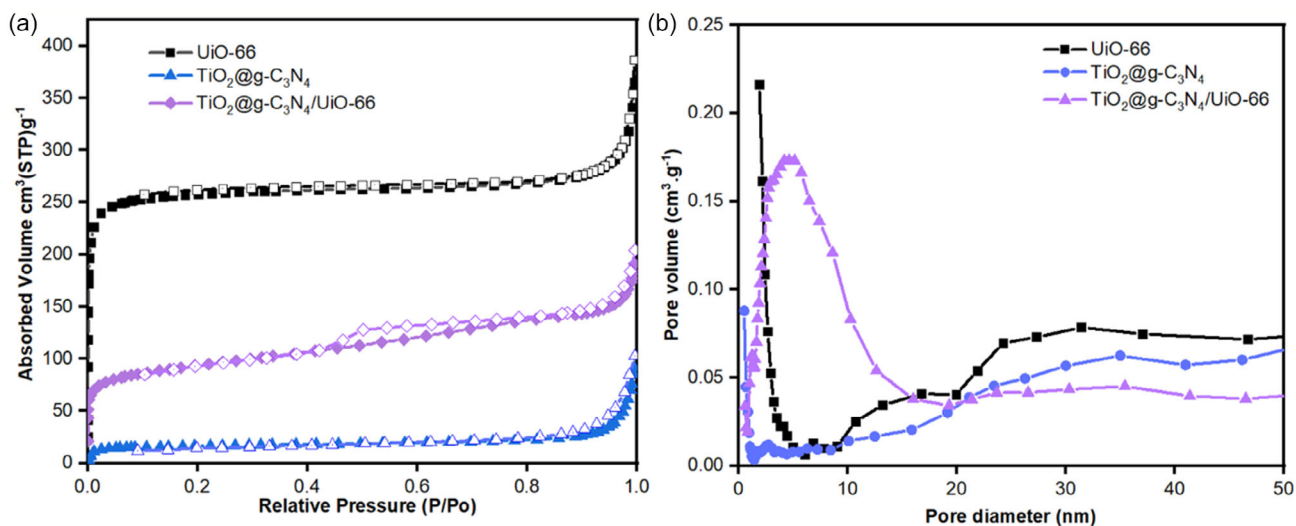


FIGURE 5 | (a) N_2 adsorption-desorption isotherms and (b) pore size distribution of UiO-66, $TiO_2@g-C_3N_4$, and the $TiO_2@g-C_3N_4/UiO-66$ composite.

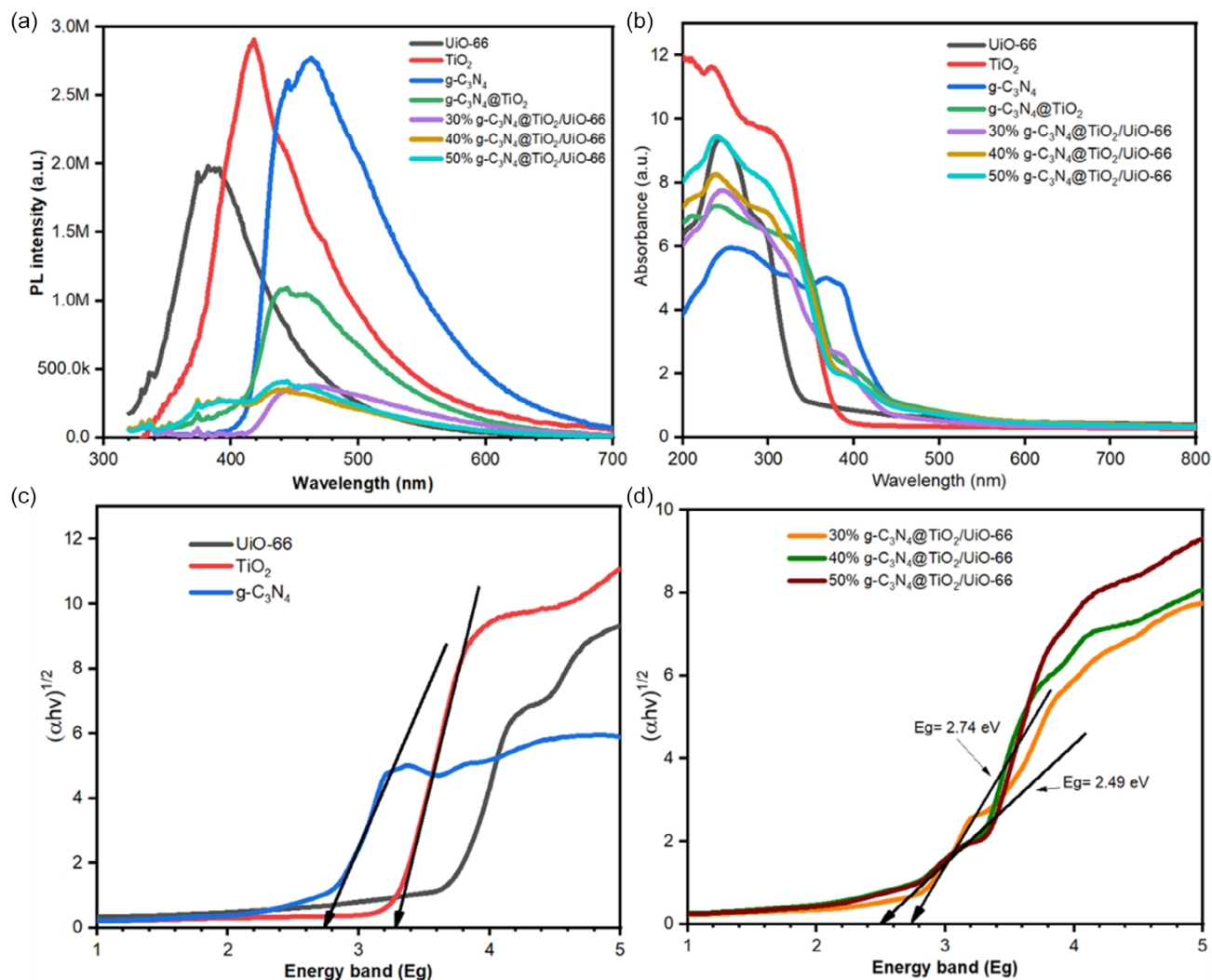


FIGURE 6 | (a) Photoluminescence spectra and (b) UV-Vis DRS of UiO-66, $g-C_3N_4$, TiO_2 , $TiO_2@g-C_3N_4$, and the 30–50% $TiO_2@g-C_3N_4/UiO-66$ composites. (c) Bandgap energy of UiO-66, $g-C_3N_4$, TiO_2 and (d) bandgap energy of 30–50% $TiO_2@g-C_3N_4/UiO-66$ composites. DRS = Diffuse reflectance spectroscopy.

