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Self-forming dynamic membrane formed on mesh filter coupled with membrane bioreactor at different sludge concentrations

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Abstract. This study attempted to evaluate the process of self-forming dynamic membrane formation on mesh filter in membrane bioreactor with a two-stage method of batch (agitation) and continues (aeration) stage at different sludge concentrations. Four concentrations of activated sludge including 6±0.4, 8±0.5, 10±0.3, 14±0.3 g/L were used to demonstrate the optimal concentration of sludge for treating municipal wastewater and reducing fouling in dynamic membrane bioreactor. The formation time and effluent turbidity were decreased in the batch stage when increasing the activated sludge concentration. The minimum values of formation time and effluent turbidity were 14 min and 43 NTU for the optimum mixed liqueur suspended solids of 8±0.5 g/L, respectively. To improve operational condition and fouling reduction in the aeration stage, critical fluxes were measured for all concentrations by flux-step method. With increasing the sludge concentration, the relevant critical fluxes reduced. The optimum subcritical flux of 30 $L/m^2/h$ was applied as operating flux in the second stage. The maximum COD removal efficiency of 98% was achieved by the concentration of 8±0.5 g/L. Compressibility index of self-forming dynamic membrane and transmembrane pressure trend remained somewhat constant until the optimal concentration of 8±0.5 g/L and thereafter they increased steeply.

Keywords: mesh filter; self-forming dynamic membrane; membrane bioreactor; critical flux; wastewater treatment; different sludge concentrations

1. Introduction

The membrane bioreactor (MBR) is a process that combines the biodegradation of activated sludge with membrane filtration (Judd 2008). The MBR allows small footprint, high effluent quality, high sludge concentration and less sludge production (Le-Clech 2010). With these advantages, MBR is being increasingly used for wastewater treatment. However, the membrane fouling mainly caused by the adsorption of solutes or colloids and deposition of sludge within/on the membrane during the operation, is a major obstacle hindering the widespread application of MBR (Hao *et al*. 2017, Meng *et al*. 2006).

One innovative and promising technology for solving this obstacle is termed as self-forming dynamic membrane bioreactors (SFDMBRs). Compared to MBRs, SFDMBRs are very cost-effective due to partly substitution of expensive polymeric MF/UF membranes by cheap filters and partly to the lower filtration resistance of SFDM (Zhou *et al*. 2008, Fuchs *et al*. 2005, Ren *et al*. 2010). Instead of using conventional micro/ultra filtration membranes, in SFDMBRs the filtration modules are made of coarse-pore materials usually non-woven fabric, nylon, mesh filters and stainless steel mesh (Kiso *et al*. 2000, Wang *et al*. 2012). The bio-fouling formed on the membrane surface, named SFDM, can play two opposite roles. Firstly, it blocks the pore of the membrane with reducing the performance of the membrane and secondly, it forms a layer that effectively rejects the finer particles. Therefore, the deposition of biomass on the membrane can promote the yield of performance while the bio-fouling is formed on the porous medium (Fan *et al*. 2002, Chu and Li 2006, Meng *et al*. 2009, Liu *et al*. 2012). Hence, biofilm formation process on membranes may result in a significant improvement of MBR performance.

Chu and Li (2006) reported that the effluent turbidity was nearly kept at stable level within 2-6 h for different concentrations of sludge by formation of dynamic membrane. In their study, the average COD and NH4–N removal reached 80 and 74%, respectively. Liu *et al*. (2012) obtained that COD and turbidity contents in the filtrate became stable after 5 h, implying that the complete formation of dynamic membrane at sludge concentration of 7540 mg/L required 5 h. Rezvani *et al*. (2014) demonstrated a new approach in SFDM formation by a mechanical and axial-flow agitator for complete suspension and appropriate settling of activated sludge on mesh filter in SFDMBR. They proved that the agitator can reduce the formation time of SFDM by creating a more proper flow pattern near the mesh filter surface. They also showed that the required time for the SFDM formation was equaled to 840s for MLSS of 8.5±0.8 g/L. The long-term filtration of mesh filter with the formed self-forming dynamic membrane in bioreactor, also, was investigated by Poostchi *et al*. (2015).

