

A new submerged membrane photocatalysis reactor (SMPR) for fulvic acid removal using a nano-structured photocatalyst

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Abstract

The study focuses on the degradation of fulvic acid by nano-structured TiO₂ in a submerged membrane photocatalysis reactor (SMPR). It has been demonstrated that the composite TiO₂ photocatalyst could be automatically settled due to its gravity and then be easily separated by MF membrane. In addition, it was more efficient to maintain high flux of membranes than that of commercial TiO₂ P25. The paper describes the effects of operational parameters on the photocatalytic degradation of fulvic acid in SMPR. It was found that the photocatalyst at 0.5 g/L and airflow at 0.06 m³/h were the optimal condition for the removal of fulvic acid (FA) and the FA degradation rate was higher at acidic condition than that at alkaline media. In order to compare the effects of different filtration duration on permeate flux rate of MF, P25 powder and nano-structured TiO₂ were employed. According to the experiments, the permeate flux rate of MF is improved and thus the membrane fouling phenomenon is reduced with the addition of nano-structured TiO₂ catalyst. Therefore, the submerged membrane photocatalysis reactor can be potentially applied in photocatalytic oxidation process during drinking water treatment.

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1. Introduction

Natural organic matter (NOM) is widely distributed in soil, natural water, and sediments and consisted of a mixture of the decomposition products of plant and animal residues [1]. The main components of NOM are humic acid (HA) and fulvic acid (FA). In water purification, the disinfection by-products (DBPs) like trihalomethanes (THMs) were formed from chlorination of drinking water containing NOM [2,3]. In Tianjin, it has been reported that more than 70% of NOM in surface waters was fulvic acid, representing a major fraction of NOM [4]. Consequently control of fulvic acid plays an important role in treating the surface water. Some studies have reported that NOM with fulvic acids is difficult to remove by the conventional treatment processes. In general, the removal efficiency of TOC in con-

ventional drinking water treatment processes is only 10–50% [5].

TiO₂-mediated photocatalytic oxidation (PCO) has been the focus of numerous investigations in recent years. In particular, owing to the formation of hydroxyl radicals (\bullet OH), PCO shows an extraordinarily oxidizing power in the reaction mixture and can thus mineralize a myriad of undesirable organic matters to CO₂, H₂O and inorganic constituents [6]. The catalyst can be applied in the form of powder suspended in a slurry or it can be immobilized on various supporting medium, such as glass, quartz or stainless steel [7,8]. Each configuration has its advantages and disadvantages. In case of catalyst fixed onto a supporting material, a drawback is mass transfer limitations of pollutants to the surface of the photocatalyst. When a suspension of catalyst is used the photocatalytic degradation efficiency is high. However, the catalyst particles have to be separated from treated water after detoxification.

A very promising method for solving the above-mentioned problems is coupling the photocatalysis and membrane techniques. The membrane would play both the role of a simple

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barrier for the catalyst and of a selective barrier for the molecules to be degraded [9]. Molinari et al. [9] immobilized P25 photocatalyst on different flat polymeric membranes, but the membranes were slightly damaged by UV irradiation depending on the membrane materials. Other researchers also tried to use a batch-recirculated slurry photoreactor associated with a hollow fiber ultrafiltration membrane unit for the degradation of humic acid [10]. The photocatalyst described in the literature was fine powder TiO₂ P25 photocatalyst. The major problems with application of powder catalyst are greater membrane flux decline and membrane fouling. In fact, it is reported that TiO₂ particles in suspension have a better efficiency than that in immobilized form [11].

In this study, a submerged membrane photocatalysis reactor (SMPR) was designed for degradation of fulvic acid by using novel nano-structured TiO₂/silica gel photocatalyst. The synthesized TiO₂/silica gel catalyst particles with an average size of 50 μm were small enough for suspension to bubble air through and to stir. On the other hand, the particles in the suspension were big enough to settle while having the TiO₂ with high activity. Thus, it could be easily separated, recovered and reused by microfiltration separation process. More significantly, it was efficient to maintain high flux of membranes. This paper presented the experimental investigation on the possibility of coupling photocatalysis and membrane for degradation of fulvic acid in aqueous solution. The effects of the operational parameters including TiO₂ loading (0–0.6 mg/L), pH (3.4–10.3), airflow (0.01–0.09 m³/h) were investigated. The possibility of using novel TiO₂ for the prevention of MF membrane fouling for water purification was tested as well.