detrimental to photocatalytic activity. In contrast, UiO-66 exhibits a significantly weaker PL signal due to its lower intrinsic photoactivity and poor charge carrier mobility. Interestingly, all composite samples exhibit a substantial suppression in PL intensity, particularly in the 30%, 40%, and 50% $\text{TiO}_2@\text{g-C}_3\text{N}_4/\text{UiO-66}$ samples. This attenuation in PL is a strong indication of reduced radiative recombination of charge carriers, implying that the heterojunction interface among $\text{g-C}_3\text{N}_4$, TiO_2 , and UiO-66 effectively facilitates charge separation and transfer [20, 29]. This enhancement can be attributed to the formation of a step scheme or type-II heterojunction (depending on band alignment), where photo-generated electrons from the conduction band (CB) of $\text{g-C}_3\text{N}_4$ and/or TiO_2 can migrate across the interface, while holes in the valence band (VB) may be retained in the component with stronger oxidative potential. This charge carrier migration promotes spatial separation, suppresses recombination, and enhances photocatalytic performance under visible-light irradiation.

The UV-Vis DRS spectra in Figure 6b provide insights into the light absorption capability of the materials. TiO_2 exhibits a sharp absorption edge near 390 nm, indicating that its intrinsic absorption is mainly in the UV region. $\text{g-C}_3\text{N}_4$ extends the absorption edge to about 460 nm, suggesting visible-light activity due to its narrower bandgap. UiO-66 absorbs minimally in the visible region. Remarkably, the ternary composites demonstrate a broadened and red-shifted absorption edge, reaching up to 500 nm, which suggests enhanced visible-light harvesting. This broadened absorption is likely due to the synergistic interaction among the three phases, which modifies the electronic structure and narrows the effective bandgap.

To quantitatively estimate the bandgap energies (E_g), Tauc plots were constructed as shown in Figure 6c,d. The E_g values were derived using the Tauc relation for indirect bandgap semiconductors

$$(\alpha h\nu)^n = A(h\nu - E_g) \quad (1)$$

where α is the absorption coefficient, $h\nu$ is the photon energy, A is a constant, and $n=2$ for indirect transitions. Based on

the extrapolation of the linear region to the photon energy axis, the estimated E_g values are ≈ 3.13 eV for TiO_2 , 3.36 eV for UiO-66, and 2.75 eV for $\text{g-C}_3\text{N}_4$. Importantly, the E_g values of the composite materials decrease progressively with increasing $\text{g-C}_3\text{N}_4$ content, with the 50% $\text{g-C}_3\text{N}_4@\text{TiO}_2/\text{UiO-66}$ sample exhibiting the lowest E_g at around 2.49 eV. This bandgap narrowing further supports the improved visible-light responsiveness of the composite materials.

2.2 | Photocatalytic Degradation of Polystyrene under Visible Light

Figure 7 presents the control experiments designed to clarify the individual roles of light irradiation and catalyst components in the degradation of PS. As shown in Figure 7a, when PS was exposed to light irradiation in the absence of any catalyst, the degradation efficiency remained nearly zero throughout the entire reaction period (0–30 h). This result clearly indicates that direct photolysis of PS under the applied light source is negligible, confirming that light irradiation alone is insufficient to induce PS degradation. Therefore, the presence of a photocatalyst is essential to activate reactive pathways for polymer breakdown. Figure 7b illustrates the adsorption behavior of PS in the presence of individual catalyst components (UiO-66, TiO_2 , and $\text{TiO}_2@\text{g-C}_3\text{N}_4$) as well as the $\text{TiO}_2@\text{g-C}_3\text{N}_4/\text{UiO-66}$ composite under dark conditions. All samples exhibit a rapid increase in PS adsorption during the initial stage, followed by a gradual stabilization, indicating the establishment of adsorption–desorption equilibrium. Among the tested materials, UiO-66 and the $\text{TiO}_2@\text{g-C}_3\text{N}_4/\text{UiO-66}$ composite show significantly higher PS adsorption capacities compared to pristine TiO_2 , which can be attributed to the high specific surface area and porous structure of the MOF component. The composite catalyst demonstrates the highest overall adsorption efficiency, suggesting a synergistic effect between UiO-66 and $\text{TiO}_2@\text{g-C}_3\text{N}_4$ that enhances PS enrichment on the catalyst surface. Importantly, no degradation occurs under dark conditions for any catalyst, despite measurable PS adsorption. This observation confirms that adsorption alone

