

# Efficient Photocatalytic Dye Degradation and Bacterial Inactivation by Graphitic Carbon Nitride and Starch-Doped Magnesium Hydroxide Nanostructures

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Cite This: *ACS Omega* 2022, 7, 39998–40008



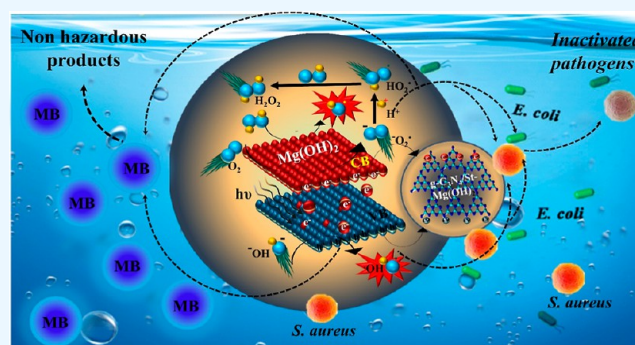
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**ABSTRACT:** The removal of hazardous pollutants from water is becoming an increasingly interesting topic of research considering their impact on the environment and the ecosystem. This work was carried out to synthesize graphitic carbon nitride (g-C<sub>3</sub>N<sub>4</sub>) and starch-doped magnesium hydroxide (g-C<sub>3</sub>N<sub>4</sub>/St-Mg(OH)<sub>2</sub>) nanostructures via a facile co-precipitation process. The focus of this study is to treat polluted water and bactericidal behavior with a ternary system (doping-dependent Mg(OH)<sub>2</sub>). Different concentrations (2 and 4 wt %) of g-C<sub>3</sub>N<sub>4</sub> were doped in a fixed amount of starch and Mg(OH)<sub>2</sub> to degrade methylene blue dye from an aqueous solution with bactericidal potential against *Escherichia coli* (*E. coli*) and *Staphylococcus aureus* (*S. aureus*) pathogens. The textural structures, morphological evolutions, and optical characteristics of the as-prepared samples were analyzed using advanced characterization techniques. X-ray diffraction confirmed the hexagonal phase of Mg(OH)<sub>2</sub> with improved crystallinity upon doping. Fourier transform infrared spectroscopy revealed Mg(OH)<sub>2</sub> stretching vibrations and other functional groups. UV–visible spectroscopy exhibited a red shift (bathochromic effect) in absorption spectra representing the decrease in energy band gap ( $E_g$ ). Photoluminescence patterns were recorded to study recombination of charge carriers (e<sup>-</sup> and h<sup>+</sup>). A significant enhancement in photodegradation efficiency (97.62%) and efficient bactericidal actions against *E. coli* (14.10 mm inhibition zone) and *S. aureus* (7.45 mm inhibition zone) were observed for higher doped specimen 4% g-C<sub>3</sub>N<sub>4</sub>/St-Mg(OH)<sub>2</sub>.



## 1. INTRODUCTION

In recent years, the continuously developing population and corresponding environmental deterioration caused by a substantial concentration of dyes, heavy metals, and microorganisms have become a serious concern of society and academia.<sup>1</sup> In developing countries, fast developments of industries give rise to industrial effluent removal into water bodies without appropriate precautions, endangering aquatic life and plant life. Specifically, organic dyes are toxic, carcinogenic, and potentially mutagenic, yet researchers do not break them down in the environment because of their structures. The significant human being's exposure to methylene blue (MB) through the food chain may cause dizziness, vomiting, shock, paralysis, cyanosis, and neurological injury in humans.<sup>2,3</sup> In addition, municipal wastewater is discharged into water reservoirs, and pathogens *Staphylococcus aureus* (*S. aureus*) and *Escherichia coli* (*E. coli*) in stagnant water storage pollute water.<sup>4,5</sup>

Hence, researchers must devise effective strategies to degrade hazardous contaminants. So far, different approaches

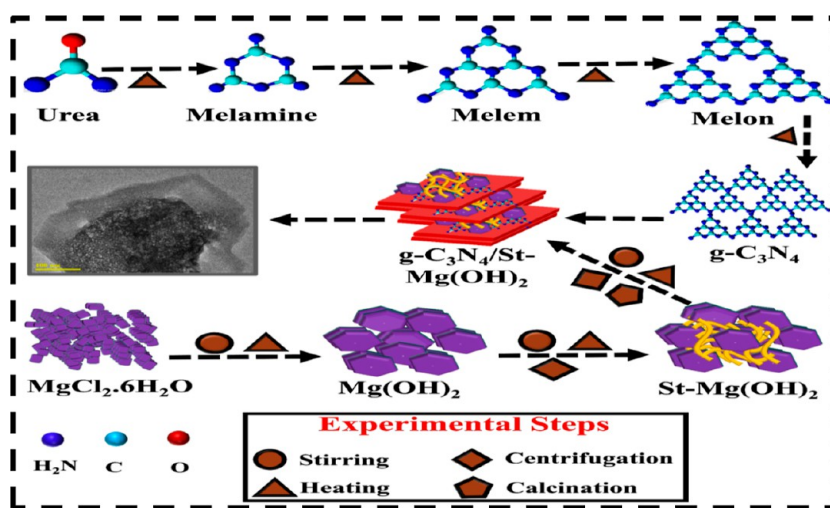
have been employed, including physicochemical and biological approaches. The main issue is the high cost of these traditional treatment techniques on a wide scale. Moreover, these processes vary in effectiveness, cost, and environmental impact. Photocatalytic degradation utilizing renewable UV/sunlight is one of the auspicious water purification methods attributed to its simplicity, low cost, and lack of secondary pollutant production.<sup>6</sup> Photocatalysis begins with photoinduced e<sup>-</sup> and h<sup>+</sup> on the catalyst's surface. Reactive oxygen species (ROS) are produced by employing photocatalysts along water vapors in aerobic circumstances. ROS are the highly reactive species that convert oxygen to O<sub>2</sub>, •O<sub>2</sub><sup>-</sup>, H<sub>2</sub>O<sub>2</sub>, and •OH. Later, these

Received: July 22, 2022

Accepted: October 13, 2022

Published: October 24, 2022





**Figure 1.** Schematic synthesis illustration of  $g\text{-C}_3\text{N}_4$  by pyrolysis and  $g\text{-C}_3\text{N}_4/\text{St-Mg}(\text{OH})_2$  by co-precipitation.

described radicals degrade dyes and disturb bacterial cell membranes.<sup>7,8</sup>

