# A novel sponge-submerged membrane bioreactor (SSMBR) for wastewater treatment and reuse

Wenshan Guo, Huu-Hao Ngo, Saravanamuthu Vigneswaran<sup>\*</sup>,

Wen Xing, Pavan Goteti

Faculty of Engineering, University of Technology, Sydney, P.O. Box 123, Broadway, NSW 2007, Australia

\* Correspondence author, tel: +61-2-9514-2641, fax: +61-2-9514-2633,

E-mail: S.Vigneswaran@uts.edu.au

**Abstract:** Membrane fouling has been regarded as one of the biggest challenges to widespread application of membrane bioreactor (MBR). This study focuses on minimizing the membrane fouling and improving the performance of submerged membrane bioreactor (SMBR) by porous sponge addition. The effects of sponge addition on sustainable flux and membrane fouling were investigated. Acclimatized sponge could significantly increase the suspended growth in SMBR with biomass of 16.7 g/L(sponge). With sponge volume fraction of 10%, SSMBR could enhance sustainable flux up to 50 L/m<sup>2</sup>.h compared with sustainable flux of SMBR (only 25 L/m<sup>2</sup>.h). SSMBR also exhibited excellent results in terms of DOC removal (over 95%), COD removal (over 97%), lower transmembrane pressure development and oxygen uptake rate. Over 89% of NH<sub>4</sub>-N and 98% of PO<sub>4</sub>-P were removed when SSMBR was operated with a MLSS concentration of 15 g/L.

*Keywords:* Membrane bioreactors; Oxygen uptake rate; Critical flux; Microbial growth; Biodegradation; Wastewater treatment

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# **INTRODUCTION**

Membrane bioreactors (MBRs) have been used as an innovative and promising option for wastewater treatment and reuse. Membrane bioreactor technology encourages wastewater reuse and improves water sustainability. This technology is simple to operate, needs modest technical support, takes up little space and can remove many contaminants from wastewater in one step [1]. MBR comprises of a suspended growth bioreactor and a filtration on porous membrane, which leads to the total retention of .biomass (high microbial concentration) and improved biological reactor operation (high sludge ages) in the bioreactor [2]. In MBR system design, the submerged membrane configuration (SMBR) can assist in significantly reducing power consumption.

Although MBR offers the effective separation of pollutants and tolerance to high or shock loadings, MBR technology is currently facing some research and development challenges such as membrane fouling, high membrane cost and pretreatment. Membrane fouling is the most difficult challenge, which increases operational cost and shortens the membrane life [3]. To overcome membrane fouling problem, various studies have been conducted to understand and minimize membrane clogging, such as using intermittent suction instead of continuous suction [4], alum and natural zeolite addition [5], association of SMBR and powdered activated carbon (PAC) [6,7] and using modified cationic polymers [8] etc. Besides, suspended carriers were used to attach activated sludge and reduce the effect of suspended solids on membrane fouling. Lee at al. [9] found out that membrane-coupled moving bed biofilm reactor (M-CMBBR) had much lower biofouling rate than a conventional MBR when using activated carbon coated polyurethane cubes as attached growth media. Yang et al. [10] also investigated a hybrid membrane bioreactor (HMBR) with porous, flexible suspended carriers to treat terephthalic acid wastewater. The HMBR was efficient in controlling membrane fouling, especially the cake layer on the membrane. In short-term experiments, the critical flux of HMBR increased by 20% and the cake resistance of HMBR decreased by 86% in comparison with conventional MBR.

Operating membrane system below critical flux is also one of the rational approaches to control membrane fouling. This concept of critical flux has been introduced in the mid 1990's with theoretical and experimental evidence. The critical flux hypothesis for microfiltration (MF) is that there exists a flux below which a decline of flux with time does not occur; above it fouling is observed [11]. Normally, two different methods are used to determine the critical flux: (i) Based on particle mass balance: By monitoring the change of particle concentration in the fluid phase, the extent and rate of particle deposition at membrane surfaces can be determined at various permeation rates. The highest flux value at which no particle deposition is observed, is taken as the critical flux; (ii) Based on the increase in transmembrane pressure (TMP) required to maintain a constant permeate flux: The TMP increases during the constant permeate flux operation in order to compensate the increase in the resistance to permeation. Accordingly, the critical flux is the flux below which there is no presence of this increase in resistance to permeation (i.e. the TMP is constant with time). In the case of SMBR, 'sub-critical' flux operation does not appear to be feasible and the challenge is determination of the 'sustainable flux', where TMP rise is tolerable before rapid fouling and increase of TMP is seen to occur [12].