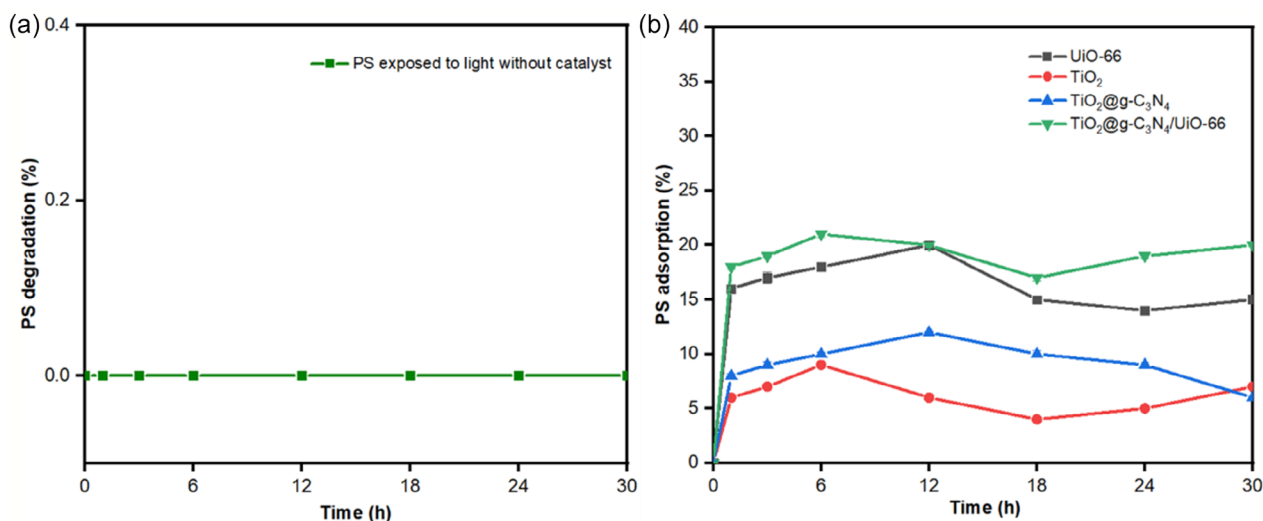


FIGURE 7 | Control experiments for PS degradation and adsorption. (a) PS degradation efficiency under light irradiation in the absence of any catalyst and (b) PS adsorption behavior in the presence of individual catalyst components (UiO-66, TiO_2 , and $\text{TiO}_2@\text{g-C}_3\text{N}_4$) and the $\text{TiO}_2@\text{g-C}_3\text{N}_4/\text{UiO-66}$ composite under dark conditions.

does not contribute to PS degradation and that photocatalytic activity is strictly light-driven. The enhanced adsorption capacity of the $\text{TiO}_2/\text{g-C}_3\text{N}_4/\text{UiO-66}$ composite is expected to promote photocatalytic degradation under illumination by increasing local PS concentration near active sites, thereby facilitating interfacial charge transfer and reactive species attack.

2.2.1 | Experimental Conditions

PS solution volume = 50 mL; catalyst dosage = 50 mg (unless otherwise specified); initial PS concentration = 200 ppm (unless otherwise specified); pH = 7; temperature = 30°C; visible-light source = 250 W xenon lamp equipped with a cutoff filter ($\lambda > 420$ nm); continuous magnetic stirring during irradiation.

2.2.1.1 | Effect of Catalyst Composition. Figure 8a compares the photocatalytic degradation efficiency of PS using UiO-66, TiO_2 , $\text{g-C}_3\text{N}_4$, $\text{TiO}_2/\text{g-C}_3\text{N}_4$, $\text{TiO}_2/\text{UiO-66}$, $\text{g-C}_3\text{N}_4/\text{UiO-66}$, and $\text{TiO}_2/\text{g-C}_3\text{N}_4/\text{UiO-66}$. Notably, pristine $\text{g-C}_3\text{N}_4$ exhibits a relatively high PS degradation efficiency ($\approx 79\%$ after 28 h), which can be attributed to its suitable bandgap (≈ 2.75 eV), strong visible-light absorption, and sufficiently negative CB potential that favors the generation of superoxide radicals

($\cdot\text{O}_2^-$). However, despite this appreciable activity, $\text{g-C}_3\text{N}_4$ suffers from rapid electron-hole recombination and limited charge transport efficiency, which restricts its long-term photocatalytic performance. In contrast, the $\text{TiO}_2/\text{g-C}_3\text{N}_4/\text{UiO-66}$ composite achieves the highest degradation efficiency (90.2% after 28 h), significantly outperforming both the individual components and binary systems. This enhancement arises from the synergistic integration of TiO_2 and UiO-66 with $\text{g-C}_3\text{N}_4$, which establishes multiple heterojunction interfaces, facilitates cascade charge transfer, suppresses recombination, and increases the steady-state concentration of reactive oxygen species. Additionally, the porous UiO-66 framework improves catalyst dispersion and adsorption of PS fragments, further promoting efficient and sustained photocatalytic degradation [30, 31].

2.2.1.2 | Effect of Catalyst Dosage. As shown in Figure 8b, the degradation efficiency increased significantly when the catalyst amount was increased from 25 mg to 50 mg, but only a slight improvement was observed at 75 mg. This indicates that 50 mg is the optimal dosage. At lower concentrations, the limited number of active sites reduces the generation of reactive radicals, while excessive catalyst loading leads to light scattering and shielding effects, thereby decreasing the effective utilization of photons [32].

