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REVIEW

Photocatalytic Degradation of Phenolic Pollutants by Nanocomposites: A Systematic Review and Pooled Analysis

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Abstract

All living things depend on water, which is a precious natural resource. Modern hydropower generation, industrial processes, and transportation increasingly rely significantly on water. Emerging contaminants are currently posing a serious threat to our environment and significantly damaging human health. To address these water-related problems, photocatalysis, one of the advanced oxidation processes, has drawn a lot of attention. The most common photocatalytic approach for the elimination of phenolic pollutants, which is one of the emerging pollutants in the aquatic environment, was determined through a systematic review of the scientific literature in the current study. Furthermore, It was thought about how independent variables like pollutant concentration, catalyst amount, radiation time, pH, and contact time might affect the process. Twelve research, all of which focused on phenolic contaminants, were included. In two studies, all 12 phenolic contaminants were eliminated. Most of the pollutants exhibited a degradation efficiency above 90%. The removal of organic contaminants from water can be accomplished with efficiency and effectiveness by using sophisticated photocatalytic treatment methods. However, the combined data support photocatalytic treatment procedures as a new technique in recent years for the removal of organic contaminants.

Keywords: Photocatalysis, Advanced oxidation processes, Organic pollutants, Emerging pollutants

1. Introduction

Water is a treasured natural resource and vital for all living creatures. In modern years water is very much essential for hydropower generation, industries, and also for conveyance purposes. Yet, water is being polluted and contaminated indiscriminately on a day-to-day basis with the disposal of untreated domestic wastewater, agricultural runoff, and hazardous toxic pollutants to the nearby freshwater ecosystem [1–5]. Recently, emerging organic pollutants are posing a serious threat of water pollution, which resulted in a shortage of drinking water and many other environmental problems [6,7]. Wholesome water is a vital prerequisite for human beings and presently its availability has grown into a major environmental issue currently due to increasing population, urbanization, and industrialization [8–12]. Generally, phenolic compounds of measurable concentrations are present in wastewater. The sources for these compounds are mainly oil refineries, hospitals, fossil fuel gasification, pharmaceutical industries, herbicide manufacturing units, pharmaceutical industries, and improper disposal of pharmaceuticals. A minimum concentration of 0.005 ppm of phenol and its derivative presence in water is not only toxic but adds an odd odor and taste [13,15]. These pollutants together cause a wide range of risks to environmental health. Water treatment is

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mostly on conventional treatment methods that consist of treatment units such as screening followed by clariflocculation, then sand filtration with disinfection. These treatment units are not efficient in the further removal of organic contaminants and their by-products which have the potential of causing damage to the ecosystem as well as other living species. and hence they limit their application [16]. Thus, it is imperative to find an eco-friendly, renewable, and effective treatment method effective, to expel these organic pollutants from water [17,18]. Due to this, Photocatalysis one of the Advanced Oxidation Processes has attracted more focus and consideration for the degradation of environmental pollutants, to solve the energy crisis, and for the conversion of a hydrocarbon to CO_2 [19–22]. Hence Photocatalysis with the application of nanocomposites is an appealing option, as it minimizes the chemical addition with reduced waste generation and can utilize renewable solar energy to drive the reactions [23–25]. In this method, a semiconductor will be illuminated by photons energy greater than the band gap energy of the electrons ($e-$). due to which electron shift from the valence band to the conduction [26–28]. Then the h^+ will react with water molecules (H_2O) to spawn hydroxyl radicals ($\cdot OH$), along with the release of superoxide radicals ($\cdot O_2$-) by reducing the dissolved O_2. These active radicals due to their excellent redox capability engage in the mineralization of organic molecules [29,30]. The study mainly aims to systematically review the most relevant works about photocatalysis using different nanocomposites for the removal of phenolic contaminants in aqueous media.

