



A systematic review on the photocatalytic efficiency of g-C₃N₄ based photocatalyst for antibiotic degradation under LED light irradiation

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ABSTRACT

In recent years, photocatalytic processes have gained attention as one of the effective methods for removing resistant pollutants, including antibiotics. This study, conducted as a systematic review, examines the studies regarding the use of graphitic carbon nitride (g-C₃N₄) based photocatalysts for the degradation of antibiotics under LED light irradiation. The process's Operational parameters were analyzed, including antibiotic concentration, photocatalyst dosage, pH, and reaction time. Factors such as light intensity, reaction kinetics, and the photocatalyst's recyclability were also investigated. Finally, 60 eligible studies were ultimately selected for this systematic review. Additionally, 56 % of the studies demonstrated antibiotic removal efficiency of over 90 %. The pH range that showed the highest efficiency was between 6 and 7. 34 % of the studies examined antibiotic concentrations in the range of 10–20 mg/L. The highest photocatalyst dosage was found in two ranges: 0.4–0.5 g/L (22 % of studies) and 0.8–1 g/L (25 % of studies). The most stable catalyst, which showed the least reduction in efficiency during reuse, exhibited only a 2.3 % decrease in efficiency from cycles 1 to 4. Overall, this review revealed that optimization of operational parameters, along with the integration of graphitic carbon nitride (g-C₃N₄) into composite photocatalysts via elemental doping or structural engineering, markedly enhances antibiotic degradation performance. The distinctive band-gap characteristics of g-C₃N₄ promote efficient visible-light harvesting and charge carrier separation, whereas LED irradiation offers a stable and energy-efficient light source ideally aligned with the photocatalytic response of g-C₃N₄-based systems.

1. Introduction

One of the major issues facing modern humanity is environmental pollution, which has dramatically grown in recent years due to the rapid expansion of industry [1,2]. Human health has been seriously threatened by the untreated release of wastewater containing organic compounds into the environment. Pharmaceutical chemicals that enter water resources and ecosystems as micro-pollutants are one of the primary causes of this effluent [3]. Among pharmaceutical chemicals, antibiotics comprise a significant portion of the medications used to treat illnesses in humans and animals. These wastewaters come from a variety of sources, including hospitals, homes, and pharmaceutical factories

[4,5]. As a result, many antibiotics, particularly those that are used excessively, wind up in wastewater [6]. Antibiotics slowly build up in the environment because of their extended half-life and high water solubility [7,8]. These substances can suppress microorganisms and have an impact on bacterial metabolism [9]. Antibiotics are currently regarded as emerging pollutants because, even at low environmental concentrations, they have substantial ecological consequences [10,11]. Antibiotic-resistant bacteria and related genes may emerge due to the persistent presence of antibiotics in the environment. Living things are thought to be seriously threatened by this issue [9,12,13]. Antibiotics are useful in the treatment of many illnesses, but they can also have serious side effects. For instance, metronidazole (MTZ) can be

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carcinogenic and fatal to living things while having a wide range of bacterial activity and being very soluble in water. Moreover, MTZ is scarcely broken down by UV radiation and is highly stable in aquatic settings [14,15]. Because of their propensity to cause cancer and the emergence of antibiotic resistance, sulfonamides (SAs) pose major health risks to people. Levofloxacin (LVX), the primary medication used to treat respiratory infections, can be found in aquatic habitats because of the body's inadequate metabolism. An antibiotic belonging to the quinolone class, ofloxacin (OFL), is used to treat bacterial infections in the skin, kidneys, and respiratory system, among other areas of the body. Low amounts of this medication might linger in water for long periods because the body does not completely metabolize it, excreting just 20 to 80 % of it in its active form [16–18]. Ceftriaxone (CTX) is used to treat bacterial illnesses caused by several kinds of bacteria, both Gram-positive and Gram-negative. Even in low quantities, this antibiotic can cause antibiotic resistance because of its extensive usage in both medical and veterinary care [12]. Metronidazole, ofloxacin, levofloxacin, and ceftriaxone are just examples of antibiotics that, in addition to treating diseases, have significant environmental and health impacts. Antibiotic-containing water and wastewater are now treated using a variety of techniques, such as adsorption, membrane filtration [1,9], biological treatment [6,19], coagulation, and flocculation [18]. These approaches, however, have limitations in real-world applications and disadvantages, including low efficacy, high operating costs, the possibility of secondary contamination, and inefficiency at low pollutant concentrations [20,21]. Therefore, to manage these kinds of contaminants, more sophisticated techniques are required. Pharmaceutical substances can be extracted from aqueous solutions using advanced oxidation processes (AOPs), such as ozonation [22], photo-Fenton [23], electro-Fenton processes [24], and photocatalytic reactions [25]. These reactions have the potential to oxidize contaminants or transform them into other innocuous chemical species, depending on the kind and quantity of reactive radical species generated [26]. Since a catalyst may help reactions happen when exposed to light, photocatalysts are now acknowledged as leaders in this sector. Among the numerous benefits of photocatalysts are their low energy needs, high quantum yield, and non-toxicity [27–29]. A suitable photocatalyst should be stable, have a large surface area, and have a low band gap [30]. Currently, one of the most promising techniques for eliminating pharmaceutical substances and dealing with the problem of wastewater treatment, including these compounds, is photocatalytic oxidation [12,31,32]. The following are the phases of the heterogeneous photocatalytic process: (1) Adsorption and trapping of visible light, (2) Electric charge excitation to create electron/hole (e^-/h^+) pairs is the step in the heterogeneous photocatalytic process, (3) Efficient separation of light collision-induced e^-/h^+ couples, and (4) Conducting redox reactions and breaking down organic contaminants on the catalyst surface [33]. Graphitic carbon nitride ($g-C_3N_4$) has garnered a lot of interest as a semiconductor among photocatalysts [34,35]. This material has joined the new generation of visible-light-sensitive photocatalysts due to its low band gap (around 2.7 eV) and strong chemical stability [36,37]. As a cost-effective, environmentally friendly, and secure photocatalyst, $g-C_3N_4$ needs sunshine and visible light to initiate photocatalytic processes. Melamine, urea, and dicyandiamide are inexpensive precursors that may be used in a straightforward thermal condensation process to create this material [13,17]. Furthermore, $g-C_3N_4$ has found extensive use because of its non-toxicity, affordability, chemical, electrical, and thermal stability, as well as its resilience to high pH levels [36,38,39]. In addition to applications where $g-C_3N_4$ is utilized alone, a variety of compounds and structures have been used in conjunction with it. Each of these has contributed unique characteristics to the synthesized composite, including enhanced light absorption, magnetization, decreased recombination of e^-/h^+ pairs, and increased catalytic surface area [27,40]. The light-emitting diode lamp (LED) is one of the light sources employed in research because of $g-C_3N_4$'s ability to absorb visible light. Conventional UV lamps are mostly used for the photocatalytic destruction of

contaminants. These lamps, which include mercury vapor lamps (at low, medium, or high pressures), are challenging to deal with since they require cooling, contain dangerous mercury, have a very limited life-span, and consume a comparatively large amount of energy. Effective use of solar light necessitates a relatively vast area, and the method's major disadvantages include expensive installation costs and restrictions on daylight hours. The development of photocatalytic reactors and the effectiveness of photocatalytic processes have been significantly affected by these aspects. Energy is present in every facet of daily life since it is crucial to the advancement of society [41–43]. Utilizing efficient energy sources to create more cost-effective and efficient processes in a variety of applications is crucial to addressing future energy challenges. Recently, tiny, inexpensive, and eco-friendly LEDs have been developed by solid-state technology. An LED is a semiconductor device that uses a form of electroluminescence to generate light in a specific spectrum. Depending on the settings and makeup of the semiconductor components, LEDs may emit light at various wavelengths [44]. An LED's light output is directly proportional to the current flowing through its active area and is directed. UV LEDs are smaller, require no warm-up time, and can be precisely adjusted for the best purification as compared to conventional lights. Furthermore, the utilization of targeted or combination multi-wavelength sources for various levels of purification is made possible by the availability of certain wavelengths. To maximize their efficacy, LED lamps need a thorough comprehension of photochemical processes, reactor design, and microbial deactivation mechanisms [45,46]. This study intends to systematically review the role of effective parameters in the removal of antibiotics using photocatalysts that incorporate $g-C_3N_4$ in their structure and use LED light as the radiation source. This is done in light of the previously mentioned points, as well as the widespread use of $g-C_3N_4$ in the removal of pollutants, especially antibiotics. The authors believe that this article will help readers better comprehend the photocatalytic processes involving $g-C_3N_4$ by providing a summary of the mechanisms that occur during these processes in the presence of $g-C_3N_4$.

2. Materials and methods

2.1. Search protocol

In this study, a comprehensive search was conducted to identify articles that utilized $g-C_3N_4$ -based photocatalysts for the removal of antibiotics. A search syntax was developed by determining relevant keywords and MeSH terms. The search syntax was reviewed and validated by a specialist. To ensure broader coverage of the literature, the light source (LED) was not included in the keywords at this step. The summary of search syntax used is in Table 1, as follows: (See Figs. 1–12.)

In Supplementary Information (S1), the search syntax is detailed for each database. The search was performed in the databases of ScienceDirect, Web of Science, Scopus, and Google Scholar. After exporting the search results to EndNote, the articles were sorted based on predefined inclusion and exclusion criteria. The timeline for the primary search results obtained was from June 30, 2015, to February 3, 2024.

2.2. Selection criteria and data extraction

Following the completion of the literature searches and receiving the EndNote format output, the results were imported into EndNote 8. During the import process, duplicate articles were removed. In the next step, articles were manually rechecked for duplicates, and other repeated items were also removed. The articles selected for this study are original, relevant articles in English; then, the following items were excluded: articles in other languages, abstracts, conference papers, review articles, books, and unrelated articles. In the next step, the screening of articles was conducted based on the use of the $g-C_3N_4$ photocatalyst in antibiotic removal by reviewing the title and abstract. To ensure consistency and comparability among the selected studies, we

Table 1
summary of search syntax in this systematic review.

