



# A pilot-scale coupling catalytic ozonation–membrane filtration system for recirculating aquaculture wastewater treatment



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## HIGHLIGHTS

- A catalytic ozonation–membrane system was designed for aquaculture water treatment.
- Ti–Mn/TiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> membranes exhibited better catalytic capability than TiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub>.
- Suspended solids, TAN, NO<sub>2</sub><sup>-</sup> and COD<sub>Mn</sub>, were removed efficiently by this system.
- The recovery ratio of the aquaculture wastewater was as high as 95.8%.

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## ABSTRACT

A pilot-scale coupling catalytic ozonation–membrane filtration system was designed for the recirculating aquaculture wastewater treatment at a fish farm. The ceramic membranes were coated with Ti–Mn/TiO<sub>2</sub> oxide for both catalytic ozonation and filtration. Scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDX) images showed that the Ti–Mn oxides were distributed smoothly over both the membrane surface and the inner walls of the pores. Compared with the TiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> membranes, the Ti–Mn/TiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> membranes exhibited better catalytic ozonation capability with enhanced membrane antifouling (a normalized permeate flux obtained after 2 h operation was 0.56) and higher removal efficiency of organic matters (52.1%). The ozone dose has an important effect on the degradation of organic matters and ammonia. At an ozone dose of 52 mg/min (stable permeate was obtained after 1 h operation), turbidity and nitrite were almost completely removed, and the concentration of total ammonia nitrogen (TAN) dropped to less than 0.1 mg/L. The permeate quality satisfied the water quality requirement for aquaculture. The recovery ratio of the aquaculture seawater was as high as 95.8%.

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## 1. Introduction

The limited suitable water supplies, the absence of land sources and the concerns over the environmental pollution are regarded as key obstacles to further expansion of traditional flow-through aquaculture systems [1]. Thus, recirculating aquaculture systems, which can remove solids, organic matters and total ammonia nitrogen, and can kill microbes to realize low water exchange compared with flow-through systems, are of great interest in aquaculture water treatment. Conventionally, a good recirculating aquaculture system should incorporate unit processes to remove suspended solids and organic matters, add oxygen, and control ammonia, nitrite, pH and pathogenic bacteria [2]. However, the present recirculating aquaculture systems, which are mainly based on the biological method, suffer some problems, such as frequent backwashing of the

biofilter [3], complex biological denitrification process [4], changeable biological nitrification beds [5], long hydraulic retention time or large area occupied, and the long start-up period (3 or 4 weeks) [6].

Alternatively, membrane filtration is effective at removing particles and organic matters from wastewater in aquaculture process [7,8]. However, the fouling of membranes caused by inorganic or organic matters retained on the membrane surface is inevitable during the operation and decreases the membrane filtration capacity [9,10]. The general approaches to solve the membrane fouling problem are chemical cleaning and back washing. Nevertheless, the periodical chemical cleaning or backwashing of the membranes can generate additional wastewater and increase economic costs. In recent years, the integration of advanced oxidation processes (AOP) in the membrane filtration process to repress membrane fouling has become a popular topic. Ozonation, one kind of AOP, is frequently used in aquaculture systems for water quality improvements and disinfection [11,12]. The combination of ozonation with the membrane filtration process is an effective

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**Table 1**  
Characteristics of membrane module.

Parameters	
Type	Tubular
Pore size	2 $\mu\text{m}$
Material	Ceramic
Porosity	40%
Effective filtration area per module	0.7 $\text{m}^2$
Initial seawater flux (20 °C)	Ti–Mn/TiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> membrane: 269.6 $\pm$ 4.6 (L/m <sup>2</sup> h bar) TiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> membrane: 340.5 $\pm$ 5.7 (L/m <sup>2</sup> h bar)
Membrane dimension	30 mm (D) $\times$ 1016 mm (L) $\times$ 19 channels

approach to mitigating membrane fouling in treating surface water [13–15]. In this process, the contaminants (suspended and colloidal inorganic or organic substances) are intercepted and deposited on the membrane surface by membrane separation, then the enriched contaminants are degraded by ozonation. In the meantime, the membrane fouling can be reduced to some extent and the water quality can be improved. Catalytic ozonation by catalysts can promote the decomposition of ozone and the formation of hydroxyl radicals ( $\cdot\text{OH}$ ) or other radicals [16,17].  $\cdot\text{OH}$  can react with almost all types of contaminants due to their high reactivity. Consequently, coating ozone catalysts on the membrane is a more promising subject for improving water quality and alleviating membrane fouling [18–20].