2. Methodology

2.1. Literature sources and search strategy

A systematic review was carried out to review the studies related to the photocatalysis process for the removal of organic pollutants by nanocomposites. Research articles published from 2011 to 2021 were selected for the review study. To fulfill this objective, a comprehensive search was made in all available information resources such as Google Scholar, Scopus, Science Direct, Springer, and PubMed. The search included the following keywords: (photocatalysis) OR (photodegradation) AND (nanocomposite) AND (organic pollutant) OR (emerging pollutant) AND (water) OR (wastewater) AND (treatment). With the following inclusion and exclusion criteria described below, we selected the most suitable studies for the review.

2.2. Inclusion and exclusion criteria

In the present study, only research articles were included, which were published in English and selected from peer-reviewed journals about emerging organic pollutant removal through different types of nanocomposites, alone or in combination with other compounds.

Also in this study, articles with duplicate content, unrelated topics, reviews, book chapters, presentations, meta-analyses, and conference papers were all excluded from the study.

2.3. Screening and selection of criteria

Initially, the duplicate research journal articles were excluded from the study manually. Furthermore, research articles were meticulously examined, and irrelevant papers were excluded from the remaining review research study. The research articles selected for the study followed this pattern: Titles, author, publication year, the nanocomposite, type of organic pollutant, pH, catalyst dosage, initial concentration of the pollutant, contact time, and percentage degradation.

3. Result and Discussion

3.1. Characteristics of literature

Firstly, a total of 482 articles were collected from Science Direct (n = 127), Scopus (n = 117), Google Scholar (n = 101), Springer (n = 98), and PubMed (n = 39) considering the mentioned search strategy. 26 documents were excluded from the study manually because of duplication. Fig. 1 depicts the PRISMA flow diagram for the search and selection of the papers. The remaining 456 research articles were investigated with their research topic, abstracts, and complete article, of which, 419 documents were relinquished considering the mentioned exclusion criteria. Finally, 12 significant research articles were selected for this systematic review of the removal of 12 phenolic pollutants by nanocomposites investigated in the consistent articles.

With increased population growth, industrial development, inflation of urban areas and have substantially increased water, food, and other natural resources demand. Global Climate change along with the overuse of water resources and reduction of water resources with man-made water pollution have heightened the concerns regarding the supply of portable or wholesome water globally. Water pollution due to anthropogenic activities is

