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Review Article

Advances in Solar-Photocatalytic Removal of Pollutants From Pharmaceutical Industrial Wastewater

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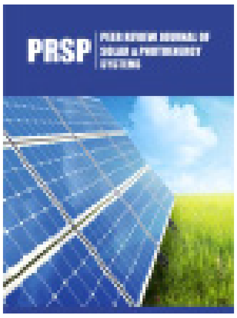

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Abstract

The countless existence of pharmaceutical pollutants discharged wastewater has been identified as a potential concern for aquatic creatures and humans. Despite the fact that their presence in drinking water has caused significant concern, little is known about their fate and environmental impact. As a result, these contaminants are inexorably pushed into our food chain even at low levels. Pharmaceutical effluents have a both direct and indirect impact on environmental and human health, especially in the vicinity of pharma industrial zones. Therefore, it is important to remove pharmaceutical pollutants from wastewater before final discharge in the environment. Among various treatment, photocatalysis has been widely regarded and gained popularity in recent years as a potential technology for removal of variety of environmental contaminants. The present mini review mainly focused on the feasibility of photocatalysis process for its potential to treat pharmaceutical wastewater. Moreover, the emphasis is given to the solar based (visible light active) photocatalyst such as C_3N_4 and its composites.

Keywords: Pharmaceutical pollutants; Photocatalysis; Nano-Composites; C_3N_4; Wastewater

Introduction

Pharmaceutical pollutants have a direct impact on human health because they have been discovered in our food chain, which includes fruits, vegetables, and drinking water [1,2]. The pharmaceutical department has emerged as one of the key benefactors of this trend, as the use of pharmaceuticals and public-health products has been made more affordable and accessible. Pharmaceuticals medicines play very important character for people life insurance and life quality because they are used in curing various health concerns for instance; pregnancy prevention, stress, both mental and physical, fever, infection and agricultural growth every year [3,4]. Every year, the rate of use of pharmaceutical drugs has been continuously increasing globally by 11.9 % such as in USA, France, Japan, United Kingdom, Spain, and Italy etc. [2,5]. Due to this, pharmaceutical contaminants are found in aquatic habitats and animal tissues [6]. The vital downside connected with such a vast utilization of pharmaceutical objects is their unlimited excretion into both marine and terrestrial ecosystem [1]. One of the most common contaminants is fluoxetine antidepressants, which may affect behavioral and physiological processes in non-target individuals [6]. Moreover, a considerable concentration of endocrine disrupting chemicals is also classified among the pollutants that are on the rise [1].

Pharmaceutical chemicals typically enter natural water systems through the overflow of different non-point water sources, such as agricultural activities or static water supplies, such as municipal and hospital wastewater treatment plants [7] (Okoye et al., 2022). In human drinking water there are possibility of presence of pharmaceutical compounds consists of various sources such as pharmaceutical production business, process of production, use and application by human and veterinary. In the above context, it is important to remove the pharmaceutical compounds from water in order to protect its secondary impact on human and environmental health. Several conventional wastewater cure methods for detach pharmaceutical compounds have been notified like activated sludge, adsorption, ozonation, wetlands, and micro-algae [1]. Biological treatment methods are wieldy applied for the treatment of industrial wastewater. The utilization of activated sludge and trickling filters for the disposal of pharmaceutical wastewater proved ineffective, resulting in the wastewater being released into the environment and constantly polluting surface, soil, and groundwater [7]. Furthermore, traditional biological wastewater treatment systems remain in short supply for entirely separating refractory toxins from pharmaceutical wastewater (Inyang et al. 2016). Although, the primary treatment techniques for pharmaceutical wastewater treatment are physio-chemical and biological methods, however, the biological treatment is less expensive, but it is less efficient in carbon-based pollutants in wastewater.

