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Sand supported TiO₂ photocatalyst in a tray photo-reactor for the removal of emerging contaminants in wastewater

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ABSTRACT

There have been rising concerns about micro-pollutants that are not efficiently removed by conventional wastewater treatment plants. TiO₂ photocatalysis is one of the most promising methods for their removal. One of the major challenges to the widespread application of TiO₂ photocatalytic treatment is the use of fine TiO₂ particles in suspension.

A tray photocatalytic reactor based on using supported TiO₂ as a photocatalyst was designed and constructed. The reactor maintains a thin water film over TiO₂ supported on sand grains. Reactor performance for the degradation of phenol as a model compound was evaluated using 3 different approaches: (1) direct immobilization using the sol gel technique, (2) coating with TiO₂/cement grout; and (3) binding with TiO₂-epoxy coating.

The recirculating tray configuration achieved photocatalyst activation, turbulent flow, avoiding treatment dead zones, and continuous water oxygenation. The reactor performance using the epoxy TiO₂ sand composite was satisfactory in terms of degradation efficiency. The water turbidity remained unchanged indicating photocatalyst resistance to abrasion. Successful operation of the tray photo-reactor in the continuous mode was also achieved. The tray reactor is suitable for scale-up and commercialization due to five distinctive features which are: modular design; integrated storage; ease of continuous mode operation; absence of need for UV transmitting components; robustness and low cost of the epoxy-TiO₂-sand composite.

1. Introduction

Emerging contaminants (ECs), are a broad range of chemicals found in water at very low levels. More than two thousands emerging contaminant have been identified in surface waters around the world [1]. Many ECs including pharmaceuticals, personal care products, and hormones were found in treated effluents in many locations around the world [2–6]. Municipal wastewater treatment plants are identified as the major source for ECs [7]. Conventional secondary wastewater treatment is ineffective in degrading these persistent non-biodegradable compounds [8].

With an estimate of more than 38,000 chemicals currently being used worldwide and additional 300 new material synthesized annually [9], the concerns about ECs are rising. ECs can pose hazards whether the treated wastewater effluent is disposed to surface water or reused in irrigation. ECs having estrogenic effects on living organisms, can mimic or oppose the hormonal effect, alter the pattern of hormonal metabolism and interfere with the normal endocrine functions of living organisms [10]. Arrival of residual antibiotics to surface water lead to

increasing of antibiotic resistance in pathogenic as well as non-pathogenic micro-organisms [11].

Advanced oxidation processes (AOPs) for water/wastewater treatment are rapidly growing worldwide for removal of persistent non-biodegradable emerging water pollutants [12]. TiO₂ photocatalysis is one of the AOPs most promising technologies. It is worth noting that positively charged holes on TiO₂ have a relative oxidation power to chlorine of 2.35 [13]. In spite of the intense academic research that has been conducted during the last four decades on the possibility of mineralization of a vast range of water pollutants using TiO₂ photocatalysis, commercial application of TiO₂ photocatalysis treatment systems is still very limited. It is postulated that using fine TiO₂ particles in suspension is hindering the wide application of TiO₂ photocatalysis in water treatment [14]. TiO₂ slurry systems require post separation of the catalyst from the effluent to avoid catalyst loss as well as water contamination with TiO₂ particles that may pose threats to aquatic life [15]. This additional post separation is expensive and renders the process economically unfeasible [16]. Glass, polymeric substrates, silica and quartz materials have been commonly used as

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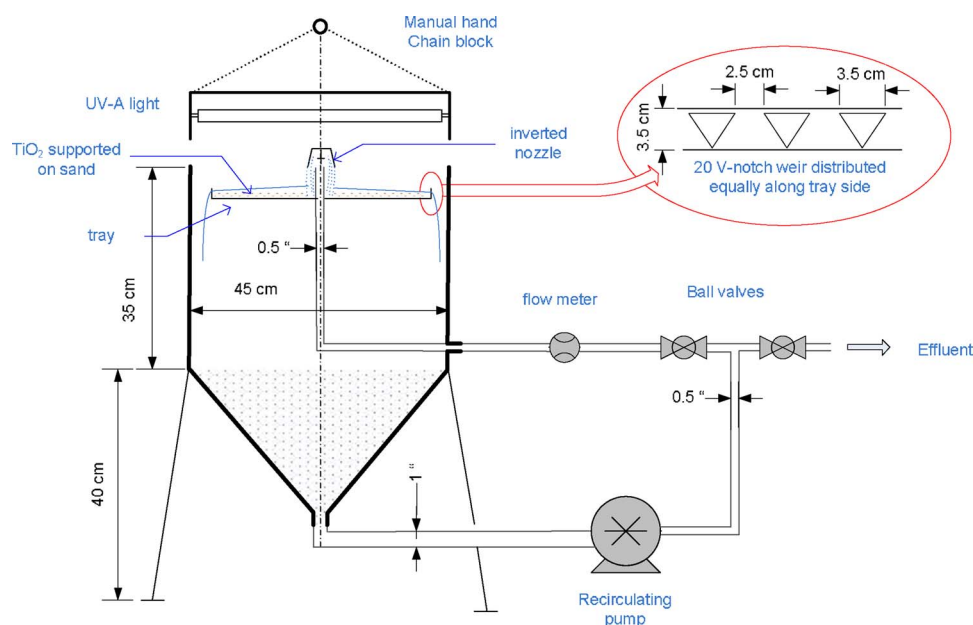


