Comparative Assessment of Continuous Flow Photocatalytic Oxidation Reactors for Organic Wastewater Degradation

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ABSTRACT

Photocatalysis is an environmentally friendly technique for removing organic pollutants such as dyes, pesticides, etc. The photo reactors could be of the slurry type and fixed bed type. Continuous flow photocatalytic reactors generally are fixed bed-type reactors. Slurry type designs like loop thin-film slurry flat-plate photoreactors, step aeration slurry reactors etc. were also tried out for continuous flow operations. Continuous flow photocatalytic reactors have become one of the most ensuring methods for the treatment of mass water. However, uniform dispersion of the photocatalyst within the wastewater volume is still existing as a challenge. Different reactor designs like immobilized bed reactors (packed bed reactor and fluidized bed reactor), annular reactor with photocatalyst coated on inner/outer cylinder, photocatalytic membrane reactors, tubular reactors, microreactors, etc. are tested for their efficiency. This review tries to provide a generalized comparison of the relative merits and demerits of these reactor designs and immobilization methods on the degradation of organic contaminants.

Keywords: Photoreactors, Advanced Oxidation Process, Photocatalysis

1 Introduction

Water is an essential natural resource covering more than 70% of the earth's surface. Since the past few decades, there has been an uninterrupted rise in the demand for raw materials related to plastic, dye, textile & fertilizer industry due to growing urbanization, technological advancements, and exploitation of non-renewable resources. Energy-related crises and environmental pollution have already reached an awful situation. Industrial wastes are non-biodegradable as compared to municipal refuse. These industrial wastes include heavy metals, grease, fat, oil, ammonia, phenol etc. Large quantities of pesticides and other chemicals have been released through agricultural and pharmaceutical effluents which are responsible for some severe diseases that are detrimental for human endocrine, causing water non potable. These contaminants in wastewater due to increased human activities should be separated or converted harmless before discharged into streams. Thus, there is a sudden need to develop novel environment friendly technologies which contributes to the complete elimination or degradation of environmental contaminants. Advanced oxidation processes (AOPs) are environmentally friendly techniques for the removal of various types of contaminants such as chlorinated hydrocarbons, petroleum-based products, pesticides, hydrocarbons, insecticides, volatile organic compounds (VOC), aromatics and other organic compounds from air and water. With one unpaired electron, AOPs produces reactive oxygen species such as superoxide radicals, hydrate radicals, hydroxyl radicals etc. They readily & vigorously react with a variety of chemical species, that would be highly complex to degrade otherwise [1]. The most suitable advanced oxidation processes (AOPs) for wastewater& water treatment are, Fenton, sonolysis, ozonation, UV photolysis, photocatalysis, wet air oxidation etc. [2]. Among these, photocatalysis is the most promising approach for addressing the difficulties created by organic contaminants in the environment.

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1.1 Photocatalysis

Photocatalysis, a green technology, is a process of photoreaction which accelerates in the presence of a catalyst along with a light source. Photocatalysts are substances that alter the rate of a chemical reaction in the presence of a source of light, and those reactions are termed as photocatalysis. Photocatalytic reactions can be basically classified into two types based on the phase of the reactants and the semiconductor material. If both the semiconductor & the reactants are in the same phase, it is termed as homogenous photocatalysis, for e.g., gas, solid, or liquid. When both are in dissimilar phases, such reactions are termed as heterogeneous photocatalysis [3]. The materials are typically characterized into metal or conductor, semiconductor and insulator with respect to their band gap. The energy disparity between the valence band and the conduction band is referred to as the band gap. Semiconductors are often used as photocatalysts due to their ability to conduct electricity even at room temperature, in the presence of light. When the photocatalyst are exposed to light of sufficient wavelength, electrons get excited from valence band to conduction band by the absorption of energy from the photons. A hole pair is generated in the valence band. Thus, an electron-hole pair get generated. The excited electron enables reduction whereas hole enables oxidation. Thus, a photocatalyst allow a redox reaction and finally degrade the pollutants. Photocatalysis is an important tool for environmental detoxification by way of visible light induced photocatalysis. The photocatalytic activity (PCA) in a photo generated catalysis mainly relies upon the capacity of the catalyst to generate electron-hole pairs, which contributes to free radical formation. These radicals act as an effective oxidizing agent in wastewater and water remediation. The development of titanium dioxide-based water electrolysis made its practical applications viable [4].