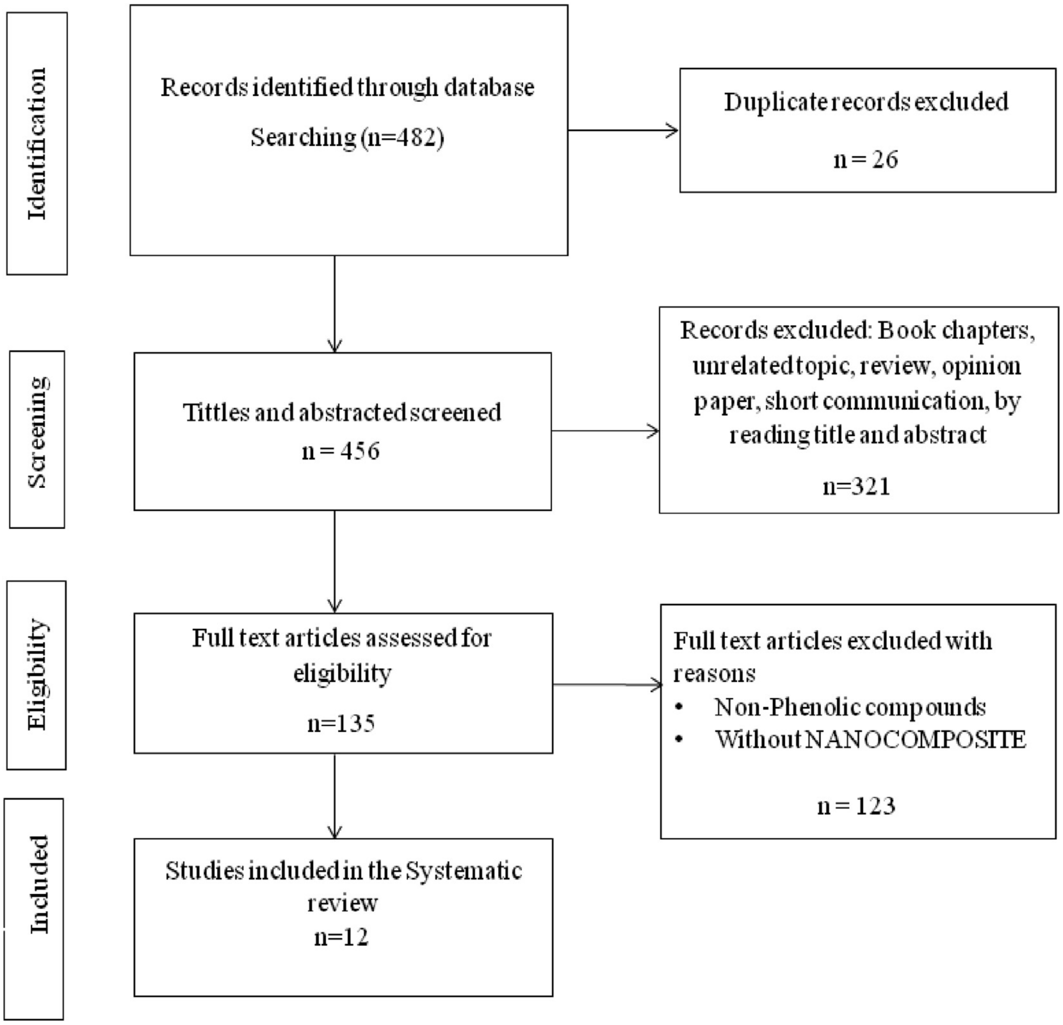


Fig. 1. PRISMA flow diagram for the search and selection of papers.

posing a threat to the natural environment and human life in danger. Wastewater released from agricultural runoff, industrial effluent, and domestic wastewater becomes a primary contributor to major environmental problems such as eutrophication which in turn leads to dissolved oxygen depletion in natural water bodies. This condition can imperil aquatic life, human health, and wildlife [30]. It is globally estimated that around 359 billion cubic meters of wastewater are produced each year and that about 80% of the global wastewater without any treatment is disposed of on a larger scale to nearby water bodies, ultimately polluting rivers, lakes, and oceans. Thus due to this increased water pollution is posing a threat to human health.

Some degree of treatment is usually essential to release these effluents or to reuse the wastewater without hampering any change to the natural ecosystem and human health. Numerous technologies have been employed for the same. Conventional water or wastewater treatment strategies, which usually consist of several steps to reach a desirable level of purification implement biological, chemical, and physical processes combined to detoxify or remove harmful nutrients, chemicals, toxic ions, and solid particles. However, conventional technologies are unable to fulfill such high expectations of treating wastewater due to their limitations. In recent years, various novel approaches have been extensively studied for wastewater treatment. A vast number of alternative materials have been recently synthesized to be used as membrane filters, adsorbents, ion exchangers, catalytic surfaces, etc. for water purification [17,25]. Nanomaterials and polymers are at the forefront of emerging materials employed for efficient wastewater treatment. They have functional characteristics that can be tuned as per the needs of different wastewater treatment strategies.

The lists of the 12 references are taken for this study based on wastewater treatment using varied nanomaterials and nanocomposites. The common physical and chemical factors picked up from these studies include Catalyst Dosage, Pollutant Concentration, pH, Contact Time and Degradation percentage are the factors that are being taken under consideration for this study shown in Table 1. Varied pollutants are considered under the study which include 2,4- dimethylphenol, 2-chlorophenol, pentachlorophenol, bisphenol A, polychlorinated biphenyls, phenol, 4-nitrophenol, 2,2′,4,4′,5,5′-hexachlorobiphenyl, 2,4-Dinitrophenol, and aniline.