Nanotechnology can be very helpful in supplying clean potable water for a growing population. Various photocatalysts, including oxides, sulfides, and halides, including  $\text{TiO}_2$ ,  $\text{ZnO}$ ,  $\text{WO}_3$ ,  $\text{MoS}_2$ ,  $\text{ZnS}$ ,  $\text{In}_2\text{S}_3$ ,  $\text{AgCl}$ , and  $\text{BiOI}$ , have been reported for hazardous pollutant remediation. However, the practical uses are restricted by swift electron–hole pair recombination, low quantum yields, and incredibly weak response to visible light.<sup>9</sup> Metal hydroxide-based nanomaterials such as  $\text{Ca}(\text{OH})_2$  and  $\text{Mg}(\text{OH})_2$  have been investigated as promising photocatalysts for MB dye decolorization.  $\text{Mg}(\text{OH})_2$  is found to be an eco-friendly, nonpoisonous, noncorrosive, and inexpensive photocatalyst and antibacterial agent for environmental decontamination. These applications are ascribed to the large surface area, biodegradability, and abundant hydroxyl (OH) groups on  $\text{Mg}(\text{OH})_2$  surfaces. Morphology and particle size significantly improve photocatalysis efficiency by permitting charge mobility, band gap modifications, surface reactivity, and adsorption. Various physiochemical methods were adopted to prepare  $\text{Mg}(\text{OH})_2$  nanostructures. Das et al. synthesized  $\text{Mg}(\text{OH})_2$  NPs using a precipitation route for the photo-degradation of MB dye. Liu et al. synthesized  $\text{Mg}(\text{OH})_2$  nanoflowers using a hydrothermal route and exhibited the highest photocatalytic degradation against MB.<sup>10–12</sup> Moreover, positively charged  $\text{Mg}(\text{OH})_2$  also electrostatically attracts bacteria, degrades cell walls' integrity, and ultimately kills pathogens. The antibacterial process was also attributed to ROS formation, which disturbs membrane function and eventually kills bacteria<sup>13,14</sup> Pan et al. prepared  $\text{Mg}(\text{OH})_2$  nanoflakes as an antibacterial agent against *E. coli*. Kumari et al. prepared hexagonal  $\text{Mg}(\text{OH})_2$  sheets that exhibited bactericidal potential against pathogens (*S. aureus* and *E. coli*).<sup>15</sup> The co-precipitation process appears to be environmentally friendly and cost-effective, which allows modifying particle size distribution, surface area, morphology, and agglomeration. The described attributes rely mostly on the reaction temperature, pH, concentration, and type of reagent. Various morphologies, including hexagonal, rods, tubes, or needle-like, have been investigated using the co-precipitation synthesis route.<sup>16</sup> To strengthen the characteristics of nanomaterials, several methods such as tuning and doping with carbon-based substances and polymers have been proposed as a promising

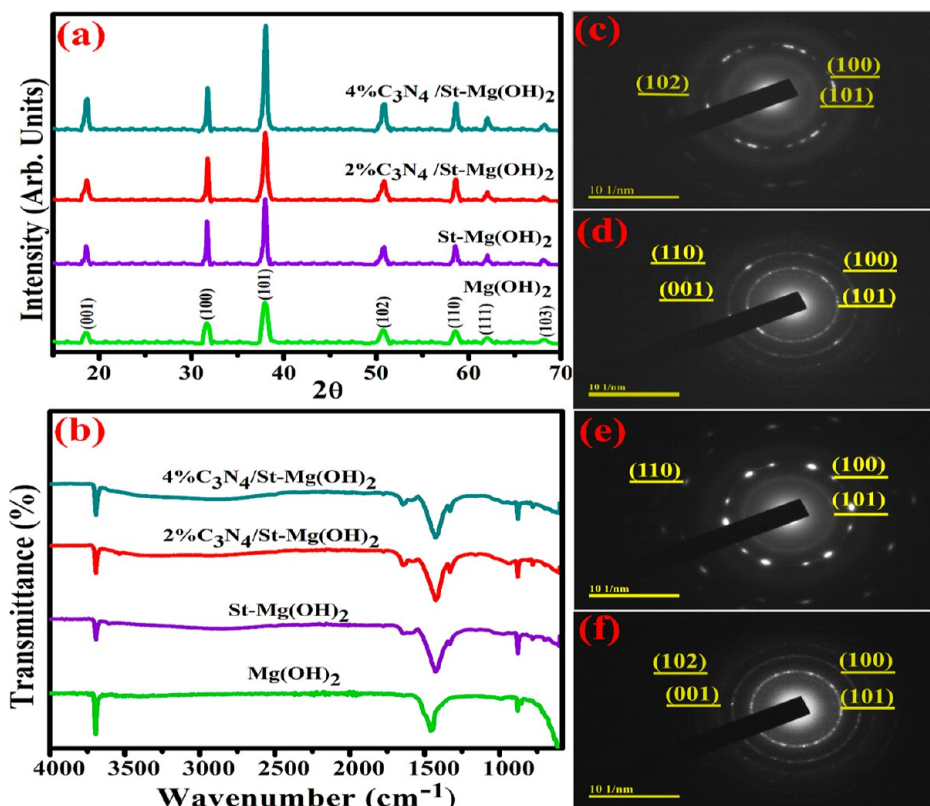
approach. By decreasing recombination probabilities of  $e^-$  and  $h^+$ , doping can significantly enhance photocatalytic activity (PCA).<sup>17,18</sup> Polymeric photocatalysts have been widely used to degrade water contaminants attributed to thermal stability, chemical inertness, and high surface areas. Various natural polymers such as chitin, chitosan, and starch have drawn great attention for environmental remediation.<sup>19,20</sup>

Starch is a highly abundant recyclable substance that has been extensively used for dye degradation because of the OH group in the polymer chain. Anionic starch has a high affinity to cationic dyes such as MB and crystal violet, introducing the carboxylate to improve selective adsorption performance.<sup>21,22</sup> Furthermore, 2D carbon-based material  $g\text{-C}_3\text{N}_4$  has drawn great interest for water splitting as a photocatalyst,  $\text{CO}_2$  reduction, pollutant remediation, and bacterial disinfection owing to its lower  $E_g$ , nontoxicity, physical stability, inexpensiveness, and chemical stability.<sup>23,24</sup> Peng and co-workers synthesized mesoporous  $\text{C}_3\text{N}_4$  to adsorb MB from the aqueous environment.<sup>25</sup> Various metal (Zn, Cu, and Mg) hydroxides have been coupled with the  $g\text{-C}_3\text{N}_4$  as highly effective photocatalysts for degradation performance by reducing charge carriers' recombination rate. Catalysts with required optical characteristics, large surface area, and stability may be produced by fabricating  $g\text{-C}_3\text{N}_4$  with  $\text{Mg}(\text{OH})_2$ . Therefore, to investigate the synergetic effect on dye degradation and bactericidal potential, polymers were doped with  $\text{Mg}(\text{OH})_2$ .<sup>26</sup> The present study demonstrates a co-precipitation technique for synthesizing  $\text{Mg}(\text{OH})_2$  nanoparticles (NPs) and  $g\text{-C}_3\text{N}_4$  layered nanosheets formed around  $\text{St-Mg}(\text{OH})_2$  NPs. The prepared samples have been used as photocatalysts for MB degradation from an aqueous solution. In addition, the bactericidal potential of prepared samples was also examined against *E. coli* and *S. aureus* via the agar diffusion technique.

## 2. EXPERIMENTAL SECTION

**2.1. Materials.** Magnesium chloride hexahydrate ( $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ , 99%), starch ( $(\text{C}_6\text{H}_{10}\text{O}_5)_n$ ), sodium hydroxide (NaOH, 99%), and urea ( $\text{CH}_4\text{N}_2\text{O}$ , 99%) were acquired from Sigma-Aldrich, Germany.

**2.2. Synthesis of  $g\text{-C}_3\text{N}_4$  and  $\text{Mg}(\text{OH})_2$ .** To prepare  $g\text{-C}_3\text{N}_4$ , the usual procedure, that is, urea pyrolysis, was adopted.<sup>27,28</sup> Desired quantity of urea was heated in a furnace



**Figure 2.** (a) XRD spectra of C<sub>3</sub>N<sub>4</sub>/St-Mg(OH)<sub>2</sub>, (b) FTIR analysis, and (c–f) SAED pattern of Mg(OH)<sub>2</sub>, St-Mg(OH)<sub>2</sub>, and (g-C<sub>3</sub>N<sub>4</sub>/St-Mg(OH)<sub>2</sub>).

at 500 °C for 5 h. Afterward, heating produced a schematic for possible intermediates at different temperature ranges forming melon, which was further heated to attain g-C<sub>3</sub>N<sub>4</sub>, as shown in Figure 1. Mg(OH)<sub>2</sub> was synthesized using 0.5 M MgCl<sub>2</sub>·6H<sub>2</sub>O through a co-precipitation technique under constant stirring at 100 °C. NaOH was incorporated as a precipitating agent into stirred solution, and precipitates appeared, indicating the formation of the compound. Afterward, the obtained sediment was centrifuged at 7500 rpm, dried at 200 °C for 12 h, and then crushed to collect the powder. Similarly, to prepare starch–Mg(OH)<sub>2</sub> and starch/g-C<sub>3</sub>N<sub>4</sub>–Mg(OH)<sub>2</sub>, a fixed quantity of starch and various concentrations of g-C<sub>3</sub>N<sub>4</sub> (0.02 and 0.04 wt %) were incorporated into Mg(OH)<sub>2</sub>, as shown in Figure 1.