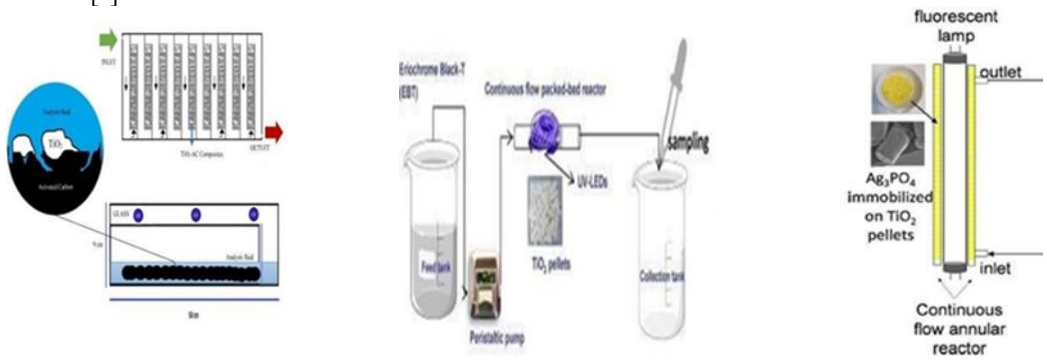
2 Photocatalytic Reactors for Wastewater Treatment

Advanced oxidation process is typically classified into Homogeneous and heterogeneous technologies based on the phase of reagents. The process is mainly based on various parameters like dosage, pH of solution, iron salt, temperature & mixing rate. The main disadvantage is pH control & high chemical intake. The major parts of the photocatalytic reactor involve photocatalyst (semiconductor), additives, auxiliaries, light irradiations etc. Semiconductor nanoparticles are widely used as an effective photocatalyst due to their wide band gap. Heterogeneous photocatalyst with sheets and flower like morphologies are highly predominant, due to their capacity to provide large surface area and high catalytic activity. The design and concept of photocatalyst mainly depends on working mechanism (dynamic or static), photocatalyst morphology (bulk or powders) and volume of liquid [5]. Reactors used for wastewater treatment are classified into two: fixed bed type reactors & slurry bed type reactors. In slurry type, suspended photocatalyst are employed. Suspended photocatalyst provides a very simple design and high surface area, whereas its disadvantages are the poor catalyst recovery and less light penetration. In fixed bed type reactors, immobilized photocatalyst are employed. The main advantages of immobilized photocatalyst is the simple operation and not requiring catalyst recovery. The main disadvantage is that of less surface area. Since intensity of light is directly proportional to the total irradiated surface area, the above key challenge can be reduced by reducing the thickness of supported photocatalytic layer to thin. The slurry type reactors are typically batch – type, whereas fixed bed types are of continuous in operation. Different types of reactors have been used in the photocatalytic water treatment, which includes downflow contactor reactor, cascade photoreactor, annular slurry photoreactor etc. [6].

2.1 Continuous Flow Photoreactors

Continuous reactors (also known as flow reactors) are those reactors in which reactants are supplied continuously into the reactor and a continuous stream of product can be obtained. Continuous flow