The present study takes into consideration four important factors across 12 studies, The Mean and Standard Deviation values for the factors being Catalyst dosage are 471.67 ± 366.73, Pollutant Concentration is 19.93 ± 14.08, pH is 6.00 ± 2.56, Contact time is 122.08 ± 66.66 (mg/L) and 95.70 ± 4.13 for Degradation percentage shown in Descriptive Statistics Table 2. Degradation is the important factor among the four factors as it provides us with the removal efficiency of the considered nanocomposite.

Table 3 represents the association among different factors considered in the study. Pearson's correlation coefficient was indicating a small association between the variables such as Catalyst Dosage and pH ($r = 0.234$), Pollutant Concentration and pH ($r = 0.117$), Catalyst Dosage and Contact Time ($r = 0.108$), pH and Contact Time ($r = 0.120$), Catalyst Dosage and Degradation ($r = 0.151$), Contact Time and Degradation ($r = 0.079$). There are small negative correlations among the remaining values which include Catalyst Dosage and Pollutant Concentration ($r = -0.019$), Pollutant Concentration and Degradation ($r = -0.138$), pH and Degradation ($r = -0.464$). A correlation of $r = 0.624$, which falls in the medium range of correlation between two factors, indicates that there is a positive correlation between Contact Time and Pollutant Concentration i.e., as Pollutant Concentration there would be an increase in Contact Time.

Degradation showed a slightly increased trend with Catalyst dosage in Fig. 2(a) and Contact time in Fig. 2(d). The fitted linear equation $\widehat{y} = 94.9 + 0.0017 (x)$; R SQUARED = 0.023, for Catalyst dosage and $\widehat{y} = 95.1 + 0.0049 (x)$; R SQUARED = 0.006 for Contact time, which indicates the percentage of change in the independent variable i.e., Degradation is explained by the dependent variable, it is comparatively smaller according to standards. There is a negative linear trend in both 2(b) and 2(c) i.e., with Pollutant Concentration and pH. From the result, the linear equations are $\widehat{y} = 96.51 - 0.04 (x)$; R

Table 1. Characteristics of studies included.

Study reference	Nanocomposite material	Light Source	Catalyst Dosage (mg)	Pollutant Concentration (mg)	Contact Time (mins)	pH	Degradation (%)
A.A. Parwaz Khan et al., (2020) [4]	20% $Bi_2O_2CO_3$/CNT/$ZnFe_2O_4$	Visible light	50	12.2	120	4	99
M. Anjuma et al., (2018) [20]	C_3N_4/TiO_2 NTs	Visible light	500	30	180	7	96.6
F.S. Arghavan et al., (2020) [21]	$FeNi_3$/SiO_2/ZnO	UV irradiation	500	10	180	3	100
Yu-Hsin Chen et al., (2021) [22]	GCN/Rgo-0.25%	Visible light	1000	5	30	7	97
Chenguang Li et al. (2021) [23]	N-Doped SiO_2-300	Visible light	940	37.5	240	11	97.3
Yun He et al., (2021) [24]	Ag/AgBr—Al-attapulgite	Visible light	100	10	120	10	86.5
M.Khatamian et al., (2011) [25]	7 wt % ZnO/HZSM-5	UV irradiation	500	20	90	5	91
Mingmei Ding et al., (2021) [26]	10% CTM/PMS	Visible light	20	20	15	7	96
J. Rashid et al., (2014) [27]	Fe_3O_4/SiO_2/TiO_2	UV irradiation	500	25	130	3	100
Ahmed Uddin et al., (2021) [28]	20%-In_2O_3/OGCN	Visible light	500	50	180	5	91
Vasudha Hasija et al., (2020) [29]	SGCN/Ni—Fe LDH	Visible light	50	18.4	120	4	98
Le Xua et al., (2018) [30]	TiO_2-x/rGO	Visible light	1000	1	60	6	96

Table 2. Descriptive statistics of all factors.

Factors	Minimum	Maximum	Mean	Std. Deviation
Catalyst Dosage (mg)	20	1000	471.67	366.73
Pollutant Concentration (mg)	1	50	19.93	14.08
pH	3	11	6.00	2.56
Contact Time (mins)	15	240	122.08	66.66
Degradation %	87	100	95.70	4.13

Table 3. Correlation matrix of different variables.

