



Membrane photoreactor treatment of 1,4-dioxane-containing textile wastewater effluent: Performance, modeling, and fouling control



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ABSTRACT

A membrane photoreactor (MPR) system was investigated for the purification of textile wastewater effluent containing 1,4-dioxane for potential reuse. The effects of TiO₂ dosage, alkalinity, particulate matter, and sunlight radiation on the removal of 1,4-dioxane from the effluent were evaluated. The photocatalytic 1,4-dioxane degradation followed pseudo-first order reaction kinetics. A considerably large dose of TiO₂ catalyst (5 g/L) was required for maximal 1,4-dioxane degradation by MPR. The high alkalinity (~550 mg/L) of the textile wastewater effluent negatively affected the 1,4-dioxane degradation rate, whereas 1,4-dioxane degradation was not impacted by the presence of particulate matter. Solar photocatalysis of 1,4-dioxane was as effective as that achieved with UV lamps in the MPR, which could lead to energy savings. Continuous flow MPR was effective for continuous degradation of 1,4-dioxane; the unsteady- and steady-state performances were well-predicted at various hydraulic residence times (HRTs). The quality of the MPR product obtained at an HRT of 8.57 h satisfied the drinking water guidelines. The transmembrane pressure (TMP) buildup, which is indicative of fouling, was dependent on the TiO₂ dose and permeate flux. The TMP buildup was marginal at low flux, but increased dramatically beyond a flux level of >60 L/m²-h. However, the fouling layer could be sufficiently removed by periodic backwashing, even at an extremely high flux of 100 L/m²-h. The membrane permeability was sustained without any sign of irreversible fouling during MPR treatment of textile wastewater effluents.

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1. Introduction

1,4-Dioxane in industrial wastewater effluents is of great concern to humans and ecosystems (Stepien et al., 2014). It was recently reported that the 1,4-dioxane level in drinking water intakes ranged from 0.35 to 100 µg/L depending on dilution factors (Lee and Choo, 2013; Simonich et al., 2013; Stepien et al., 2014). De facto wastewater reuse, which can expose humans to hazardous substances, is real and significant as reported in a survey across the United States of America (Rice and Westerhoff, 2014). Hence, it is urgent and important to develop innovative methods to either treat or recycle wastewater containing 1,4-dioxane and thereby prevent contamination of drinking water sources (Lee and Choo, 2014).

Several attempts have been made to remove 1,4-dioxane from the aqueous phase; however, its removal by carbon adsorption or air stripping is difficult because of its hydrophilic nature, and it is

not readily biodegradable because of its heterocyclic structure (Adams et al., 1994; Chitra et al., 2012; Lee et al., 2011; Rietman and Verliefe, 2010; Zenker et al., 2003). Thus, advanced oxidation processes (AOPs) have been tested as efficient methods for the removal of 1,4-dioxane (Barndök et al., 2014a; Kwon et al., 2012; Stefan and Bolton, 1998). Several researchers have investigated various combinations of AOPs for 1,4-dioxane degradation including the following: hydrogen peroxide and ferrous iron as well as ozone and hydrogen peroxide (Adams et al., 1994); hydrogen peroxide (H₂O₂) and zero-valent iron (Fe⁰) in the presence of UV light (Son et al., 2009; Stefan and Bolton, 1998); Fe⁰, Fe²⁺, and S₂O₈²⁻ with sonication (Beckett and Hua, 2003; Son et al., 2006); ozone and electrolysis (Barndök et al., 2014b; Kishimoto et al., 2008); and photocatalytic degradation using TiO₂ and UV light (Coleman et al., 2007; Lam et al., 2007; Lee and Choo, 2013; Nakajima et al., 2004; Vescovi et al., 2010; Yanagida et al., 2006). Combining AOPs with membrane separation processes has been demonstrated for the removal of emerging contaminants and dissolved organic matter as follows: chemical oxidation (by using ozone, chlorine, O₃/H₂O₂, UV, or UV/H₂O₂) followed by

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nanofiltration (Acero et al., 2015); electrochemical oxidation with microfiltration (Park et al., 2013); Fenton's reagent with microfiltration and nanofiltration (Moravia et al., 2013); and combined nanofiltration and solar photo-Fenton (Miralles-Cuevas et al., 2014).

A membrane photoreactor (MPR), which can effectively degrade toxic micropollutants as well as separate particulate matter, is also attractive for 1,4-dioxane removal and zero liquid discharge (Choo et al., 2008b; Wang et al., 2013). A slurry reactor system is preferentially used instead of a fixed-catalyst reactor to increase the photocatalytic activity of TiO₂ (Chin et al., 2006; Molinari et al., 2002) because the high specific surface area of dispersed TiO₂ powder enhances the overall mass transfer and reaction rate (Balasubramanian et al., 2004; Fu et al., 2006; Mozia, 2010; Sarasidis et al., 2011). The membrane in the MPR affects both the recovery of catalysts and production of good quality effluents (Balasubramanian et al., 2004; Lee et al., 2001). Use of photocatalytic membranes, which have photoactive layers on a porous support, provides dual functionalities of photocatalysis and separation (Goei et al., 2013; Goei and Lim, 2014; Papageorgiou et al., 2012; Syaferi et al., 2008). However, light radiation onto the membrane surface is a critical issue to be resolved in practical applications. MPRs still have advantages over other types of photocatalytic reactors including the utilization of simple suspended catalysts, maximal use of active catalyst surfaces, ease of light radiation, and efficient control of hydraulic and solids residence times (Lim et al., 2011; Mozia et al., 2005, 2006; Sarasidis et al., 2011).