↓OR Photocatalytic Photocatalyst	AND⇒	↓OR Graphitic carbon nitride	AND⇒	↓OR Removal Degradation Mineralization	AND⇒	↓OR Anti-Bacterial Agents Compounds, Anti-Bacterial Agents, Bacteriocidal Bactericide	AND⇒	↓OR Wastewater Waste Water Aqueous
Photocatalysis		g-C ₃ N ₄		Decomposition		Agents, Anti- Mycobacterial Antibiotic		Water

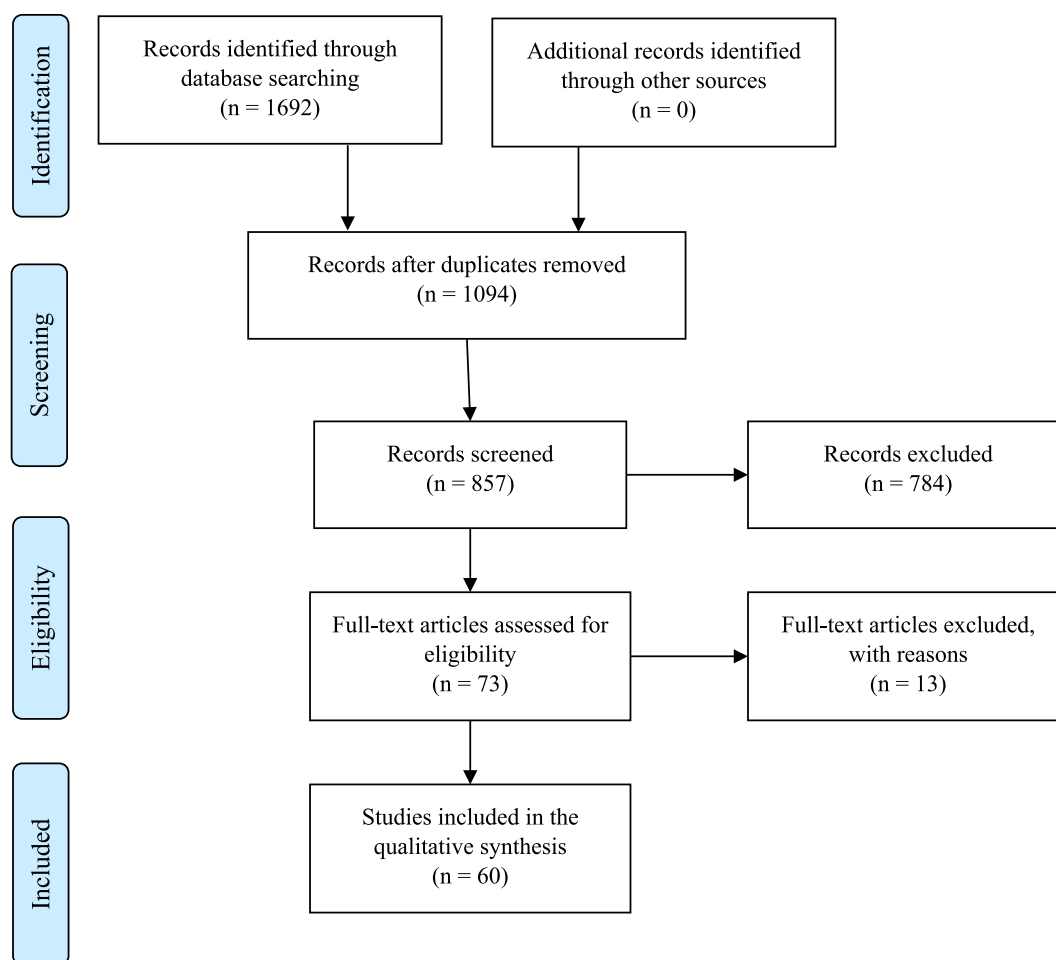


Fig. 1. Description of the PRISMA flow diagram applied to the selection and search of databases.

further restricted the inclusion to articles that employed LED as the irradiation source. This restriction was applied because LED light has gained growing attention in recent years as an energy-efficient alternative to conventional UV or xenon lamps. Accordingly, studies using other irradiation sources were excluded at the full-text screening stage. In the final step, data extraction was carried out based on the criteria examined in the study, which included the following: author name, year of publication, article title, journal name, antibiotics studied in the articles, type of photocatalyst, antibiotic concentration, photocatalyst dosage, pH, contact time, maximum efficiency of degradation, reuse of the catalyst (number of cycles and its efficiency in the final cycle), reaction kinetics, and characteristics of the radiation source. The completed data extraction results are available in Supplementary Information (S2).

3. Results and discussion

3.1. Literature characteristics

The results obtained from the database search are presented as follows: PubMed ($n = 73$), Web of Science ($n = 828$), Scopus ($n = 509$), and Google Scholar ($n = 282$). Consequently, 1692 articles were identified. Following the transfer of these results to EndNote 8 software, duplicate entries were systematically removed, leaving 1094 articles at this stage. An initial screening of the articles was conducted to eliminate those deemed unrelated to the review based on their titles and abstracts. This led to the exclusion of 237 articles due to irrelevance. Ultimately, following the established criteria for this study, 60 articles were selected as the most relevant for inclusion in the systematic review. It is noteworthy that one of the limiting factors for the articles was the light source, with LED being the designated light source for the study; thus, articles utilizing alternative light sources were excluded during the second screening phase. The process of article selection and exclusion is

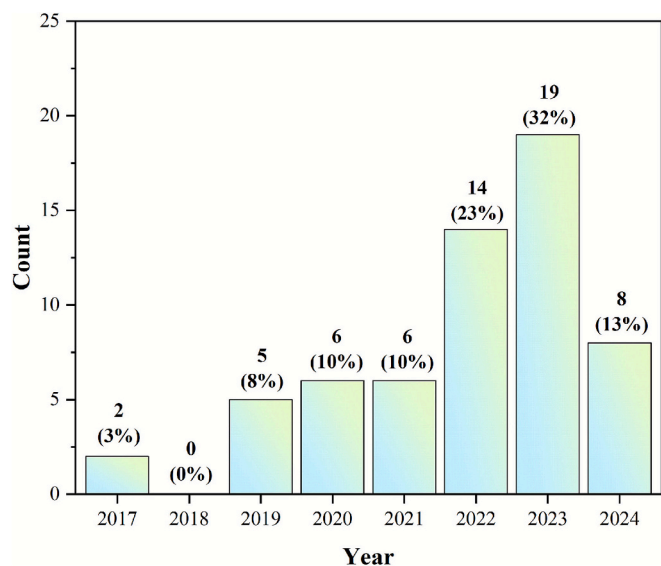


Fig. 2. Number of reported studies per year.

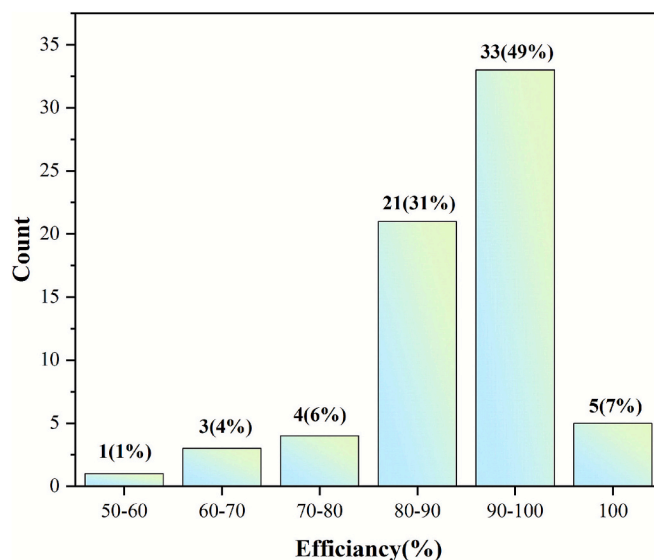


Fig. 4. Maximum efficiency observed for the catalytic reaction.

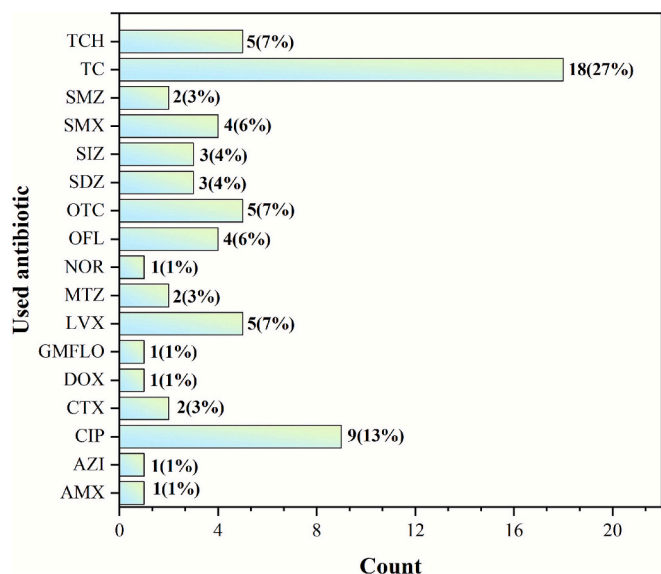


Fig. 3. Distribution of the studied antibiotics.