Factors	Catalyst Dosage (mg)	Pollutant Concentration (mg)	pH	Contact Time (mins)	Degradation %
Catalyst Dosage (mg)	1				
Pollutant Concentration (mg)	−0.019	1			
pH	0.234	0.117	1		
Contact Time (mins)	0.108	0.624	0.120	1	
Degradation %	0.151	−0.138	−0.464	0.079	1

SQUARED = 0.019 for contact time and $\hat{y} = 100-0.75\ (x)$; R SQUARED = 0.216 for pH, which indicates a 1.9% change in degradation is explained by Pollutant Concentration and 21.6% by pH respectively.

3.2. Photodegradation of phenolic pollutants by nanocomposites

Phenolic compounds categorized in this class include 2-Chlorophenol, Bisphenol A, Phenol, 2,4-dimethylphenol, Pentachlorophenol, Polychlorinated biphenyl, 4-nitrophenol, 2,4- dinitrophenol and Aniline.

3.2.1. 2-Chlorophenol

Among these 20 studies, 4 studies were conducted on the degradation of 2- chlorophenol. The maximum and minimum levels of degradation efficiency of 2-chlorophenol were found to be 100% and 80%, respectively. Total degradation of this pollutant was recorded using $Fe_3O_4/SiO_2/TiO_2$ and TiO_2eRGOeCoOnanocomposites respectively [12,22]. Furthermore, 96.6% degradation efficiency was obtained by C_3N_4/TiO_2 nanocomposite [12]. The least degradation efficiency was 80% obtained by using Ni $(OH)_2$/graphene oxide/TiO_2 [13].

3.2.2. Bisphenol A

The degradation of Bisphenol A using different nanocomposites was carried out in 4 different studies. The highest level of degradation efficiency of Bisphenol A was found to be 97% by GCN/rGO-0.25% [9]. Whereas the lowest degradation efficiency of 91% occurred by 20%-In2O3/OGCN[28]. However, 94% degradation was obtained by ZnO@FeHCF [14], and 96% degradation efficiency was observed by TiO2−x/rGO nanocomposite [30].

