engineering proceedings

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Proceeding Paper

A Systematic Review of Metal–Organic Framework (MOF)-Based Nanocomposites and Their Application in Photocatalytic Degradation of Pharmaceutical Compounds †

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† Presented at the 3rd International Electronic Conference on Processes—Green and Sustainable Process Engineering and Process Systems Engineering (ECP 2024), 29–31 May 2024; Available online: https://sciforum.net/event/ECP2024.

Abstract: Photocatalytic degradation has emerged as a promising and sustainable solution to the rising environmental concerns caused by the emergence of pharmaceutical contaminants in aquatic bodies. Given its sustainability, nontoxicity, and energy-efficient attributes, extensive research has been conducted to investigate this approach and explore the synthesis of various photocatalysts. Metal–organic framework (MOF)-based nanocomposites exhibit a broad spectrum of applications across various fields due to their substantial surface area and versatile functionality. In these fields, MOF-based nanocomposites exhibit a broad spectrum of applications across various fields due to their substantial surface area and versatile functionality. In these fields, MOF-based nanocomposites have proven to be excellent photocatalysts in water treatments, particularly in the breakdown of pharmaceutical pollutants in aquatic environments. This systematic review provides an overview of MOF-based nanocomposites and their applications in the photocatalytic degradation of pharmaceutical products. Several studies have explored different synthesis techniques for various MOF-based nanocomposites, and their degradation efficiencies against pollutants, under varying conditions, such as catalyst loading, pollutant concentration, light intensity, and exposure time, among others, have also been reported. Moreover, modifications to these synthesis techniques have been made to enhance the degradation capability and stability of MOF-based nanocomposites. Meanwhile, the integration of other metals and biomaterials into MOF-based nanocomposites has been explored to improve the stability and recyclability of these photocatalysts. This paper also highlights the existing gaps in the current research to direct future investigations on photocatalytic degradation using MOF-based nanocomposites.

Keywords: metal organic framework; nanocomposite; photocatalytic degradation; pharmaceuticals; pollutant; MOF-based nanocomposite

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Citation: Bautista, K.A.; Mata, E.J.D.C.; Mercado, C.D.B.; Placio, R.J.C.; Alano, V.H.O.; Soriano, A.N.; Rubi, R.V.C. A Systematic Review of Metal–Organic Framework (MOF)-Based Nanocomposites and Their Application in Photocatalytic Degradation of Pharmaceutical Compounds. *Eng. Proc.* **2024**, *67*, 33. https://doi.org/10.3390/engproc2024067033

Academic Editor: Juan Francisco García Martín

Published: 6 September 2024

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1. Introduction

The growing concern about the emergence of pharmaceutical products in aquatic environments, with concentrations ranging from ng/L to µg/L [1], which may pose health risks for both human and aquatic life, highlights the need for efficient and sustainable water treatment technologies. One such technology is photocatalytic degradation, known as an

advanced oxidation process (AOP). In this method, radicals, produced from photocatalysis, break down organic pollutants into less harmful substances [2]. Moreover, photocatalytic degradation is considered as a promising and environmentally friendly method due to its sustainability, nontoxicity, and energy-saving attributes [3]. This makes it well-suited to treating wastewater containing pollutants that exhibit poor biodegradability and high concentrations [4].

Organic pollutants, such as pharmaceutical products, are widely degraded using a photocatalyst, and one of the many arising photocatalysts being studied and utilized is metal–organic frameworks (MOFs). These new materials have garnered significant interest from scientists and material engineers due to their unique properties, making them highly promising for diverse applications across various fields. Yaghi [5], a prominent figure in the advancement of MOF chemistry, defines MOFs as structures consisting of metal ions or clusters (known as secondary building units or SBUs) linked together by charged organic linkers or units. Typically, MOFs are synthesized using metal ions including alkaline earth metals (such as Ca or Mg) or transition metals (such as Fe, Mn, Al, Zr, and Ti), along with organic linkers that often include dicarboxylic acid, tricarboxylic acid, tetracarboxylic acid, and imidazolate [6]. Furthermore, MOFs are commonly doped or integrated with additional substances to enhance their properties, including structural integrity and catalytic performance.

This paper will explore current research on MOF-based nanocomposites for the photocatalytic degradation of pharmaceutical products, identifying existing research gaps to provide directions for future research.