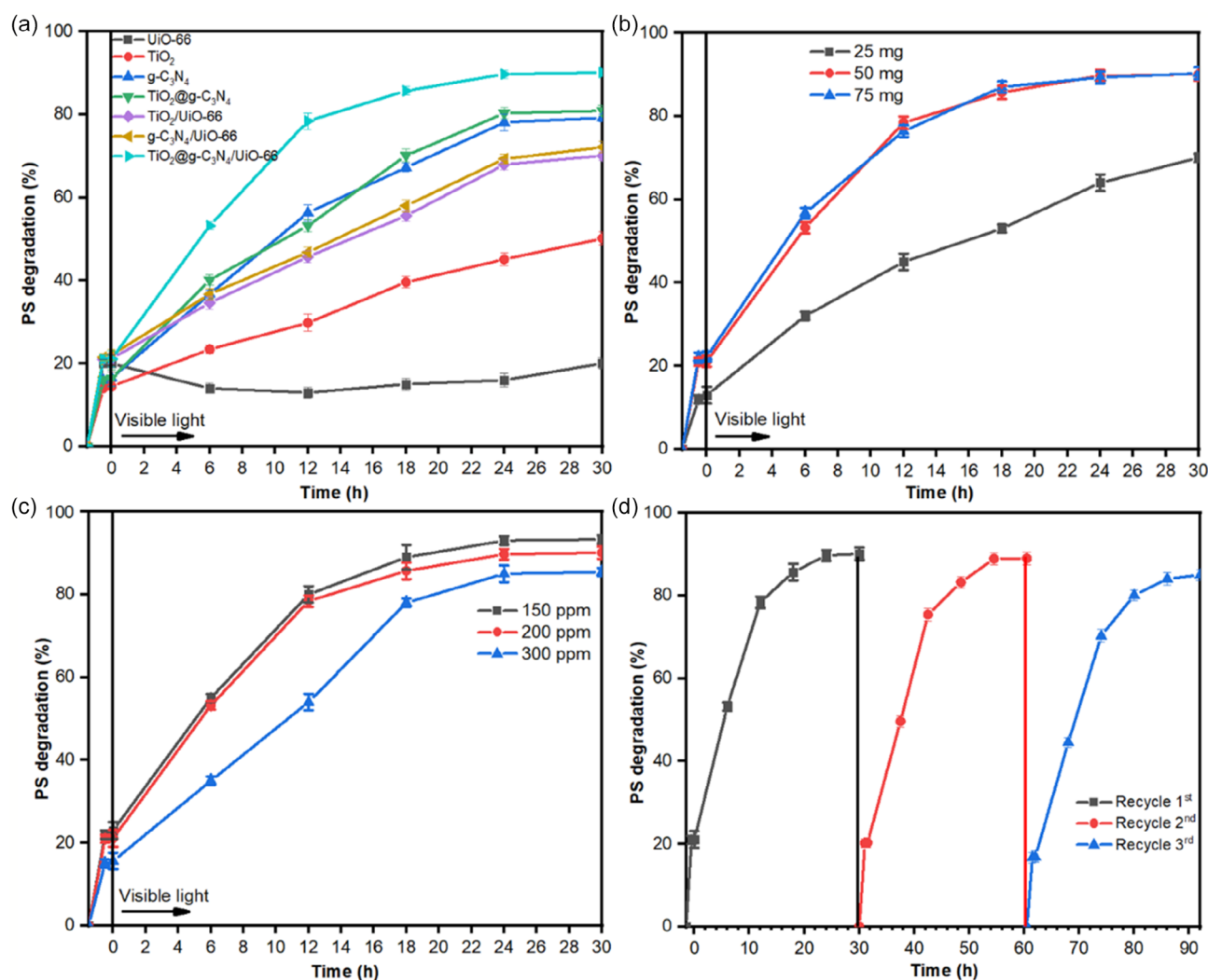


FIGURE 8 | (a) Photocatalytic degradation of PS using UiO-66, $\text{g-C}_3\text{N}_4$, TiO_2 , $\text{TiO}_2/\text{g-C}_3\text{N}_4$, and $\text{TiO}_2/\text{g-C}_3\text{N}_4/\text{UiO-66}$ composites, (b) effect of $\text{TiO}_2/\text{g-C}_3\text{N}_4/\text{UiO-66}$ dosage, (c) effect of PS concentration, and (d) recyclability of $\text{TiO}_2/\text{g-C}_3\text{N}_4/\text{UiO-66}$ in the photodegradation of PS.

2.2.1.3 | Effect of Polystyrene Concentration. Figure 8c illustrates that the degradation efficiency decreased with increasing PS concentration. At 150 ppm, nearly 95% of PS was degraded, while at 300 ppm, the efficiency dropped to 75%. The decline in activity at higher concentrations is due to the reduced light penetration and competition between PS molecules for reactive radicals, leading to incomplete degradation [33].

2.2.1.4 | Reusability and Stability of the Catalyst. The recyclability of $\text{TiO}_2\text{@g-C}_3\text{N}_4/\text{UiO-66}$ was tested over three consecutive cycles (Figure 8d). The degradation efficiency slightly decreased from 95% to 85% after three runs, indicating good structural stability and resistance to photocorrosion. The minor loss in activity could be related to catalyst loss during recovery or partial surface fouling by intermediate products.

The contribution of different reactive species in the photocatalytic degradation of PS over the $\text{TiO}_2\text{@g-C}_3\text{N}_4/\text{UiO-66}$ catalyst was investigated through active species trapping experiments under visible-light irradiation (Figure 8). Specific scavengers were employed to quench targeted species, including tert-butanol (TBA) for hydroxyl radicals ($\bullet\text{OH}$), benzoquinone (BQ) for superoxide radicals ($\bullet\text{O}_2^-$), ammonium oxalate (AO) for photogenerated holes (h^+), and potassium dichromate ($\text{K}_2\text{Cr}_2\text{O}_7$) for electrons (e^-).

In the absence of scavengers, the degradation efficiency of PS reached 90.2% after 30 h of under visible-light irradiation, confirming the excellent photocatalytic activity of the $\text{TiO}_2\text{@g-C}_3\text{N}_4/\text{UiO-66}$ composite. When TBA was introduced, the degradation efficiency markedly decreased to about 47.8%, indicating that hydroxyl radicals ($\bullet\text{OH}$) play a crucial role in the oxidation of PS. Similarly, the addition of BQ caused a significant reduction in photocatalytic activity (about 49%), revealing that superoxide radicals ($\bullet\text{O}_2^-$) are also key active species involved in the degradation process. In contrast, the inhibition observed with AO and $\text{K}_2\text{Cr}_2\text{O}_7$ was relatively moderate, suggesting that photogenerated holes and electrons mainly serve as precursors for the formation of reactive oxygen species rather than acting as the direct oxidizing agents. These results demonstrate that both $\bullet\text{OH}$ and $\bullet\text{O}_2^-$ radicals are the dominant oxidative species contributing to PS

degradation, while the synergistic heterojunction interface among TiO_2 , $\text{g-C}_3\text{N}_4$, and UiO-66 effectively promotes charge separation and transfer. The improved separation efficiency facilitates the simultaneous generation of $\bullet\text{OH}$ and $\bullet\text{O}_2^-$ radicals, which synergistically attack the polymer backbone, leading to efficient PS decomposition under visible-light irradiation.