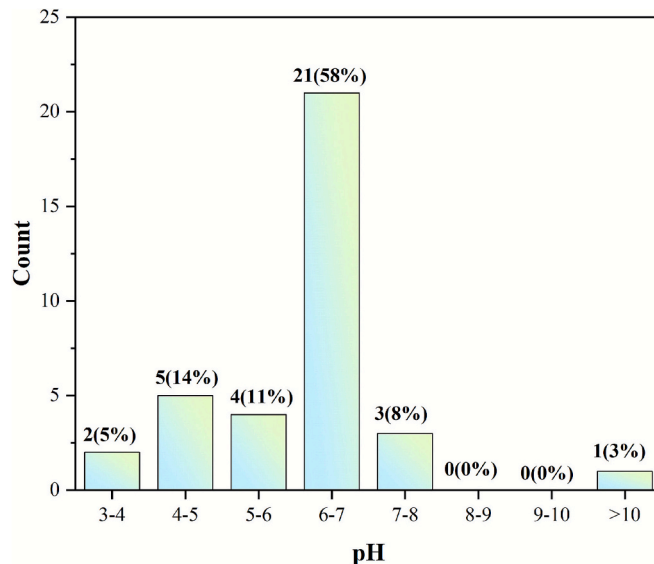


Fig. 5. Investigation of pH distribution in selected studies.

illustrated using the PRISMA flow diagram in Fig. (1).

3.2. Characteristics of the selected studies

The articles included in this study were published between 2017 and 2024. The findings indicate that, before 2017, studies on removing antibiotics using gC_3N_4 -containing photocatalysts did not utilize LED as a radiation source. However, there has been a notable increase in the number of relevant studies over time, culminating in 2023, which accounted for 32 % of the total studies. The distribution of studies across different years is as follows: 2017 ($n = 3$), 2019 ($n = 5$), 2020 ($n = 6$), 2021 ($n = 6$), 2022 ($n = 14$), 2023 ($n = 19$), and 2024 ($n = 8$). As illustrated in Fig. (2), no publications in this field were recorded in 2018. For ease of reference, abbreviations for the antibiotics mentioned in the text are detailed in Table 2.

3.3. Photocatalytic process for antibiotic degradation

In this study, the efficacy of gC_3N_4 -based photocatalysts for

removing antibiotics was systematically examined, utilizing LED as a light source. The investigation also involved a comprehensive analysis of the parameters influencing the photocatalytic process. The antibiotics most frequently studied were TC (18 articles) and CIP (13 articles), which represented 27 % and 13 % of the total antibiotics examined, respectively. The least studied antibiotics, each with a frequency of 1.0, include AMX, AZI, DOX, GMFLO, and NOR (See Fig. (3)). According to Fig. (4), the highest observed efficiency for antibiotic removal ranged from 90 % to 100 %, encompassing 49 % of the antibiotics investigated. Conversely, the antibiotic SMX exhibited the lowest removal efficiency at 54 %. Generally, it is evident that, despite the utilization of LED technology, approximately half of the studies demonstrate antibiotic degradation efficiencies exceeding 90 %, with 7.0 % of articles achieving complete degradation at 100 %. The antibiotics and photocatalysts used in studies that achieved 100 % removal efficiency of antibiotics were TC- gC_3N_4 [47], CIP- gC_3N_4 [48], SIZ- CCN [49], NOR- BNRU [50] and SMZ- $\beta\text{-ZnMoO}_4\text{-180/g-C}_3\text{N}_4\text{-2}$ % [51].

The results, including the obtained antibiotic removal efficiencies and the corresponding experimental conditions, are summarized in

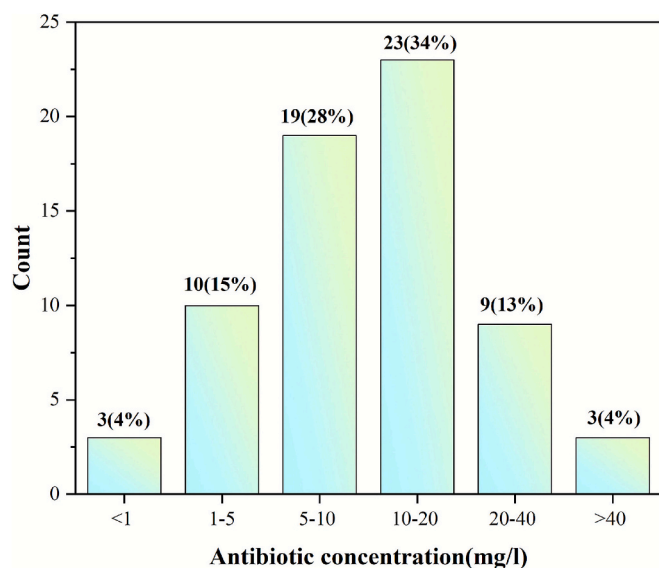


Fig. 6. Investigation of the antibiotic concentration distribution in selected studies.

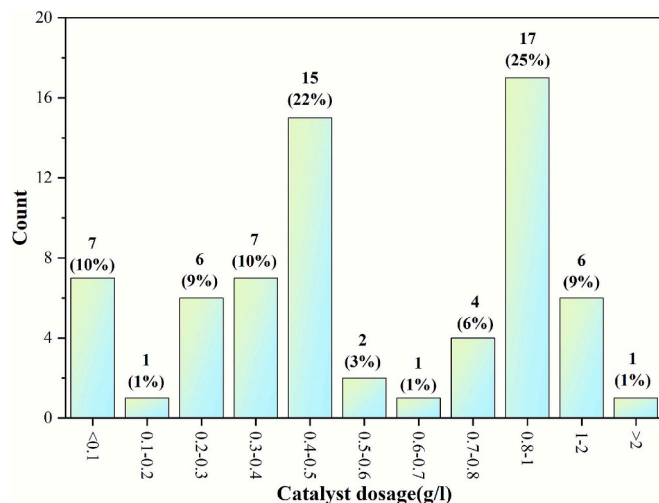


Fig. 7. Investigation of the catalyst dosage distribution in selected studies.

Table 3. It should be noted that the reported efficiencies are not necessarily optimal, as 38 of the included studies focused solely on evaluating the photocatalytic mechanism using antibiotics as model pollutants. To provide a comprehensive overview of photocatalytic processes, studies that did not investigate the optimization of operational parameters were also considered in this review.

3.4. Effect of key factors on the antibiotics degradation by photocatalyst

3.4.1. pH

31 articles studied pH as one of the key parameters affecting the performance of the antibiotic degradation process. Among these, 2.0 articles studied more than one antibiotic. Samadi-Maybodi et al.: TC, DOX [36] and Elbalkiny et al.: LVX, AZI, CTX [17]. So, a total of 36 pH cases were reported. The lowest and highest pH were 3.0 [16] and 10.5 [12], respectively, with the antibiotic removal efficiency for these studies being 96 % (SIZ) and 92.55 % (CTX), respectively. 58 % of the studies that examined pH as an effective parameter considered pH = 6.0–7.0 as optimal or the default pH in their research. 14 studies reported pH = 7 (Fig. (5)). Among the 31 mentioned articles, 22 evaluated

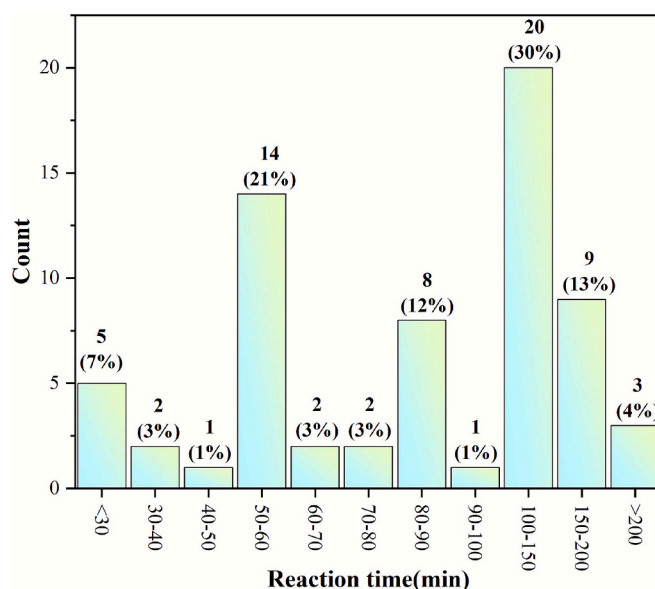


Fig. 8. Investigation of the reaction time distribution in selected studies.

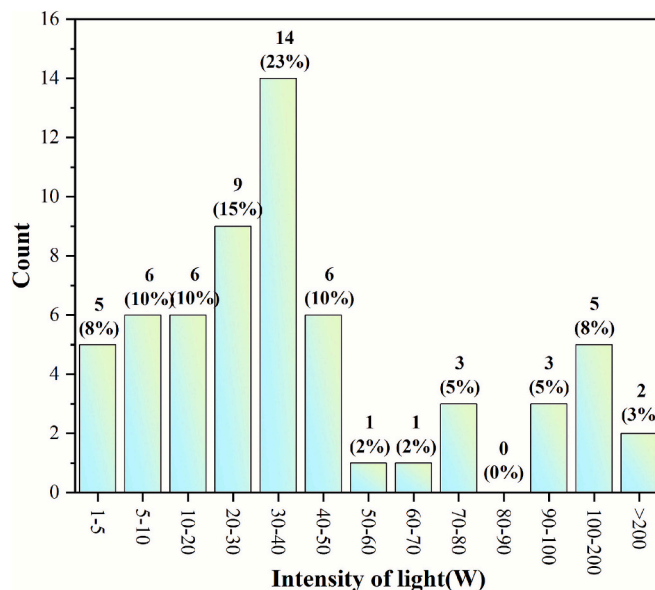


Fig. 9. Investigation of the light intensity distribution in selected studies.

pH at different levels and ultimately declared the optimal pH. However, in other studies, pH was chosen as a fixed and default value. One of the effects of pH is the change in the surface charge of photocatalysts, which affects the interaction between environmental pollutants (such as antibiotics) and the photocatalyst [17]. The point of zero charge is an indicator through which the surface charge of the photocatalyst can be determined [55]. At this pH, the surface charge of the photocatalyst is neutral, and when the environmental pH is less than or greater than the pH_{pzc} , the surface charge of the catalyst becomes positively or negatively charged, respectively [90]. It showed that changes in the pH of the solution can alter the surface charge of the catalyst, thereby affecting its catalytic activity [91]. In addition to the effect of pH on the surface charge of photocatalysts, pH variations can also affect the ionic form of antibiotics, so their solubility and reactivity can differ at various pH levels. Kaushal et al. stated that pH can influence the rate of decomposition of organic substances, including antibiotics, by affecting the amount and generation of hydroxyl radical species [92]. Therefore, the