In order to better control membrane fouling and

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maintain sustainable operation, the concept of critical flux was proposed by Field *et al*. (1995) in MBRs. They defined the critical flux as the flux below which the decline of flux or the increase of trans-membrane pressure (TMP) does not occur; however, above that level, fouling is observed. Since then, a lot of studies have focused on the concept of critical flux (Le-Clech *et al*. 2003), including the effect of sludge concentration on critical flux (Le-Clech *et al*. 2003). In order to improve operation condition and reduce the fouling rate, the operative flux considered to be lower than critical flux. It has been suggested that submerged MBR should be operated at a flux below the ‘‘critical flux’’, so-called subcritical flux, to maintain a sustainable permeability (Guglielmi *et al*. 2007). Poostchi *et al*. (2012) obtained the critical flux of 71 lm^{-2} h^{-1}(LMH) for MLSS of 6.1-8.7 g/L by the flux-step method.

Accordingly, this study continues and confirms our previous research (Rezvani *et al*. 2014), by investigating the effect of four different concentrations of mixed liquor suspended solids (MLSS) on the formation and performance of SFDM along with their critical fluxes in MBR. A two-stage method is considered to settle the suspended particles on the mesh filter in the batch stage and complete the process of SFDM formation in the continues or operation step. The flux-step method is applied to estimate the critical fluxes of different MLSSs to determine the optimal sub-critical flux which is applied as operative flux in aeration stage. Hence, the optimal MLSS is evaluated on the basis of the formation of a stable and uniform SFDM, COD removal efficiency, effluent turbidity reduction, the maximum sub-critical flux and minimum TMP variations for the fouling reduction.

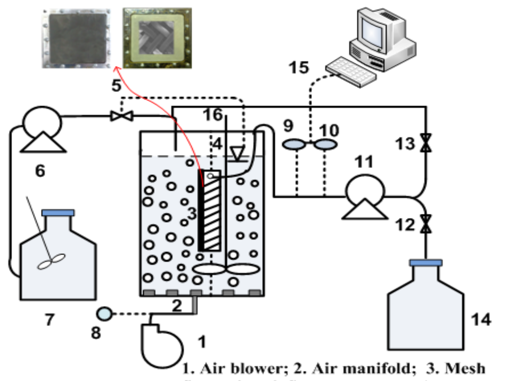
2. Material and methods

2.1 Experimental set-up

The experimental setup is illustrated in Fig. 1. The rectangular Plexiglas tank is used as the bioreactor with 0.15 m width, 0.2 m length and 0.4 m height. A polyester flat sheet mono-filamentous mesh filter is used with mean pore size of 30 μm. The area of the mesh is roughly about 0.016 m^2. The filter medium with its holding frame is vertically submerged in the centre of the bioreactor. Pressure drop across the filter medium is measured by an on-line pressure sensor. To adjust the permeate flow rate a peristaltic pump is used. The bioreactor is equipped with four fine bubbles air pipe distributers evenly placed at the bottom of the bioreactor, one of which is exactly below the filter module for intermittent cleaning of filter surface. A stainless steel four-blade agitator is used to suspend the solid while the dynamic membrane is being formed. The mechanical-driven agitator is placed at a distance of 3 cm below the filter to provide appropriate flow-pattern.

2.2 Experimental plan

The working volume of the bioreactor is 6 L. The dissolved oxygen concentrations are in the range of 3.7-5.8 mg/L. The bioreactor temperature is set in the range of 23-25^{o}C as the temperature fluctuation affects the performance of the activated sludge. The hydraulic retention time (HRT) and sludge residence time (SRT) are set on 12.5 hours and 32 days respectively. The bioreactor operates with synthetic wastewater to avoid any fluctuation in the influent loading and provide a continuous source of biodegradable pollutants. The COD:N:P ratio of influent is 100:5:1 containing 1125 mg/L glucose, 231.76 mg/L ammonium sulfate and 51.09 mg/L ammonium phosphate. The influent turbidity and COD are 288 NTU and 1200 mg/L, respectively, which is similar to municipal waste water. $NaHCO_3$ and H_2SO_4 were used to set pH in bioreactor neutral. Seed sludge is taken from the operating MBR plant (installed by Huber technology), which is dedicated for treating municipal wastewater. Four concentration of sludge used in bioreactor are selected on the basis of previous studies 6± 0.4, 8±0.5, 10±0.3, 14±0.3 g/L. At the beginning of study, the flux-step method is applied to estimate the critical fluxes. The highest sub-critical flux applied as operation flux in aeration stage is about 30 l m^{-2} h^{-1}.