The particle size distribution of PS before and after photocatalytic degradation using $\text{TiO}_2\text{@g-C}_3\text{N}_4/\text{UiO-66}$ is shown in Figure 9a,b. Before the reaction (Figure 10a), PS exhibited a single, narrow peak centered around 500 nm, with a low polydispersity index ($\text{PDI}=0.153$), indicating a uniform and stable dispersion of PS particles in the suspension. After photocatalytic treatment (Figure 10b), the DLS curve revealed two distinct peaks located at $\approx 20\text{--}30\text{ nm}$ and $60\text{--}80\text{ nm}$, while the PDI markedly increased to 0.564. This bimodal distribution demonstrates that PS underwent significant size reduction and structural fragmentation during photocatalytic degradation. The appearance of smaller particles is attributed to chain scission reactions induced by photogenerated reactive oxygen species (ROS), such as hydroxyl radicals ($\bullet\text{OH}$) and superoxide radicals ($\bullet\text{O}_2^-$), which attack the $\text{C}\text{--}\text{C}$ backbone of PS chains [34]. The higher PDI value indicates a broader particle size distribution and increased heterogeneity of the degraded products, which is typical of nonuniform oxidative fragmentation processes in polymer photocatalysis [35]. Additionally, the small-size peak ($20\text{--}30\text{ nm}$) may also correspond to residual photocatalyst nanoparticles ($\text{TiO}_2\text{@g-C}_3\text{N}_4/\text{UiO-66}$) remaining in the dispersion, contributing to the observed bimodal distribution.

The Nyquist plots of UiO-66, $\text{g-C}_3\text{N}_4$, TiO_2 , $\text{TiO}_2\text{@g-C}_3\text{N}_4$, and $\text{TiO}_2\text{@g-C}_3\text{N}_4/\text{UiO-66}$ are shown in Figure 11. All samples display a depressed semicircle in the high-frequency region, which is characteristic of the charge-transfer resistance (R_{ct}) at the electrode-electrolyte interface [36]. Notably, the $\text{TiO}_2\text{@g-C}_3\text{N}_4/\text{UiO-66}$ composite exhibits a significantly smaller semicircle than pristine $\text{g-C}_3\text{N}_4$ and TiO_2 , and a comparable R_{ct} value to UiO-66, indicating substantially improved interfacial charge transport upon formation of the ternary heterojunction. Although UiO-66 alone shows a similar R_{ct} value, its photocatalytic activity is limited by the absence of efficient visible-light absorption and reactive redox sites [37]. The binary $\text{TiO}_2\text{@g-C}_3\text{N}_4$ heterostructure shows an intermediate R_{ct} , confirming that coupling TiO_2 with $\text{g-C}_3\text{N}_4$ partially enhances electron transport [38]. In contrast, the ternary $\text{TiO}_2\text{@g-C}_3\text{N}_4/\text{UiO-66}$ system integrates efficient charge mobility with favorable band alignment and abundant interfacial contact, which collectively promote charge separation and utilization in photocatalytic PS degradation rather than relying solely on absolute R_{ct} values.

Electrochemical impedance spectroscopy (EIS) provides strong support for the proposed electron transfer behavior. As shown in Figure 11, the $\text{TiO}_2\text{@g-C}_3\text{N}_4/\text{UiO-66}$ composite exhibits the smallest semicircle radius in the Nyquist plot, corresponding to the lowest R_{ct} among all samples. This behavior indicates highly efficient interfacial charge transport and suppressed electron-hole recombination, which are widely recognized as essential prerequisites for effective heterojunction-driven photocatalytic degradation of polymeric MPs. Similar correlations between reduced R_{ct} , enhanced charge mobility, and accelerated microplastic oxidation have been reported in recent photocatalytic systems targeting persistent polymer pollutants [39, 40].

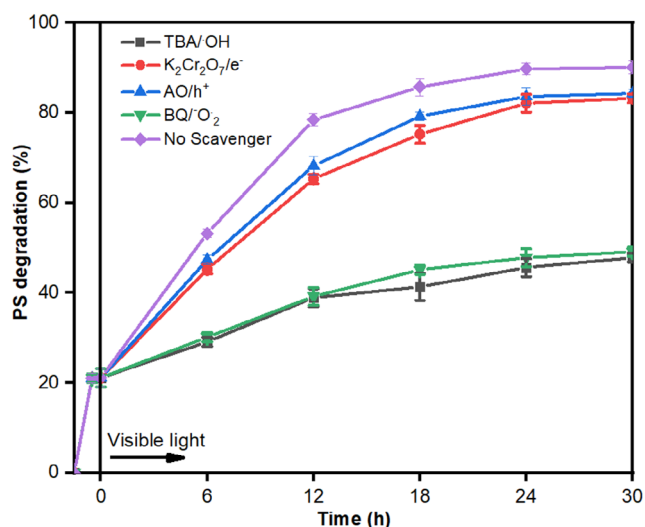


FIGURE 9 | The active species trapping experiments for the degradation of PS over the $\text{TiO}_2\text{@g-C}_3\text{N}_4/\text{UiO-66}$ photocatalyst.