**2.4. Photocatalysis Assay.** MB dye was selected to evaluate the degradation potential of prepared samples. In a typical procedure, MB solution (5 mg/500 mL) was prepared in DI water and pH (4–12) was adjusted using NaOH and H<sub>2</sub>SO<sub>4</sub> solutions to observe dye degradation in acidic and basic media. Afterward, 10 mg of synthesized samples was incorporated in 30 mL of MB solution and magnetically stirred in the dark to obtain adsorption–desorption equilibrium among photocatalysts and contaminants. Furthermore, the photocatalytic reaction was preceded for 120 min under a Hg lamp (400 W, 20 cm), and ~3 mL of the irradiating suspension was excluded every 30 min. The optimum absorbance of MB was calculated before and after the light irradiation using the following degradation equation.

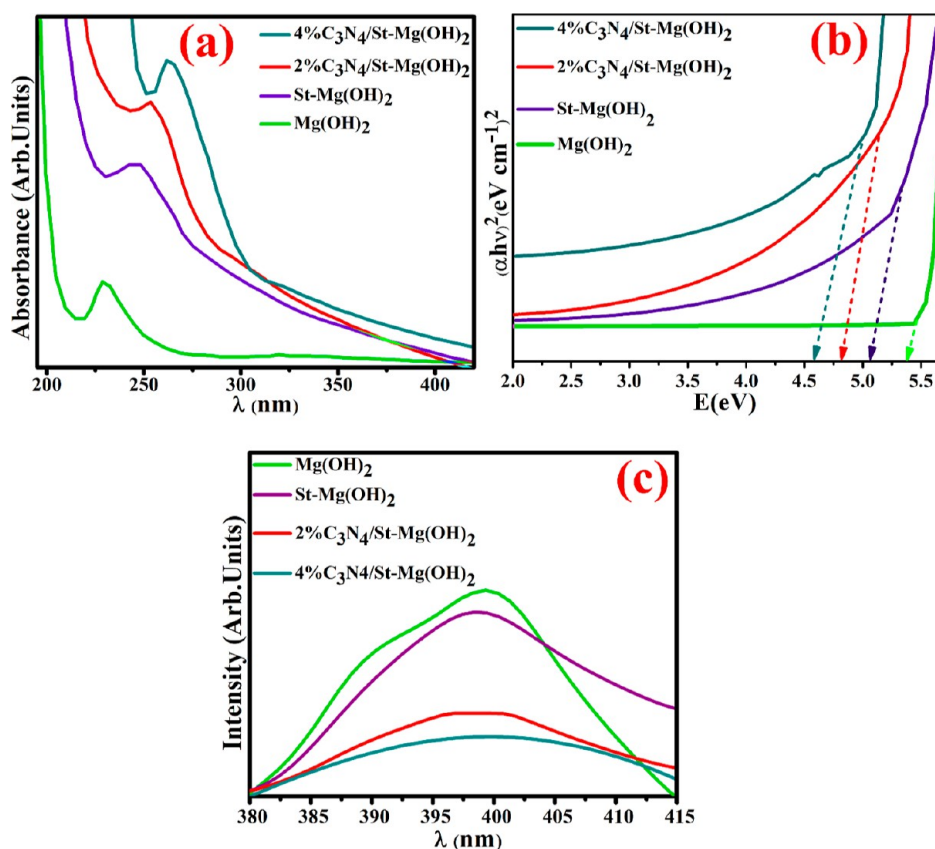
$$\text{Degradation \%} = (C_0 - C_t)/C_0 \times 100 \quad (1)$$

where C<sub>0</sub> and C<sub>t</sub> correspond to MB concentrations before and after light irradiation, respectively.

**2.5. Bactericidal Evaluation.** In Punjab and Pakistan, mastitis-positive sheep milk samples were collected from neighborhood farms and veterinary clinics, refined on 5% blood agar (SBA), and matured at 37 °C for 24 h. Colonies were formed on McFarland on mannitol salt agar (MSA) and MacConkey (MA) in order to isolate Gram-positive (*S. aureus*) and Gram-negative (*E. coli*) bacteria, respectively. Gram staining and pharmacological (catalase and coagulase) methods were used to identify these specific colonies. A sterile cork borer was used under aseptic conditions to drill 6 mm-diameter wells on MSA and MA plates with varying sample doses into each well as the minimum and maximum (0.5 and 1.0 mg/0.05 mL) of pure and g-C<sub>3</sub>N<sub>4</sub>/starch-doped Mg(OH)<sub>2</sub>. DI water (0.055 mL) and ciprofloxacin (0.005 mg/0.05 mL) were used as positive and negative controls, respectively. Furthermore, to determine antibacterial performance, the diameter of the inhibition zone was assessed using a Vernier caliper.

**2.6. Radical Scavenging Assay.** Free radical active species and antioxidant activity of the fabricated nanostructures were evaluated by a modified version of DPPH scavenging experiment. Pristine and g-C<sub>3</sub>N<sub>4</sub>–St-doped Mg(OH)<sub>2</sub> NPs (25–300 μg/mL) were mixed with an equal volume of (0.1 mM) DPPH solution. This mixture was vortexed and incubated for 30 min at ambient temperature in the dark. A standard solution of ascorbic acid was employed as a reference sample. The degradation of DPPH solution (λ = 517 nm) was employed to calculate the scavenging rate (%) of each sample using eq 2

$$\text{Scavenging rate (\%)} = A_0 - A_1/A_0 \times 100 \quad (2)$$



**Figure 3.** (a) Absorption spectra of C<sub>3</sub>N<sub>4</sub>/St-Mg(OH)<sub>2</sub> and (b)  $E_g$  and (c) PL spectra of synthesized samples.

Here,  $A_0$  and  $A_1$  represent control absorbance and standard absorbance, respectively.

**2.7. Material Characterization Techniques.** XRD with Cu K $\alpha$  radiation ( $\lambda = 0.0154$  nm and  $2\theta = 15\text{--}70^\circ$ ) was utilized to identify the crystal structure and phase of synthesized materials. Fourier transform infrared spectroscopy (FTIR) analyzed functional groups present in materials in the range 4000–580 cm<sup>-1</sup>. Surface topography and interplanar spacing were inspected via energy-dispersive X-ray spectrometry and high-resolution transmission electron microscopy (HR-TEM, JEOL JEM 2100F). Absorbance spectra were determined using a UV–vis spectrophotometer (Genesys 10S) ranging from 195 to 420 nm. To elucidate photogenerated electron–hole pairs, recombination of prepared photocatalysts PL was employed.