Using MBR to removal nutrients is also a main focus of advanced wastewater treatment technology. In aerobic MBRs, almost complete nitrification can be achieved, while denitrification needs the addition of an anaerobic tank prior to the aeration tank with conventional recycle [13]. To solve this problem, aerated MBR systems could either be coupled with chemical treatment process such as coagulation and adsorption [14, 15], or be associated with a separated anoxic tank for denitrification [16, 17]. In present situation, although these MBR systems have shown an improvement of nitrogen removal, phosphorus has not been removed significantly through these systems. Thus, anaerobic condition was added to enhance phosphorus removal. Ahn et al., [16] reported that approximately 93% phosphorus was removed in an improved sequencing anoxic/anaerobic MBR. Zhang et al. [18] examined a sequencing batch membrane bioreactor (SBMBR) in alternating aerobic and anoxic/anaerobic condition for enhancing nitrogen and phosphorus removal up to approximately 90%. Meanwhile, Sponge has been considered as an ideal attached growth media because it can act as a mobile carrier for active biomass, reduce the cake layers formed on the surface of membrane and provide large porous support which increases the possibility of contact between microorganism and the organic substrate [19, 20, 21, 22].

In this study, a novel sponge-submerged membrane bioreactor (SSMBR) has been developed for alleviating membrane fouling, enhancing permeate flux and improving phosphorus removal. The objective is to investigate the performance of SSMBR for treating a synthetic domestic wastewater as well as reducing membrane fouling in terms of sustainable flux. The effect of the mixed liquor suspended solids (MLSS) was also studied.

# MATERIALS AND METHODS

#### Wastewater

The experiments were conducted using a synthetic wastewater to avoid any fluctuation in the feed concentration and provide a continuous source of biodegradable organic pollutants. It was used to simulate high strength domestic wastewater (just after primary treatment process). The synthetic wastewater has dissolved organic carbon (DOC) of 120-130 mg/L, chemical oxygen demand (COD) of 330-360 mg/L, ammonium nitrogen (NH<sub>4</sub>-N) of 12-15 mg/L and orthophosphate (PO<sub>4</sub>-P) of 3.3-3.5 mg/L (COD: N: P = 100:5:1). The composition of synthetic wastewater is given in Table 1 [2]. Basically, NaHCO<sub>3</sub> or H<sub>2</sub>SO<sub>4</sub> were added to the wastewater to maintain a constant pH of 7.

Table 1. Constituents of the Synthetic Wastewater

## Sponge-Submerged Membrane Bioreactor (SSMBR) Set-up

A polyethylene hollow fiber membrane module was used with the pore size of 0.1  $\mu$ m and surface area of 0.195 m<sup>2</sup> (Mitsubishi-Rayon, Japan). The schematic diagram of the SSMBR is shown in Fig. 1. Synthetic wastewater was pumped into the reactor using a feeding pump to control the feed rate while the effluent flow rate was controlled by a suction pump. Level sensor was used to control the wastewater volume in the reactor. A pressure gauge was used to measure the TMP and a soaker hose air diffuser was used to maintain a high air flow rate (9 L/min or 2.77 m<sup>3</sup>/m<sup>2</sup><sub>(membrane area)</sub>.h). For physical cleaning of membranes, filtrate backwash was used at a backwash rate of 30 L/m<sup>2</sup>.h. SSMBR was filled with sludge from a local Wastewater Treatment Plant and acclimatized to synthetic wastewater. The reticulated

porous polyester-polyurethane sponge (PUS) was used in sponge-SMBR system. The PUS has density of 28-30 kg/m<sup>3</sup> with cell count of 45 cells/in. The dimensions of the sponge cubes are 10 mm, 10 mm and 10 mm in length, width and thickness respectively. Before running the experiments, the sponge cubes were acclimatized to synthetic wastewater.