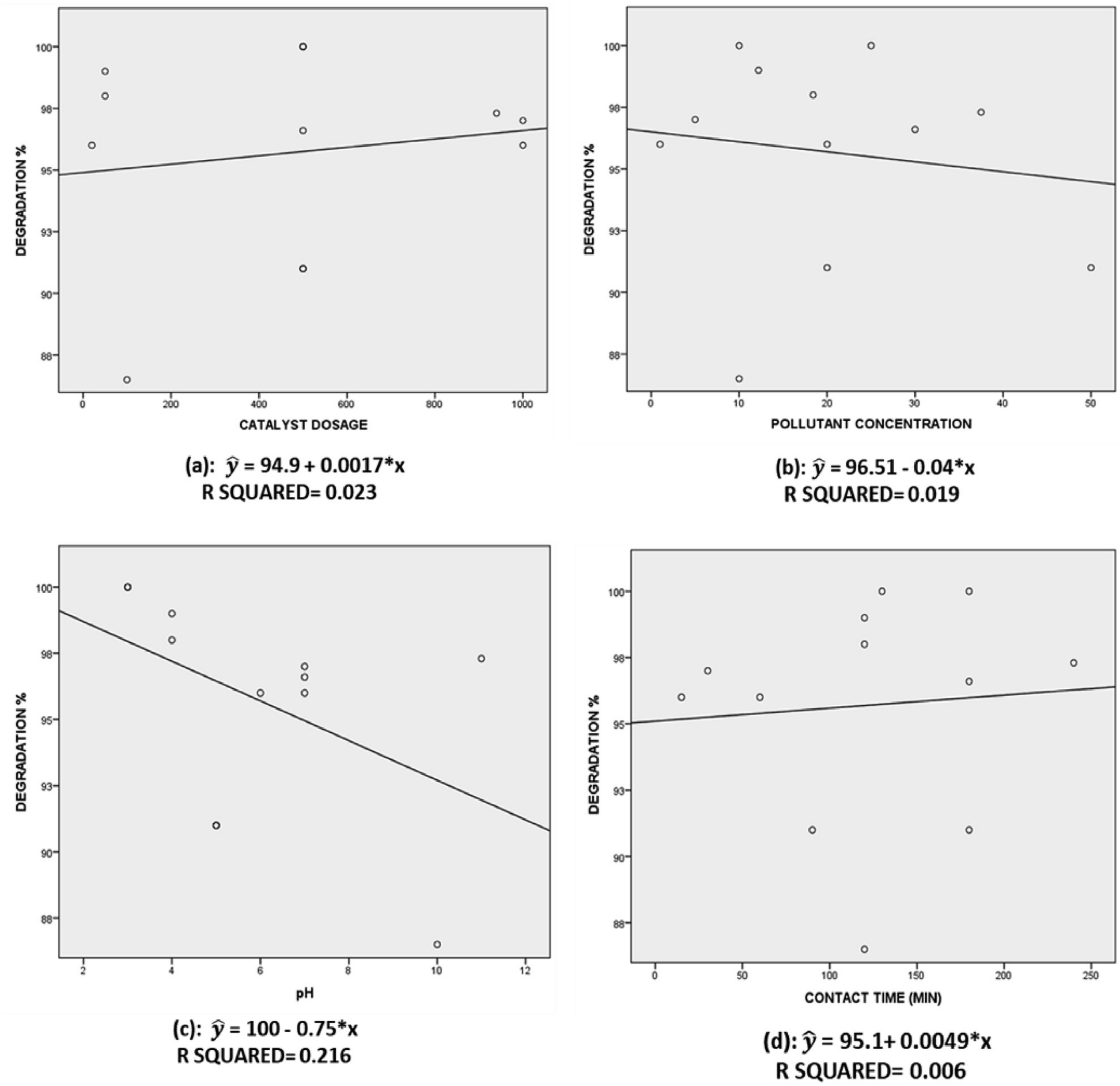
3.2.3. Phenol

Among twenty studies that are included in the phenolic pollutant study, 5 studies were found to be conducted on phenol as a target pollutant in water and wastewater. 20 mg of FDU-PdPcS nanocomposite showed the highest photodegradation efficiency of 98% in alkaline conditions with a time duration of 660 min [15]. Moreover, 10% CTM/PMS, Ag/AgBr—Al-attapulgite, ZnO—bentonite and Fe3O4cluster@SiO2@TiO2—N nanocomposites achieved 96%, 86.5%, 70% and 46 ± 1.5% accordingly [16,17,24,26].

3.2.4. Polychlorinated biphenyl

Polychlorinated biphenyl is one of the phenolic compounds which is often found in wastewaters. Here, two studies reported the photocatalytic degradation of polychlorinated biphenyl. N-doped SiO_2 nanocomposite exhibited 97.6% of degradation efficiency of (PCB-209) at an alkaline medium of pH 11 [23]. Whereas, 96.5% of 2,2′,4,4′,5,5′- hexachlorobiphenyl (PCB 153) was degraded by $Fe_3O_4@SiO_2@TiO_2$ at the highest catalyst dosage at an acidic medium of pH 5 [18].

In addition to these pollutants, five studies were conducted by the researchers on five different phenolic pollutants. The 20% $Bi_2O_2CO_3/CNT/ZnFe_2O_4$ nanocomposite exhibited 99% degradation efficiency of 2,4-dimethylphenol [4]. A complete degradation of Pentachlorophenol was achieved by $FeNi_3/SiO_2/ZnO$ nanocomposite [21]. 91% of 4-nitrophenol was degraded by 7 wt.% ZnO/HZSM-5 nanocomposite [25]. SGCN/Ni—Fe LDH exhibited 98% degradation efficiency of 2,4-Dinitrophenol



(a): $\hat{y} = 94.9 + 0.0017*x$
R SQUARED= 0.023

(b): $\hat{y} = 96.51 - 0.04*x$
R SQUARED= 0.019

(c): $\hat{y} = 100 - 0.75*x$
R SQUARED= 0.216

(d): $\hat{y} = 95.1 + 0.0049*x$
R SQUARED= 0.006

Fig. 2. Regression analysis of degradation with other factors.