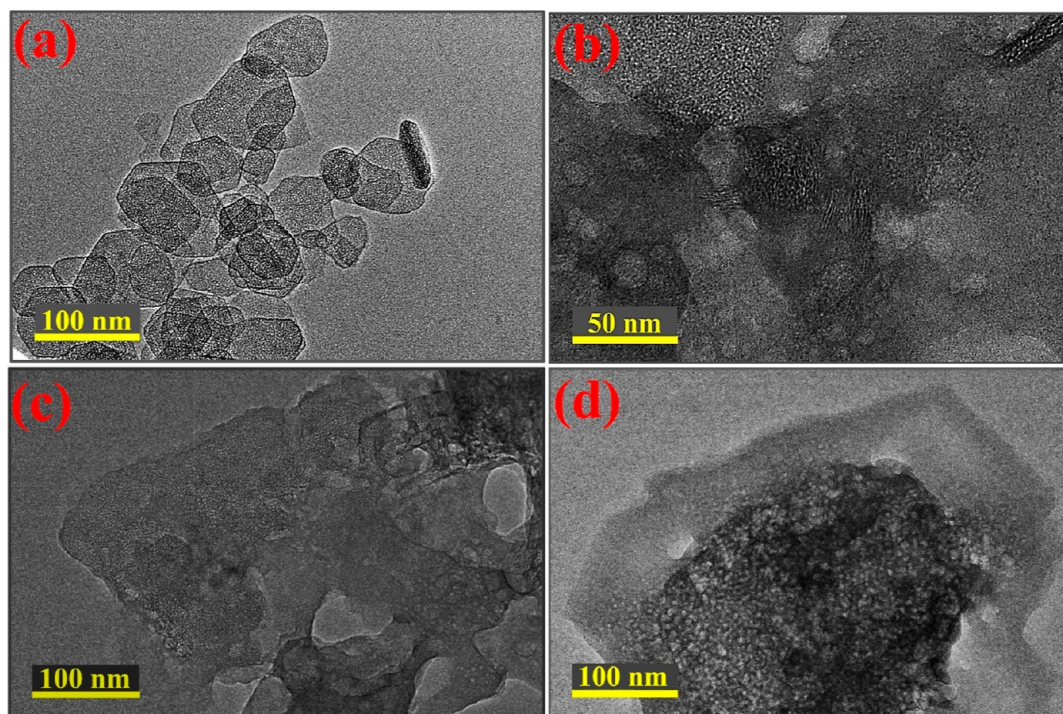
### 3. RESULTS AND DISCUSSION

The phase composition, crystallinity, and lattice structure of Mg(OH)<sub>2</sub> and g-C<sub>3</sub>N<sub>4</sub>/St-Mg(OH)<sub>2</sub> were identified via XRD in the 10–70° range, Figure 2a. The peaks at 18.5° (001), 31.7° (100), 37.9° (101), 50.7° (102), 58.6° (110), 62° (111), and 68° (103) correspond to a hexagonal Mg(OH)<sub>2</sub> crystal reported in JCPDS no. 7–239. The sharp peaks confirm crystallinity, while broad peaks indicate the small particle size of Mg(OH)<sub>2</sub>.<sup>29</sup> Furthermore, upon starch doping, crystallinity has been increased, revealing that Mg(OH)<sub>2</sub> crystals enhanced the crystallization by polymer matrixes.<sup>13,30</sup> Eventually, all the peaks were identified in pure and doped Mg(OH)<sub>2</sub> diffractograms without impurities. To access functional groups, FTIR spectral analysis was carried out, and related patterns were plotted among 4000–500 cm<sup>-1</sup>, as illustrated in Figure 2b.

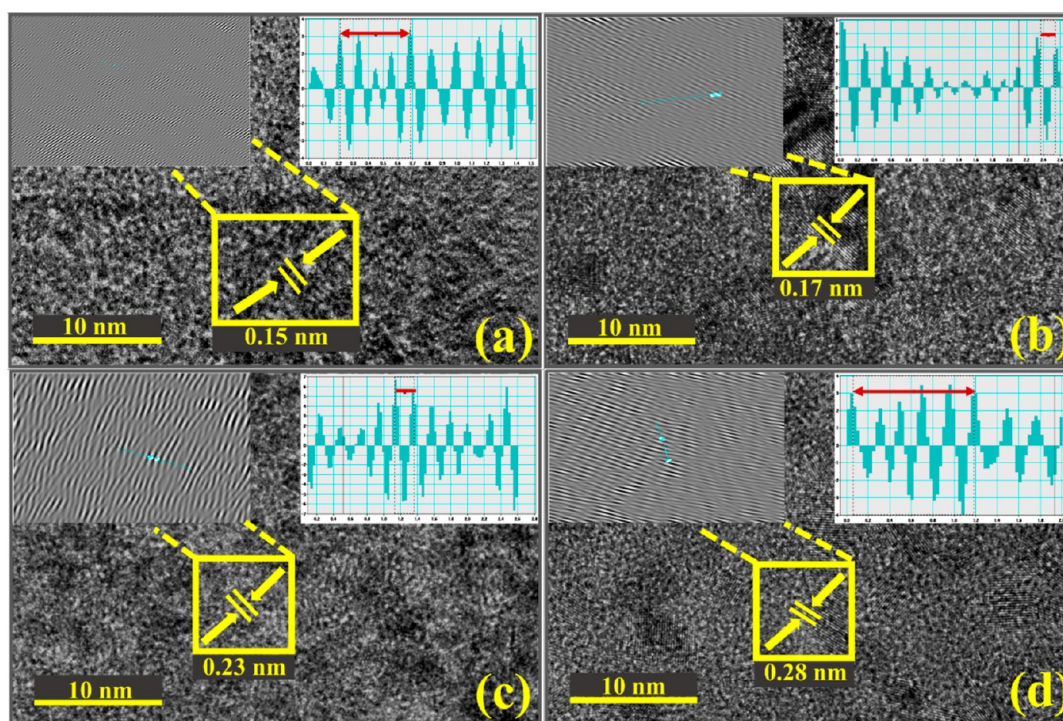
Pure Mg(OH)<sub>2</sub> NP transmittance spectra were found around 3699, 1450, and 867 cm<sup>-1</sup>, indicating O–H, C–O, and Mg–O/Mg–OH stretching vibrations correspondingly (see Figure 2b).<sup>31–33</sup> With starch doping, the transmittance peak appears at 1640 cm<sup>-1</sup>, corresponding to intramolecular hydrogen bonding, indicating the existence of starch on the Mg(OH)<sub>2</sub> surface, while there was no discernible shift upon g-C<sub>3</sub>N<sub>4</sub> doping.<sup>34,35</sup> The selected area electron diffraction (SAED) pattern was observed by directing electron beams on the sample lattice, revealing discrete diffraction rings and the crystalline structure of the synthesized samples, as illustrated in Figure 2c–f. Figure 2c,d reveals the polycrystalline nature of pure and St-Mg(OH)<sub>2</sub> NPs.<sup>36</sup> Figure 2e,f shows that upon 2% g-C<sub>3</sub>N<sub>4</sub> doping, polycrystalline nature with a few bright spots appeared, while with 4% g-C<sub>3</sub>N<sub>4</sub> doping, bright spots disappeared and displayed concentric circles.

A UV–vis spectrophotometer was employed to access optical characteristics in the  $\lambda = 200\text{--}430$  nm range. As illustrated in Figure 3a, the optical absorption of Mg(OH)<sub>2</sub> indicates strong absorption peaks around 230 nm.<sup>37</sup> Absorption intensity mainly relies on particle size, charge carriers' concentration, and dielectric characteristics of the surrounding medium.<sup>38</sup> Upon doping, the absorption band is shifted toward the higher wavelength (230–260 nm) accompanied by the red shift. According to Figure 3b representation of Tauc's relation, the estimated band gap energy ( $E_g$ ) of the Mg(OH)<sub>2</sub> sample was computed.  $E_g$  for Mg(OH)<sub>2</sub> was measured to be 5.4 eV,<sup>13</sup> and it was reduced to 4.6 eV for St/g-C<sub>3</sub>N<sub>4</sub>-Mg(OH)<sub>2</sub>.