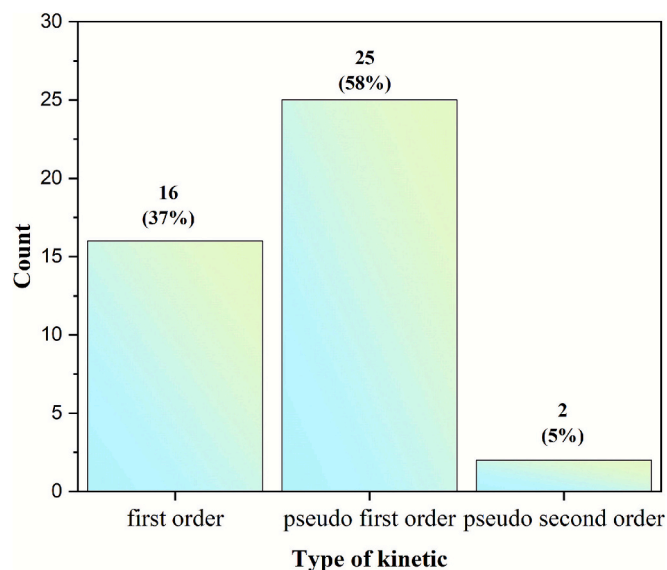


Fig. 10. The kinetic models' distribution in selected studies.

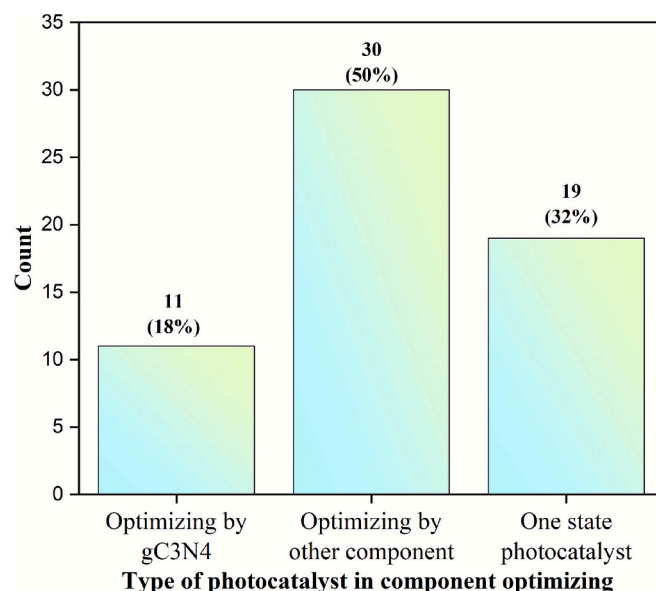


Fig. 12. Types of photocatalysts in component optimizing distribution.

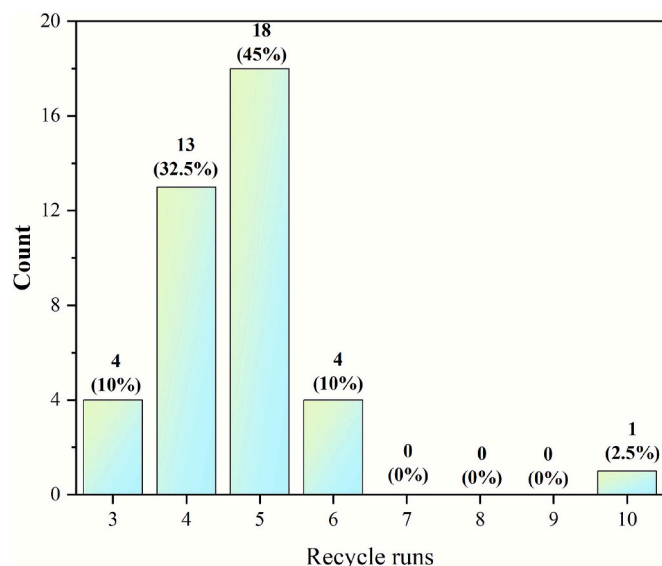


Fig. 11. The distribution of the number of recycling runs in the selected study.

solubility and ionization state of antibiotics can change with pH, which can either increase or decrease their removal efficiency. For example, under acidic conditions, if an antibiotic has greater solubility, it may be more available for photocatalytic degradation. Elkdous et al. emphasized that the initial pH can significantly affect the removal rate of antibiotics and also play a role in the adsorption of antibiotics onto the photocatalyst [93]. Furthermore, photocatalytic processes are not solely dependent on pH; other factors, including the characteristics of the antibiotics and the specifications of the photocatalyst, can also influence the process [19].

Considering the above explanations, the effect of pH is due to its impact on surface charge and the creation of repulsive and attractive conditions between components. In several studies, pH = 7.0 was chosen as optimal. In the study by Feng et al. [54], the effect of pH on the surface charge of CIP and ErFeO₃/g-C₃N₄ (2-EFC) was reported as follows: CIP forms different species under various pH levels [94]. At pH < 5.9, it has a positive charge, and at pH > 8.9, it has a negative charge, appearing as a zwitterion between pH 5.9 and 8.9. The point of zero charge ErFeO₃/g-C₃N₄ was 6.6. Therefore, pH > 6.6, it has a negative

Table 2

Description of the antibiotic's name.

Full name of antibiotics	Abbreviation
Metronidazole	MNZ
Amoxicillin	AMX
Azithromycin	AZI
Ciprofloxacin	CIP
Ceftriaxone	CTX
Doxycycline	DOX
Gemifloxacin mesylate	GMFLO
Levofloxacin	LVX
Norfloxacin	NOR
Ofloxacin	OFL
Oxytetracycline	OTC
Sulfadiazine	SDZ
Sulfisoxazole	SIZ
Sulfamethoxazole	SMX
Sulfamethazine	SMZ
Tetracycline	TC
Tetracycline hydrochloride	TCH

charge, and at pH < 6.6, it has a positive charge. At pH < 6.0, a repulsive force was created between CIP and ErFeO₃/g-C₃N₄, but at neutral pH, the catalyst showed the highest adsorption capacity, leading to increased photocatalytic activity. Similarly, Hasanvandian et al. [58] also reported the highest antibiotic removal efficiency at pH = 7.0. pH < 6.0 and pH > 6.0, the dominant surface charge for VO-UCN/CCO-40 was negative and positive, respectively. The surface charge of LVX was also zwitterionic in the range of 5.59 < pH < 7.94, and at pH = 7.0, an attractive force was created between the antibiotic and the catalyst. Similar mechanisms were observed in studies referenced in this article, which found neutral pH as optimal, for example, using catalysts and antibiotics: g-C₃N₄/γ-Fe₂O₃/ZnO and LVX [38], C, N codoped-TiO₂/g-C₃N₄ and TC [19], and Bi₂WO₆/Fe₃O₄/g-C₃N₄ and LVX [61]. In two studies, optimal conditions were obtained at extremely acidic and alkaline pH levels. Zhong et al. [16] declared pH = 3.0 as optimal. In that study, SIZ at pH = 1.0 had a positive charge and pK_a = 1.5, and considering pH_{pzc} = 3.5, PBCN0.05 also had a positive charge, creating repulsion between the pollutant and the catalyst. As pH increased from 1.0 to 3.0, the surface charge of the catalyst moved towards positive, and the surface charge of SIZ moved towards negative (anionic form), ultimately reaching the highest efficiency at pH = 3.0, and it was even observed that under neutral and alkaline conditions, the degradation of

Table 3

The characteristics and process conditions of selected papers.

No	Photocatalyst	Antibiotic	Process conditions	Efficiency (%)	Ref.
1	TiO ₂ /Ag ₃ PO ₄ /g-C ₃ N ₄	MNZ	C _A = 8.2 ppm, catalyst = 1.2 g/L, pH = 6.05, time = 60 min, LI = 24 W, (blue light)	97.18	[9]
2	50 % CdS/g-C ₃ N ₄	LVX	C _A = 5 ppm, catalyst = 0.4 g/L, time = 200 min, LI = 455 nm	64.42	[52]
3	20 %BiFO/BiCN	OFL	C _A = 10 ppm, catalyst = 0.25 g/L, pH = 8, time = 90 min, LI = 100 W	95.5	[6]
4	ZnO/CN-15	TC	C _A = 10 ppm, catalyst = 0.3 g/L, time = 120 min, LI = 80 W	95	[27]
5	CdS-g-C ₃ N ₄	CTX	C _A = 15 ppm, catalyst = 0.06 g/L, pH = 10.5, time = 81 min, LI = 300 W	92.55	[12]
6	CN-F 35	SMX	C _A = 30 ppm, catalyst = 1 g/L, pH = 6.5, time = 120 min, LI = 4*1150 W.m ⁻²	54	[3]
7	g-C ₃ N ₄ /ZnO	OFL	C _A = 10 ppm, catalyst = 1.6 g/L, time = 180 min, LI = 150 W	63.4	[18]
8	g-C ₃ N ₄ /PrFeO ₃	TC	C _A = 30 ppm, catalyst = 0.1 g/L, pH = 6.5, time = 60 min, LI = 24 W	89	[32]
9	CNU-SA-2	TC	C _A = 20 ppm, catalyst = 0.5 g/L, pH = 5.53, time = 180 min	82.3	[13]
10	g-C ₃ N ₄	LVX	C _A = 0.5 ppm, catalyst = 0.71 g/L, pH = 6.6, time = 86.4 min, LI = 35 W	92.6	[17]
		AZI	C _A = 50 ppm, catalyst = 1 g/L, pH = 4, time = 90 min, LI = 50 W	98.5	
		CTX	C _A = 50 ppm, catalyst = 1 g/L, pH = 4, time = 90 min, LI = 50 W	99.1	
11	L-Arginine C, N codoped-TiO ₂ /g-C ₃ N ₄	MTZ	C _A = 50 ppm, catalyst = 1 g/L, pH = 7, time = 40 min, LI = 50 W	99.9	[19]
		OTC	C _A = 50 ppm, catalyst = 1 g/L, pH = 7, time = 40 min, LI = 50 W	99	
12	Pt/chitosan/g-C ₃ N ₄	GMFLO	C _A = 20 ppm, catalyst = 1 g/L, time = 25 min, LI = 250 W	93.3	[53]
		CIP	C _A = 4 ppm, catalyst = 1 g/L, time = 60 min, LI = 40 W (white LED)	83	
13	Cu ₂ V ₂ O ₇ /Cu ₃ V ₂ O ₈ /g-C ₃ N ₄ (CVCs-2)	TC	C _A = 40 ppm, catalyst = 1 g/L, time = 60 min, LI = 40 W (white LED)	86	[1]
		OTC	C _A = 20 ppm, catalyst = 1 g/L, time = 60 min, LI = 40 W (white LED)	80.5	