photocatalytic reactor generally uses an immobilized thin film photocatalyst in which the photocatalyst is fixed on a support. It has an advantage of simple operation strategy and catalytic recuperation. However, uniform dispersion of the photocatalyst within the wastewater remains a challenge. The various effects of operational parameters are studied to obtain the most suitable design for a reactor to enhance the photocatalytic degradation. According to Coronel et al., [7], the TiO_2 supported activated carbon was employed for the photocatalytic degradation of cyanide in a continuous flow UV reactor, and a considerable degradation efficiency is obtained.as shown in figure 1 (a). The photo- reactor was fabricated with glass and covered with a wooden box to separate the fluid of external conditions. Photocatalytic and adsorption tests were conducted independently & 97% of CN degradation efficiency was obtained within 24 h due to the combined absorption & photocatalytic oxidation as compared to their individual performance at the end of the study [7]. Zeitoun et al., [8] on the study examined the performance efficiency of a developed photocatalytic membrane reactor (PMR) for the degradation of organic dye waste by membrane distillation process. The setup consists of a continuous stirred type photocatalytic feed tank, & it consists of a slurry titanium dioxide article that enacted by using UV irradiation at 365 nm & a poly - vinylidene fluoride (PVDF) layer membrane cell. The experimental operation was differentiated into two stages. The PVDF layer was manufactured & characterized to study its shape, surface charge & hydrophobicity, an electron surface zeta potential filter, contact point tests & a magnifying lens separately during the first stage. The effects of using various TiO_2 photocatalyst concentrations & nourish (e.g., concentration, colour) were also examined. PMR can accomplish pure permeation & 100% dye removal efficiency is acquired under specific circumstances [8].



(a) (b) (c)

Figure 1: (a) *Arrangement of immobilized beds of Activated Carbon or TiO_2-AC during the adsorption processand photocatalytic degradation of the cyanide in continuous flow photoreactor developed by Coronel[7].* **(b)** *Laboratory scale setup developed by Vaiano [10].* **(c)** *A continuous flow annular reactor which consist of Ag_3PO_4 immobilizes on pellets TiO_2 developed by Petala [12].*

Ghanbari et al., [9] investigated the photocatalytic efficacy of removing Cr (VI)and azo dyes by developing a novel fixed bed continuous reactor. N-Fe-co-doped TiO_2/SiO_2 nanocomposites were prepared. To obtain the best degrading efficiency, operating parameters, including PH, flow rate, angle against sunlight, were designed. A complex composition of pollutants, including BY- 51, Cr (VI), BB-41 & BR-29, were assessed under the influence of visible light and sunlight. The efficiencies of degradation were discussed in table 1. Under natural climatic conditions, the novel photoreactor and nanocomposites showed promising activity for the photocatalytic remediation of water pollutants [9]. As shown in figure 1(b), Vaiano et al., [10] designed a continuous flow photocatalytic packed-bed reactor. The reactor, when exposed to UV-LED light contributes to the degradation of two harmful anionic azo dyes. Anatase TiO_2 pellets were used as the catalytic material. The effect of the liquid flow rate on the performance of the reactor was studied in the range of 0.5-2.1 mL/min. The peak efficiency of the reactor was attained through a liquid flow rate of 0.5

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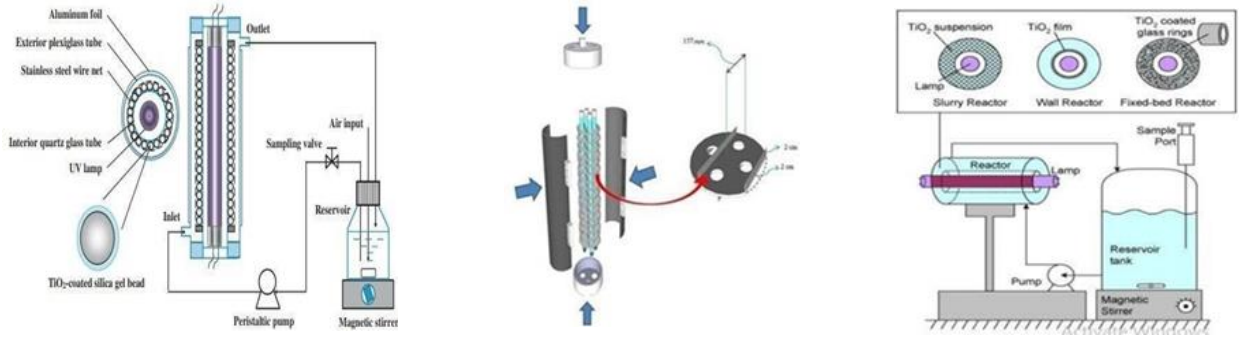
mL/min in distilled water. Complete decolorization of EBT was obtained in tap water under the same conditions. Methyl orange (MeO) degradation was 70%. The major benefit of this technology is that, by using a packed-bed reactor, the toxicity of EBT can be completely removed. MeO required the use of powdered activated carbon filtration in order to fully remove the toxicity [10]. For the evaluation of newly synthesized UiO-66(Ti)-Fe_3O_4-WO_3 magnetic photocatalysts and to examine their photocatalytic efficiency for the degradation of ammonia, Bahmani et al., [11] developed a flow-loop thin-film slurry reactor. The synthesized heterojunction possesses high stability and acceptable reusability which are the best part of the system. The system offers about 91.80% degradation efficiency under LED light source [11]. Petala et al., [12] designed, a continuous flow annular photoreactor for evaluating the photocatalytic effectiveness of immobilized Ag_3PO_4 photocatalyst for the degradation of micro-pollutants with as a supporting material as shown in figure 1(c). Degradation (75%) was also observed to primarily alter flow rates, micro-pollutant concentrations, and molecules. The system shows stability when exposed to inorganic ions, is one of the key advantages. But, failed to function effectively when humic acid is introduced to the feed [12].

