Journal of Water Process Engineering 21 (2018) 133–142

Contents lists available at ScienceDirect

Journal of Water Process Engineering

journal homepage: www.elsevier.com/locate/jwpe

Searching for ultrafiltration membrane molecular weight cut-off for water treatment in recirculating aquaculture system

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ARTICLE INFO

Keywords:
Recirculating aquaculture system
Ultrafiltration
Fouling
Molecular weight cut-off

ABSTRACT

Ultrafiltration (UF) has been proposed as a promising technology in recirculating aquaculture systems (RAS) to remove both organic contaminants and other fine contaminants including viruses and pathogenic bacteria. However, fouling is still a severe problem during this application. This paper investigated the fouling behavior of three different UF membranes examined using five different aquaculture contaminants. The experiments were performed using UF membranes with molecular weight cut-off 10, 50, and 100 kDa. Humic acid, shrimp feed, *Spirulina sp.*, *Vibrio harveyi* and IHHNV were used as contaminant models. Scanning electron microscope was used to visualize the presence of foulant on the membrane surface. The results showed that fouling behavior was affected by both membrane cut-off (pore size) and foulant type. Two fouling behaviors were observed: (i) rapid flux decline at the early stage of filtration followed by relatively constant permeate flux until experiments finished, and (ii) rapid flux decline at the early stage of filtration followed by a gradual decrease in permeate flux. Due to its reliable flux value and high rejection, 100 kDa UF membrane should be considered as the most suitable UF membrane for RAS application.

1. Introduction

Aquaculture, the farming of aquatic organisms and plants in both coastal and inland areas, is very important sector in achieving world food security. Therefore, much attention has been paid for a sustainable aquaculture on the one hand. Recirculating aquaculture system (RAS), which recirculates or reuses water after undergoing treatment, is one of the developments towards sustainable aquaculture. On the other hand, a severe problem found in aquaculture system is diminishing or decreasing of animals as a result of disease or lack of oxygen. This problem is found in conventional aquaculture systems, recirculating aquaculture system (RAS) and hatchery systems. More attention should be paid for the RAS, where water is continuously reused via recycling process [1–5].

The presence of excess organic compounds in an aquaculture system lowers the dissolved oxygen (DO) content (both chemical and biological) that has an impact on the lack of oxygen supply to the animals respiratory. Organic compounds in aquaculture system can be derived from the water used itself, the residual of feed, and the metabolism of animals. It was reported that eighty percent (dry weight) of feed used in the aquaculture system is released as an animal excretion [6]. As an example, production of 100 kg catfish produced 1190 kg of dry solids, 60 kg of nitrogen compounds and 12 kg of phosphorus compounds [7]. In the more recent study [8], it was reported that 25% of the feed applied to the aquaculture system will end up as suspended solid. Therefore, the organic compounds should be reduced to the safe level for culture growth of aquatic animals such as fish and shrimp. In addition, viruses, microalgae, and pathogenic bacteria are also frequently found in aquaculture systems in which the presence of these contaminants can cause the destruction of animals.

To avoid the destruction of aquatic organisms, removal of organic compounds and other contaminants from aquaculture systems has to be conducted. This removal is usually performed by the centrifugation, clarification, gravity filtration or precipitation process, screening with different pore sizes (60–200 microns), biofilter, and biological processes [9–13]. The fundamental weakness of these techniques is the leakage of fine particles or contaminants including pathogenic bacteria and viruses [14]. In addition, biological processes have other problems such as their complexity especially for denitrification [15], high hydraulic retention time (HRT) [16], and high backwash intensity of the

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https://doi.org/10.1016/j.jwpe.2017.12.006
Received 31 August 2017; Received in revised form 8 November 2017; Accepted 9 December 2017

biofilter [17].

Foam fractionation has been proposed to remove those fine particles and can be combined with other processes such ozone to enhance the separation efficiency [18,19]. Nevertheless, this technology is not suitable for large and primary particles removal. Phytoremediation has also been proposed for treating aquaculture wastewater [20]. However, the use of plants in aquaculture system will block photosynthesis and consequently, the oxygen content in water will decrease. To solve these problems, application of membrane technologies in aquaculture systems has been proposed by many researchers [3,21–25]. In a recent study, it was shown that the integration of membrane filtration into RAS increased the quality of water effluent [2]. Further, several membrane technologies have been applied in small aquaculture industries [3,26]. A coupling catalytic ozonation-membrane filtration system for the recirculating aquaculture wastewater treatment using membranes coated by Ti-Mn/TiO_2 oxide was reported in a recent study by Chen et al. [27]. They claimed that their coupling system results in good permeate quality, short start-up period, and the stable operating conditions. However, special attention should be paid for the residual ozone, which is harmful to the aquatic organisms. It was reported that algae paste, decaying rotifers, and dry feed seemed to contribute to the most of membrane fouling [28].

In general, the results of both laboratory and field experiments indicated that membrane technologies deliver a promising potential for removing fine contaminants including organic compounds, viruses, and pathogenic bacteria. However, as in other applications, fouling–the decline of flux over time due to the deposition of suspended or dissolved substances on external surface, at the pore openings or within its pore [29,30]–reduces significantly the performance of the process and consequently prevents a more widespread commercial applicability in aquaculture system.