2. Methodology

In this review article, the systematic literature review protocol posed by (Kashem et al. 2024) [7] was utilized as shown in Figure 1. The process of collecting and screening data was shown in Figure 1 where there are four distinct stages: identification, screening, eligibility, and inclusion. The initial stage of conducting a systematic literature review involves the process of identification, which is manually examining significant publications and locating relevant articles that fit with the objective of the study. Screening processes are necessary to ensure the inclusion of research that meets predetermined criteria following the identification phase. The primary objective of this screening method is to reduce the likelihood of bias in the studies that are finally chosen. To see if the papers were eligible, the full texts were looked at to see how well they addressed the study issue, met the specific goals of the research review, and used a good research method. Lastly, the inclusion criteria encompassed research that extracted and synthesized data from the selected studies, thus providing a concise summary of the most pertinent information.

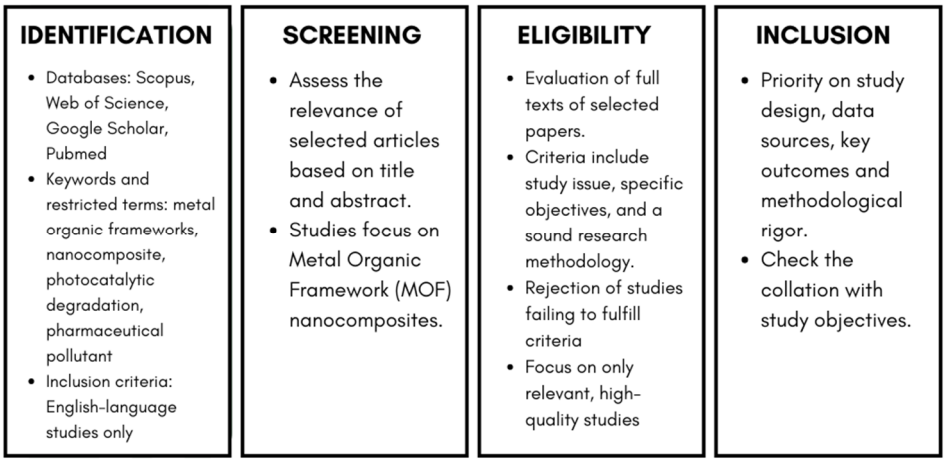


Figure 1. Flowchart of systematic literature review adapted from Kashem, Shamsuddoha, and Nasir.

The Preferred Reporting Items for Systematic reviews and Meta-Analyses (PRISMA) framework was also used to determine the search strategy and assess the quality of the studies, as shown in Figure 2. A comprehensive screening process was conducted to guarantee that only pertinent and significant publications were included in this study. A total of 612 papers were discovered from internet databases. An initial screening was conducted using the keywords 'metal organic framework', 'photocatalytic degradation', 'nanocomposite', and 'pharmaceuticals'. Duplicate publications were manually excluded from the database, as well as those that did not include significant information on the specified topic. Two papers were removed from this retrieval procedure (n = 2). The final assessment of eligibility was carried out by individually examining the abstracts of the screened papers. Ten papers were considered ineligible and hence excluded. The reason for this is that four papers did not explicitly utilize metal organic framework (MOF) as the nanocomposites for the degradation of pharmaceutical pollutants. Additionally, six journal articles solely presented the recent progress on metal organic framework without providing the study's results. To select the appropriate papers, several factors were considered, including the presence of pharmaceutical pollutants, the utilization of metal organic framework as a nanocomposite, synthesis process, the light source, and the effectiveness and duration of degradation.