1. Air blower; 2. Air manifold; 3. Mesh filter with biofilm membrane ; 4. Level switch; 5. Solenoid valve ; 6. Feed pump; 7. Feed tank; 8, 9. Flow indicator; 10. Pressure indicator; 11. Permeate pump ; 12. Permeate outlet valve ; 13. Recycle valve; 14. Permeate tank; 15. Computer; 16. Agitator.

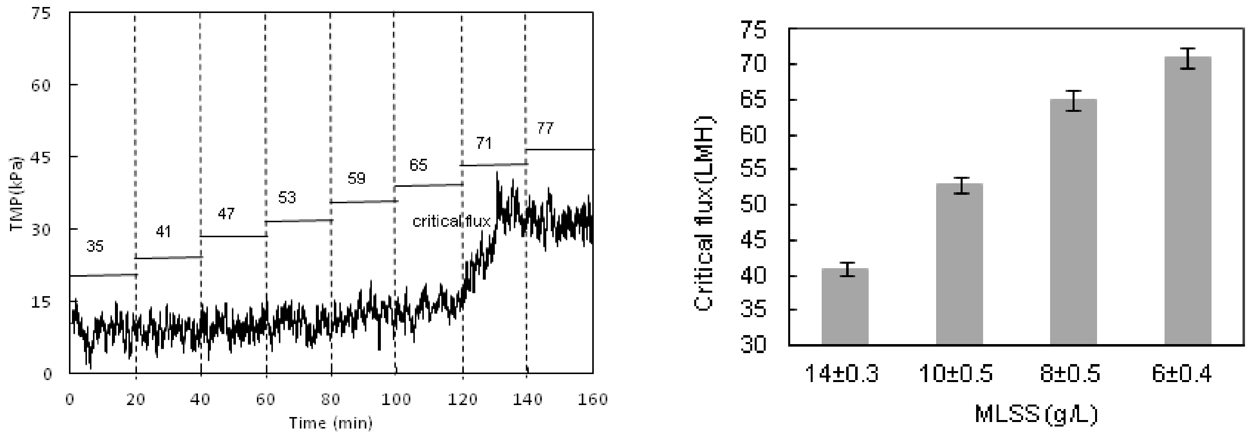
Fig. 1 Schematic diagram of self-forming dynamic membrane bioreactor system

2.3 Analytical procedure

The analytical parameters such as COD, turbidity and MLSS are analyzed using Merck reagents, according to APHA standard methods (APHA 2005). DO and temperature are measured with multi-meter (WTW 340 i, Germany). Mass of suspended solids is measured with balance (Tecator, Germany) at a resolution of 0.0001g. In order to minimize the errors due to the experimental conditions, all results are duplicated in two times and the average of the results is considered.

2.4 Field emission scanning electronic microscopic (FESEM)

The surface of fresh mesh filter and surface of cake layer formed on the used mesh filter with corresponding cross-section structure were observed with scanning electron microscopy (S4160, Hitachi, Japan). At each sampling, the suction pump is stopped and the permeate valve is normally closed to prevent the back flow of the



(a) Critical flux determination of continuous SFDMBR for MLSS of 8±0.5 g/L

(b) Critical flux versus MLSS

Fig. 2 Critical flux determination by flux stepping method

filtrate due to driving force owing to relatively high pressure difference between filter module and suction line. The filter with the SFDM is separated from the holding frame. The sample is fixed with 2% (v/v) glutaraldehyde in 0.1 M phosphate buffer at pH 7.2 for 2 h and then washed for 10 min and again immersed for 1 h in 0.1M phosphate buffer. The fixed sample is dehydrated with ethanol. When the drying is accomplished, the sample of dynamic membrane is precisely obtained and it is accurately fixed to the welfare of SEM.

3. Result and discussion

3.1 Critical flux measurement

Critical flux is measured by flux stepping method (Poostchi *et al*. 2012) for different concentrations of 6±0.4, 8±0.5, 10±0.3, 14±0.3 g/L in order to improve operating conditions and reduce fouling in MBR. The steps begin with flux of 35 LMH and continue by 6 LMH per step. Each step is about 20 minutes. To determine the critical flux for MLSS of 8±0.5 g/L, the TMP variation versus time at different imposed fluxes is shown in Fig. 2(a). Since the trend of TMP under imposed fluxes for other concentrations were similar to the TMP trend of 8±0.5 g/L (Fig. 2(a)), the relevant figures are not shown. The values of critical fluxes for each MLSS are shown in Fig. 2(b).