Numerous studies on the application of UF membrane in RAS have been reported. However, different results were observed among different reports. It is certainly due to different process condition, membrane property, contaminant, and feed water characteristic are involved. As a result of this situation, it is hard to determine the best molecular weight cut-off (MWCO) of UF membrane that should be used in an aquaculture system. It is important to mention that MWCO is a parameter usually used to express the retention and membrane pore size of ultrafiltration membrane. To the best of our knowledge, no systematic study has been performed to determine the most suitable UF membrane pore size that should be applied in RAS. It should be kept in our mind that the performance of a process using membrane is strongly influenced by membrane pore size or MWCO [31–33]. Systematic fouling study using seawater as the feed has been performed [34]. Nevertheless, their study was dedicated to the pretreatment of RO membrane for desalination.

As consequences of various contaminants with different characteristics exist in aquaculture systems and various commercial UF membranes with different MWCOs are available, selection of UF membrane based on their MWCO is a very important step in practical application of UF membrane in RAS. The main aim of this study is to determine the suitable MWCO of UF membrane in RAS application considering the fouling mechanism study. It is important to point out that for the practical application; the use of different MWCO membrane for the removal of different contaminants is not possible. Furthermore, the membranes used in RAS cannot be chosen only based on the manufacturer specification. The best MWCO determination is based on the efficiency and fouling behavior, which corresponds to the resulting flux. Various model contaminants of aquaculture systems, i.e. humic acid, residual shrimp feed, microalgae (*Spirulina* sp.), pathogenic bacteria (*Vibrio harveyi*) and virus (IHHNV, Infection hypodermal and hematopoietic necrosis virus) were used in this study. These five contaminants were expected to be able to represent natural organic matters (NOMs), feed residuals, microalgae, bacteria and viruses; respectively, which usually exist in aquaculture system.

2. Materials and methods

2.1. Materials

Since the consistency of membrane properties manufactured industrially is typically better than lab-made membrane, the commercial UF membranes (supplied by Alfa Laval, Denmark) were used in this study. The membranes included GR81PP (10 kDa, polyethersulfone), GR51PP (50 kDa, polysulfone) and GR40PP (100 kDa, polysulfone). The contaminant models used were shrimp feed (SF) with the composition was (%wt) protein > 36%, crude fiber < 4%, fat > 5%, water < 12% and ash < 12% (obtained from P.T. Gold Coin, Indonesia), humic acid (HA, purchased from Sigma-Aldrich), *Spirulina platensis* (MA, obtained from Center for Bioprocess and Renewable Energy, Diponegoro University), marine bacterium *Vibrio harveyi* (VH, obtained from BBPBAP, Jepara, Indonesia), and IHHNV (obtained from BBPBAP, Jepara, Indonesia). The experiment was also performed using mixture of these contaminant models (SMH). The usage of these contaminants was based on the results of preliminary analysis of water quality from the real aquaculture system as well as previous literatures [1–5]. Real aquaculture water (RAW) with total dissolved solids ~4.1 g/L was obtained from the aquaculture pond in Jepara, Indonesia. Sodium chloride and HCl were purchased from Merck. Purified water produced from home-made RO-ion exchange system was used for all experiments. In order to apply the UF membranes in RAS, all foulant solutions were prepared by dissolving the contaminant models in 10 g/L of sodium chloride solution.

2.2. Methods

2.2.1. Investigation of fouling behavior

Study on fouling behavior was performed by investigation of membrane-solute interactions (adsorptive fouling) and membrane-solute–solute interactions (ultrafiltration fouling). The investigation of adsorptive fouling was carried out by using a dead-end stirred cell filtration system (Amicon cell model 8010 from Millipore) and followed our earlier reported work [35]. Briefly, pure water flux (J_0) was measured for each membrane sample. The pure water flux was measured at a pressure of 300 kPa for 10 kDa UF membrane and 100 kPa for both 50 and 100 kDa UF membranes. A model of contaminant solution with certain concentration (SF (1 g/L), MA (1 OD in water with 10 g/L TDS), HA (1 g/L), VH (10^8 CFU), IHHNH (10/500 mL/mL)) was added to the cell. SMH composed of 0.25 g/L SF, 0.5 OD MA and 0.25 g/L HA was also used. Thereafter, the outer membrane surface was exposed for 3 h without any flux at a stirring rate of 300 rpm and atmospheric condition. The preliminary experiments showed that 3 h of adsorption was sufficient to achieve saturation of the surface adsorption capacity for all foulant models. This experiment condition was also supported by our previous studies [30,36]. Afterward, the solution was removed, and the membrane surface was rinsed twice by filling the cell with pure water (5 mL) and shaking it for 30 s. Pure water flux (J_a) was again measured. The extent of adsorptive fouling was expressed in term of relative water flux reduction (RFR; cf. Eq (1)), which was calculated from the water fluxes at the same pressure before and after adsorptive fouling. The effect of contaminant concentration on RFR was also investigated using SF, HA and MA as contaminant models. The concentration of SF and HA was varied from 0 to 5 g/L, whereas the concentration of MA was varied from 0 to 1 OD.

$$RFR = \frac{J_0 - J_a}{J_0} \tag{1}$$

A home-made laboratory scale for cross-flow filtration test was used in all ultrafiltration experiments [30]. The set-up consisted of a feed tank (3 L volume), a flow indicator, a pump, a pressure indicator connected to feed side of the membrane to determine the trans-membrane pressure and a flat–sheet membrane cell. A simplified diagram of the set-up