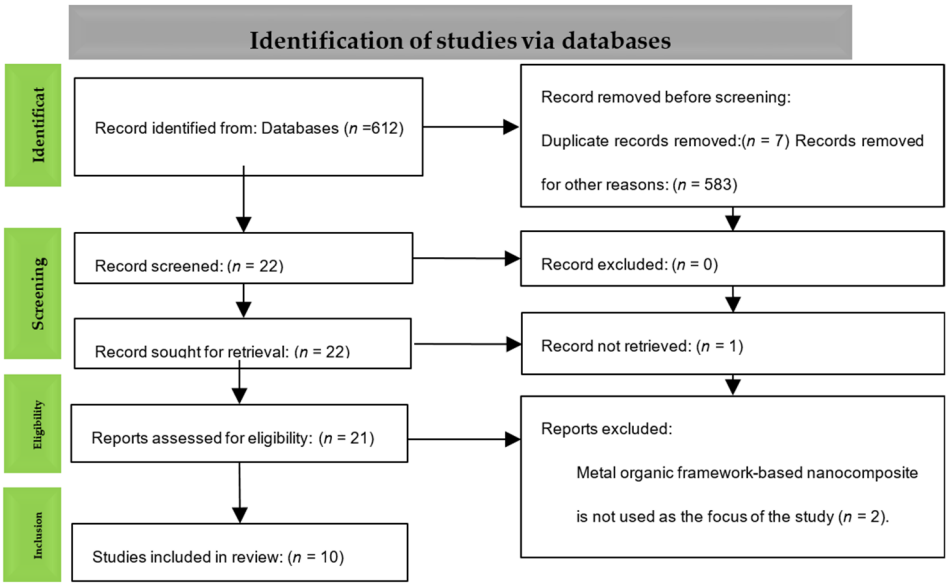


Figure 2. Adapted PRISMA protocol used in the study.

3. Mechanism of Photocatalytic Degradation

Photocatalytic degradation is an advanced oxidation process (AOP) that utilizes light and a photocatalyst to drive pollutant degradation as depicted in Figure 3. It is known as an environmentally friendly, sustainable, and energy saving technique where solar energy is harnessed by photocatalysts to degrade pollutants. Pollutant molecules break down through oxidation and hydrolysis when they absorb photons from three electromagnetic regions: visible (vis), ultraviolet (UV), and infrared (IR). When the photocatalytic material is exposed to UV/vis/IR light, electrons move from the valence to the conduction band, creating electron and hole pairs through a photo-induced process [2]. In this phenomenon, reactive oxygen species (ROS) are formed due to the reaction between photo-induced electrons and holes with oxygen (O_2), water (H_2O), and hydroxyl groups. These ROS are hydroxyl radicals (·OH) and superoxide radical anions ($\cdot O_2^-$) with strong oxidation abilities that serve as the major responsible species for the degradation of persistent organic pollutants in wastewater. These radicals can interact with pollutant molecules, resulting in the complete or partial degradation of persistent pollutants.

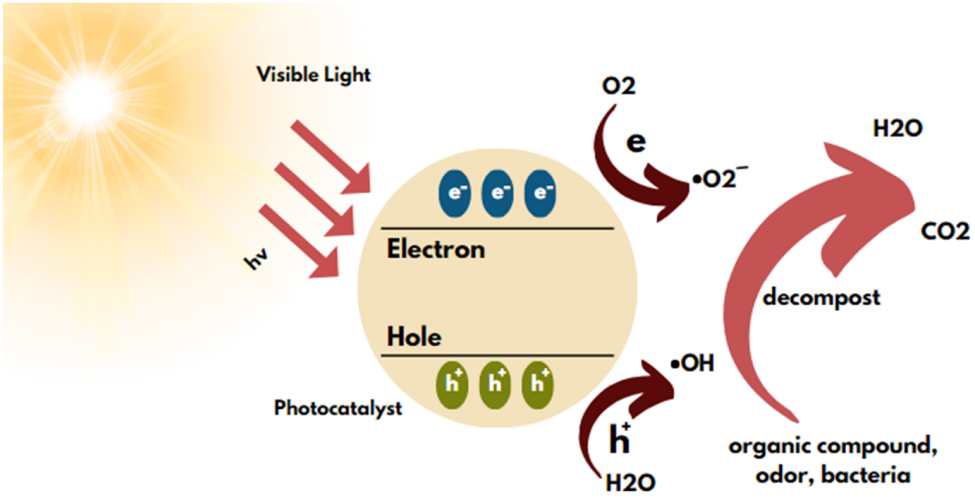


Figure 3. Photocatalytic degradation mechanism.

4. MOF-Based Nanocomposites for the Photocatalytic Degradation of Pharmaceutical Products

4.1. Method of Synthesis

The intriguing properties of metal–organic frameworks (MOFs), such as significant surface area, flexible porosity, numerous pores, versatile functionality, compatibility with biological systems, and high efficiency in drug delivery [7,8], have attracted researchers to extensively explore the development of MOFs. They are highly versatile materials known for their tunable structural, physical, chemical, and electrical properties, making them ideal for a range of applications. Structurally, MOFs are composed of metal ions or clusters connected by organic linkers, allowing for variable pore sizes and shapes that can be synthesized for specific uses like gas storage and separation. Their chemical properties, including catalytic activity, can be enhanced through structural modifications, making them effective in industrial processes and environmentally friendly applications. Additionally, the electrical properties of MOFs, such as conductivity and band gaps, can be influenced by the incorporation of guest molecules, positioning them as promising candidates for sensors, electronic devices, and energy storage systems. This combination of adaptable characteristics underscores the potential of MOFs in advancing various technological fields [9].