PL was carried out to elucidate photogenerated electron–hole pair recombination of prepared photocatalysts presented



**Figure 4.** (a–d) TEM micrographs of (a)  $\text{Mg}(\text{OH})_2$  NPs, (b)  $\text{St-Mg}(\text{OH})_2$  NPs, and (c,d) 2 and 4%  $\text{g-C}_3\text{N}_4$ -loaded  $\text{St-Mg}(\text{OH})_2$  nanosheets.



**Figure 5.** (a–d) HR-TEM images of pure and doped synthesized samples.

in Figure 3c. The strong and broad emission band was observed around 400 nm which was related to oxygen vacancies in  $\text{Mg}(\text{OH})_2$ .<sup>15,31</sup>  $\text{St-Mg}(\text{OH})_2$  exhibits a decrease in emission intensity, while upon  $\text{g-C}_3\text{N}_4$  doping, intensity reduction resulted in improving electron–hole separation due to the junction formed between the  $\text{Mg}(\text{OH})_2$  and  $\text{g-C}_3\text{N}_4$ .<sup>26</sup> These results reveal the enhanced degradation performance. Moreover, the emission band shape and position are the same

except for intensity. This may be due to the same conditions under which all the nanomaterials were synthesized.

The morphology and surface topography of prepared samples were examined using TEM. Figure 4a reveals the randomly oriented hexagonal  $\text{Mg}(\text{OH})_2$  NPs. Figure 4b depicts the addition of starch and shows interpenetrated agglomeration that may be ascribed to the chemical compatibility between starch and  $\text{Mg}(\text{OH})_2$ . The hydrogen bonding between the exposed OH group on the  $\text{Mg}(\text{OH})_2$

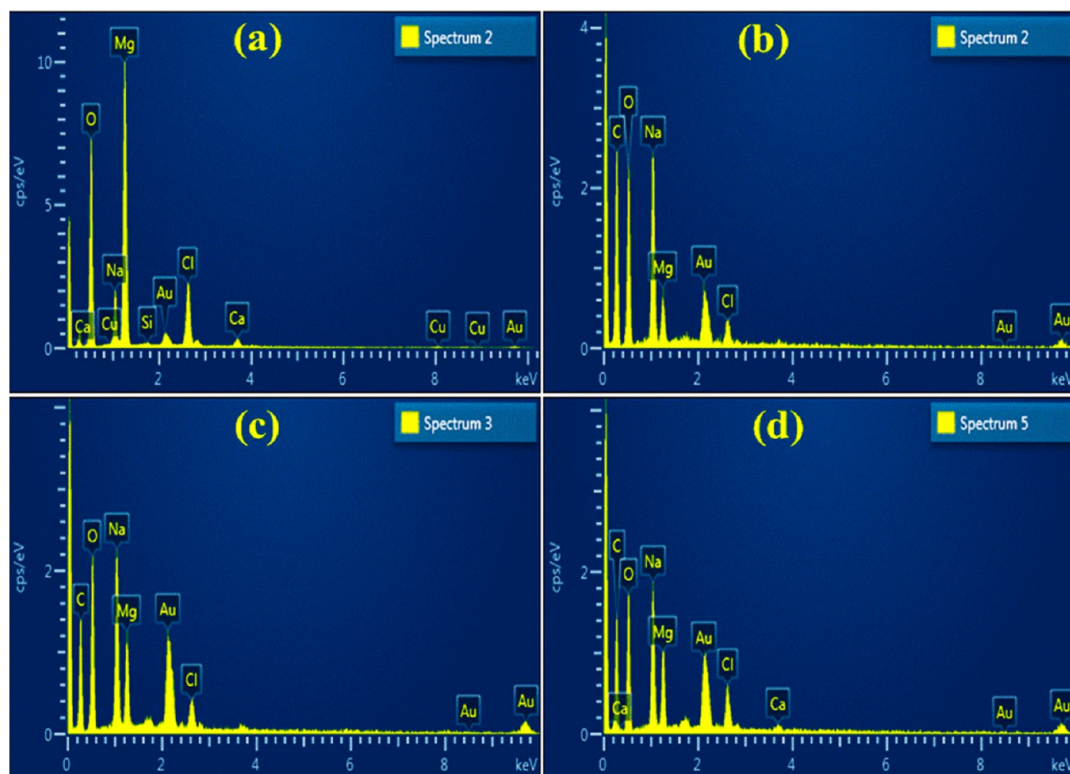


Figure 6. (a–d) EDS pattern of synthesized pure and  $g\text{-C}_3\text{N}_4/\text{St}$ -doped  $\text{Mg}(\text{OH})_2$  samples.

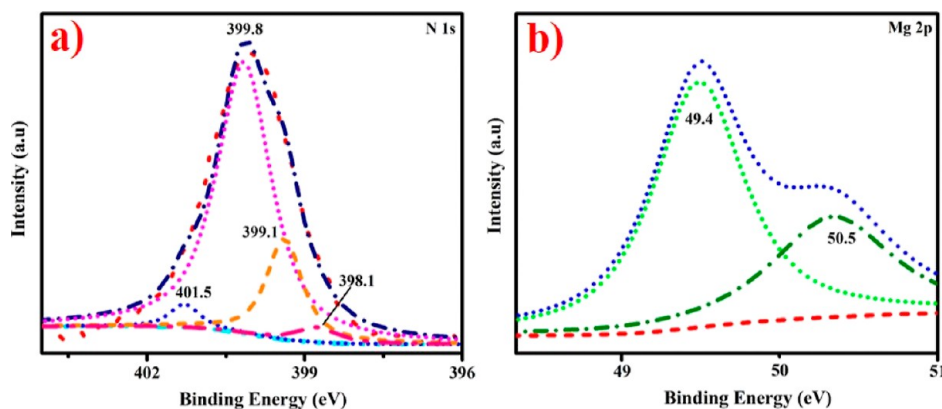


Figure 7. XPS spectra of fabricated  $g\text{-C}_3\text{N}_4/\text{St}$ -doped  $\text{Mg}(\text{OH})_2$  samples (a) N 1s and (b) Mg 2p.

surface and the starch macromolecules may cause the interfacial bonding. Figure 4c,d represents well-dispersed and wrapped St- $\text{Mg}(\text{OH})_2$  NPs by layered structured  $g\text{-C}_3\text{N}_4$  nanosheets. This may contribute to light absorption and the well-contact interaction of reactants with catalytic sites.

The lattice fringes of the synthesized product in the HR-TEM image are separated by an interplanar distance and calculated using Gatan software, as shown in Figure 5a–d. The pristine  $\text{Mg}(\text{OH})_2$  has an interlayer  $d$ -spacing of  $\sim 0.15$  nm (Figure 5a), compatible with the XRD result. Furthermore, adding polymers into  $\text{Mg}(\text{OH})_2$  shows considerable interplanar spacing from  $\sim 0.15$  to  $0.28$  nm (Figure 5c,d).