Figure 1. Experimental set-up of SSMBR

#### Analysis

DOC of the influent and effluent was measured using the Analytikjena Multi N/C 2000. The analysis of COD and the measurement of mixed liquor suspended solids (MLSS) and biomass (monitored as mixed liquor volatile suspended solids, MLVSS) were according to Standard Methods [23]. For measuring MLSS and biomass, three samples were taken each time and the average values were then calculated. NH<sub>4</sub>-N and PO<sub>4</sub>-P were measured by photometric method called Spectroquant® Cell Test (NOVA 60, Merck). The bacterial activity during operation of MBR can be evaluated by measuring the oxygen consumption (by respirometric procedure). YSI 5300 Biological Oxygen Monitor was used to measure specific oxygen uptake rate (SOUR). It is a useful tool for measuring respiration, oxidative activity, and cellular metabolism. The oxygen consumption measurement can be achieved through use of oxygen electrode with oxygen permeable Teflon membrane. Voltage generated from the reaction is proportional to the oxygen concentration of the sample and produces oxygen uptake or evolution curves in 2 to 15 minutes. Total viable counts and total coliform counts were carried out using spread plate technique on nutrient agar and MacConkey agars as media respectively. All samples were diluted using 0.1%

bacteriological peptone water. Nutrient agar, MacConkey agar and bacteriological peptone were obtained from OXOID<sup>®</sup>.

# **RESULTS AND DISCUSSION**

#### **Attached Biomass Growth on Sponge During Acclimatization**

The polyester-polyurethane sponge (PUS) cubes (1.5 L) were acclimatized to the activated sludge in SMBR in a 10 L aeration tank with an initial MLSS of 5 g/L before running with membrane. The average concentrations of MLSS and biomass (MLVSS) were measured and the results are shown in Fig. 2. The MLSS and biomass on the sponge reached stable growth phase (around 18.1 and 16.7 g/L(sponge) respectively) after 15-day acclimatization. A quantitative microbiological analysis was carried out with acclimatized sponge. High degree of growth was noticed in sponge and bacterial numbers increased up to  $2.1 \times 10^7$  cfu/ml(sponge) after 25 days of acclimatization. The mixed liquor in aeration tank also had the viable count of  $2.6 \times 10^5$  cfu/ml and total coliform of  $4.0 \times 10^3$  cfu/ml. SOUR results also indicated that the microbial activity was strong at the first 10 days which corresponded to the fastest equilibrium of SOUR (97.5% on the 5<sup>th</sup> day and 97% on the 7<sup>th</sup> day within 8 minutes respectively). SOUR was then remained constant with much lower equilibrium rate (96% within 26 minutes) (Fig. 3.).

Figure 2. The attached growth on sponge during acclimatization

Figure 3. SOUR variation of attached growth on sponge during acclimatization

## Sustainable Flux of Sponge-SMBR System

Acclimatized sponge cubes were added in the SMBR system with certain volume (percent of effective SMBR volume of 6 L). Sustainable fluxes were measured in the

sponge-SMBR system (SSMBR) with the same initial MLSS of 10 g/L. Sponge volume fraction in the reactor was varied at 0% (no sponge), 10% and 20% (Fig. 4. and Table 2). After every 1 hour flux-step, 1 minute- backwash was provided at a backwash rate of 30 L/m<sup>2</sup>.h using membrane filtrate. The purpose of backwash was mainly to minimize the TMP increase due to reversible fouling during every experimental flux-step, which could lead to TMP developments. As can be seen in Fig. 4(b), suspended sponge could significantly reduce the membrane fouling and enhance sustainable flux (two times increase in flux with the sponge volume fraction of 10%). A slight decline of sustainable flux was observed for 20% of sponge fraction. This is mainly due to the reduction of sponge cube mobility in the reactor. The SSMBR system could achieve higher quality effluent with a total organic carbon removal efficiency of over 95% in all cases.

*Figure 4.* Constant filtration fluxes versus TMP of SSMBR (LMH =  $L/m^2$ .h)

Table 2. Sustainable flux and effluent quality in SSMBR

### **Comparison of Different Sludge Concentrations**

### DOC and COD removal

The SSMBR system was operated at different sludge concentrations in terms of constant MLSS concentration from 5 g/L to 15 g/L. The permeate flux was kept constant at 30 L/m<sup>2</sup>.h with effective SSMBR volume of 7 L. Figs. 5. and 6. show the DOC and COD removal efficiencies during 7 days of operation. The results indicated that SSMBR system achieved superior DOC removal efficiencies (over 95%) for all three MLSS concentrations studied. COD removals were over 97% at MLSS concentrations of 10 g/L and 15 g/L, while lower COD removal values were obtained at the lowest MLSS concentration (5g/L). Table 3 presents the SOUR of mixed liquor

in SSMBR on 2<sup>nd</sup> and 5<sup>th</sup> day of operation, suggesting the higher MLSS concentration

could achieve higher oxygen consumption rate in the system.