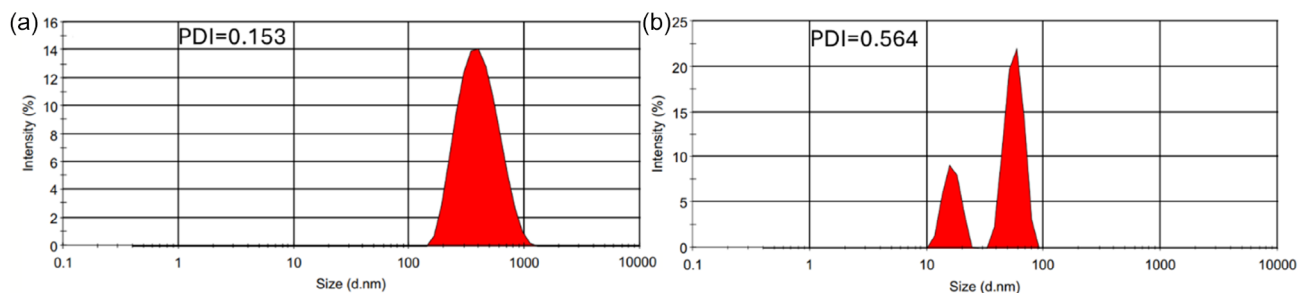


FIGURE 10 | DLS of (a) PS initial and (b) PS after reaction. DLS = Dynamic light scattering.

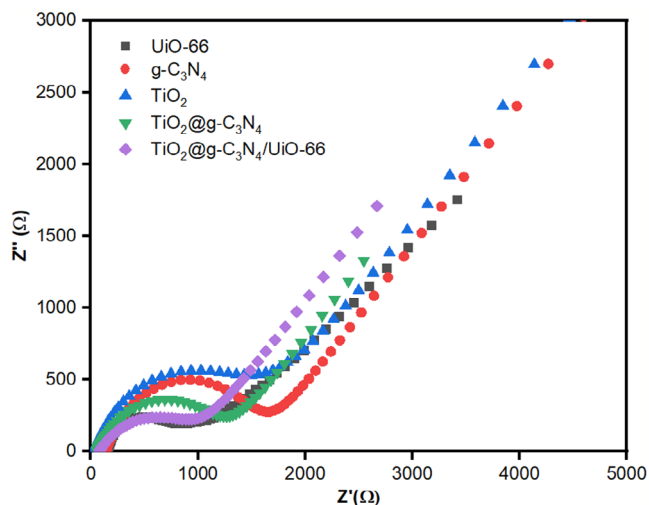


FIGURE 11 | EIS Nyquist plots of UiO-66, g-C₃N₄, TiO₂, TiO₂@g-C₃N₄, and TiO₂@g-C₃N₄/UiO-66 samples in a 0.1 M KCl containing 5 mM K₃[Fe(CN)₆] and K₄[Fe(CN)₆]. EIS = Electrochemical impedance spectroscopy.

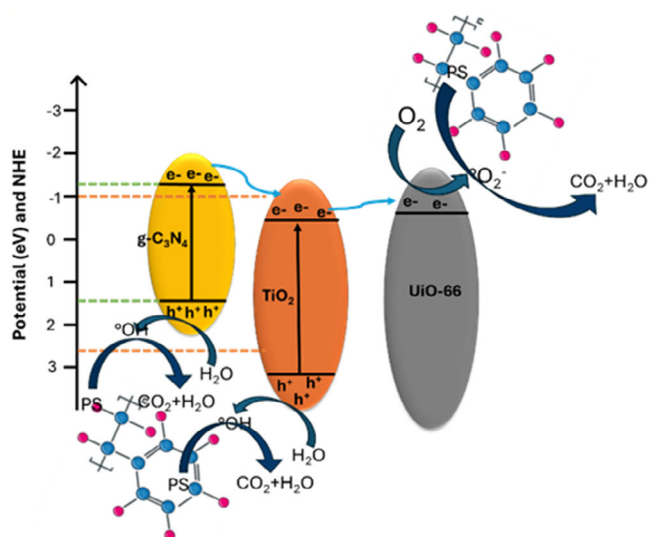


FIGURE 12 | Proposed schematic illustration of the photocatalytic mechanism of PS degradation over the TiO₂@g-C₃N₄/UiO-66 composite.

Based on the relative band edge positions, a cascade electron transfer pathway is proposed (Figure 12). Upon light irradiation, photogenerated electrons in the CB of g-C₃N₄ (−1.1 eV) are thermodynamically driven to migrate to the CB of TiO₂

(−0.3 eV) and subsequently to UiO-66 (−0.6 eV). UiO-66 thus acts as an effective electron acceptor and reservoir, facilitating the reduction of adsorbed O₂ molecules to superoxide radicals (•O₂[−]). Meanwhile, the photogenerated holes remaining in the VBs of g-C₃N₄ and particularly TiO₂ (2.83 eV) possess sufficient oxidative potential to oxidize surface hydroxyl groups or water molecules, generating highly reactive hydroxyl radicals (•OH). These reactive oxygen species (•O₂[−], •OH), together with photo-generated holes (h⁺), initiate oxidative attacks on the C≡C and C≡H bonds of PS chains, leading to polymer chain scission and progressive fragmentation into lower-molecular-weight intermediates, consistent with recently reported photocatalytic microplastic degradation pathways [40–42]. The significantly reduced R_{ct} value observed in EIS confirms that the ternary heterojunction effectively promotes charge separation and prolongs carrier lifetime, thereby increasing the steady-state concentration of reactive oxygen species and enhancing PS degradation efficiency. In addition to its electronic role, the porous UiO-66 framework contributes structurally by improving the dispersion of TiO₂@g-C₃N₄ and providing abundant adsorption sites for PS fragments and dissolved oxygen. This adsorption–reaction synergy has been identified as a critical factor for sustaining photocatalytic oxidation of MPs in heterogeneous systems [43]. Therefore, the synergistic integration of favorable band alignment, efficient charge mobility, and structural confinement collectively substantiates the proposed photocatalytic mechanism for PS degradation.