Abdel et al., [13], developed a continuous flow photoreactor (heterogenous) to destroy organic pollutants in wastewater. The degradation of Phenol and MO under UV light by PS/ZnO (NCs) membrane (I) was 72% and 16.5%, & under visible light was 30% and 11%, respectively. Under UV, the PS/ TiO_2/SiO_2 (NCs) membrane (II) was able to degrade phenol by 18.1% and 40.3%. The degradation of MO and phenol reached 97% and 95% under visible light, under membrane(ll), and the performance was increased by increasing the oxygen concentration, through the addition Of H_2O_2. The major disadvantage is that the performance of NCs can only be increased by the addition of H_2O_2 i.e., by increasing O_2 content [13]. Sacco et al. [14] proposed a continuous flow micro-reactor that utilizes UV-LED irradiation for the photocatalytic degradation of crystal violet dye. Using Spherical Zeolites Pellets (ZEO) with immobilized Zinc Oxide (ZnO) as catalyst, have benefits such as maximum photocatalytic exposure to light sources, uniform illumination of the entire solution volume, and improved mas transfer phenomena. Higher dye removal efficiency 93% was obtained under UV-LEDs by using this setup [14].

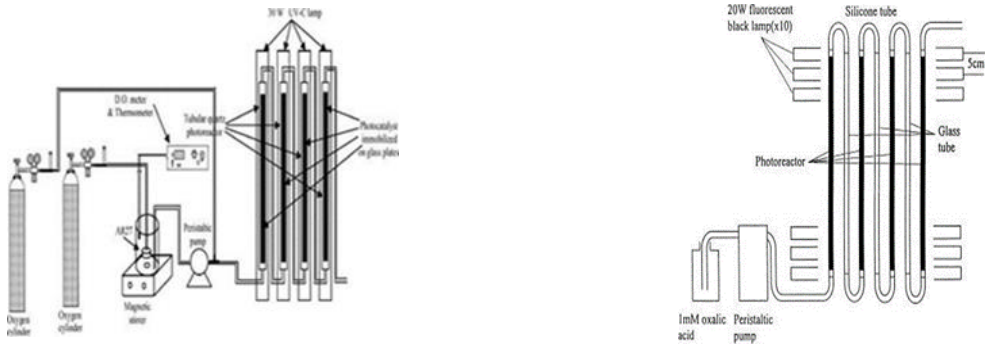


(a) (b) (c)

Figure 2: (a) *Schematic diagram of silica gel beads coated by TiO_2 immobilized with double photoreactive shell (DCS) and photocatalyst system developed by Li [16].* **(b)** *Arrangement & dimension of the photoreactor designed by Rezaei [17].* **(c)** *Schematic representation of the experimental setup for photocatalytic disinfection of E.coli suspensions in wall, slurry & fixed-bed reactor types developed by Van [18].*