[29]. Also, RGO/ZnO/MoS_2 showed complete removal of Aniline in the reaction medium [19].

3.3. Degradation pathway of phenolic pollutant

With the aid of a photocatalyst, the phenolic contaminants are broken down systematically by UV and visible light. After absorbing the proper photons, the catalyst typically undergoes photo-activation. Reactive oxidants are produced by the photocatalyst's redox reactions at the conduction and valence bands, which act on the phenolic pollutant to break down the molecule and eventually form simple organic compounds like water and carbon dioxide. The process of phenolic pollutant photodegradation using nanoparticles or nanocomposites is shown in Figs. 3 and 4 with UV light and visible light sources respectively. The dehalogenation and ring fragmentation, which create a few intermediates and ultimately lead to intermediates, water, and carbon dioxide, are other potential pathways for degradation. In the present review out of 12 studies, 3 studies are with a UV light source and the remaining 9 studies are with a visible light source which is tabulated in Table 1, and the corresponding degradation pathway is

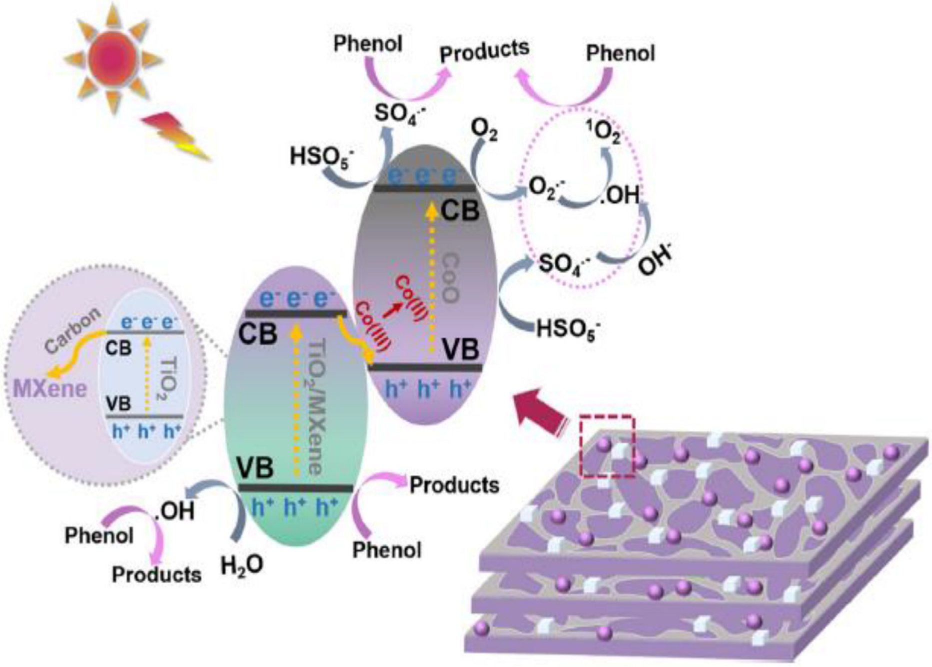


Fig. 3. Catalytic degradation mechanism in CTM/PMS/Vis system. (Reused from [26] with permission from Elsevier, Licence No. 5557580868598).

explained in all the 12 studies for the phenolic pollutant degradation.