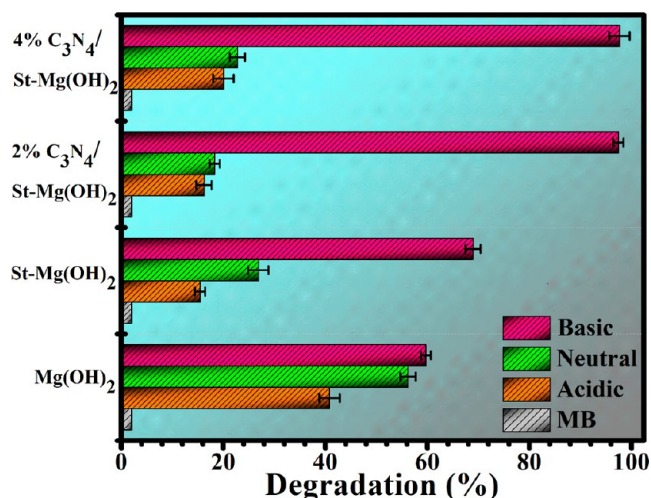
The elemental composition of  $\text{Mg}(\text{OH})_2$  and St/ $g\text{-C}_3\text{N}_4$ - $\text{Mg}(\text{OH})_2$  was assessed via EDS analysis. The results indicate that all elements present in the compound and the peaks of Mg, Cl, and O confirm the synthesis of  $\text{Mg}(\text{OH})_2$  from the  $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$  precursor (Figure 6a–d). In addition, Cu, Na,

and Au peaks were designated to the copper grid, used NaOH during synthesis as a precipitating agent, and gold coating.<sup>39</sup> The carbon peak may be attributed to St/ $g\text{-C}_3\text{N}_4$  dopants.

XPS was used to investigate the elemental and surface composition and binding energy variations of  $g\text{-C}_3\text{N}_4/\text{St}$ -doped  $\text{Mg}(\text{OH})_2$  nanostructures, as represented in Figure 7a,b. The spectra of the N 1s core level indicate a wide peak positioned at 399.1 eV with a shoulder at greater BE. Deconvolution of data for fabricated nanostructures revealed four peaks with energies of 398.1, 399.1, 399.8, and 401.5 eV, separately. The predominant nitrogen signal, focused at 399.1 eV, is derived from the  $\text{sp}^2$ -bonded nitrogen in triazine rings ( $-\text{C}=\text{NH}$ ). The peak at lower BE at 398.1 eV typically alludes to quaternary nitrogen, that is, “graphitic” nitrogen when nitrogen atom substitutes carbon in the graphene layer.<sup>40</sup> The bands at 399.8 and 401.5 eV correspond to tertiary nitrogen ( $\text{N}-(\text{C})_3$ ) and amino functional units containing

hydrogen atoms (C–N–H), respectively.<sup>41,42</sup> The Mg 2p spectrum comprised two peaks: one at 49.4 eV was corresponding to Mg(OH)<sub>2</sub> and MgCO<sub>3</sub>,<sup>43</sup> while other at 50.5 eV was related to MgO.<sup>44–47</sup>

The photodegradation rate of organic dyes is significantly influenced by several parameters such as pH, concentration of dyes, photocatalyst particle size and its concentration, reaction temperature, and light intensity.<sup>48</sup> The pH alternation may shift the valence band (VB) and conduction band (CB) redox potentials and affect the interfacial charge transfer. The pH effect on PCA is generally attributed to electrostatic interaction among charges on the surface of the catalyst and dye molecule.<sup>49</sup> Figure 8 represents that initially, MB was not



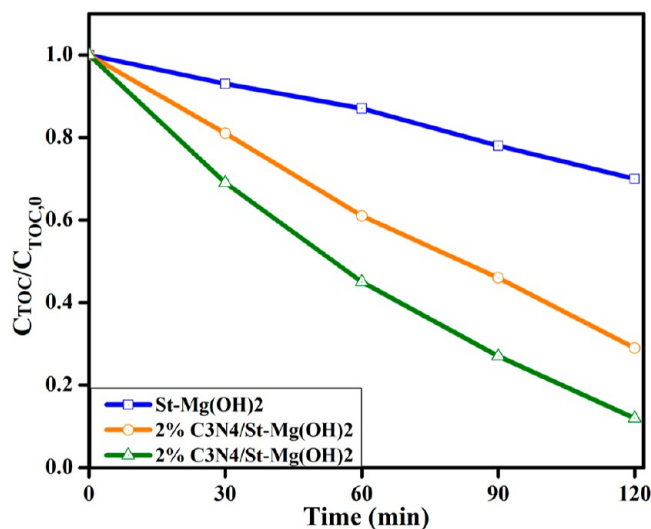
**Figure 8.** Degradation efficiency (%) of prepared pristine and g-C<sub>3</sub>N<sub>4</sub>-St-doped Mg(OH)<sub>2</sub> samples.

decolorized under light without catalysts, implying that MB has relative stability under irradiation. Therefore, MB dye was degraded with un-doped and doped Mg(OH)<sub>2</sub> photocatalysts showing 40.82, 15.45, 16.22, and 20.04% in acidic medium (pH = 4), 56.17, 26.87, 18.28, and 22.76% in neutral medium (pH = 7), and 59.69, 68.93, 97.42, and 97.62% in basic medium (pH = 12) correspondingly. It is clear that the pH degradation rate also increases because high pH encourages cationic MB dye adsorption on the catalyst surface. In the acidic medium, the positively charged Mg(OH)<sub>2</sub> catalyst tended to oppose cationic MB dye adsorption. In an alkaline medium, the catalyst's surface acquires a negative charge; therefore, dye degradation rate is enhanced. Similarly, the St-Mg(OH)<sub>2</sub> photocatalyst shows improved cationic dye degradation because in an alkaline medium, hydroxide ions could more readily attract starch and even ionize its protons, acquiring negative charges for starch molecules. Additionally, in higher doped specimen g-C<sub>3</sub>N<sub>4</sub>/St-Mg(OH)<sub>2</sub>, superior photocatalytic efficacy can be ascribed to more active sites to enhance reaction provided by larger surface area.<sup>26</sup> Ge et al. synthesized g-C<sub>3</sub>N<sub>4</sub>/MgO nanosheets and found surface areas of 110.1 and 8.1 m<sup>2</sup>/g for MgO and g-C<sub>3</sub>N<sub>4</sub>, respectively. The nanosheets of g-C<sub>3</sub>N<sub>4</sub>/MgO showed a synergetic effect for both cationic (MB) and anionic (methyl orange) dye degradation due to an increase in surface area.<sup>50</sup>

In this work, the results were compared with Degussa P25, and optimum MB dye degradation of 97.62% was observed for 4% g-C<sub>3</sub>N<sub>4</sub>/St-Mg(OH)<sub>2</sub>. Degussa (Evonik) P25 is an

extensively used titania photocatalyst that consists of 75% anatase and 25% rutile. Tichapondwa et al. performed MB degradation using Degussa (Evonik) P25 and obtained 81.4% degradation efficiency.<sup>51,52</sup>

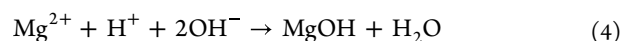
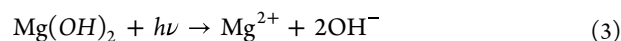
A total organic carbon (TOC) assessment of treated water was conducted to estimate the dye (MB) degree of mineralization. The study was performed on St-Mg(OH)<sub>2</sub> and g-C<sub>3</sub>N<sub>4</sub>/St-Mg(OH)<sub>2</sub> (2 and 4%) using varying time intervals up to 120 min. This analysis (Figure 9) demonstrated



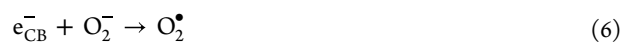
**Figure 9.** Variations in the TOC content of MB solution during photodegradation.