Table 3 (continued)

No	Photocatalyst	Antibiotic	Process conditions	Efficiency (%)	Ref.
14	ErFeO ₃ /g-C ₃ N ₄ (2-EFC)	CIP	time = 60 min, LI = 40 W (white LED)		
15	g-C ₃ N ₄ /γ-Fe ₂ O ₃ /ZnO	LVX	C _A = 5 ppm, catalyst = 0.5 g/L, pH = 7, time = 200 min, LI = 40 W	87.8	[54]
16	MMH/g-C ₃ N ₄ /MFC3	SDZ	C _A = 10 ppm, catalyst = 0.6 g/L, pH = 7, time = 25 min, LI = 170 W (green LED)	80	[38]
17	4EGCN	TC	C _A = 37.5 ppm, catalyst = 0.5 g/L, pH = 6.5, time = 130 min, LI = 15 W	93	[55]
18	g-C ₃ N ₄ /RGO/In ₂ S ₃	TCH	C _A = 20 ppm, catalyst = 1 g/L, pH = 4.7, time = 120 min, LI = 50 W (white LED)	78	[56]
19	VO-UCN/CCO-40	LVX	C _A = 20 ppm, catalyst = 0.5 g/L, time = 60 min, LI = 1.5 W	95.6	[57]
20	CNNBr20	OTC	C _A = 20 ppm, catalyst = 0.7 g/L, pH = 7, time = 120 min, LI = 100 W	95	[58]
21	SnO _{2-x} /g-C ₃ N ₄	TCH	C _A = 10 ppm, catalyst = 1 g/L, time = 150 min, LI = 38.5 W (white LED)	75	[7]
22	g-C ₃ N ₄	TC	C _A = 20 ppm, catalyst = 1 g/L, time = 120 min, LI = 35 W	83	[59]
23	0.1AGCN	CIP	C _A = 10 ppm, catalyst = 0.4 g/L, time = 60 min, LI = 10 W (white LED)	100	[47]
24	Bi ₂ WO ₆ /Fe ₃ O ₄ /g-C ₃ N ₄	LVX	C _A = 10 ppm, catalyst = 2 g/L, time = 60 min, LI = 5 W (white LED)	84	[60]
25	P16-ZrO ₂ /gCN	TC	C _A = 25 ppm, catalyst = 0.45 g/L, pH = 7, time = 50 min, LI = 150 W	84.5	[61]
26	g-C ₃ N ₄	CIP	C _A = 40 ppm, catalyst = 0.4 g/L, time = 140 min, LI = 5 W (white LED)	80	[62]
27	Ag@CN-LDH	CIP	C _A = 4 ppm, catalyst = 0.22 g/L, time = 150 min, LI = 40 W (white LED)	~100	[48]
28	CoFe-LDH/g-C ₃ N ₄	TC	C _A = 20 ppm, catalyst = 2 g/L, time = 90 min, LI = 50 W	97	[63]
			C _A = 40 ppm, catalyst = 0.5 g/L, time = 180 min, LI = 5 W	83.8	[64]

(continued on next page)

Table 3 (continued)

No	Photocatalyst	Antibiotic	Process conditions	Efficiency (%)	Ref.
29	TiO ₂ /g-C ₃ N ₄	TC	C _A = 100 ppm, catalyst = 0.5 g/L, pH = 7.7, time = 100 min, LI = 20 W	79.11	[65]
30	Fe ³⁺ /g-C ₃ N ₄ and biofilm	SMX	C _A = 2 ppm, catalyst = 0.5 g/L, time = 600 min, LI = 80 W	96.27	[66]
31	Fe ³⁺ -doped g-C ₃ N ₄	SMX	C _A = 10 ppm, catalyst = 0.5 g/L, time = 720 min, LI = 80 W	81.2	[67]
32	g-C ₃ N ₄ nanosheet/CuBi ₂ O ₄ /Bi ₃ ClO ₄	TCH	C _A = 20 ppm, catalyst = 1 g/L, time = 30 min, LI = 50 W	99	[68]
33	g-C ₃ N ₄ /CoMoO ₄ (6:4)	TC	C _A = 5 ppm, catalyst = 4 g/L, pH = 7, time = 180 min, LI = 20 W	95.93	[69]
34	ZnO/NiO/g-C ₃ N ₄	TC	C _A = 20 ppm, catalyst = 0.05 g/L, time = 300 min, LI = 40 W (white LED)	78.43	[70]
35	g-C ₃ N ₄ /Fe ₃ O ₄ /Bi ₂ WO ₆ /Bi ₂ S ₃	TC	C _A = 20 ppm, catalyst = 1 g/L, pH = 7.2, time = 105 min, LI = 140 W	98	[70]
36	Fe ³⁺ -doped g-C ₃ N ₄	SDZ	C _A = 2 ppm, catalyst = 0.1 g/L, time = 90 min, LI = 3 W (white LED)	99.8	[71]
37	Py-PO ₃ CN	OFL	C _A = 20 ppm, catalyst = 0.3 g/L, time = 120 min	95	[72]
		AMX	C _A = 20 ppm, catalyst = 0.3 g/L, time = 120 min	89	
38	CN-MCA3	OFL	C _A = 20 ppm, catalyst = 0.3 g/L, time = 120 min	96.5	[73]
39	MIL-MIL101Fe (NH ₂)/g-C ₃ N ₄ @CoFe ₂ O ₄ /GO	TC	C _A = 20 ppm, catalyst = 2 g/L, pH = 5, time = 65 min, LI = 30 W	96.3	[36]
		DOX	C _A = 20 ppm, catalyst = 1 g/L, time = 180 min, LI = 35 W	80.5	
40	g-C ₃ N ₄ /ZnO(1:3)	TCH	C _A = 5 ppm, catalyst = 1 g/L, time = 60 min, LI = 50 W	89.33	[74]
41	g-C ₃ N ₄ /BiOF _{0.4} Br _{0.5} I _{0.1}	TC	C _A = 10 ppm, catalyst = 1 g/L, time = 60 min, LI = 50 W	92.6	[75]
42	2 %V ₂ O ₅ /g-C ₃ N ₄	CIP	C _A = 50 ppm, catalyst = 0.4 g/L, pH = 7, time = 150 min, LI = 100 W	90.17	[76]
43	g-C ₃ N ₄ (CNx)	TC	C _A = 10 ppm, catalyst = 0.5 g/L, pH = 7, time = 60 min, LI = 30 W	85	[77]
44	2-ZnFe ₂ O ₄ /g-C ₃ N ₄	CIP	C _A = 10 ppm, catalyst = 0.5 g/L, pH = 7, time = 100 W	97	[78]

Table 3 (continued)

No	Photocatalyst	Antibiotic	Process conditions	Efficiency (%)	Ref.
45	PCN	SIZ	= 60 min, LI = 64 W C _A = 10 ppm, catalyst = 0.4 g/L, time = 30 min, LI = 9 W (blue LED)	99	[79]
46	HPCN	SMX	C _A = 10 ppm, catalyst = 0.6 g/L, pH = 5.56, time = 75 min, LI = 9 W	90.1	[80]
47	MnTiO ₃ /Ag/g-C ₃ N ₄	TC	C _A = 10 ppm, catalyst = 0.1 g/L, time = 120 min, LI = 30 W	61	[81]
48	g-C ₃ N ₄ /BiOBr(1:2)	SDZ	C _A = 30 ppm, catalyst = 1 g/L, pH = 5, time = 150 min, LI = 150 W	~98	[82]
49	Bi ₃ TaO ₇ (QDs)/g-C ₃ N ₄ (NSs)	CIP	C _A = 10 ppm, catalyst = 0.5 g/L, time = 120 min, LI = 86 W	91	[83]
50	ErFeO ₃ /porous g-C ₃ N ₄ nanosheets	OTC	C _A = 20 ppm, catalyst = 0.5 g/L, pH = 5, time = 80 min, LI = 40 W	99.7	[84]
51	CCN	SIZ	C _A = 10 ppm, catalyst = 0.4 g/L, pH = 5.69, time = 40 min, LI = 9 W	100	[49]
52	MgO-CN	TCH	C _A = 30 ppm, catalyst = 1 g/L, time = 150 min, LI = 12 W	82	[85]
53	BNRU	NOR	C _A = 10 ppm, catalyst = 0.5 g/L, pH = 7, time = 15 min, LI = 9 W	100	[50]
54	LaFeO ₃ /g-C ₃ N ₄	OTC	C _A = 20 ppm, catalyst = 0.5 g/L, pH = 7, time = 120 min, LI = 40 W (white LED)	90	[37]
55	CDs/BiOI/g-C ₃ N ₄	TC	C _A = 20 ppm, catalyst = 0.35 g/L, pH = 7, time = 60 min, LI = 30 W	82.7	[86]
56	Gd _{0.12} -Co _{0.05} /g-C ₃ N ₄	CIP	C _A = 20 ppm, catalyst = 0.8 g/L, pH = 7, time = 150 min, LI = 30 W	85	[87]
57	DS/g-C ₃ N ₄	SMZ	C _A = 2 ppm, catalyst = 0.05 g/L, pH = 6, time = 180 min, LI = 18 W	85	[88]
58	β-ZnMoO ₄ -180/g-C ₃ N ₄ -2 %	SMZ	C _A = 2 ppm, catalyst = 0.5 g/L, time = 180 min, LI = 18 W	100	[51]
59	1 %FePc/UPCN	TC	C _A = 20 ppm, catalyst = 0.02 g/L, pH = 7, time = 60 min, LI = 30 W	99	[89]