To further elucidate the PS degradation mechanism over the TiO₂@g-C₃N₄/UiO-66 composite, the FT-IR spectra of the initial PS solution and that after 30 h of photocatalytic degradation were recorded. As shown in Figure 13, the initial PS solution exhibits characteristic absorption bands at 3019 and 2898 cm^{−1}, corresponding to aromatic and aliphatic C≡H stretching vibrations, along with bands at 1499, 1535, and 1402 cm^{−1} attributed to aromatic ring stretching modes. The absorption bands at 841 and 749 cm^{−1} are associated with out-of-plane C–H bending of mono-substituted benzene rings, confirming the intact PS backbone before photocatalytic treatment [44]. After photocatalytic degradation, several notable spectral changes are observed. Most importantly, a broad absorption band centered at ≈3291 cm^{−1} emerges, which can be assigned to O–H stretching vibrations of hydroxyl and hydroperoxide groups, indicating oxidative modification of PS chains [45]. In addition, the appearance and enhancement of the band at 1637 cm^{−1} suggest the formation of carbonyl-containing oxidation products or conjugated C=C structures, which are widely recognized as fingerprints of polymer photooxidation and chain scission processes [46]. Furthermore, the attenuation and distortion of characteristic aromatic C–H and ring vibration bands imply partial disruption of

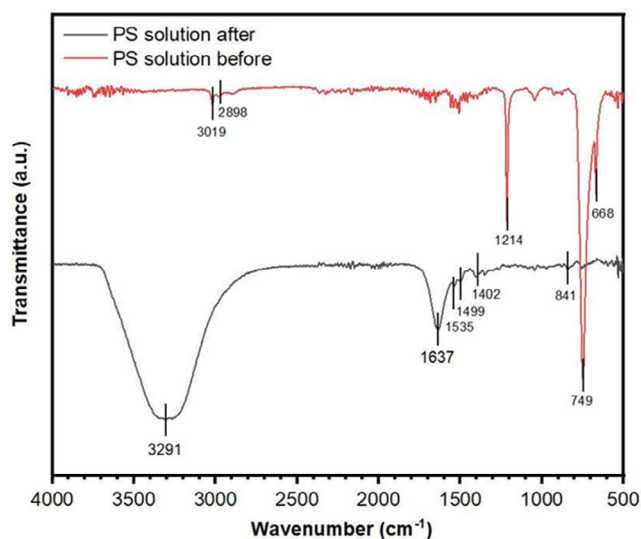


FIGURE 13 | FT-IR spectra of the initial PS solution and the solution following a 30-h reaction. FT-IR = Fourier transform infrared.

the PS backbone, consistent with reactive oxygen species ($\bullet\text{OH}$, $\text{O}_2\bullet^-$, and h^+)-induced chain scission during photocatalytic degradation [47]. Such FT-IR signatures have been extensively reported as reliable chemical evidence of oxidative degradation of PS and other aromatic polymers, even in the absence of nano-scale morphological imaging [46].

As shown in the FT-IR spectra of the $\text{TiO}_2/\text{g-C}_3\text{N}_4/\text{UiO-66}$ composite before and after photocatalytic degradation (Figure 14), the characteristic vibrational bands of the catalyst framework remain essentially unchanged after the reaction. In particular, the typical bands of UiO-66 (Zr–O stretching), $\text{g-C}_3\text{N}_4$ ($\text{C}\equiv\text{N}$ and $\text{C}=\text{N}$ stretching in the $1200\text{--}1650\text{ cm}^{-1}$ region), and TiO_2 (Ti–O–Ti vibrations below 800 cm^{-1}) are well preserved, indicating the excellent structural stability of the composite during the photocatalytic process. Importantly, no additional absorption bands corresponding to PS are observed in the postreaction

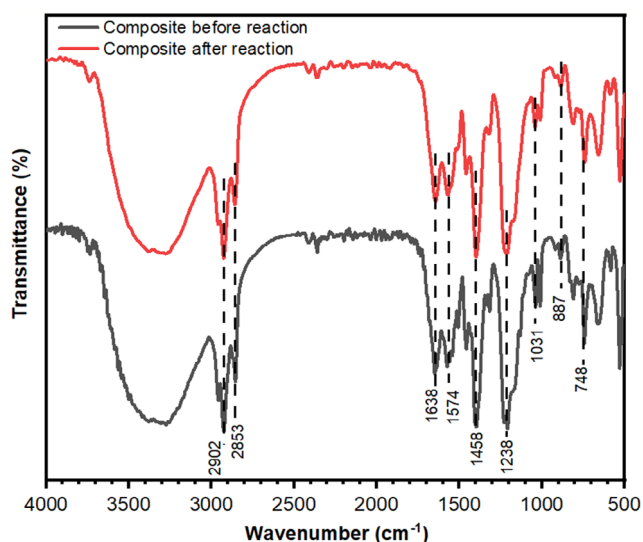


FIGURE 14 | FT-IR spectra of $\text{TiO}_2/\text{g-C}_3\text{N}_4/\text{UiO-66}$ composite before and after photocatalytic degradation of PS. FT-IR = Fourier transform infrared.

spectrum. The characteristic PS vibrational features, including the aliphatic C-H stretching bands at 2920 and 2850 cm^{-1} , the aromatic $\text{C}=\text{C}$ stretching vibrations at $1600\text{--}1500\text{ cm}^{-1}$, and the out-of-plane aromatic C-H bending modes at $750\text{--}700\text{ cm}^{-1}$, are absent. This confirms that no PS particles or polymeric residues are adsorbed or leached onto the catalyst surface after photocatalytic degradation. Therefore, the FT-IR results demonstrate both the chemical robustness of the $\text{TiO}_2/\text{g-C}_3\text{N}_4/\text{UiO-66}$ composite and the absence of residual PS contamination on the catalyst, supporting the conclusion that PS degradation occurs predominantly in the reaction medium rather than through simple surface accumulation.

3 | Conclusion

The $\text{TiO}_2/\text{g-C}_3\text{N}_4/\text{UiO-66}$ composite exhibited remarkable performance in degrading PS-MPs, attributed to its synergistic heterojunction structure and efficient charge transport pathways. The porous UiO-66 framework enhanced adsorption and dispersion of PS particles, while the $\text{TiO}_2/\text{g-C}_3\text{N}_4$ coupling extended light absorption and facilitated charge separation. As revealed by EIS, the low R_{ct} and strong visible-light activity promoted abundant ROS generation, accelerating oxidative fragmentation of PS into smaller, less persistent fragments. This study demonstrates a dual benefit: upcycling PET waste into a functional photocatalyst and efficiently degrading MPs under visible light, offering a sustainable approach to plastic pollution mitigation.

