Available online at www.sciencedirect.com

ScienceDirect

Desalination 245 (2009) 169–182

DESALINATION

www.elsevier.com/locate/desal

Application of TiO_2 photocatalytic oxidation and non-woven membrane filtration hybrid system for degradation of 4-chlorophenol

Ren-Yang Horng[a,b], Chihpin Huang[a*], Min-Chao Chang[b], Hsin Shao[b], Be-Lain Shiau[b], Yen-Jung Hu[c]

[a]*Institute of Environmental Engineering, National Chiao Tung University, Hsinchu, Taiwan*
Tel. +886 (3) 572-6463; Fax: +886 (3) 572-5958; email: cphuang@mail.nctu.edu.tw
[b]*Energy and Environment Research Laboratories, Industrial Technology Research Institute, Hsinchu, Taiwan*
[c]*KNH Enterprise Co., Ltd., Taipei, Taiwan*

Received 13 November 2007; Accepted 10 June 2008

Abstract

A system coupling photocatalytic oxidation of titanium dioxide particles separated by non-woven membrane for the degradation of 4-chlorophenol in an aqueous solution was studied. Non-woven membranes with three different pore sizes were applied to compare their specific flux and permeate turbidity. The results showed that when the pore size was slightly smaller than secondary particles of photocatalyst, higher specific flux and lower turbidity permeate were obtained. Four different applied fluxes, 0.25, 0.5, 1.0 and 2.0 $m^3/m^2/d$, were used with a 2.0 μm pore size non-woven membrane in a continuous system. The results revealed that the transmembrane pressure (TMP) was less than 10 kPa in all applied fluxes, due to the formation of a porous dynamic cake layer on the surface of the membrane. Based on the filtration characteristics of non-woven membranes, we found that cake formation, rather than pore blocking or pore narrowing, was the dominant factor. This phenomenon also resulted in permeate turbidity ranging from 0.5 to 1.5 NTU and non-detectable concentration of TiO_2 in the permeate. At the same time the experimental results indicated that the concentrations of TOC and 4-chlorophenol decreased with decreasing applied flux, and that the concentration of chloride ions increased with decreasing applied flux in a 4-chlorophenol photodegradation test. It showed that 4-chlorophenol could be effectively degraded by this hybrid system with lower TMP and stable applied flux.

Keywords: Non-woven membrane; 4-chlorophenol; TMP; Flux; Photocatalytic

1. Introduction

In the past three decades, heterogeneous photocatalytic processes have been considered as one of the most promising advanced oxidation processes (AOP) for water and wastewater treatment containing refractory organic pollutants. This technology can be applied in two different ways, i.e. suspended and fixed type. The suspended photocatalyst has a more effective

---

*Corresponding author.

doi:10.1016/j.desal.2008.06.018

photocatalytic activity than the fixed type due to having many active surface sites. However, the effective separation of suspended photocatalysts from aqueous solution is still an issue. In recent years, the separation of the photocatalysts from the liquid phase has been tried by using MF [1], UF [2] or even NF [3] membranes. However, problems such as low flux, high operational pressure and membrane fouling have not been overcome yet.

It is well known that non-woven fabric material with random structures is cheaper and has extensive application as a filter material for water treatment [4]. In our laboratory, the successful application of submerged non-woven fabric membrane bioreactor technology for industrial wastewater treatment [5] and wasted sludge reduction [6] has been developed. Meanwhile, a side stream of non-woven membrane to separate photocatalysts has been established in our laboratory for the degradation of methylene blue [7]. A hybrid system combining photocatalytic oxidation by titanium dioxide with particles separated by a submerged non-woven membrane was also studied. Hence the aim of this study was to investigate the performance and filtration characteristics of membrane fouling of hybrid systems with various applied fluxes, and the subsequent photodegradation of 4-chlorophenol (4-CP) as a model compound in water.

2. Methods and materials

2.1. Membranes, TiO_2 and 4-chlorophenol characteristics

Three different non-woven membranes, A, B and C, with pore sizes of 0.2, 2.0, and 20.0 μm, respectively, manufactured by KNH, Taiwan, were used in a batch study. Nominal pore sizes of the membranes were determined by air bubble method (Automated perm porometer, Porous Materials, U.S.A.). Total area of each membrane was 0.02 m^2 for both sides of membrane. A spacer was installed inside the membrane to provide a conduit for the permeate. The membrane's support layer was made of polyester. In the continuous system, membrane B was selected with total surface area of 0.045 m^2 for both sides.

Degussa P25 TiO_2 powder with a primary particle size of 20–30 nm was selected. Average aggregated secondary particle size after aggregation in the liquid phase was measured by a zeta potential analyzer (Zetaplus, Brookhaven Instruments, USA) and ranged from 3 to 8 μm, depending on different pH values and aeration intensities in the solution.

The residual turbidity of the permeate from different pore size membranes and applied fluxes was measured by turbidity meter (2100P Turbidimeter, Hach, USA). The 4-chlorophenol was analyzed by HPLC (Incelligent 500 and UV-2075 Incelligent UV/Vis detector), mobile phase ($CH_3OH/H_2O=50\ 50$, added 1% CH_3COOH), and Column: YMC-Pack Pro C_{18} R.S. (150 mm× 4.6 mm, ID S-5 μm). The chloride ion was quantified by IC (ICS-90 chromatography system, Dionex). Organic carbon was detected by TOC (Liqui TOC, Eelementar).

2.2. Experimental operations and conditions

The schematic diagram of our non-woven membrane system used in this study is shown in Fig. 1. Our system consisted of a reactor in which the membrane module was submerged. Three reactors were installed in parallel and each rectangular tank had a working volume of 8 L. The membrane module containing two parts of non-woven membrane plate was made of polyester. The pore size of non-woven membrane tested was 0.2 μm (membrane A), 2.0 μm (membrane B) and 20.0 μm (membrane C), respectively. The effective filtration area was 0.02 m^2 for each flat membrane. The aeration units were installed at the bottom part of the reactor to maintain suspended particles in reactor, and also induce a cross flow along the membrane surface. The