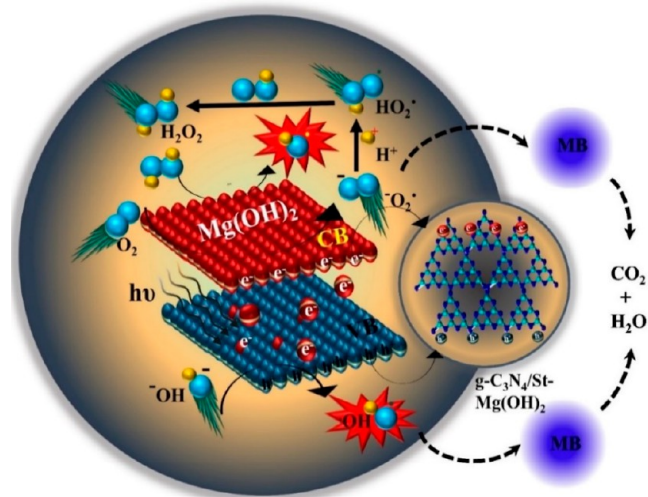
that the TOC of the MB solution treated with fabricated compounds under visible light irradiation reduced continuously with reaction time, and a significant amount of mineralization of the dye was found after 120 min in 4% g-C<sub>3</sub>N<sub>4</sub>/St-Mg(OH)<sub>2</sub>. The study also indicates that the dye may be converted into various intermediate forms and that degradation of the dye after 120 min may culminate in its total mineralization.

In general, photocatalysis involves adsorption–desorption, e<sup>−</sup> h<sup>+</sup> pair production, electron pair recombination, and chemical reaction.<sup>53,54</sup> The possible mechanism was described as follows to determine the degradation performance of Mg(OH)<sub>2</sub> NPs and the impact of ROS on the degradation of MB dye. PCA begins with irradiation of photon energy ( $h\nu \geq E_g$ ) to excite electrons (e<sup>−</sup>) from the lower VB to the upper CB, leaving behind a hole (h<sup>+</sup>) in the VB (Figure 10). Under aqueous conditions, by the dissolution reabsorption process, Mg<sup>2+</sup> species are generated (eq 2). MgOH formation is faster compared to ionization of Mg(OH)<sub>2</sub> (eq 3).

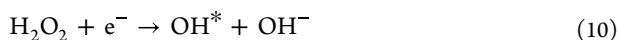
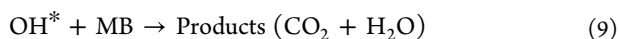
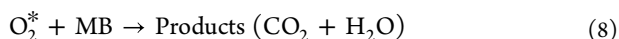
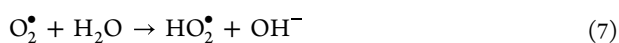


Later, these formed e<sup>−</sup> and h<sup>+</sup> react with O<sub>2</sub><sup>−</sup> and OH<sup>−</sup> to generate ROS, superoxide (O<sub>2</sub><sup>−</sup>), and hydroxyl (OH<sup>\*</sup>) radicals that can strongly oxidize organic dye (MB) into nonhazardous products (CO<sub>2</sub> and H<sub>2</sub>O).





**Figure 10.** Photocatalysis mechanism of undoped and g-C<sub>3</sub>N<sub>4</sub>-St-doped Mg(OH)<sub>2</sub> nanostructures.

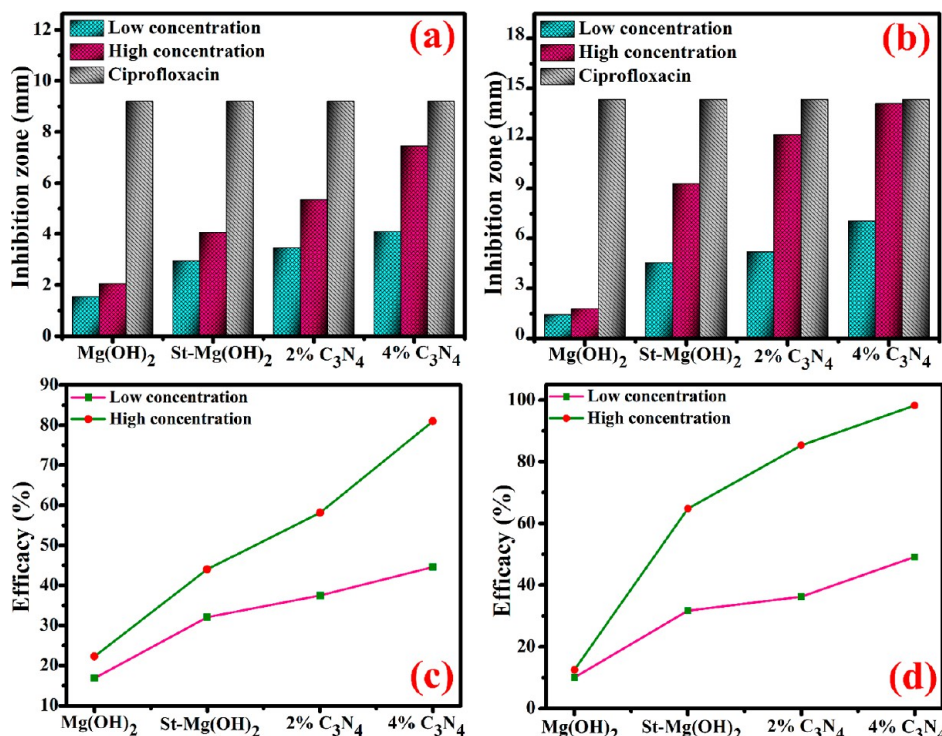


The results denoted that, in the photocatalytic oxidation of MB, OH\* performs the main role compared to O<sub>2</sub>\*.<sup>13</sup>

The bactericidal behavior of pristine and doped Mg(OH)<sub>2</sub> nanostructures against *S. aureus* and *E. coli* was determined via the agar diffusion technique. At minimum and maximum

concentration, significant zones of inhibition were seen as 1.55–4.10 mm and 2.05–7.45 mm for *S. aureus* and 1.45–7.05 mm and 1.80–14.10 mm for *E. coli* (Figure 11a,b). The results have been compared through DI water (0 mm) as the –ve control and ciprofloxacin as the +ve control with inhibitory areas (9.20 mm) for *S. aureus* and (14.35 mm) *E. coli*, as illustrated in Table 1. According to these observations, Mg(OH)<sub>2</sub> NPs show improved bactericidal performance toward *S. aureus* than *E. coli* because of the cell wall difference. This observation is attributed to the cell wall difference between G + ve and G – ve pathogens.<sup>11</sup> Upon starch and g-C<sub>3</sub>N<sub>4</sub> doping into Mg(OH)<sub>2</sub>, bactericidal potential enhanced toward *E. coli* gradually attributed to the thicker cell wall of *S. aureus* than *E. coli*.<sup>55</sup> Furthermore, (Figure 11c,d) shows that the % age efficacy of Mg(OH)<sub>2</sub> enhances upon doping for *S. aureus* as 16.84–44.56% and 22.28–80.97% at a minimum and higher concentrations, respectively. Similarly, the % age efficacy for *E. coli* also increases as 10.10–94.12% and 12.54–98.25% correspondingly.