4 | Experimental Methods

4.1 | Materials and Chemicals

PET waste bottles were collected, thoroughly washed with distilled water, and dried before use. Zirconium(IV) oxychloride octahydrate ($\text{ZrOCl}_2 \cdot 8\text{H}_2\text{O}$, $\geq 98\%$), TPA (BDC, 99%), N, N-dimethylformamide (DMF, 99.8%), titanium tetraisopropoxide (TTIP, 97%), ethanol (EtOH, 99.5%), acetic acid (CH_3COOH , 99.8%), urea ($\text{CO}(\text{NH}_2)_2$, $\geq 99\%$), and PS-MPs ($M_w = 192,000\text{ g/mol}$) were purchased from Sigma-Aldrich and used without further purification. Distilled water was used in all synthesis processes.

4.2 | Synthesis of Terephthalic Acid from Polyethylene Terephthalate

The recovery process of TPA (BDC) from waste PET was conducted according to the procedure previously reported by our research group [48].

4.3 | Synthesis UiO-66

The UiO-66 MOF was synthesized via a solvothermal method. Initially, a 0.3 M ligand stock solution was prepared by dissolving TPA (BDC) in 20 mL of DMF, followed by stirring at 300 rpm for 5 h and sonication for 30 min . Concurrently, 0.35 g of zirconium(IV) oxychloride octahydrate ($\text{ZrOCl}_2 \cdot 8\text{H}_2\text{O}$) was dissolved in 15 mL of DMF, stirred for 3 h , and sonicated for 30 min . To this solution, 0.3 mL of distilled water and 1.3 mL of acetic acid were added, and the mixture was further stirred

and sonicated before being stirred at room temperature for 24 h. For the synthesis, 5 mL of the BDC stock solution was slowly added to the zirconium precursor solution, and the mixture was stirred for 5 h. The resulting solution was then transferred to an autoclave and heated at 120°C for 36 h. The as-synthesized product was purified by washing three times with DMF and once with ethanol. The sample was separated after each washing step by centrifugation or vacuum filtration. Finally, the purified white powder was dried at 100°C for 24 h.

4.4 | Synthesis TiO₂

To synthesize TiO₂, 20 mL of TTIP was added to 32 mL of ethanol and stirred for 1 h to form a homogeneous solution. A second homogeneous solution was prepared by mixing 8 mL of water, 32 mL of ethanol, and 16 mL of acetic acid. The first solution was then slowly added to the second solution and stirred continuously for 24 h to form a gel. This gel was subsequently dried at 100°C for 12 h to obtain TiO₂.

4.5 | Synthesis of TiO₂@g-C₃N₄

To synthesize the TiO₂@g-C₃N₄ composite, 2.8 g of the prepared TiO₂ was added to 50 mL of a urea solution, maintaining a molar ratio of N: Ti at 3:1. The mixture was stirred for 1 h and then sonicated for 30 min. The resulting suspension was dried at 100°C for 12 h. Finally, it was calcined at 450°C with a heating rate of 5°C/min for 3 h to obtain the TiO₂@g-C₃N₄ composite.

4.6 | Synthesis TiO₂@g-C₃N₄/UiO-66

First, 0.5 g of TiO₂@g-C₃N₄ was added to 50 mL of ethanol, stirred thoroughly, and then sonicated for 30 min to create mixture A. Similarly, 0.5 g of UiO-66 was added to 50 mL of ethanol, stirred, and sonicated for 30 min to form mixture B. Next, mixture A and mixture B were combined, stirred for 1 h, and then sonicated for an additional 30 min. The resulting solution is transferred into an autoclave and heated at 120°C for 24 h. The final product was filtered and dried at 80°C to yield the TiO₂@g-C₃N₄/UiO-66 composite.

4.7 | Characterization

The synthesized materials were characterized using various analytical techniques. The crystalline structure and phase composition were identified by XRD (Bruker, Germany) using Cu K α radiation ($\lambda = 0.154$ nm) at a scanning rate of 0.2°s⁻¹. FT-IR spectra were recorded on a Jasco 4700 spectrometer to identify functional groups and bonding characteristics. The morphology and particle size distribution were examined by SEM (S-4800, Hitachi).

The optical properties were investigated by UV-Vis DRS (Shimadzu) and PL spectroscopy using a Cary Eclipse fluorescence spectrophotometer (Varian). The BET-specific surface area and pore characteristics were determined from N₂ adsorption-desorption isotherms at liquid nitrogen temperature (77 K) using a Tristar-3030 analyzer (Micromeritics, USA). The surface

chemical states and elemental compositions were further analyzed by XPS (ESCALab 250, Thermo VG, UK).

4.8 | Investigation of Photocatalytic Activity

The photocatalytic performance of the synthesized materials was evaluated for the degradation of PS-MPs in the liquid phase. In a typical experiment, 50 mg of photocatalyst was dispersed in 50 mL of PS suspension (200 ppm) under continuous magnetic stirring at 500 rpm. The suspension was first kept in the dark for 1 h to achieve adsorption-desorption equilibrium between PS particles and the catalyst surface. After equilibrium, the system was irradiated with a 250 W xenon lamp for 30 h. During the reaction, 3 mL aliquots of the suspension were withdrawn every 6 h, followed by filtration to remove the catalyst. The concentration of PS in the filtrate (C) was determined using a UV-Vis spectrophotometer (Shimadzu UV-2450) at the maximum absorption wavelength of 290 nm ($\lambda_{\text{max,PS}} = 290$ nm). The degradation efficiency of PS was calculated using the following equation

$$H\% = \frac{(C_0 - C)}{C_0} \times 100 \quad (2)$$

where C_0 and C represent the initial and time-dependent concentrations of PS, respectively.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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