Combined catalytic ozonation and membrane filtration were applied for surface water [13,14] or tertiary effluent [15] treatment. However, no special attention was paid to the removal of TAN and nitrite nitrogen ( $\text{NO}_2^-$ -N) in this process. TAN and  $\text{NO}_2^-$ -N are toxic to fish even at a low concentration. Long-term exposure of TAN or  $\text{NO}_2^-$ -N would significantly inhibit the growth of fish because of a decrease in food intake. Although TAN,  $\text{NO}_2^-$ -N and dissolved organic matters are difficult to be removed by membrane filtration alone, they are expected to be removed by catalytic ozonation. There have been no reports about the combination of catalytic ozonation with membrane separation for aquaculture wastewater treatment till now. In the present study, a pilot-scale coupling catalytic ozonation–ceramic membrane filtration system was established for recirculating aquaculture wastewater treatment at a fish farm. This study focused on the feasibility of the coupling system, the permeate flux of catalytic membranes, the recovery ratio of the aquaculture wastewater and the water quality which should meet the water quality requirement for aquaculture. The performances of Ti–Mn/TiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> and TiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> catalytic membranes in terms of

antifouling and removal of total ammonia nitrogen and chemical oxygen demand (permanganate index,  $\text{COD}_{\text{Mn}}$ ) were also investigated.

## 2. Experimental section

### 2.1. Membrane preparation

#### 2.1.1. Preparation of intermediate layer (TiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> membranes)

Commercial tubular ceramic membranes ( $\alpha$ -Al<sub>2</sub>O<sub>3</sub>, 30 mm (D)  $\times$  1016 mm (L)  $\times$  19 channels, Jiexi Lishun Technology, Inc., China) were first coated with rutile TiO<sub>2</sub> (with a particle size of approximately 200 nm) (JinZhou PengDa Titanium Dioxide Manufacturing Co., Ltd., China) by a dip-coating method to form the TiO<sub>2</sub> intermediate layer. A home-made dip-coating system was used to coat the membranes. In the coating procedure, a cylindrical vessel was first filled with TiO<sub>2</sub> suspension (8 wt.%), and then the tubular ceramic membranes were immersed in the suspension for 2 min. Afterwards, the TiO<sub>2</sub> suspension was discharged by a peristaltic pump at a velocity of 200  $\mu\text{m/s}$ . This process was repeated twice. Subsequently, the coated membranes were dried at 80 °C for 12 h and sintered at 1000 °C for 1 h in air with a heating and cooling ramp of 10 °C/min.

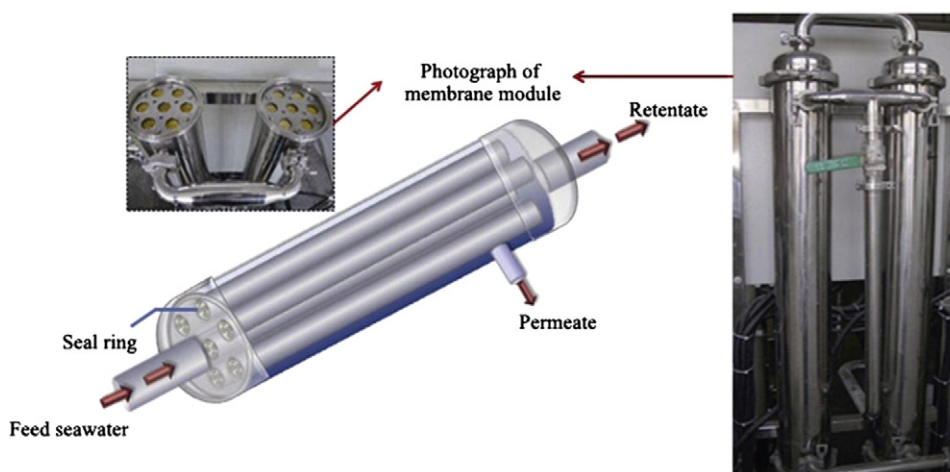
#### 2.1.2. Preparation of Ti–Mn oxide layer (Ti–Mn/TiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> membranes)

The Ti–Mn oxide layer was prepared as follows: Ti–Mn sol was prepared as introduced in our previous work [21]. Pluronic F127 (45 g) was dissolved in 1000 mL ethanol, and then 250 mL titanium (IV) isopropoxide, 71.5 mL acetic acid, 38.45 mL manganese (II) nitrate solution (50%), and 75 mL pure water were added to the solution dropwise under vigorous stirring to obtain a light yellow transparent Ti–Mn sol with a Ti/Mn molar ratio of 1:0.1. The TiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> membrane channels were filled with Ti–Mn sol for 2 min, and then the Ti–Mn sol was extracted at a velocity of 200  $\mu\text{m/s}$ . This dip-coating process was also repeated twice. After coating, the Ti–Mn/TiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> membranes were sintered in air at 550 °C for 1 h to fuse the Ti–Mn catalyst to the underlying TiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ceramic membranes.

### 2.2. Membrane module

The characteristics of the membrane module are listed in Table 1. Every tubular ceramic membrane had a filtration area of 0.1  $\text{m}^2$ , and seven membranes were assembled into one module (Fig. 1). Two modules were connected in parallel to obtain the desired treatment capacity.

A schematic diagram and photographs of the membrane module are shown in Fig. 1. A cross-flow filtration mode at a flow velocity of  $\sim$ 1.04 m/s was adopted in all membrane tests.



**Fig. 1.** A schematic diagram and photographs of the membrane module.

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