A chemical-less Visible-UV photochemical continuous-flow reactor was developed by Moussavi et al., [15] for the direct oxidation of contaminated water for the removal of ammonium to N_2 gas. The reactor consists of a 400 mm-tall Pyrex tube column with a 30 mm inner diameter & the reactor was installed vertically. The photoreactor has a working volume of 135 mL. A 5.7 W low-pressure mercury UV was used as the light source. The operations were conducted in continuous as well semi- batch mode. 100% oxidation

of ammonium was detected for 50 mg/L sample [15]. A novel double-cylindrical-shell photoreactor for the degradation of rhodamine B (RhB) and methyl orange was designed & fabricated by Li et al. [16]. The reactor was immobilized with monolayer TiO_2-coated silica gel beads. Inner quartz glass tube of the reactor was coated with TiO_2 immobilized silica gel particles on the exterior surface as shown in figure 2 (a). Compared to slurry and thin-film photoreactors for the degradation of Rhodamine B and MeO, the novel photoreactor exhibited better repetitive operating performance, reduced energy consumption & a higher efficiency [16]. Rezaei et al., [17] introduced a continuous flow immobilized TiO_2 photoreactor on the same year and it consists of four quartz tubes contained in an aluminum tube as shown in figure 2 (b). Four UV lamps, each with a maximum wavelength of 254 nm were placed at the axis of the quartz tubes. For the fluid flow along the length of the reactor, twelve stainless steel circular baffles coated with particles are mounted inside the reactor in a zigzag pattern. This design offers a high mass transfer coefficient. Under optimum processing conditions, 75.50% of phenol was observed to degrade [17]. Van et al., [18] developed a Wall and fixed bed type reactors for the deactivation of Escherichia coli as illustrated in Figure 2 (c). The immobilization of TiO_2 in an annular reactor can be done in two different ways, either on the surface of the glass rings used in the packed bed reactor or on the interior reactor. The effect of increase in film thickness & its effects on the degradation efficiency were studied. The main drawbacks of the arrangement are its less photocatalytic activity as compared with a slurry system & resistance to the inhibition by organic matter [18]. Behnajady et al., [19] designed a tubular continuous flow photoreactor which immobilized on glass plates for the photocatalytic degradation of C.I. Acid Red 27 (AR27), an anionic mono azo dye of the acid class, in aqueous solution as depicted in figure 3 (a). photoreactor consists of four quartz tubes, connected serially from top to bottom by transparent polyethylene tubes. Each quartz tube contains three glass plates with P_{25} TiO_2. As irradiation sources, four low pressure mercury UV lamps were used. The removal efficiency rises linearly with light intensity. It was shown that final COD was extremely low with an increasing flow rate [19]. A fixed-film continuous-flow bioreactor with three components, a top, transparent tubes, and bottom. A green Sulphur bacteria called Chlorobium thiosulfatophilum was utilized to extract hydrogen sulphide from synthetic industrial wastewater and transform it into elemental Sulphur. 21.2 mL total volume of reactor were formed with active part of twenty 150 mm x 3 mm ID Tygon tubes & the recovery rates of elemental Sulphur ranged between 75–95% and 82– 100%, respectively. The high bacterial density and light intensity (light/volume) also contribute to higher efficiency of the reactor [20].



(a) (b)

Figure 3: (a) *Schematic diagram of tubular continuous-flow photoreactor developed by Behnajady* [19]. **(b)** *Continuous flow photoreactor developed by Kobayakawa et al. [21], packed with TiO_2 immobilized on large silica gel beads [21].*

A tubular continuous flow photoreactor for the degradation of oxalic acid was developed by Kobayakawa et al., [21]. It comprises of an 8mm diameter Pyrex glass tube filled with titanium dioxide photocatalyst immobilized on 2 mm diameter silica gel beads shown in figure 3 (b). The tubular reactor was filled with