Photocatalysis has gained popularity in recent years as a potential technology for resolving vitality issues and preventing environmental pollution [8-13]. Photo-catalysis is a form of chemical reaction that is extremely advantageous due to its ease of use, inexpensive safety, effective degradation, and perfect stability. The primary application areas in catalysis, particularly in wastewater treatment, include photocatalytic electrolysis, environmental protection, solar cells, storage apparatus, and others. In case of wastewater treatment, TiO_2 photo catalysis has been proved to efficiently degrade the organic pollutants [2,14]. The catalytic activity of TiO_2 is based on its photoelectric characteristics and electrical structure. The photo-catalytic response principle is defined using the band theory in which the catalyst is activated under the continuous illumination of light with the energy equal to or higher than the band gap energy of catalyst, resulting in generation of a hole-electronic pair for oxidation and reduction of pollutants.

Environmental Impacts of Pharmaceutical Pollutants

Pharmaceutical substances are utilized for a variety of beneficial uses in industrial life, but they often release highly toxic chemicals into the atmosphere, either directly or as a result of chemical changes [15]. Pharmaceutical medications, both for medical and veterinary medicine, are posing a threat to the environment. Analgesics, antibiotics, antiepileptics, antiseptics, beta-blockers, antihypertensive, hormones, contraceptives, psycho therapeutics, and antivirals are some of the pharmaceuticals that have been listed. Any medications in the atmosphere even at low concentrations will damage aquatic species [16]. Diclofenac was then extensively researched in other bird species (pigeons and hens), with death rates of 0.25mg kg1 in pigeons and 2.5mg kg^1 in chickens [17]. It's thought that diclofenac's toxicity causes an increase in reactive oxygen species formation and, as a result, a shift in uric acid metabolism (creation and elimination) [18] Fish acute toxicity values are expected to reach 100mg l 1. During a 28-day test, however, it was revealed that at a dose of 1mg l 1, harmful effects on rainbow trout may already have begun. River trout have been shown to be harmed by 50mg/1 dosages (Hoeger et al. 2005). The conversion of these medications to compounds with features similar to their maternal molecules and the ability to bio-accumulate in the tissues is also a concern for the environment [19]. Under laboratory conditions, carbamazepine has an effect on a zebra mussel *(Dreissena polymorpha)* [20]. They discovered that diclofenac in the vultures' food web contributed to their extinction [21]. Pharmaceuticals enter the atmosphere through processing units and patient effluents, as well as applications for land (e.g., bio-solids and water reuse).

Sewage treatment services, on the other hand, aren't always effective in separating active chemicals from waste water. As a result, pharmaceuticals end up in the marine ecosystem, where they have direct effects on aquatic life and can enter food chains. Several drugs were detected in extremely high amounts (mg/L) in effluents from a local wastewater treatment plant near Visakhapatnam, India, in a recent report [15]. Trace level concentrations have also been found in microorganisms, fish, mollusks, and rodents, but in fewer publications. The pharmaceutical industry's waste water has a strong tint, a pungent taste, a low BOD and a high COD.

The various wastewater channels from the active pharmaceutical sector, unpackaged drugs, and associated pharmaceuticals, all of which use a lot of water, must be recognized, and the best technologies for eliminating them must be analyzed in order to increase public quality and water supplies [7]. Researchers have identified high quantities of pharmaceutical chemicals in a variety of wastewater types, including ground and surface waters, as well as drinking water sources (Yang et al.,2014). As a result, the environmental influence and public health threats associated with these forms of wastewater are being studied by experts all over the world.

Pharmaceutical compounds typically enter natural water supplies through source of non-point water such as agronomic activity or source of constant water such as wastewater treatment plants in municipalities and hospitals [7] (Okoye et al., 2022). Massive concentrations of heavy metal ions, radionuclides, toxins, products of personal care, and chronic chemical chemicals are eventually released into the natural world as a result of the rapid growth of industry, urban growth, and cultivation.

