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Control of submerged hollow fiber membrane fouling caused by fine particles in photocatalytic membrane reactors using bubbly flow: Shear stress and particle forces analysis

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ABSTRACT

In current work, bubbly flow was applied to control membrane fouling in photocatalytic membrane reactors (PMRs) under different aeration rate. The fine particles of TiO_2 ($d_{0.5}$ = 1.75 μm) in combination with natural organic matter (NOM) in suspended solutions were employed to conduct membrane fouling experiments in a submerged hollow fiber membrane tank. The results revealed that bubbly flow was an efficient approach to mitigate membrane fouling, leading to a rapidly decreasing fouling rate (dTMP/dt) (0.0908–0.0069 kPa/min) with the aeration increase. Additionally, computational fluid dynamics (CFD) simulations were implemented to calculated shear stress distribution on the spatial point of the membrane surface. Based on the detailed CFD simulation results, it was observed that the mean shear stress (0.505–2.111 Pa) increased rapidly with the increase of air flow rate (0–3.2 L/min). Furthermore, the forces exerted on the fine particles were analyzed in terms of calculated shear stress. It was quantitatively demonstrated that the streaming particles were prone to deposit on the vicinity of the layer and the particles deposited on the layer cannot be removed by hydrodynamics forces. It is expected that this study would be basis for further improvement of hydrodynamic condition design induced by bubbly flow in PMRs.

1. Introduction

Nano-sized TiO_2 has been studied extensively [1–4] because TiO_2-based photocatalysts with large surface area exhibit high photocatalytic efficiency. However, besides membrane process, it is very difficult to find conventional separation methods to separate TiO_2 particles from treated solutions. Hence, photocatalytic membrane reactors (PMRs) have shown great potential for use in energy-efficient water purification and wastewater treatment as they combine physical membrane separation and organic degradation by photocatalysis in a single unit [5–9].

Implementation of membrane process is mainly limited by membrane fouling [10,11]. Membrane fouling usually occurs as a result of the deposition and accumulation of unwanted impurities, such as organic compounds, inorganic species and microorganisms [12]. Specially, in a PMRs system, photocatalysis allows decomposition and mineralization of organic pollutants present in water because of the action of highly reactive hydroxyl radicals ($OH^{\cdot}$ radical dot) generated during advanced oxidation processes [9]. In spite of PMRs progress, membrane fouling remains the primary hindrance for its universal and large scale applications (the summary of latest membrane fouling in the field of PMRs could be seen in Table S1). In 2016 year, Zhang et al. [13] reviewed the membrane fouling in PMRs for water and wastewater treatment. Nearly 20 published reports in the literature about membrane fouling in PMRs were also summarized in this review. In principle, PMRs involves various photocatalysis reactions between the two main compositions in the system: organic pollutants and TiO_2 photocatalysts. These reactions and their interaction with membrane surface play a significant role in fouling formation during membrane filtration [13].

Abbreviations: UF, ultrafiltration; PMRs, photocatalytic membrane reactors; CFD, computational fluid dynamics; DLS, dynamic light scattering; TMP, transmembrane pressure (Pa); PVC, polyvinyl chloride; MWCO, molecular weight cutoff; HA, humic acid; CFV, cross-flow velocity; SEM, scanning electron microscopy.

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The cleaning of the membranes cannot be avoided, but its application can be minimized by the proper choice of operating conditions. Hydraulic cleaning (e.g. cross-flow velocity (CFV), air sparging) can be conducted to alleviate the membrane fouling in the PMRs system [6,14–16]. Ong et al. [6] found that the average membrane flux increased with the increasing air bubble flow rate from 0 to 5 L/min during oily wastewater treatment process using a submerged PMRs. The reason was due to the generation of circulation flow in the PMRs could limit the oil adsorbed onto the membrane surface. Choo et al. [14] reported an improvement of 60% in flux (initial water permeability: 183–219 L/(m^2 h)) as a consequence of the increased tangential velocity from 0.19 to 1.45 m/s in a PMRs, and it was because flow regimes could govern the deposition of TiO_2 particles on the membrane surface. Zhang et al. [15] demonstrated that specific cake resistance (the slop of the fitting line) was affected by CFV (0.5–2.0 m/s) during hollow fiber membranes (HFM) PMRs. Mozia et al. [16] demonstrated that selection of a proper feed cross flow velocity was a more efficient way to improve permeate flux through the microfiltration (MF) membrane. For details, at the feed CFV of 6 m/s the permeate flux was higher than the pure water flux for ca. 1650 dm^3/(m^2 h), whereas at 4.5 m/s–for ca. 510 dm^3/(m^2 h). Mozia et al. [17] also studied the stability of polyethersulfone ultrafiltration membranes in a PMRs. The results presented that when the feed CFV was changed from 0.8 to 0.4 m/s while TMP remained at 3 bar, the permeate flux decreased significantly and reached 62% of pure water flux.