2. Experiments

2.1. Materials

The nano-structured TiO₂/silica gel photocatalyst employed in this study was obtained from Kunland University, Australia, with a BET surface area of 250 m²/g and an average particle size of 50 μm. Titanium dioxide P25 (Degussa, Germany), mainly anatase (ca. 70%) under the shape of non-porous polyhedral particles of ca. 30 nm mean size with a surface area of 50 m²/g. Fulvic acids were provided by Beijing Chem. Corp. The concentration of FA used in the experiment was 11.95 TOC mg/L. The microfiltration membrane module made of polyvinylidene (PVDF) hollow fiber membranes with a pore size of 0.2 μm and a filtration area of 0.2 m² was provided by Mo Tian Mo Corp., Tianjin. The pH of the solutions was adjusted by the addition of either H₂SO₄ or NaOH. All reagents utilized were of analytical purity.

2.2. Photoreactor

The SMPR process, shown in Fig. 1, was designed and conducted in a laboratory-scale. The reactor was made of plexiglass with a working volume of 3200 cm³. A light tight baffle separated the reactor into two parts: the photocatalytic oxidation zone and the membrane separation zone, which were connected by a bottom flow channel and an overflow channel. The low pressure UV lamp (11 W, Philip) emitting a wavelength of predominantly 253.7 nm was suspended vertically inside the quartz glass cylinder in the middle of the photocatalytic zone. Air was supplied from a porous titanium plate directly below the membrane

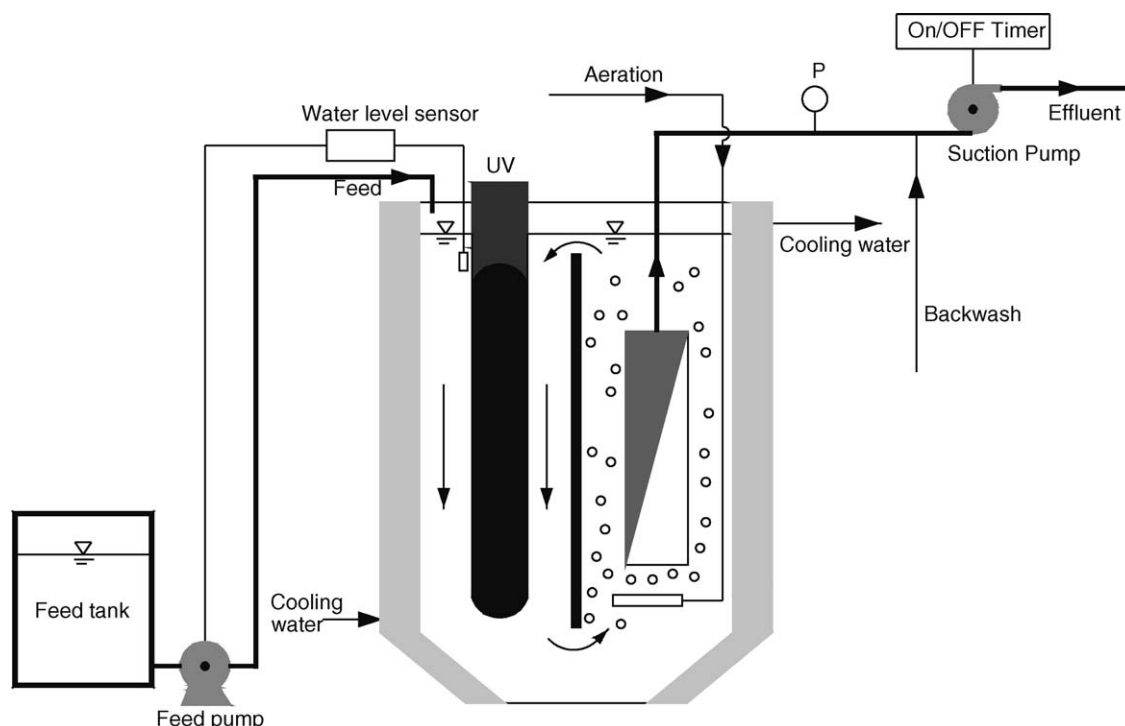


Fig. 1. Schematic diagram of the SMPR system.

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