Photocatalysis Process

Photo-catalysis has been widely regarded in recent years as a potential technology for resolving vitality issues and preventing environmental pollution. Photo-catalytic electrolysis of water, environmental protection, solar cells, storehouse apparatus, and so on are the main application areas in catalysis, especially in wastewater treatment. Organic pollutants such as pesticides, dyes, and chemical waste can be found of large quantities in wastewater, causing severe damage to biological health and ecological practices. Organic contaminants that are particularly toxic and slow to degrade (such as contaminants, fertilizer, and dyes) have a long half-life, and even their exploration may cause biological differences. As a green catalytic mechanism, TiO_2 photo-catalysis will almost fully degrade all organic pollutants. TiO_2 photo-catalytic methods can degenerate more than 3000 forms of problems decomposing organic composites, according to research [22].

The examination of visible-light active semiconductor photo-catalysts has obtained great use in environmental nanotechnology. Several conventional wastewater cure methods for detach PhACs and EDCs have been notified such as activated sludge, adsorption, wetlands, ozonation, and micro-algae [1]. Photo-catalysis is a form of chemical reaction that is extremely advantageous due to its ease of use, low cost, safety, high degradation efficiency and perfect stability. TiO_2's catalytic activity is based on its electronic structure and photoelectric properties. The photo-catalytic response principle can be defined using the band theory. The photo-catalytic response principle can be defined using the band theory. When exterior TiO_2 is illuminated with light that is consistently equal to or greater than the band gap energy of TiO_2, the surface is stimulated, and a hole-electronic pair is formed, which allows for oxidation and reduction. The general mechanism of the photocatalysis process is shown in Figure 1.

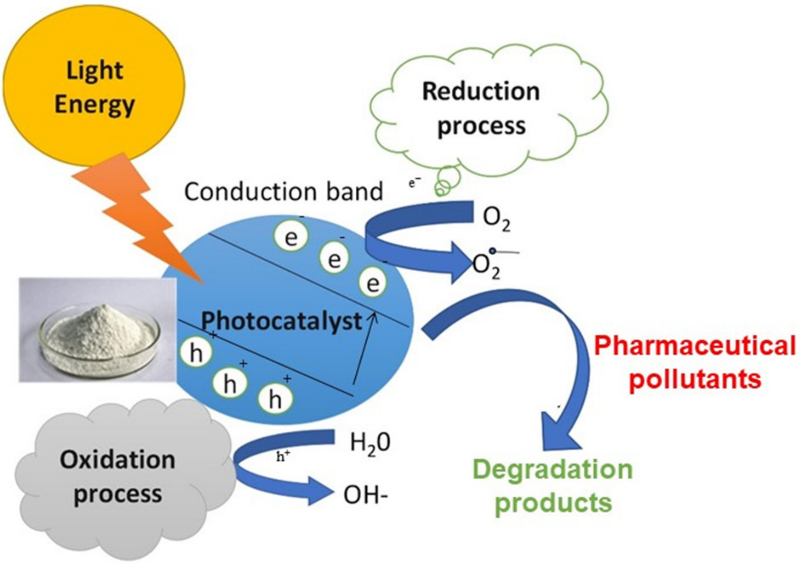

Figure 1: Photocatalysis general mechanism.

C_3N_4 Based Photocatalysts

Since its ground breaking use in photo-catalysis in 2009, graphitic phase carbon nitride (g-C_3N_4) has become a research hotspot as a novel kind of nonmetal and stable polymeric semiconductor content [23,24]. Exfoliation of the bulk structure of g-C_3N_4 is one of the techniques for improving its photo-catalytic performance. Exfoliation increases the basic surface area of these g-C_3N_4 structures, which improves their photo-catalytic activity greatly [25]. The use of g-C_3N_4 for photo-catalytic deterioration of the anti-inflammatory pharmaceuticals (DIC, PAR, and IBU) is still being studied. Diclofenac degradation is reported in just a few articles [26]. The g-C_3N_4 as a traditional synthetic material has become a hot topic in the field of chemistry and material science, particularly for photo-catalysis, due to its low cost, ease of preparation, strong stability, and unusual physicochemical properties [27,28]. The g-C_3N_4 materials are successfully formed by air flow rate increases with small organic molecules that are nitrogen-rich precursors, such as urea, cyanimide, melamine, dicyanamide and thiourea, and the form of tectonic units depends primarily on the reaction processes [29]. The most successful method for purifying wastewater contaminated by organic contaminants is the adsorption and photo-catalytic oxidation of organic molecules under visible light irradiation. The structure properties and adsorption capacities, in general, dominate photo-catalytic capacity. As a result, the majority of the literature on the elimination of organic compounds focused on adsorption and photo-catalytic degradation. The photocatalytic performance of various C_3N_4 based photocatalysts is summarized in Table 1; [30-37].