*Figure 5.* DOC profile of SSMBR system at different sludge concentrations (filtration flux =  $30 \text{ L/m}^2$ .h; backwash rate =  $30 \text{ L/m}^2$ .h; backwash = 1 minute every half an hour; HRT = 1.2 hours)

*Figure 6.* COD profile of SSMBR system at different sludge concentrations (filtration flux =  $30 \text{ L/m}^2$ .h; backwash rate =  $30 \text{ L/m}^2$ .h; backwash = 1 minute every half an hour; HRT = 1.2 hours)

Table 3. SOUR of mixed liquor in SSMBR

## TMP development

The variation of TMP values were measured during the operation of SSMBR at different sludge concentrations (Fig. 7.). As can be seen from the results, the lowest TMP development (29.5 kPa) was observed when SSMBR was operated with a sludge concentration of 15 g/L. The higher the MLSS, the lower TMP development could achieve when the MLSS concentrations varying from 5 to 15 g/L in sponge-SMBR system. Thus, MLSS concentration could be considered as one of the key elements for evaluating TMP development.

*Figure 7.* TMP development of SSMBR system at different sludge concentrations (filtration flux =  $30 \text{ L/m}^2$ .h; backwash rate =  $30 \text{ L/m}^2$ .h; backwash = 1 minute every half an hour; HRT = 1.2 hours)

### NH<sub>4</sub>-N and PO<sub>4</sub>-P removal

Nutrients removal in the SSMBR was investigated in terms of ammonium nitrogen  $(NH_4-N)$  and orthophosphate  $(PO_4-P)$ . 89% of  $NH_4-N$  was removed with MLSS concentration of 15 g/L while there was only 75 % of  $NH_4-N$  removal with MLSS concentration of 5 g/L (Fig. 8.). Normally, an anaerobic/aerobic (or anoxic) sequence is necessary to improve biological phosphorus removal and phosphorus removal

increases with the increasing of sludge retention time (SRT) in anaerobic/anoxic sequencing batch reactor [24]. With three different MLSS concentrations, the SRTs were 70 days, 60 days and 35 days respectively for MLSS of 5 g/L, 10 g/L and 15 g/L. However, the SSMBR system could reach very high PO<sub>4</sub>-P removal efficiencies in all three cases with notable SRT variations (Fig. 9). Over 98% of PO<sub>4</sub>-P was removed and PO<sub>4</sub>-P concentration of the effluent was less than 0.1 mg/L in all three occasions. This is due to the sponge provide good anoxic condition around the surface of sponge and anaerobic condition inside the sponge which make aerobic SMBR able to get higher removal efficiency of PO<sub>4</sub>-P. The quantitative microbiological analysis also showed that total coliform were not found in acclimatized sponge, which may prove the sponge had an anoxic/anaerobic condition around and inside the sponge.

*Figure 8.* NH<sub>4</sub>-N profile of sponge-SMBR system at different sludge concentrations (filtration flux =  $30 \text{ L/m}^2$ .h; backwash rate =  $30 \text{ L/m}^2$ .h; backwash = 1 minute every half an hour; HRT = 1 hour)

*Figure 9.* PO<sub>4</sub>-P profile of sponge-SMBR system at different sludge concentrations (filtration flux =  $30 \text{ L/m}^2$ .h; backwash rate =  $30 \text{ L/m}^2$ .h; backwash = 1 minute every half an hour; HRT = 1 hour)

# CONCLUSIONS

Sponge addition in the SMBR could significantly improve the sustainable flux and reduce membrane fouling. The acclimatized sponge could hold 16.7 g/L(sponge) biomass which significantly increased the suspended growth in SMBR. With sponge volume fraction of 10%, SSMBR was found to give superior result that could improve sustainable flux by 2 times than that of SMBR alone.