According to the analysis of BPA mineralization and degradation presented, the majority of BPA molecules under the conditions used in the experiment were degraded into the intermediates. In order to further analyse the treated solution, LCMS-IT-TOF was used at the selected irradiation times (0, 30, and 60 minutes). Bisphenol A ($C_{15}H_{16}O_2$), product A ($C_9H_{12}O$), product B-1

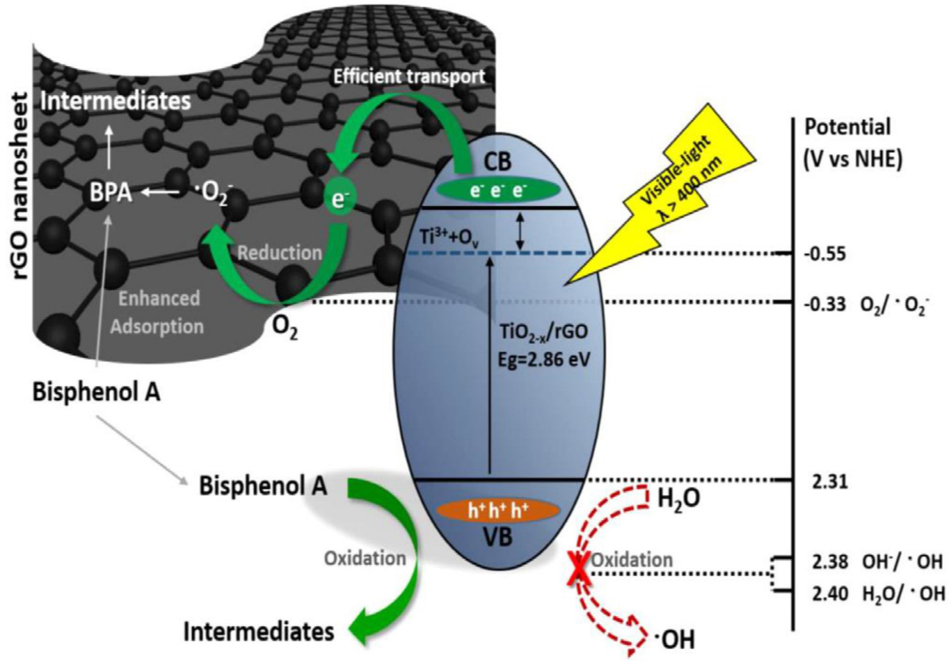


Fig. 4. Schematic illustration of the possible mechanism of enhanced BPA degradation by TiO2−x/rGO under visible-light photocatalytic activity (Reused from [30] with permission from Elsevier, Licence No. 5557580271085.

($C_9H_{12}O$), product B-2 ($C_8H_8O_2$), product C ($C_{15}H_{16}O_3$), product D ($C_{15}H_{14}O_3$), product E ($C_{14}H_{14}O_2$), product F ($C_{13}H_{12}O_2$), and product G ($C_9H_{10}O_2$) were the intermediates under visible light which is also shown in Fig. 4. Additionally, using a magnetic nanocomposite made of $FeNi_3/SiO_2/ZnO$, the PCP was removed using UV irradiation. This procedure produced the intermediate chemicals (tetrachlorophenol), TTCP, (tetrachlorocatechol), TTCC, and (trichlorophenol) with CO_2 and H_2O(1).

4. Conclusion

In the present study, a systematic pooled analysis review was furnished for the assessment of the photocatalytic degradation process of emerging organic pollutants from the aqueous solution using different nanocomposites. The independent factors such as the amount of photocatalyst, pH, irradiation time, the concentration of pollutant, and contact time that have a crucial role in photocatalytic degradation processes were reviewed. In this study, 12 studies were included which particularly consisted of phenolic pollutants. Among 12 phenolic pollutants, 2 studies reported complete removal of pollutants. Most of the pollutants exhibited a degradation efficiency above 90%. Recently, major developments in the utilization of advanced photocatalytic treatment for the elimination of organic pollutants present in any aqueous solutions have been used. Thus, it can be concluded that the advanced photocatalytic treatment processes are efficient and effective in the removal of organic pollutants from the water. However, the pooled analysis confirms the removal of organic pollutants or pesticides by the photocatalytic treatment processes as an upcoming technology in recent years.

Conflict of interest

There is no conflict of interest.

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