Table 1: Photocatalytic removal of various pollutants by modified C_3N_4 photocatalysts.

Photocatalysts	Light Source	Pharmaceutical Pollutant	Removal (%)	References
g-C_3N_4/Bi_2MoO_6/Bi_2WO_6	Visible light	tetracycline	98	Sun et al. [30]
Cu doped TiO_2/g-C_3N_4	Ultra violet	Methylene blue	98	Liyanaarachchi H et al. [31]
Fe-Mo-O doping g-C_3N_4	Visible light	Rhodamine B	99	Kong H et al. [32]
Bi_2WO_6/pg-C_3N_4	Visible light	17 β-estradiol f	99.5	Qing et al. [33]
g-C_3N_4@CdO/ZnO	Ultra violet	Methyl red	97.7	Berhanu et al. [34]
B-doped g-C_3N_4	NR	4-chlorophenol	100	Jing et al. [35]

$g\text{-}C_3N_4/ZnS$	Solar light	Bisphenol A	92.1	Ma et al. [36]
TiO_2/Zr-doped $SiO_2/g\text{-}C_3N_4$	Solar light	Berberine hydrochloride	98.11	Yu et al. [37]
		Tetracycline	80.76	
		Oxytetracycline	84.84	

Modification of Catalyst for Visible Light (Solar) Photocatalysis

Since it can induce the formation of hydroxyl radicals from H_2O_2, graphitic carbon nitride ($g\text{-}C_3N_4$) is regarded as a Fenton-like catalyst. $g\text{-}C_3N_4$ is also a widely used photo-catalyst for water splitting and organic degradation (Li et al., 2016; Dong et al., 2017). However, the poor separation performance of charge carriers as photo-catalysts and the low utilization efficiency of H_2O_2 as a Fenton-like catalyst limit g-use C_3N_4's [38,39]. While alkalinization and metal ions alteration will improve the Fenton-like catalytic and photo-catalytic capability of $g\text{-}C_3N_4$, the redox potential is still limited from a thermodynamic perspective. The use of Z-scheme hetero-junctions can increase the spectrum of light absorption, facilitate charge carrier separation, and improve photo-catalyst redox efficiency. Numerous Z-scheme hetero-junctions based on $g\text{-}C_3N_4$, such as $g\text{-}C_3N_4$@C-TiO_2 and $g\text{-}C_3N_4/Ag_3PO_4$/AgI, have been developed [40,41]. Replication of holes (h+) in the Valence Band (VB) of $g\text{-}C_3N_4$ with Electrons (e-) in the Conduction Band (CB) of another semiconductor gives the e- in the CB of $g\text{-}C_3N_4$ and the h+ in the VB of another semiconductor super removing and oxidizing capabilities, simultaneously. Using a calcination - impregnation process, we formed a novel Z-scheme MnO_2/Mn-modified alkalinized $g\text{-}C_3N_4$ catalyst (MnO_2/CNK-OH-Mn). MnO_2/CNK-OH-Mn displayed strong Fenton-like photo-catalytic activity, with high Tetracycline (TC) degradation and TOC removal performance, as well as COD extraction efficiency in pharmaceutical wastewater [42].