SSMBR achieved high DOC (over 95% at MLSS concentrations of 5,10 and 15 g/L) and COD removal efficiencies (over 97% at MLSS concentrations of 10 and 15 g/L) when running 7-day experiment at filtration flux of 30 L/m<sup>2</sup>.h. The MLSS

concentration is one of the main factors for TMP development. With higher MLSS (up to 15 g/L), TMP development was lower. In addition, SSMBR revealed outstanding PO<sub>4</sub>-P removal and the effluent PO<sub>4</sub>-P concentration of SSMBR was lower than 0.1 mg/L. Therefore, sponge addition to submerged membrane bioreactor can be an excellent solution to reduce membrane fouling, enhance permeate flux and improve phosphorus removal. Further studies on the improvement of complete phosphorus and nitrogen removal simultaneously in SSMBR are necessary.

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Compounds	Molecular weight	Concentration
	(g/mol)	(mg/L)
Organics and nutrients		
Glucose ( $C_6H_{12}O_6$ )	180.0	280
Ammonium sulfate $((NH_4)_2SO_4)$	132.1	72
Potassium phosphate (KH <sub>2</sub> PO <sub>4</sub> )	136.1	13.2
Trace nutrients		
Calcium chloride (CaCl <sub>2</sub> ·2H <sub>2</sub> O)	147.0	0.368
Magnesium sulfate (MgSO <sub>4</sub> ·7H <sub>2</sub> O)	246.5	5.07
Manganese chloride (MnCl <sub>2</sub> ·4H <sub>2</sub> O)	197.9	0.275
Zinc sulfate ( $ZnSO_4 \cdot 7H_2O$ )	287.5	0.44
Ferric chloride anhydrous (FeCl <sub>3</sub> )	162.2	1.45
Cupric sulfate (CuSO <sub>4</sub> ·5H <sub>2</sub> O)	249.7	0.391
Cobalt chloride (CoCl <sub>2</sub> · $6H_2O$ )	237.9	0.42
Sodium molybdate dihydrate (Na <sub>2</sub> MoO <sub>4</sub> ·2H <sub>2</sub> O)	242.0	1.26
Yeast extract		30

Table 1. Constituents of the Synthetic Wastewater

Sponge volume (%)	Sustainable flux (L/m <sup>2</sup> .h)	Effluent DOC (mg/L)
0	25	< 6
10	50	< 4
20	45	< 5

Table 2. Sustainable flux and effluent quality in SSMBR

MLSS of SSMBR	Days of	DO concentration	Equilibrium time
(g/L)	operation	(%)	(min)
5		96.7	12
10	2	96.8	10
15		97.8	6
5		97.1	14
10	5	98.5	10
15		97.0	6

Table 3. SOUR of mixed liquor in SSMBR



Figure 1. Experimental set-up of SSMBR



Figure 2. The attached growth on sponge during acclimatization



Figure 3. SOUR variation of attached growth on sponge during acclimatization



Time (min) (c) sponge-SMBR (Sponge volume = 20 %)

*Figure 4.* Constant filtration fluxes versus TMP of SSMBR  $(LMH = L/m^2.h)$ 



*Figure 5.* DOC profile of SSMBR system at different sludge concentrations (filtration flux =  $30 \text{ L/m}^2$ .h; backwash rate =  $30 \text{ L/m}^2$ .h; backwash = 1 minute every half an hour; HRT = 1.2 hours)



*Figure 6.* COD profile of SSMBR system at different sludge concentrations (filtration flux =  $30 \text{ L/m}^2$ .h; backwash rate =  $30 \text{ L/m}^2$ .h; backwash = 1 minute every half an hour; HRT = 1.2 hours)



*Figure 7.* TMP development of SSMBR system at different sludge concentrations (filtration flux =  $30 \text{ L/m}^2$ .h; backwash rate =  $30 \text{ L/m}^2$ .h; backwash = 1 minute every half an hour; HRT = 1.2 hours)



*Figure 8.* NH<sub>4</sub>-N profile of sponge-SMBR system at different sludge concentrations (filtration flux =  $30 \text{ L/m}^2$ .h; backwash rate =  $30 \text{ L/m}^2$ .h; backwash = 1 minute every half an hour; HRT = 1 hour)



*Figure 9.* PO<sub>4</sub>-P profile of sponge-SMBR system at different sludge concentrations (filtration flux =  $30 \text{ L/m}^2$ .h; backwash rate =  $30 \text{ L/m}^2$ .h; backwash = 1 minute every half an hour; HRT = 1 hour)