Previous studies demonstrated the photocatalytic degradation of toxicants (e.g., 1,4-dioxane, trichloroethylene, and 4-chlorobenzoic acid) in relatively clean aqueous media or in air (without particulate matter) using catalytic slurries (Jo and Lee, 2013; Lee and Choo, 2013), films (Balasubramanian et al., 2004; Rojviroon et al., 2014), and membranes (Goei et al., 2013; Goei and Lim, 2014; Syaferi et al., 2008); however, the potential of MPRs for actual wastewater treatment and reuse has rarely been reported. Therefore, the present study involves a systematic investigation of the MPR treatment of real textile wastewater effluent containing 1,4-dioxane under various operating conditions. The effects of TiO₂ dose, alkalinity, and particulate matter on the kinetics of 1,4-dioxane degradation were evaluated in a batch reactor. Photocatalytic 1,4-dioxane degradation by MPR activated under actual sunlight was compared to that achieved with UV lamps. The performance and modeling of 1,4-dioxane degradation in continuous flow MPRs were investigated at various hydraulic retention times. The degree and control of membrane fouling in batch and continuous flow MPRs were evaluated and compared.

2. Materials and methods

2.1. Feed wastewater

The wastewater samples used for this study were taken from the effluent stream of a textile manufacturing factory in Korea. The key characteristics of the textile wastewater effluent are provided in Table 1. The effluent contained a high concentration of 1,4-dioxane after biological treatment. To check the effect of alkalinity on 1,4-dioxane degradation efficiency, the alkalinity of the original effluent sample was reduced to 4.5 mg/L as CaCO₃ by purging the sample with N₂ gas for 15 min after adjusting the solution to pH 4.5 using H₂SO₄. After the alkalinity adjustment, the solution pH was returned to its original level using NaOH. Further examinations of the effects of alkalinity on 1,4-dioxane removal were conducted while adjusting the alkalinity level within the range of 43–1000 mg/L as CaCO₃ by adding NaHCO₃ (Merck, Germany).

Table 1

Properties of the textile wastewater effluent (feed) used and MPR product obtained at an HRT of 8.57 h (corresponding to a flux of 10 L/m²-h).

Parameter	Feed	Product
pH	8.5	8.7
Turbidity, NTU	2.53	0.23
Conductivity, μS/cm	1450	1450
COD, mg/L	8.0	5.7
DOC, mg/L	4.4	1.6
UV ₂₅₄ absorbance, cm ⁻¹	0.0572	0.0239
Alkalinity, mg/L (as CaCO ₃)	550	525
Hardness, mg/L (as CaCO ₃)	62	30.8
Suspended solids, mg/L	4.2	ND
1,4-Dioxane, μg/L	850	42
Cl ⁻ , mg/L	78.8	80.2
NO ₃ ⁻ -N, mg/L	5.4	6.9
SO ₄ ²⁻ , mg/L	12.8	13.6
Chloroform, mg/L	0.003	ND
Cu, mg/L	0.008	ND
Fe, mg/L	0.01	ND
B, mg/L	0.01	0.01

ND: not detected.

2.2. Photocatalyst and membrane

Commercially available TiO₂ particles (Degussa P25, Germany) were used as photocatalysts in all experimental runs; the particles had an average grain diameter of 3 μm in aqueous solution and surface area of 50 m²/g. The TiO₂ dosage was varied within the range of 1.0–10 g/L. The membranes were polyvinylidene difluoride hollow fibers (Philos, Korea) with an average pore size of 0.4 μm and an effective bench-scale module surface area of 0.07 m².

2.3. Membrane photoreactor operation

The MPR used in this study comprised a reactor with a working volume of 6.0 L, six 8 W black light blue UV lamps (Philips, Holland), an air diffuser, and a submerged MF membrane fiber module (Fig. 1). The energy consumption of a UV lamp measured was 6.54 W each, whereas the measured UV radiation intensity at the reactor was 0.58 W/L. Air was supplied at 18 L/min (velocity gradient = 490 s⁻¹) to agitate the reactor containing the TiO₂ particles. A 1 L glass reactor was alternatively used for solar photocatalysis of 1,4-dioxane, as described elsewhere (Lee and Choo, 2013). The measured transmission efficiency of solar UV light (365 nm) through the glass reactor was approximately 82%. The UV intensity of the solar light was monitored using a light-receiver sensor facing the sun altitude. The contents of the glass reactor

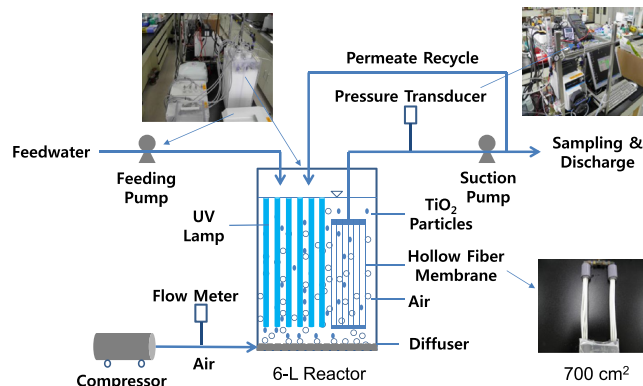


Fig. 1. Schematic diagram and photos of the bench-scale membrane photoreactor with submerged membranes used for the treatment of textile wastewater effluent.

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