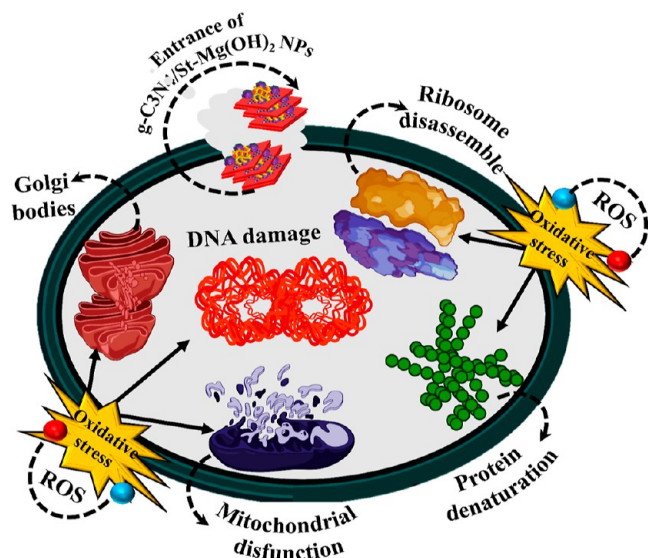
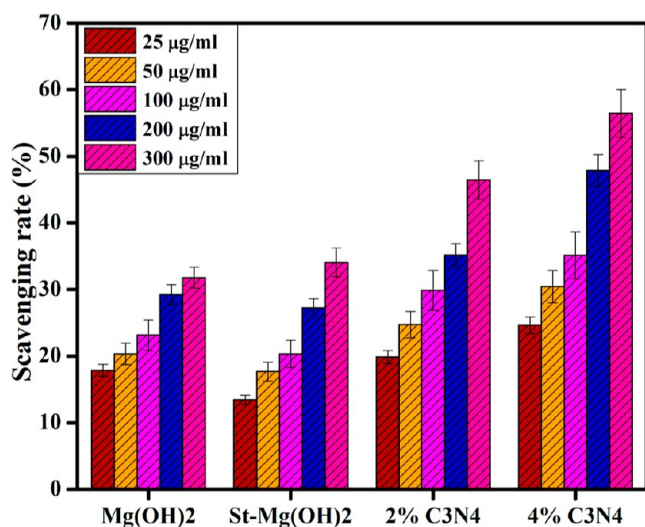
The mechanistic antibacterial activity of the hexagonal Mg(OH)<sub>2</sub> is attributed to different types of NPS–bacteria interactions, including electrostatic interaction between the bacterial cell and NP surface, ROS generation attributed to oxygen vacancies on the surface of Mg(OH)<sub>2</sub> by redox reaction, and bacterial cell inactivation by cell membrane disruption and internal cellular leakage like; DNA, cytoplasm, and ribosomes resulting bacterial cell death, as shown in Figure 12.<sup>11</sup> In addition, the Mg(OH)<sub>2</sub> surface becomes alkaline because, in an alkaline environment, O<sub>2</sub><sup>–</sup> and OH<sup>–</sup> layers have high chemical stability.<sup>56</sup> DPPH scavenging was used to examine antioxidant effects of active radical species (Figure 13). Antioxidant activity of compounds is interrelated with their potential to transfer hydrogen or electrons to the DPPH



**Figure 11.** In vitro bactericidal potential of pure and g-C<sub>3</sub>N<sub>4</sub>/St-loaded Mg(OH)<sub>2</sub> against (a) *S. aureus* (b) *E. coli* and corresponding efficacy (%) for these bacteria in (c,d).

Table 1. Antibacterial Activity of Pristine and g-C<sub>3</sub>N<sub>4</sub>/St-Doped Mg(OH)<sub>2</sub>

Samples	<i>S. aureus</i> inhibition region (mm)		<i>E. coli</i> inhibition region (mm)	
	0.5 mg/50 $\mu$ L	1.0 mg/50 $\mu$ L	0.5 mg/50 $\mu$ L	1.0 mg/50 $\mu$ L
Mg(OH) <sub>2</sub>	1.55 $\pm$ 0.35	2.05 $\pm$ 1.15	1.45 $\pm$ 0.24	1.80 $\pm$ 0.46
St-Mg(OH) <sub>2</sub>	2.95 $\pm$ 0.78	4.05 $\pm$ 1.27	4.55 $\pm$ 1.97	9.30 $\pm$ 3.12
2% g-C <sub>3</sub> N <sub>4</sub> /St-Mg(OH) <sub>2</sub>	3.45 $\pm$ 1.34	5.35 $\pm$ 2.16	5.20 $\pm$ 2.10	12.25 $\pm$ 4.22
4% g-C <sub>3</sub> N <sub>4</sub> /St-Mg(OH) <sub>2</sub>	4.10 $\pm$ 1.76	7.45 $\pm$ 2.94	7.05 $\pm$ 2.51	14.10 $\pm$ 4.92
Ciprofloxacin	9.20 $\pm$ 0.0	9.20 $\pm$ 0.0	14.35 $\pm$ 0.0	14.35 $\pm$ 0.0
DI water	0 $\pm$ 0.0	0 $\pm$ 0.0	0 $\pm$ 0.0	0 $\pm$ 0.0

Figure 12. Bactericidal potential of pure and g-C<sub>3</sub>N<sub>4</sub>/St-loaded Mg(OH)<sub>2</sub>.Figure 13. Antioxidant potential of pure and g-C<sub>3</sub>N<sub>4</sub>/St-loaded Mg(OH)<sub>2</sub>.

free radical, resulting in stable diamagnetic compounds. All prepared samples exhibited a dose-dependent behavior; 4% g-C<sub>3</sub>N<sub>4</sub>/St-Mg(OH)<sub>2</sub> displayed the highest scavenging performance up to 56.45% at a 300  $\mu$ g/mL concentration and scavenged DPPH radicals by donating hydrogen atoms. The formation of highly reactive  $\cdot$ OH and  $\cdot$ O<sub>2</sub> radical species can interact with DPPH free radicals and result in bacterial cell death, which is highly correlated with the standard (ascorbic acid).

The comparison table of MB dye degradation and bactericidal action with the literature is shown in Table 2.

#### 4. CONCLUSIONS

Novel pristine and g-C<sub>3</sub>N<sub>4</sub>/St-doped Mg(OH)<sub>2</sub> nanostructures were synthesized using the co-precipitation technique to examine photocatalytic and antibacterial activity. Several structural and optical characterization techniques were employed to examine the characteristics of synthesized nanostructures. The XRD pattern endorsed the hexagonal crystal structure of the pristine sample and improved crystallinity by incorporating dopants. Meanwhile, the presence of functional groups in synthesized NPs and metal hydroxide spectra was identified at 867  $\text{cm}^{-1}$  using FTIR. TEM micrographs revealed hexagonal morphology of Mg(OH)<sub>2</sub> NPs. Polymers doping g-C<sub>3</sub>N<sub>4</sub> and nanosheets wrapped around St-Mg(OH)<sub>2</sub> were investigated. Pure and doped Mg(OH)<sub>2</sub> exhibited interlayer spacing (0.15–0.28 nm), consistent with HR-TEM. EDS confirmed the elemental composition of pure and doped samples. UV spectra revealed that the red shift with doping led to gradually reduce  $E_g$  (5.4–4.6 eV). The efficiency of MB dye degradation implies the improved light-driven PCA upon polymer doping into Mg(OH)<sub>2</sub>. Moreover, the agar diffusion technique assessed the bactericidal behavior of synthesized samples against *S. aureus* and *E. coli* pathogens. The optimum dye degradation efficiency (97.62%) and bactericidal solid action against *E. coli* (14.10 mm inhibition zone) was observed with incorporation of g-C<sub>3</sub>N<sub>4</sub> into St-Mg(OH)<sub>2</sub>. This research indicates that Mg(OH)<sub>2</sub> NPs with polymer doping might be environmentally and economically effective against pathogens and the dye degrader.

Table 2. Comparison Table of MB Degradation and Bactericidal Action with the Literature

materials	synthesis routes	MB degradation	antibacterial	references
Mg(OH) <sub>2</sub> sheets	precipitation	55%		13
Mg(OH) <sub>2</sub> flakes	co-precipitation		88% inhibition against <i>E. coli</i>	57
Mg(OH) <sub>2</sub> NPs	precipitation		6.5 mm <i>S. aureus</i> inhibition	58
4% C <sub>3</sub> N <sub>4</sub> /St-Mg(OH) <sub>2</sub>	co-precipitation	97.62%	<i>S. aureus</i> inhibition (7.45 mm) and <i>E. coli</i> (14.10 mm)	present work

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### Notes

The authors declare no competing financial interest.

Data available on demand.

## ACKNOWLEDGMENTS

The authors are thankful to HEC, Pakistan via National Research Program for Universities (NRPU) project-20-17615.

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