The Cambridge Crystal Database Collection already documents more than 100,000 distinct MOF structures, a number that continues to grow daily [5]. Consequently, there is a growing interest in their application across a range of fields, particularly in their use in the photocatalytic degradation of emerging pharmaceutical pollutants. Synthesizing MOFs requires the input of energy to facilitate the binding between metal ions and organic linkers, and typically occurs at room temperature and up to 250 °C in order to increase the rate of MOF formation without organic linker decomposition [6,9]. Several popular methods have already been employed in the synthesis of MOF-based nanocomposites, which will be briefly presented and discussed with chosen representative existing studies accordingly.

Among all the methods used to synthesize MOF-based nanocomposites, the conventional solvothermal method is the simplest and most widely used. This involves the use of a solvent under moderate to high pressure and temperature, typically ranging from 100 °C to 1000 °C, to facilitate precursor interactions in a closed reaction vessel. Zhang et al. [10] used the solvothermal approach to assemble MIL-53(Fe), while Pattappan et al. [11] used it to synthesize a bimetallic metal–organic framework (MOF) consisting of iron and cobalt (FeCo). When the solvent used in the process is water, it is referred to as the hydrothermal method. Typically, this method is performed at relatively mild temperatures below 300 °C [12]. Fawzy et al. [13] have employed hydrothermal synthesis in the creation of

MIL-53(Al)/ZnO nanocomposites. Both methods offer similar advantages including ease of synthesis, good crystallinity, and composition control.

In addition to traditional thermal processes, calcination treatments have been gaining traction in the synthesis of MOF-based nanocomposites. In fact, in the study of Liu et al. [14], they introduced the calcination treatment of virgin MIL-53(Fe) at 400 °C, which successfully yielded a magnetic photocatalyst known as Fe_3O_4@MIL-53(Fe). Similarly, calcination was used as an enhancement technique in the study of Liu et al. [15], wherein they created a photo-Fenton catalyst using a UIO-66 (Zr) metal–organic framework (MOF) dispersed in diatomite via a single-step solvothermal method for hydroxylation (HO-UIO-66). To enhance stability, the photocatalysts were then subjected to calcination at different temperatures of 300 °C, 400 °C, and 500 °C. Generally, calcination refers to heating the material at high temperatures. In addition, in the study of Tsai et al. [16], a novel Cu_xO/MOF nanocomposite was introduced with a CuO–Cu_2O–carbon structure via carbonization process at 300 °C. This process typically transforms MOFs into carbonaceous material, allowing Cu_xO nanoparticles to deposit onto the C-skeleton of MOF. In general, these processes can bring about several improvements in the synthesized MOF-based nanocomposite, including increased surface area, enhanced absorption ability, improved stability, and ultimately, enhanced degradation efficiency [12,13].

Researchers are developing new advanced methods for creating MOF-based nanocomposites that aim to achieve faster synthesis, more uniform crystals, higher yields, and minimize environmental impact [8,15]. In the study of Li et al. [17], Fe_3O_4@MIL-100(Fe) was synthesized through a rapid microwave heating process. Microwave synthesis offers a promising approach for MOF synthesis by converting electromagnetic waves into heat, allowing for shorter reaction times and superior yields compared to conventional methods. In addition, Salimi et al. [18] synthesized solar-light-driven photocatalysts, MIL-125(Ti) mixed linker and different quantities of g-C_3N_4, through ultrasonication and solvothermal approaches. Ultrasonication, a process that uses sound waves with frequencies higher than the human auditory range of more than 20 kHz, assisted the synthesis process and ultimately enhanced the stability, photocatalytic activity, and biocompatibility of the synthesized nanocomposites. Furthermore, facile methods for MOF synthesis are gaining significant attention in research due to their focus on simplicity, efficiency, and accessibility. Zhao et al. [19] demonstrated the facile synthesis of Co-MOF with layered CoO_x as the precursor and with the use of metal salts. This led to a high concentration of metal ions around the precursor particles, resulting in the formation of a densely stacked lamellate, Co-MOF. In general, facile synthesis methods are also believed to improve the stability of MOFs prepared with metal salts, as reported in this study.