TiO_2 modified C_3N_4

The photocatalytic degradation of paracetamol, ibuprofen, and diclofenac was investigated using TiO_2 nanoparticles as well as exfoliated $g\text{-}C_3N_4$. Their aqueous solutions were first exposed for 2 hours to UV and VIS radiation. When the adsorption–desorption equilibrium between medicines and nanomaterials was achieved, the photocatalytic studies began (after dark). The medicines did not undergo photolysis [43]. Because of low toxicity, high durability, low price, and environmental advantages, photo-catalysis has gotten a lot of publicity. As previously said, titanium dioxide (TiO_2) is a prototype photo-catalyst that has been used to decompose a wide range of pollutants [44,45]. However, because of their ease of agglomeration, limited UV activity, and high recombination rate of photo-generated e-h+ pairs, pristine TiO_2 nanoparticles have some limitations in practical use [46,47] (Li et al., 2018a). As a result, the semiconductor/TiO_2 construction may be considered to increase photo-catalytic activity.

This photo-catalyst is powered by visible light and has a suitable band gap (2.70eV) [48]. This carbon-doped supramolecule-based $g\text{-}C_3N_4$ (BCCN) and TiO_2 hybrids can therefore be used to improve visible-light-driven photo-degradation pharmaceuticals. LED lamps, on the other hand, seem to be promising sites of pollutant degradation [49,50]. An in-situ process was used to fabricate an eco-friendly 2D hetero-junction photo-catalyst composites (BCCNT) made up of carbon-doped supra-molecule-based $g\text{-}C_3N_4$ (BCCN) layers and TiO_2 nano particles (Hu et al., 2019). The photo-catalytic degradation of paracetamol (PAR), ibuprofen (IBU), and diclofenac was studied using exfoliated graphitic carbon nitride ($g\text{-}C_3N_4$) and two commercially usable titanium dioxide nano materials (P25 and CG300) (DIC) [43].

Due to the breakdown of intermediate intermediates, a slightly pinkish solution was noticed after 2–3h during diclofenac photodegradation, which disappeared with a longer irradiation period. There was no discernible effect in the case of paracetamol. In the sequence $g\text{-}C_3N_4$ CG300 P25, photocatalyst degradation effciency for all medicines increased. When considering the wavelength of 368nm and the band gap energies of TiO_2 nanoparticles, this was not surprising. Because of its rapid photoinduced electron-hole recombination, $g\text{-}C_3N_4$ has the lowest photodegradation activity. The degradation effciency of $g\text{-}C_3N_4$ was around 77 percent (VIS) and 7% (UV), which corresponded to a band gap energy of 2.70eV. (459nm). As a result, $g\text{-}C_3N_4$ should be active in both UV (368nm) and VIS (446nm). Because none of these compounds absorbed visible light at 446nm, the photodegradation in the presence of TiO_2 could not be attributable to sensitization by the adsorbed medicines, as was commonly reported in the case of different dyes. The VIS light absorption of both TiO_2 nanomaterials, as a result of structural flaws such as oxygen vacancies, most likely the cause. The oxygen vacancies are inherent defects that produce intermediate energy levels inside the TiO_2 band gap and function as a recombination site for photo induced electron and hole recombination. The absorption spectra of TiO_2 suspensions, both pure TiO_2 nanomaterial and their mixtures, with the medication are displayed [43]. In order to get the right quantifiable absorbance, the P25 solution was diluted 60 times and the CG300 suspension was diluted 15. Before the photocatalytic process began, the spectra were collected after 1 hour of darkness. TiO_2 absorbed visible light in both cases, most likely due to electron transfer from their valence band to the intermediate energy level of oxygen vacancies.

Conclusion

In the current environment, modern water technologies are required to maintain high water quality, remove chemical and biological contaminants, and accelerate industrial waste production processes. Photocatalysis is one of the best alternatives for advanced wastewater treatment in this regard. For wastewater treatment, several nano-materials have been effectively produced and researched. Under the influence of visible light, a novel visible light photocatalyst C_3N_4 effectively synthesized, with the capacity to degrade antibiotics and remove pathogens from pharmaceutical industrial wastewater. Nano-particle photo-catalysts can be utilized to treat both hazardous contaminants and heavy metals, with the capacity to employ visible sunlight instead of expensive artificial UV radiation due to changes in catalyst material. The catalyst C_3N_4 demonstrated excellent photo-catalytic activity as well as good stability, thus can be used for effective treatment of wastewater.

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