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Submerged microfiltration-catalysis hybrid reactor treatment: Photocatalytic inactivation of bacteria in secondary wastewater effluent

Lili Jiang [a], Xiaolei Zhang [a], Kwang-Ho Choo [a,b,*]

[a] Department of Environmental Engineering, Kyungpook National University, 80 Daehak-ro, Buk-gu, 41566, Republic of Korea
[b] Advanced Institute of Water Industry, Kyungpook National University, 80 Daehak-ro, Buk-gu, 41566, Republic of Korea

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ABSTRACT

This study investigated the performance of a membrane photocatalytic reactor (MPR) with respect to the removal of bacteria from secondary effluents. The MPR achieved disinfection through several mechanisms: rejection by a membrane, inactivation by direct UV radiation, adsorption onto photocatalysts, and oxidation by reactive oxygen species. Bacterial removal by the MPR reached a maximum at a TiO_2 dose of 1.0 g/L, with the TiO_2 dose ranging between 0 and 5 g/L. The optimal TiO_2 dose for bacterial removal appeared to be the result of the trade-off between accelerated catalytic reactions and the light obstruction by surplus catalysts. Continuous aeration required for membrane fouling control had a negative impact on bacterial removal. Although intermittent backpulsing reduced bacterial removal, its impact was not as significant as that of continuous aeration. Backpulsing frequencies and durations did not play a significant role, but the dynamic cake layer formed on the membrane was responsible for the bactericidal behavior. An increase in membrane flux deteriorated the bacterial removal performance because it resulted in shorter reaction times, even though the formation of a thicker cake layer was possible at higher fluxes. The MPR can guarantee >2.5 log removal in total bacterial count, given that the right conditions are maintained. This is significantly larger than the removal achieved by microfiltration alone (ca. 0.5 log removal).

1. Introduction

Photocatalytic disinfection with UV illumination onto TiO_2 particles is notable because it generates OH radicals on the surface of the catalyst [1–4]. Photocatalytic degradation of toxic and refractory micropollutants under UV and/or visible light is another advantage of this method [5,6]. Photocatalysis generates byproducts (e.g., trihalomethanes) as well, although extended photoreactions can reduce them dramatically [7]. TiO_2 photocatalysis inactivates MS-2 phage and *E. coli* with the generation of OH radicals, although the former is mainly disinfected by the bulk phase free hydroxyl radicals [8]. The efficiency of photocatalytic disinfection increases in the presence of additional biocidal agents, such as Cu and Ag [1]. The AgBr-Ag-Bi_2WO_6 nano-junction catalyst exhibited a higher *E. coli* disinfection performance than the other photocatalysts that were also activated by visible light [9]. Sulfur-doped TiO_2 photocatalysts were found to have a significant bactericidal effect on *M. lylae* (a gram-positive bacterium) under visible light [10]. Bacterial regrowth after photocatalytic disinfection was observed, but it slowed down after UV irradiation with TiO_2, at an acidic pH range [4]. In addition to bacteria, fungi strains were destroyed by 2-h photocatalysis, using a 400-W sodium lamp [11]. Solar photocatalysis with a parabolic collector and immobilized titanium dioxide was tested at pilot scale and found to be effective in disinfecting *E. coli* and *F. solani* [12–15]. However, it was found that solar photocatalytic disinfection was ineffective at eliminating the cyst stage of the protozoa *A. polyphaga*, despite the fact that its trophozoite stage was well-inactivated [16]. With the TiO_2 photocatalyst modified with reduced graphene oxide, the *E. coli* inactivation efficiency remained unchanged under visible light (i.e. when the solar UVA was cut-off), although the original P25 photocatalytic performance decreased significantly [17].

An electrospun membrane that consists of Ag nanoparticles and TiO_2 nanofibers was reported to have an excellent antibacterial performance (3 log removal) under a 30-min solar irradiation [18]. A similar approach with TiO_2 photocatalytic membranes, covered with Ag, was carried out and it achieved a 7 log removal of *E. coli* [19]. A photocatalyst-coated ceramic membrane was also applied to the inactivation of bacteriophage P22 [20]. It approximately achieved a 5 log virus removal, which was better than that

* Corresponding author at: Department of Environmental Engineering, Kyungpook National University, 80 Daehak-ro, Buk-gu, 41566, Republic of Korea.
E-mail address: chookh@knu.ac.kr (K.-H. Choo).

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of serial processes of UV disinfection followed by microfiltration (MF) (approximately 2 log removal). The use of photocatalytically active membranes is still at an early stage, as it is limited by light bombardment onto the membrane [21].

A membrane photocatalytic reactor (MPR), which is a hybrid of photocatalysis and membrane filtration, was developed in order to facilitate the separation of suspended photocatalysts with efficient light illumination [22,23]. It has been demonstrated that the MPR was able to degrade a variety of micropollutants and refractory organic matter in water, while managing the effective confinement of the photocatalysts inside the reactor [24–31]. In addition, virus removal (bacteriophage f2) by the MPR was achieved, showing the importance of OH radicals and electrons generated [32]. Viral inactivation was found to occur mainly by photocatalysis (as opposed to membrane filtration and TiO_2 adsorption). However, the bacterial disinfection of water and wastewater samples by the MPR has not been well investigated.