(continued on next page)

Table 3 (continued)

No	Photocatalyst	Antibiotic	Process conditions	Efficiency (%)	Ref.
60	BCN/PCN	SSZ	C _A = 10 ppm, catalyst = 0.2 g/L, pH = 3, time = 60 min	96	[16]

SIZ stopped. The study by Attari Khasraghi et al. [12], which examined CTX and CdS-g-C₃N₄, showed that pH = 10.5 had the highest efficiency and was considered the optimal pH. In that study, this was attributed to the production of a high concentration of OH⁻, which facilitated the degradation process. From the studies reviewed in this article, it was concluded that determining the point of zero charge of the catalyst and the pK_a of the pollutant can help define the appropriate pH range and ultimately achieve optimal pH. This allows for the identification of the surface charge of the catalyst and the ionic form of the pollutant, leading to a more precise understanding of the pH effect mechanism.

3.4.2. Antibiotic concentration

In all 60 articles, the concentration was reported. Considering that 5 articles examined more than 1.0, the number of antibiotic concentrations extracted from the articles is 67. The used antibiotic concentrations ranged from 0.5 to 100 ppm. 21 articles investigated the concentration at different levels, while in other studies, the reported concentration was a fixed value without considering the concentration range. In two studies, the lowest concentration measured was 0.5 ppm, which related to the antibiotics CTX, LVX, and AZI [17], and the highest concentration was for TC (100 ppm), reported in another study [65]. TC had the highest prevalence among the antibiotics, comprising 27 % of the studies, with a concentration range of 10 to 100 ppm. Fig. (6), shows the distribution of antibiotic concentration in selected studies.

Another factor affecting the efficiency of photocatalytic processes is the role of pollutant concentration, such as antibiotics. Typically, there is an inverse relationship between the increase in antibiotic concentration and its removal efficiency. Various reasons can be cited for this effect, including the saturation of the photocatalyst surface and reduced access of the catalyst to light [12,58,77]. As the concentration of antibiotics increases, the active sites on the photocatalyst may become saturated, leading to a decrease in efficiency due to fewer available active sites [95,96]. Additionally, at high concentrations of antibiotics, some of the light is absorbed by the antibiotic molecules, reducing the amount of light received by the photocatalyst and consequently disrupting the production of reactive species [69,70]. Furthermore, at high concentrations and in the presence of intermediate compounds resulting from degradation, competition arises among pollutant molecules for the active sites on the catalyst surface [80,84]. The competition can prevent the effective adsorption of pollutants onto the surface of the photocatalyst [95,96]. On the other hand, as a result of antibiotic degradation and the formation of intermediate compounds, the degradation products may be more toxic than the original pollutant, affecting the photocatalytic process and limiting the performance of the photocatalyst [97].

After extracting the antibiotic concentration in the studies included in this systematic review, the results showed that as the antibiotic concentration increased, the removal efficiency decreased. Researchers in their studies have presented various reasons for the obtained results. Abbasi-Asl et al. [9] show that as the concentration of MTZ increased from 4.0 to 20 ppm, the efficiency decreased from 100 % to 73 %. Ultimately, according to process optimization with CCD, an MTZ concentration of 8.2 ppm was considered optimal with a removal efficiency of 97.18 %. Hasanvandian et al. [58], with LVX concentrations of 20, 25, and 30 ppm, using the VO-UCN/CCO-40 catalyst, reported efficiencies of 95 %, 80 %, and 68 %, respectively. They cited the following reasons for these results: first, the occupation of active sites on the catalyst or

contaminants reduces the production of e⁻/h⁺ pairs by the catalyst, limiting the process. Second, the production of intermediate compounds competes with LVX molecules. Third, some photons are absorbed by antibiotic molecules, reducing their absorption by the catalyst, ultimately decreasing the production of active species [98]. Samadi-Maybodi et al. [36] also attributed the decrease in the removal efficiency of TC and DOX to the constant dose of the catalyst and the limited active sites on the catalyst's surface.

By survey of other studies also revealed similar conditions and results, showing a decrease in efficiency with increasing antibiotic concentration: a reduction in efficiency from 100 % to 50 % was reported with an increase in TC concentration from 5.0 to 40 ppm in the presence of the CNFBB catalyst in the study by Okab et al. [70]. The reduction in photocatalytic efficiency at higher antibiotic concentrations is primarily due to light absorption by the antibiotic molecules, which limits the light available to the catalyst. Analysis of the effect of antibiotic concentration on photocatalytic performance indicates that, irrespective of the antibiotic type or photocatalyst, common factors such as pollutant accessibility and free radical availability control degradation efficiency. Lower pollutant concentrations increase the availability of active sites and free radicals, enhancing degradation. Additionally, catalyst dosage influences degradation efficiency, with its effect depending on both the catalyst structure and the applied dose across different antibiotic concentrations. This relationship will be further discussed in the context of photocatalytic antibiotic removal.

3.4.3. Photocatalyst dosage

In the articles, the dose of photocatalyst ranged from 0.02 to 4 g/L, with minimum and maximum values applied for the photocatalysts FePc/UPCN [89] and g-C₃N₄/CoMoO₄ (6:4) [69], which achieved 99 % removal of MTZ and 95.93 % removal of SIZ at the mentioned doses, respectively. Among the 60 articles, only 18 examined the catalyst dose at different levels and reported the optimal amount, while other studies used a fixed dose to investigate the photocatalytic process. Doses of 1.0 g/L and 0.5 g/L were evaluated in 17 and 14 studies, respectively, making them the most frequently used doses by researchers. Fig. (7), shows the catalyst dosage distribution in selected studies.

In photocatalytic processes, the dosage of the photocatalyst has a significant impact on the efficiency of pollutant removal. Increasing the dosage of the photocatalyst up to a certain point can play a role in providing active sites for reactions, thereby enhancing the interaction between antibiotic molecules and the catalyst [9,56,67]. Abbasi-Asl et al. attributed the increase in MTZ removal efficiency to the increase in the dose of TiO₂/Ag₃PO₄/g-C₃N₄ from 0.25 to 0.75 g/L [9]. The importance of this, in fact, with an increase in active sites, the possibility of antibiotic adsorption on the surface of the photocatalyst also increases, leading to improved efficiency in the removal of antibiotics [58,99]. Additionally, with an increase in the dosage of the photocatalyst, the capacity for light absorption by the catalyst also increases, as the activation of photocatalysts occurs in the presence of light to generate reactive oxygen species (ROS) that are essential for the degradation of antibiotics [19,58]. Thus, by increasing the catalyst dosage, the potential for the generation of more electron-hole pairs is provided, ultimately leading to the production of hydroxyl radicals and other reactive species [19,100]. However, it is necessary to note that if the dosage of the photocatalyst exceeds a certain limit, it can lead to a decrease in the efficiency of antibiotic removal. This reduction can be attributed to various factors. At higher dosages, the phenomenon of light shielding can occur, where some catalyst particles act as a barrier to light reaching other catalyst particles, thereby reducing the light penetration into the solution and subsequently decreasing photocatalytic activity [9,12,86]. This phenomenon can also occur due to the saturation of the photocatalyst, where additional catalyst particles may lead to agglomeration instead of assisting in pollutant removal [70,101].

Therefore, although increasing the dosage of the photocatalyst can effectively enhance the process's efficiency, determining the optimal

dosage becomes crucial due to the inverse effects of increased dosage at higher levels. Consequently, while higher catalyst dosages can enhance the degradation of antibiotics by providing more active sites, improving light absorption, and increasing ROS production, there is a threshold beyond which efficiency may decline due to light shielding, saturation of active sites, particle aggregation, and inhibition by intermediate compounds. Thus, precise optimization of catalyst dosage is essential for maximizing the effectiveness of photocatalytic processes. In confirmation of the points mentioned above, the studies conducted among the articles that examined the catalyst dose showed that by increasing it to a certain level, the antibiotic removal efficiency increased, and then a decrease in the antibiotic removal efficiency occurred, based on the reasons stated before. In Yin et al.'s study [86] indicate that with an increase in the dose of CDs@BiOI/g-C₃N₄ from 0.05 to 0.35 g/L, the efficiency increased, but after increasing it from 0.35 to 0.65 g/L, the removal efficiency of TC decreased. Therefore, the dose of 0.35 g/L was reported as the optimal dose. In other studies using HPCN [80] and ErFeO₃/porous g-C₃N₄ nanosheets [84] an increase in the dose from 0.6 to 0.8 g/L was reported. Attari Khasraghi et al. [12] reported the decrease in efficiency with an increase in catalyst dose due to catalyst agglomeration and selected a dose of 0.6 g/L as optimal, leading to a removal efficiency of 92.55 % for CTX. Ghosh et al. [56] reached an increased removal efficiency of TC with an increase in dose from 0.5 to 2.0 g/L, attributing this to the increase in active sites on the surface of the catalyst.

3.4.4. Reaction time

The reaction time ranged widely from 8.0 to 720 min. The minimum was related to the removal of SIZ using the BCN/PCN catalyst, which achieved an efficiency of 96 % [16], while the maximum time was for the removal of SMX using the Fe³⁺-doped g-C₃N₄ catalyst, reaching an efficiency of 81.2 % [102]. 14 articles used a 60 min contact time, which was the most frequent. Fig. (8), shows the time reaction distribution in selected articles.