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fine powder attached to 30–40 mesh silica gel that was prepared by blending a concentrated slurry. Two sets of 10 W, 20 W black, fluorescent UV lamps were used for irradiation. The light tubes were spaced 20 cm apart in flat arrays. As a result of using layers of silica gel beads coated with higher efficiency in the breakdown of oxalic acid was achieved. High resistance to the flow of solution is identified as the major disadvantage. This is due to the densely packed granular particles of silica gel [21]. Ali & Kim, 2018 designed a continuous flow photo- reactor. Cylindrical anionized TiO_2 nanotubes (TNTs) worked as an excellent photocatalyst against methylene blue dye. The colour removal efficiency with a UV light irradiation is about 89%. The usage of visible light shows better results. Also, TNTs & modified TNTs shows better catalytic reusability & treatment of wastewater in textile industry [22]. Chekir et al., [23] developed a tubular photocatalytic reactor under solar irradiation. In this study, the catalytic efficiencies of TiO_2 & ZnO for the degradation of methylene blue is observed among which shows 98% degradation efficiency. The concept of solar oxidation & its feasibility is well explained with the reactor [23].

Table 1: *Comparison of efficiencies of different continuous flow photoreactor*

Reactor Type	Pollutant	Catalyst	Efficiency	Light source	References
Continuous flow UV glass photo-reactor	Cyanide	$Ti0_2$-AC	97%	UV	[7]
Photocatalytic membrane reactor	Organic dye	TiO_2	100%	UV (365 nm)	[8]
Novel fixed bed continuous reactor	BB-41 BR-29 BY-51 Cr (VI)	N-Fe-co doped. TiO_2/SiO_2	87% 85.64% 58.59% 91.735		[9]
CF photocatalytic packed-bed reactor	EBT, MeO	Anatase $Ti0_2$pellets	70%	UV-LEDs	[10]
Flow- loop thin-film slurry flat-plate photoreactor	Ammonia	UiO-66(Ti)-Fe_3O_4- WO_3	91.80%	LED	[11]
Continuous flow annular photoreactor	sulfamethoxazole	Ag_3PO_4	75%	Fluorescent Lamp	[12]
Continuous flow photoreactor	Crystal Violet dye	Zeolites pellets with immobilized ZnO	93%	UV-LEDs	[14]
Double-cylindrical-shell (DCS)	RhB, MO	silica gel beads with coating	75.50%	UV lamps (254 nm)	[16]
Wall & Fixed bed Type	Escherichia coli	TiO_2	1CFU/ml	UV-A	[18]
Tubular continuous flow photoreactor	C.I. Acid Red 27 (AR27), mono azo dye of acid class	TiO_2	95.24%	UV	[19]
Fixed-film, continuous-flow bioreactor	Sulphide	Chlorobium Thiosulfatophilum(a green Sulphur	82%– 100%	UV	[20]

		Bacterium)			
Continuous-flow Photocatalytic reactor	Methylene Blue	TiO_2 nanotubes coating of 4 x 5m^2	89%	UVC lamp of 16 W	[22]
Solar tubular reactor	Methylene Blue	TiO_2/ZnO	~99%	Sunlight	[23]
Continuous flow photoreactor	Phenol MO	(PS/TiO_2/ZnO) membrane	97% 95%	Visible light	[13]

Different types of continuous flow reactors such as photocatalytic membrane reactor, fixed bed continuous reactor, continuous flow annular reactor, double cylindrical shell photoreactor, tubular continuous flow photoreactor etc. are discussed in table 1. Most of the photoreactors works with TiO_2 and its composites as catalyst. By the comparative analysis of different type of continuous flow photoreactors, photocatalytic membrane reactor with as catalyst shows the best results for the degradation of organic dye compounds. Various intensities of light sources such as UV, Visible light, sunlight, fluorescent light etc. were used for different reactor types.

3 Conclusion

Photocatalysis is one of the most promising technologies for eliminating pathogenic microorganisms & degrading toxic industrial pollutants. Various researches are carrying out to implement this technology in large scale with time and cost effective. The main issue is to design an optimal reactor for the process. TiO_2 & its composite photocatalysts shows best results as compared to others. Continuous flow photocatalytic reactors have various advantages over others on the catalytic recovery. Various continuous photocatalytic reactors were discussed and among that the recent technique, photocatalytic membrane reactor (PMR) showed 100% organic dye removal efficiency. By optimizing the reactor design, the remaining challenges can be rectified in the future.

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