In our previous study, an MPR showed the enhanced degradation of secondary effluent organic matter with low fouling propensity [33]. The focus of this study was thus to extend the investigation of the membrane-photocatalyst hybrid performance with respect to bacterial inactivation in the secondary effluents obtained from the biological activated sludge process. The contribution of key reactor components (e.g., the catalyst and the membrane) to bacterial removal was evaluated systematically. The effects of catalyst dosage, physical cleaning, and membrane permeability on bacterial removal were examined while monitoring the time profiles of membrane assisted photocatalytic disinfection.

2. Materials and methods

2.1. Feed wastewater

Secondary effluent samples taken from the Shincheon Wastewater Treatment Plant in Daegu, Korea were used as a feed solution for all the experiments. The key characteristics of secondary effluent are given in Table 1. The total bacterial count of the secondary effluent was relatively low compared to that present in the average secondary effluent. The reasons for this lie in the fact that the sample was moved to the laboratory and stored in a refrigerator at 4 °C before carrying out the experiments. Hence, before each experiment, the secondary effluent was placed in an incubator at 35 °C for 2 d, in order to allow the bacteria in the feed solution to proliferate instead of spiking specific cultivated bacteria.

2.2. Membrane, photocatalyst, and UV lamp

The microfiltration (MF) hollow fiber membrane used, was made of hydrophilized polyethylene, with a nominal pore diameter of 0.4 μm and an effective surface area of 60 cm^2 (KMS, Korea). TiO_2 particles (P25, Degussa, Germany) were used as photocatalysts, which had an average aggregate particle diameter of 3 μm and a surface area of 50 m^2/g. An 8-W blacklight blue UV lamp (Sankyo, Japan) was used for the activation of TiO_2 photocatalyst. The UV lamp had a maximum wavelength of 360 nm, with a photon flux of 4 mW/cm^2.

Table 1
Qualities of secondary effluent.

Parameter	Value
pH	7.0–7.3
Chemical oxygen demand, mg/L	10.3–15.0
Total organic carbon, mg/L	2.1–2.4
UV_{254}, cm^{-1}	0.064–0.065
Total bacteria count, cfu/mL	420–460
Specific UV absorbance, L/mg-m	2.7–2.9
Conductivity, μS/cm	748
Turbidity, NTU	1.72
Total suspended solids, mg/L	6
Alkalinity, mg/L as $CaCO_3$	87
Hardness, mg/L as $CaCO_3$	146

2.3. Reactor configuration and operation

The MPR used in this study consisted of an 8-W UV lamp and an outside-in submerged MF membrane module (Fig. S1), which had a working volume of 700 mL, as shown by previous tests conducted in other studies [23,33]. Peristaltic pumps (Model 7253-40, Cole Parmer, USA) were used for feeding and suction with MaterFlex tubings. Permeate samples were collected using a Universal Fraction Collector (Eldex, USA), after backpulsing if it was employed.

The MPR was operated at a continuously stirred tank reactor, while the secondary effluent was fed at the same flow rate as the permeate was discharged during continuous runs. Initially, the feed and TiO_2 powders were mixed in the reactor (which was defined as −60 min in the time scale) and agitated for 60 min for equilibration before membrane filtration. There was no direct wastage or discharge from the reactor other than the membrane permeate. The reactor volume was maintained at the constant level (700 mL) throughout the experiments, with the use of a level sensor. The membrane was operated at a constant flux between 50 and 100 L/m^2-h (corresponding to a flow rate of 5–10 mL/min), so that the hydraulic residence time (HRT) in the reactor varied between 140 and 70 min. Continuous experiments were run for 8 h in order to monitor the reactor performance (e.g., the removal of bacteria) for the treatment of secondary effluent amounts corresponding to more than three times the reactor volume (instead of three batch tests). The transmembrane pressure (TMP) was monitored continuously using a pressure transducer (ZSE40F, SMC, Japan) and it was recorded on a personal computer, which was connected to the transducer via a multimeter (M-3850D, METEX, Korea). Occasionally, the MPR was operated in a batch mode under the same conditions, and no feed was added. The hydraulic filtration resistance during membrane operation was monitored and evaluated based on the resistance-in-series model (see Supplementary Material).

2.4. Membrane cleaning protocols

Two different physical membrane cleaning methods were employed in this study. First, a continuous and extensive supply of air (which is normally adopted for a membrane bioreactor to control fouling) was provided. Air sparging (2 L/min corresponding to a velocity gradient of 479 s^{-1}) was provided using an air diffuser placed underneath the membrane module. Second, a novel backpulsing method was devised (Fig. S2), which could achieve almost a total water recovery. Compressed nitrogen gas (0.5 bar) was supplied to the lumen side of the membrane with different filtration cycles and durations, as follows: membrane filtration cycle, 30–60 min; backpulsing duration, 30–105 s.

2.5. Analytical methods

The total bacterial count was determined through cultivation on Petrifilm plates (3M® Petrifilm™ Aerobic Count Plates, USA) in an incubator (MIR-553, Sanyo, Japan) at 35 ± 2 °C for 48 h. The UV absorbance at 254 nm and the dichromate chemical oxygen demand were measured using a UV–VIS spectrophotometer (DR/4000U, Hach, USA), while the measurement of total organic carbon (TOC) concentration was carried out with the use of a TOC analyzer (Model 820, Sievers, USA). The UV absorbance of water samples was measured after filtering out the particulate

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