In brief, the major reason for hydrodynamic conditions reducing membrane fouling is because of the induced shear stress [18–22], which can promote turbulence and instabilities of fluid and enhance the convective flow of particles away from the membrane. Although the application of air sparging for membrane fouling control during PMRs operation has been studied, insufficient attention has been devoted to a fundamental understanding of quantitative assessment of the stress on the spatial point of the membrane surface. A good understanding of shear stress distribution on membrane wall is of significant benefit for effectively designing the operating parameters of PMRs. Computational fluid dynamics (CFD) is a powerful tool to investigate the effect of shear stress on membrane fouling in the fields of ultrafiltration(UF) [18,20,22–24]. For two-phase flow of membrane process, the shear stress is related to bubble flow. Nonetheless, little is known about the detailed hydrodynamic characteristics on the whole membrane surface of HFM in the field of PMRs research.

Furthermore, based on the detailed shear stress distribution on the vicinity of HFM, the forces exerted on the particles would be estimated to predict the particle deposition behavior on the cake layer. Altmann et al. [25] divided particles during the membrane filtration into two parts: streaming particles and deposited particles. Forces on a streaming particle determine the particle transport to the layer, while forces on a deposited particle are linked with the tendency that if the attachment of particle to the cake layer is an irreversible process. It is noted that, besides drag force and lift force, the forces exerted on the deposited particles include the Waals forces and electrostatic interactions, which is due to the stabilizing propensity in presence of organic compounds [26,27]. The addition of HA would impart negative charges to particle surface and therefore increased the electrical double-layer (EDL) repulsive energy between particles [28]. Even Zhang et al. [15] provided a comprehensive understanding of the forces exerted on the streaming TiO_2 particle in CFV mode, an analysis of forces on a deposited particle was still lacking.

In this work, bubbly flow was implemented to alleviate membrane fouling of fine TiO_2 particles in combination with natural organic matter (NOM) during PMRs. Special attention was drawn to analyzing the shear stress distribution on membrane surface through CFD and to determining how the trans-membrane pressure (TMP) profiles varied with an increase of air flow rate. Moreover, we quantitatively examined the forces exerted on the streaming particle and deposited particle, which determinated the cake layer formation or deformation. The eventual aim of our work was to provide perspectives on the optimal bubbly flow hydrodynamic design of HFM PMRs.

2. Materials and methods

2.1. Experimental setup and membrane

A schematic diagram of the submerged ultrafiltration process is illustrated in Fig. 1. The system consisted of an ultrafiltration unit and an aeration unit. The untreated suspension was pumped into the 2.4 L membrane reactor from a feed tank. A hollow-fiber UF membrane module (3 fibers) with an effective surface area of 0.0042 m^2 was mounted vertically in the submerged membrane reactor. Permeate was continuously drawn from the membrane module by a peristaltic pump, and UF was performed with a given time operation. The Vacuum degree inside the membrane was measured using a vacuum transducer, which was connected to a PC for data logging. Simultaneously, the aeration at various intensity levels was provided to reduce the fouling. The air flow rate was monitored by a rotameter. Bubbles were generated from aerators, which were located at the bottom of the membrane tank and connected to the aeration tube.

UF membrane was composed of polyvinyl chloride (PVC) (Litree, China) with a molecular weight cutoff (MWCO) of 100 kDa. The inner and outer diameters of the membrane fibers were 1.4 and 1.8 mm, respectively. All membranes were preconditioned prior to filtration tests. To remove preservative materials on new membranes, UF membrane modules were washed with deionized (DI) water, submerged in 200 ppm sodium hypochlorite for 1 h, and then stored in DI water.

2.2. Membrane foulant and deposition experiments

The suspension studied in this work was considered to more closely represent the photocatalyst with NOM processed by ultrafiltration in the aqueous solution. TiO_2 suspensions were prepared by the addition of TiO_2 powder (EvonikP25) into deionized water. EvonikP25 (Aeroxide®, Germany) was obtained from Evonik Degussa Corporation (Parsippany, NJ). The photo catalyst P25 is a mixture of two crystalline phases, anatase (80%) and rutile (20%), and its purity is in excess of 99.5%. According to the provider, the specific BET-surface area, average particle size and density at 20 °C are 50 m^2 g^{-1}, 21 nm and 3.8 g/cm^3, respectively. The organic model compounds chosen for preparing the synthetic UF feed water were humic acid (HA, Aldrich), as representatives of NOM

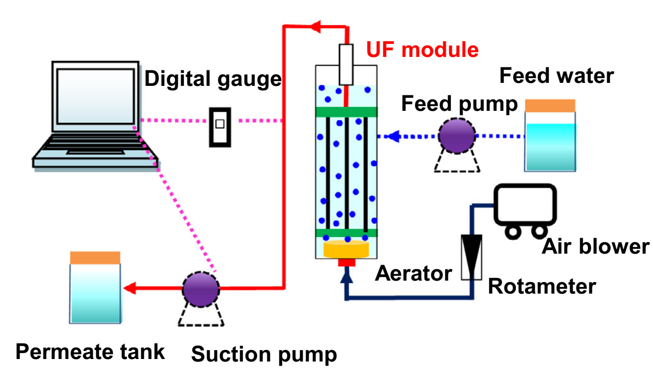

Fig. 1. Schematic diagram of experimental set-up.