Finally, the integration of biomaterials in the synthesis of MOFs is also an emerging interest among scientists researching photocatalytic degradation. These materials can enhance the properties of MOFs, particularly their degradation efficiency. Español et al. [20] recently utilized ultrasound-assisted biomimetic synthesis to create MIL-100(Fe)/HAp nanocomposites in a 10xSBF (simulated body fluid)-like solution. Biomimetic synthesis mimics biochemical processes to create materials with properties that are similar to, or enhance, those of naturally occurring biological materials. It often utilizes simulated body fluid (SBF) or electrolyte solutions that can induce the apatite growth of calcium phosphate on the surface of MOFs. As a result, the MOF-based nanocomposites exhibit promising physicochemical features and improved degrading efficiency.

4.2. Comparison of Photocatalytic Performance

The studies reviewed discuss various approaches to enhance the photocatalytic degradation of organic pollutants using metal–organic frameworks (MOFs) and their composites. As presented in Table 1, Zhang et al. [10] utilized MIL-53(Fe) combined with sulfate radical-based advanced oxidation processes (SR-AOPs) via a solvothermal approach to achieve 99.7% degradation of tetracycline hydrochloride (TC-HCl) within 80 min under visible light, attributing the catalytic effect to MIL-53(Fe) generating $SO_4^{\bullet -}$. On the other hand,

Fawzy et al. [13] synthesized MIL-53(Al)/ZnO nanocomposites using a hydrothermal approach for amoxicillin (AMX) degradation, optimizing operational parameters like initial AMX concentration and pH, achieving 100% efficiency within 60 min under visible light. In the study by Liu et al. [14], calcination treatment was introduced into hydrothermal synthesis to create Fe_3O_4@MIL-53(Fe) composites for enhanced photocatalytic activity in ibuprofen (IBP) degradation. The results showed 99% degradation of IBP within 60 min under visible light. Another study by Liu et al. [15] focused on stability enhancement through calcination integrated into solvothermal synthesis, utilizing a UIO-66 (Zr) MOF for sulfamethoxazole (SMX) degradation. This resulted in 93.8% degradation of SMX under visible light. Lastly, Pattappan et al. [11] utilized sunlight to degrade acetaminophen (AAP) and 2,4-dichlorophenoxyacetic acid (2,4-D) using a bimetallic FeCo-MOF synthesized using the solvothermal approach, exhibiting superior conversion rates compared to individual Fe- or Co-MOFs.

Table 1. Summary of commonly used and emerging synthesis approach and conditions for MOF synthesis.

Synthesis Method	Energy	Temperature	Reference
Solvothermal	Thermal Energy	120–150 °C	[10,13,14,17]
Hydrothermal	Thermal Energy	150–220 °C	[11,14,20]
Calcination	Thermal Energy	300–600 °C	[14,15]
Microwave	Microwave Radiation	30–150 °C	[16]
Ultrasonication/Solvothermal	Ultrasonic Radiation	30–80 °C	[17]
Ultrasonification/Biomimetic	Ultrasonic Radiation	30–40 °C	[20]
Facile Synthesis	No external Energy	30–40 °C	[19]
Carbonization	Thermal Energy	300 °C	[21]

In addition to conventional approaches, various synthesis methods as shown in Table 1, have emerged and been explored to speed up the formation process, create uniform crystals, increase yield, and adopt environmentally friendly practices [8,21]. Li et al. [17] employed rapid microwave heating to synthesize Fe_3O_4@MIL-100(Fe) for diclofenac (DCF) degradation, achieving more than 99.4% removal within 180 min under visible light. On the other hand, Salimi et al. [18] utilized both solvothermal and ultrasound-assisted approaches to develop MIL-125(Ti) mixed linker/g-C_3N_4 nanocomposites for cefixime (CXM) degradation under simulated solar light, showing 98% degradation after 120 min. Another study by Español et al. [20] employed ultrasound-assisted biomimetic synthesis to create MIL-100(Fe)/HAp nanocomposites for metformin (MET) degradation, achieving an average of 82.25% degradation within 120 min under ultraviolet light. Zhao et al. [19] prepared lamellate Co-MOFs via facile synthesis for ofloxacin (OFL) degradation, exhibiting 96.2% degradation efficiency, which is higher than that of the control photocatalyst. Lastly, Tsai et al. [16] developed Cu_xO/MOF nanocomposites via carbonization for ciprofloxacin (CIP) photodegradation, achieving up to 92% degradation efficiency under visible light for 120 min as shown in Table 2.

Table 2. Summary of MOF-based nanocomposites as photocatalysts for the photocatalytic degradation of pharmaceutical products.