Fig. 1. Pilot-scale supported TiO_2 tray photocatalytic reactor.

supports for immobilizing TiO_2 [14]. Other supports including perlite granules [17], black sand [18], volcanic porous stones [19], light expanded clay aggregates [20], ceramic spheres [21], and porcelain-gres tiles [22] have also been tested. However, a thorough review of the literature [23] revealed that commonly used reactor configurations for TiO_2 photocatalysis suffer from limitations that hamper their commercial application. For instance, long exposure of glass tubes of compound parabolic concentrators (CPCs) and tubular reactors to solar radiation cause glass tubes aging that lead to UV transmittance reduction [24]; regular tube replacement is necessary, incurring high operational cost. CPCs and tubular reactors are closed reactors that need continuous purging of the reaction solution with oxygen or addition of oxidants [25]; increasing the operational cost as well as the need for a thorough process control to avoid an uneven O_2 /oxidants distribution throughout the reactor [26]. Another issue with CPCs and tubular reactors is the difficulty of using supported TiO_2 since they frequently encounter problems of reactor clogging [27], catalyst fouling [28,29], and high pumping energy requirement to compensate for the pressure drop across the reactor [19].

Falling film reactors (FFRs) operate in a laminar flow regime [30], thus experiencing limited treatment efficiency due to mass transfer limitations between the contaminants and the photocatalyst [31]. Large reactor area is required to attain acceptable treatment efficiencies [32]. In addition, FFRs suffer from operation disturbances during windy weather where parts of the reactor surface are dry; thereby reducing the reactor effective surface area [33]. A cost effective technique for immobilizing TiO_2 on large flat surfaces that is resistant to photocatalyst stripping off is still needed [34,35].

Flat plate reactors [36], step photoreactor [37] and double-skin sheet reactor (DSSR) [38] are enclosed reactors having a UV transmissive glazing. For enclosing large reactors, the glazing thickness should be sufficient to withstand the operating pressures. Increasing the glazing thickness reduce the UV transmissivity as well as add to the reactor capital cost [16]. In addition to the continuous need for purging the reaction solution with oxygen or addition of oxidants, the UV glazing encounters filming problems, known as window fouling where photocatalyst/water contaminants stick on the reactor surface [39] leading to further decrease in UV transmissivity and lowering treatment efficiency. For shallow pond/shallow tank reactors either using slurry TiO_2 [40], TiO_2 immobilized on fluidized supports [21], fixed support [41] or packed bed [42], maintaining sufficient light penetration to activate the catalyst is crucial for attaining high treatment efficiency

[43]. Another type is the photocatalytic membrane reactors where two configurations are used; TiO_2 in suspension and membrane is used for catalyst separation after treatment, this type experience permeate flux deterioration and membrane fouling, the other type has TiO_2 immobilized on the membrane, using a membrane that is resistant to UV deterioration as well as attack of hydroxyl radicals is still challenging [44].

This research aims to develop a photocatalytic reactor using immobilized TiO_2 that can be activated by an external light source either artificial UV-A radiation or sunlight to be used for the removal of emerging contaminants. Phenol is chosen as a model contaminant in this study since it is a precursor for various emerging contaminants in the environment [45]. Phenols and phenol derivatives easily donate a free electron forming phenoxy radicals that can penetrate living cells, damaging their mitochondria, endoplasmic reticulum membranes, nucleus, nucleic acids and enzymes [46].

This work involves: [1] design and manufacturing of pilot-scale photo-reactor using supported TiO_2 ; [2] fabrication of supported TiO_2 photocatalyst; and [3] photo-reactor testing using the synthesized supported photocatalyst to evaluate the system performance with an emphasis on stability of immobilized TiO_2 and its resistance to abrasion.

2. Experimental methods

2.1. Pilot-scale photocatalytic reactor

The pilot-scale photo-reactor is illustrated in Fig. 1. The reactor is mainly a stainless steel tank, 45 cm in diameter, equipped with a tray having a diameter of 34 cm for supporting the immobilized TiO_2 . The tray is mounted on the tank edges. The reactor is operated in a recirculating batch mode. The water is withdrawn from the tank conical bottom, and pumped by the recirculation pump into the central vertical feeder that passes through the tray center. The vertical feeder terminates at its top with a capped nozzle. The capped nozzle concentrates the exiting water in a ring-shaped impact area around the vertical feeder. This spray pattern is intended to allow a radial flow of water along the tray. The tray side has 20 v-notch weirs, each weir with a width of 3.5 cm, and depth of 3 cm. Weirs are 2.5 cm apart from each other. The v-notch weirs maintain an appropriate depth of water above the tray. Recirculation flow rate is controlled such that the thickness of the water film over the supported catalyst does not hinder light

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