Time is one of the factors and parameters that, with its increase, primarily enhances the efficiency of photocatalytic reactions for the removal of pollutants such as antibiotics. Photocatalytic reactions start through the absorption of light by the photocatalyst, and the conditions for the formation of e⁻/h⁺ pairs are facilitated by the absorption of photons. With an increase in time, there is more opportunity for light penetration and its absorption by the photocatalyst [103]. On the other hand, increasing contact time leads to the production of more free radicals, such as hydroxyl and superoxide, which, due to the high reactivity of these species, will effectively degrade antibiotics [104]. Research has shown that with continued irradiation, the concentration of active species significantly increases, resulting in enhanced antibiotic removal [105]. As photocatalytic reactions progress, the concentration of soluble antibiotics gradually decreases, and with the reduction of pollutant concentration, the reaction environment becomes more favorable for the degradation of the remaining antibiotics. Additionally, the photocatalyst can interact with other intermediate compounds resulting from the degradation of antibiotics, and with increased time, the possibility of further degradation of antibiotic molecules and the intermediate compounds also increases [106]. One of the reasons that is important in increasing reaction time is the stabilization of reaction conditions. This means that over time, aggregation that may arise during the experiment is balanced, leading to better performance of the photocatalyst [107]. In conducted studies, time has also been reported as one of the parameters that has a direct effect on improving antibiotic removal. Moreover, in the study by Eskandari et al. [19], it was stated that with increased residence time and the increased degradation of intermediate compounds, the biodegradability of these intermediate compounds increases. In fact, it can be concluded that with increased residence time, the toxicity of the intermediate compounds decreases. However, when selecting the optimal time, it should be considered that the passage of time does not negatively affect the stability of the

photocatalyst, and the time chosen should be feasible for conducting experiments under real conditions.

3.4.5. Intensity of light source

One of the considerations in this study is the selection of LED as the radiation source. The range of light intensities used was 1.5–300 W. The lowest intensity (1.5 W) was used for the removal of TCH with the g-C₃N₄/RGO/In₂S catalyst [57], while the highest intensity (300 W) was for the removal of CTX with the CdS-g-C₃N₄ catalyst [12] achieving efficiencies of 95.6 % and 92.55 %, respectively. Among 60 articles, 3.0 used blue light LED, 1.0 used green light LED, and 12 used white light LED, while other articles did not mention the color of the LED. 23 % of the articles used a light intensity of 30–40 W, which was the most frequent. No study used a light intensity of 80–90 W. The lowest frequency was related to light intensities greater than 200 W, which included 2.0 studies. 4.0 studies reported light intensity in mW/cm², and 1.0 study did not mention light intensity at all. Fig. (9), shows the distribution of light intensity in selected articles.

It should be noted that among the reviewed studies, detailed reporting of LED characteristics such as wavelength range, irradiation distance, and methods for controlling light uniformity was not standardized. While some studies specified the color of the LED source (e.g., white, blue, green), most did not provide precise spectral data. This lack of uniform reporting limited the possibility of establishing a direct correlation between the LED wavelength and the photocatalytic degradation efficiency. Nevertheless, the rationality of LED light source selection in the reviewed literature can be justified by the close correspondence between the emission spectra of visible-light LEDs (400–700 nm, 1.8–3.1 eV) and the band-gap energy of g-C₃N₄ (2.6–2.7 eV), which ensures effective photoexcitation of the catalyst.

The effectiveness of eliminating antibiotics in photocatalytic processes is significantly affected by the intensity and type of LED lamps used in these processes. Various mechanisms improve the photocatalytic decomposition of antibiotics and increase the intensity of LED lamps. Higher light intensity leads to increased light absorption by photocatalysts, which also increases the production of electron-hole pairs. This phenomenon is very important because their access is directly related to photocatalytic activity [108,109]. In addition, the high intensity of light exacerbates the production of free radicals, which are essential for the decomposition of contaminants such as antibiotics. The presence of these free radicals facilitates the breakdown of antibiotic molecules into less harmful byproducts, thereby improving the overall efficiency of removal [110]. In addition, increasing the intensity of the lamp reduces the time it takes to achieve a balance in the photocatalytic reaction. This reduced reaction time is useful because it allows for faster decomposition of antibiotics and increases the overall efficiency of the process [111]. The type of LED lamp also plays a vital role in the photocatalytic removal of antibiotics. The different colors of LED lamps emit different wavelengths of light, which can significantly affect the efficiency of the photocatalytic process. White LED lamps that cover a wide range of wavelengths are particularly effective at breaking down a range of pollutants, including antibiotics [108,112]. In contrast, violet LED lamps that emit shorter wavelengths are very effective in producing free radicals and thus enhance the decomposition of antibiotics [109]. Blue LED lamps can also help with the photocatalytic process, though with slightly less efficiency than violet bulbs [113]. Green LED lamps, on the other hand, are generally less effective at removing antibiotics because of their longer wavelengths that are not well absorbed by many photocatalysts [109,110]. As a result, optimizing both the intensity and type of LED lamps is essential to increase the efficiency of eliminating antibiotics in photocatalytic processes. The use of purple and white LED lamps with appropriate intensities is recommended to achieve optimal results in the decomposition of antibiotics. These compounds provide maximum light absorption and production of free radicals while minimizing reaction time.

3.4.6. Kinetics

Calculations related to reaction kinetics in photocatalytic processes are essential due to their role in determining the mechanisms of adsorption and reaction rates. Assessing the reaction kinetics is crucial for identifying the optimal conditions for pollutant removal, given the adsorptive properties of the catalyst [114,115]. Kinetic models are derived using linear or nonlinear regression analyses in batch photocatalytic experiments [116]. In the studies reviewed in this research, the kinetic models examined included first-order, second-order, pseudo-first-order, and pseudo-second-order. Out of the 60 articles, 43 focused on reaction kinetics. Fifty-eight percent of these articles adhered to pseudo-first-order kinetics. At the same time, none followed second-order kinetics (Fig. (10)). Results indicated that in the majority of articles (25 out of 43) adhering to pseudo-first-order kinetics, the degradation rate of antibiotics was directly proportional to the antibiotic concentration, suggesting that as the antibiotic was consumed, the reaction rate decreased [9]. Additionally, the studies demonstrate that the components of the catalyst capable of adsorbing the antibiotic also influence the adsorption rate and reaction speed. In the study conducted by Hoang An et al., researchers concluded that an increase in one of the catalyst components (ZnO) led to a decrease in the amount of antibiotics adsorbed on TC, indicating that in ZnO@CN-25, the reaction rate constant decreased compared to ZnO@CN-5. This was attributed to the fact that a larger proportion of ZnO partially reduced the active sites on CN, diminishing the contribution and role of CN in the catalyst [27]. In Li et al.'s study, the pseudo-first-order kinetic model ($\ln(C_0/C) = kt$) was utilized for further analysis of TC photodegradation. The degradation rate (k) for CoFe-LDH/g-C₃N₄ (0.43 h⁻¹) was found to be 1.87 and 2.26 times greater than that of the original g-C₃N₄ and CoFe-LDH, respectively. Finally, it is worth noting that the photocatalytic performance of CoFe-LDH/g-C₃N₄ was superior compared to other reported LDHs/g-C₃N₄ [64].

3.4.7. Reusability of photocatalyst

In photocatalytic processes, catalyst reuse can be facilitated by incorporating magnetic components into the photocatalyst structure. However, during consecutive cycles, a gradual decrease in antibiotic degradation efficiency is commonly observed. This decline is attributed to several factors, including the deposition of impurities on the catalyst surface, which reduces pollutant accessibility to active sites, and the accumulation of undecomposed compounds and oxidation intermediates, which obstruct light penetration and hinder the generation of electron-hole pairs [76,117]. Light absorption is one of the effective factors in activating photocatalysts, and over time, the light absorption capacity of the catalyst decreases, leading to a reduction in hydroxyl radical production [118]. In some cases, the catalyst itself gradually experiences wear and tear over consecutive cycles, reducing its initial capability. This wear can result from both the surface erosion of the particles and the disintegration of the interconnected components of the catalyst particles, which in the case of magnetic nanoparticles may lead to a reduction in the magnetic properties of the photocatalyst and disrupt its separation [118,119]. The adhesion and agglomeration of the photocatalyst can also be another reason for the decrease in the efficiency of antibiotic degradation in consecutive cycles [117]. Considering that after each stage of catalyst separation and preparation for the next cycle, part of the catalyst may be lost due to leakage from the system, and the method of preparing the catalyst for the next cycle may lead to the loss of its particles [73], all these factors can contribute to the reduction of antibiotic degradation efficiency in consecutive cycles. In this study, 38 out of 60 articles examined the reuse of photocatalysts. As shown in Fig. (11), the number of cycles tested for reuse ranged from 3.0 to 10 cycles. However, the number of runs from 7.0 to 9.0 was not measured in any study. Although the high degradation efficiency of the photocatalyst is a significant factor affecting its performance, maintaining efficiency over consecutive cycles is considered a positive aspect of the process. Among the studies reviewed in this systematic review,

Chen et al. showed the least reduction in efficiency during consecutive cycles (4.0 cycles) using the CNU-SA-2 photocatalyst, with only a 2.3 % decrease in efficiency for TC degradation from cycle 1.0 to 4.0. The mentioned catalyst was not magnetic, and its separation was performed by centrifugation [13]. Additionally, Liu et al. demonstrated that during 3.0 cycles, the photocatalyst Fe³⁺/g-C₃N₄ and biofilm showed the highest reduction in efficiency for SMX degradation, which was 42.67 %, with efficiency decreasing from 96.27 % in cycle 1.0 to 53.6 % in cycle 3.0. The researchers explained this significant reduction in efficiency as likely due to the loss of the photocatalyst during separation from the reaction solution in each cycle, with 0.036 g and 0.012 g of the photocatalyst lost in the first and second cycles, respectively [66]. More details are provided in Table 4.