MOF	Synthesis Method	Pollutant	Light Source	Degradation Efficiency and Time	Reference
MIL-53(Fe)	Solvothermal	Tetracycline Hydrochloride	Visible light ($420\ nm \leq \lambda \leq 780\ nm$)	99.7% in 80 min	[14]
MIL-53(Al)/ZnO	Hydrothermal	Amoxicillin	Visible light ($\lambda_{max} = 510\ nm$)	100% in 60 min	[13]

Table 2. *Cont.*

MOF	Synthesis Method	Pollutant	Light Source	Degradation Efficiency and Time	Reference
Fe_3O_4@MIL-53(Fe)	Hydrothermal/Calcination	Ibuprofen	Visible light ($\lambda > 420$ nm)	99% in 60 min	[14]
Diatomite-supported Hydroxyl-modified UIO-66(Zr)	Solvothermal/Calcination	Sulfamethoxazole	Visible light ($\lambda > 420$ nm)	93.8% in 120 min	[15]
FeCo-MOF	Solvothermal	Acetaminophen	Sunlight (75,000–80,000 lux)	97.4% in 180 min	[11]
		2,4-dichlorophenoxyacetic acid		79.8% in 180 min	
Fe_3O_4@MIL-100(Fe)	Microwave	Diclofenac	Visible light ($\lambda > 420$ nm)	99.4% in 180 min	[17]
MIL-125(Ti) mixed linker/g-C_3N_4	Ultrasonication/Solvothermal	Cefixime	UV-vis light ($320 \text{ nm} \leq \lambda \leq 780$ nm)	98% in 120 min	[18]
MIL-100(Fe)/HAp	Ultrasonication/Biomimetic	Metformin	Ultraviolet light ($\lambda_{max} = 460$ nm)	82.25% in 120 min	[20]
Co-MOF	Facile synthesis	Ofloxacin	Visible light ($\lambda > 420$ nm)	96.2% in 10 min	[19]
Cu_xO/MOF	Carbonization	Ciprofloxacin	Visible light ($\lambda = 465 \pm 40$ nm)	92% in 120 min	[16]

5. Conclusions

Research on MOF-based nanocomposites has undoubtedly demonstrated their excellent capabilities in the photocatalytic degradation of pharmaceutical products. Various synthesis methods, including conventional techniques like hydrothermal and solvothermal processes, as well as advanced approaches such as calcination, rapid microwave heating, and ultrasonication, have been employed to enhance the formation, uniformity, and yield of these nanocomposites. These efforts have led to improvements in photodegradation efficiency, chemical stability, and reusability of the nanocomposites against specific pharmaceutical pollutants. However, optimizing MOF-based nanocomposites still requires careful consideration of operational parameters like catalyst loading, pollutant concentration, pH, light source, light intensity, and exposure time.

Despite progress in this field, further investigation is necessary to fully harness the potential of MOF-based nanocomposites as photocatalysts. Enhancing their structural robustness and stability under diverse environmental conditions, ensuring sustained photocatalytic efficiency over multiple cycles, and reducing operational costs are critical areas for future research. Moreover, addressing challenges related to complex synthesis processes and low efficiency against certain pollutants will require innovative approaches to produce low-cost, highly durable nanocomposites.

Looking forward, future trends in the photocatalytic degradation of pharmaceutical drugs will likely involve the development of advanced MOF-based photocatalysts with improved properties, such as higher surface area, better light absorption, and greater stability under visible light. The integration of MOFs with other advanced oxidation processes and water treatment technologies may create hybrid systems offering synergistic effects for more effective degradation. Additionally, there is a growing interest in utilizing renewable energy sources, particularly solar energy, to drive photocatalysis, which could significantly reduce the environmental footprint of these technologies. The application of machine learning and AI to optimize photocatalytic processes, predict outcomes, and design new materials could also revolutionize the field.

Finally, the practical application of MOF-based nanocomposites for large-scale water treatment remains a challenge, necessitating further investigation into scalability and economic feasibility in pilot-scale systems. Through continued research and comprehensive exploration, it is anticipated that MOF-based nanocomposites will emerge as promising photocatalysts with significant potential to mitigate environmental challenges in the foreseeable future.

Author Contributions: Planning and Conceptualization, R.V.C.R. and A.N.S.; writing—original draft preparation, V.H.O.A., K.A.B., E.J.D.C.M., C.D.B.M. and R.J.C.P.; writing—review and editing, A.N.S.; supervision, R.V.C.R.; project administration, R.V.C.R. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

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

Conflicts of Interest: The authors declare no conflict of interest.

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