3.4.8. Role of g-C₃N₄ in photocatalyst structure

In photocatalytic processes, photocatalysts are synthesized with the primary goals of enhancing pollutant degradation efficiency, optimizing catalytic performance, and elucidating the underlying mechanisms. Research efforts typically focus on three main aspects: (1) investigating the mechanism of action using a model pollutant, where the efficiency of degradation is evaluated under controlled parameters and environmental conditions to assess catalyst activity and stability; (2) synthesizing catalysts with varying component ratios often not limited to g-C₃N₄ to identify optimal formulations and assess their pollutant removal performance; and (3) examining the influence of operational parameters, such as pH, light intensity, and catalyst dosage, on the degradation process, in order to understand their impact on photocatalytic efficiency.

In this systematic review, as shown in Fig. (12), out of 60, 11 studies synthesized g-C₃N₄ in different ratios. 30 studies synthesized another component of the catalyst in various ratios, and 19 studies only examined the synthesis of the catalyst in one format. In the study by Nguyen et al. [69], the photocatalyst g-C₃N₄/CoMoO₄ was synthesized, and both components were synthesized in different ratios, including ratios of 9.0:1.0, 7.0:3.0, 8.0:2.0, and 6.0:4.0, with the optimal ratio identified as 6.0:4.0. This ratio was found to maximize the photocatalytic activity under specific experimental conditions.

The efficient photocatalyst can also be tracked using the analyses conducted for the photocatalyst. In the study by Tan et al. DRS analysis showed that the presence of g-C₃N₄ improved the visible light absorption capability in the PCN catalyst. This is crucial for enhancing the overall efficiency of photocatalytic reactions. One of the features of pure g-C₃N₄ is its ability to absorb visible light, but when GCN is used in a modified form, this capability becomes more stable and enhanced. Researchers are continuously looking for ways to improve the stability and efficiency of photocatalysts [79]. Modification and co-doping can increase the light absorption capacity [120]. This strategy has opened new avenues for optimizing photocatalytic materials to achieve better performance. On the other hand, increasing the percentage of one component in the photocatalyst does not necessarily improve its efficiency. This has been proven in various studies, indicating an optimal range for each component in the catalyst formulation.

Zhang et al., worked on FePc/UPCN, where ratios of 0.5–5.0 % of FePc were used in the catalyst structure, and the results were observed as BCN < UPCN < 0.5 % FePc/UPCN < 5 % FePc/UPCN < 3 % FePc/UPCN < 1 % FePc/UPCN, with 1.0 % FePc considered as its optimal percentage. These findings highlight the importance of carefully selecting component ratios to achieve the desired catalytic properties [89].

The results of evaluating the performance of catalysts in different ratios and comparative experiments are available in Supplementary Information (S3). This comprehensive data serves as a valuable resource for future research in the field. The importance of g-C₃N₄, which was also addressed in the introduction of the article, is shown in these studies where its presence, even as a non-primary component, has had acceptable efficacy. This reinforces the potential of g-C₃N₄ in various photocatalytic applications, paving the way for further innovations in

Table 4

Information related to the reuse of the photocatalyst.

No	Photocatalyst	Antibiotic	Run	Time (min)	Efficiency (%) first run	Efficiency (%) last run	ref
1	TiO ₂ /Ag ₃ PO ₄ /g-C ₃ N ₄	MNZ	5	80	97.18	58.3	[9]
2	50 % CdS/g-C ₃ N ₄	LVX	5	200	64.42	65.54	[52]
3	20 %BiFO/BiCN	OFL	5	90	95.5	90.7	[6]
4	ZnO/CN-15	TC	5	120	95	90	[27]
5	g-C ₃ N ₄ /PrFeO ₃	TC	5	60	89	80	[32]
6	CNU-SA-2	TC	4	180	82.3	80	[13]
7	C, N codoped-TiO ₂ /g-C ₃ N ₄	MTZ	6	100	99.9	82	[19]
		OTC		40	99	92	
8	Cu ₂ V ₂ O ₇ /Cu ₃ V ₂ O ₈ /g-C ₃ N ₄ (CVCs-2)	CIP	4	60	83	75	[1]
9	ErFeO ₃ /g-C ₃ N ₄ (2-EFC)	CIP	3	200	87.8	~80	[54]
10	g-C ₃ N ₄ /γ-Fe ₂ O ₃ /ZnO	LVX	5	60	80	76	[38]
11	MMH/g-C ₃ N ₄ /MFC3	SDZ	10	130	93	88	[55]
12	4EGCN	TC	5	120	78	~60	[56]
13	VO-UCN/CCO-40	LVX	6	120	95	90	[58]
14	SnO _{2-x} /g-C ₃ N ₄	TCH	4	120	83	75	[59]
15	Bi ₂ WO ₆ /Fe ₃ O ₄ /g-C ₃ N ₄	LVX	5	50	84.5	70.9	[61]
16	P16-ZrO ₂ /gCN	TC	5	140	80	70	[62]
17	Ag@CN-LDH	CIP	5	90	97	94.66	[63]
18	CoFe-LDH/g-C ₃ N ₄	TC	3	180	83.8	~73.6	[64]
19	TiO ₂ /g-C ₃ N ₄	TC	5	100	79.11	62.79	[65]
20	Fe ³⁺ /g-C ₃ N ₄ and biofilm	SMX	3	600	96.27	53.6	[66]
21	ZnO/NiO/g-C ₃ N ₄	TC	4	300	78.43	73.52	[70]
22	g-C ₃ N ₄ /Fe ₃ O ₄ /Bi ₂ WO ₆ /Bi ₂ S ₃	TC	6	105	98	86	[70]
23	CN-MCA3	OFL	5	120	96.5	72	[73]
		TC		65	96.3	65.8	
24	MIL-MIL101Fe (NH ₂) /g-C ₃ N ₄ @CoFe ₂ O ₄ /GO	DOX	5	65	80.5	60.41	[36]
25	g-C ₃ N ₄ /ZnO	TCH	5	180	89.33	~81	[74]
26	V ₂ O ₅ /g-C ₃ N ₄	CIP	4	150	90.17	~65	[76]
27	g-C ₃ N ₄ (CNx)	TC	3	60	85	79	[77]
28	ZnFe ₂ O ₄ /g-C ₃ N ₄	CIP	5	60	97	90	[78]
29	PCN	SIZ	4	30	99	92	[79]
30	HPCN	SMX	4	75	90.1	85	[80]
31	Bi ₃ TaO ₇ (QDs)/g-C ₃ N ₄ (NSs)	CIP	4	120	91	84	[83]
32	CCN	SIZ	4	40	100	85	[49]
33	MgO-CN	TCH	5	150	82	63.5	[85]
34	BNRU	NOR	4	15	100	95	[50]
35	CDs/BiOI/g-C ₃ N ₄	TC	4	60	82.7	80	[86]
36	Gd _{0.12} -Co _{0.05} /g-C ₃ N ₄	CIP	5	150	85	80	[87]
37	FePc/UPCN	TC	4	60	99	95	[89]
38	BCN/PCN	SSZ	4	8	96	91	[16]

environmental technology.

One of the factors in choosing the source of radiation is its ability to activate the catalyst. The components of each catalyst also have the ability to absorb different wavelengths according to their band gap. In studies conducted in this systematic review, it focus on the use of LED as a source of radiation and the activator of catalysts containing gC₃N₄. To activate the catalyst, the light source must coordinate the energy of the photon with the band gap of the catalyst, and to transfer electrons from the valence band to the conduction band, it must receive a photonic electron whose energy is equal to or greater than the band gap energy. One of the methods for identifying and detecting the matching of band gap photocatalyst and radiation source energy is using the results of UV-vis and DRS analyses. In the present study, the results showed that in the overall analysis of UV-vis analyses, the absorption edges for gC₃N₄ typically appear in the range of 460–450 nm. The gC₃N₄ band gap is also about 2.6–2.7 eV, which, according to the results, is in the visible light region. The combination of metal components or other semiconductors, such as Ag₃PO₄, Fe³⁺, or LDH structures, can narrow the band gap to 2–2.4 eV. This modification to the photocatalyst structure enables performance in the visible light range. Considering that the studies studied LED radiation is between 400 and 700 nm (1.8–3.1 eV), the emission spectrum overlaps with the gC₃N₄-based band gap [9].

4. Conclusion

In this systematic review, the effect of g-C₃N₄-containing photocatalysts on the removal of antibiotics was examined. Given its

advantage in absorbing visible light and the possibility of using visible light as a radiation source, LED light sources were also considered as a factor in the selection of studies. Therefore, in this study, the use of modified g-C₃N₄ showed the highest efficiency in all studies. Therefore, it is essential to pay attention to the fact that researchers can be guided in the synthesis of various new and more effective catalysts using the results presented in the studies. In this study, the role of parameters such as pH, antibiotic concentration, photocatalyst dose, reaction time, and light source intensity as factors affecting the efficiency of antibiotic removal was examined. The reusability of the photocatalyst in consecutive cycles was also investigated. An important point in the reuse of the catalyst was the stability of the photocatalyst after its use in consecutive cycles. A photocatalyst with greater stability, which showed the least reduction in the efficiency of antibiotic degradation during reuse, is more reliable than a photocatalyst that loses its efficiency over consecutive cycles, resulting in a significant difference in antibiotic removal efficiency between the first and last cycles. Finally, it is suggested that researchers address the existing gaps, which include the lack of use of photocatalysts in real scales and real samples. They should also explore methods for separating catalysts on practical scales.

CRediT authorship contribution statement

Bahareh Mirzahedayat: Writing – original draft, Data curation. **Esrafil Asgari:** Writing – original draft, Supervision, Methodology. **Mehran Mohammadian Fazli:** Validation, Supervision, Methodology, Conceptualization. **Koorosh Kamali:** Methodology, Investigation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.rechem.2025.102917>.

Data availability

The data that has been used is confidential.

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