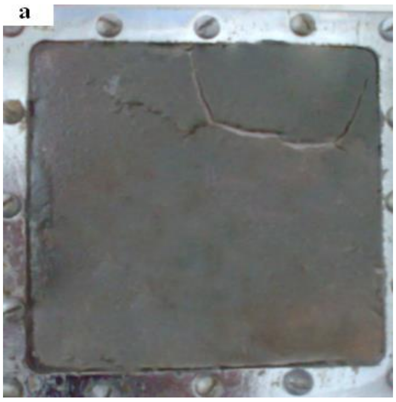
As can be seen from Fig. 2(a), there is a jump between the TMP of 65 and 71 LMH which states that the flux of 65 LMH is the critical flux for MLSS of 8±0.5 g/L. According to the Fig. 2(b), MLSS increment have caused a decreasing trend in the critical fluxes and an increasing trend in pressure drop. The minimum and maximum amounts of critical fluxes for concentrations of 14±0.3 and 6±0.4 g/L are 41 LMH and 71 LMH, respectively. Increasing the sludge concentration resulted in adhering more particles on the mesh filter, which lead to more fouling and more pressure drop (Guglielmi *et al*. 2007, Chang and Kim 2005). The obtained results are consistent with the study reported by Le-Clech *et al*. (2003) in which variations in the MLSS concentration ranging from 4 to 8 g/L have no significant effect on critical flux. In contrast, with increasing the MLSS more than 6±0.4 g/L, a significant increase is observed in the critical up to the MLSS of 12 g/L. According to the results, the constant permeate flux of 30 LMH is selected for operative flux in aeration stage which is known as sub-critical flux (Guglielmi *et al*. 2007).

3.2 Formation of SFDM

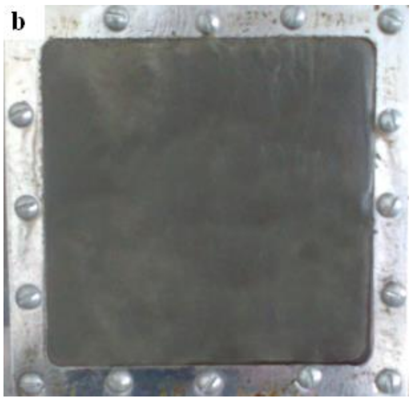
Formation process of SFDM is similar to our previous research (Rezvani *et al*. 2014). In short, the process has two stages in which biofilm is formed in the first one and gradually completed in the second one. The first one named agitation stage starts when the drag force of the pump lets pass the permeated through the filter media. During this process more particles adhere to the mesh filter surface due to the permeation drag force. The flux of the first stage is about 150 LMH. Recycle flow is supplied by the permeate flow, thus the level of bioreactor remains constant. The accumulation of particles gradually forms the biofilm which is known as the dynamic membrane (Fan and Hung 2002). To accelerate formation of self-forming dynamic membrane and also suspend the activated sludge in the bioreactor, an axial-flow agitator is used with the rotational speed of 350 rpm. The time in which the effluent quality becomes stable is known as the required time for SFDM formation at the agitation stage (Rezvani *et al*. 2014). The second one is related to operation stage in which the agitator is switched off and aeration is turned on. After the dynamic membrane is formed at the agitation stage as above-mentioned, the rest particles are gradually deposited at the aeration stage under the drag force of permeate flow contributing to membrane thickness incensement. During the operating period, the permeate flux is set to 30 LMH for long-term operation of SFDMBR. The photography pictures of SFDMs at several concentrations are shown in Figs. 3(a)-3(d).

3.3 Compressibility index of SFDM

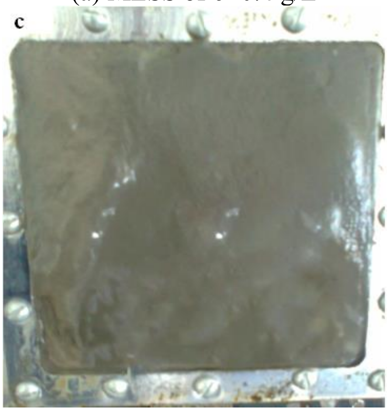
The cross-sectional SEM images of SDFM for different concentrations are depicted in Figs. 4 (a)-4(d). According to



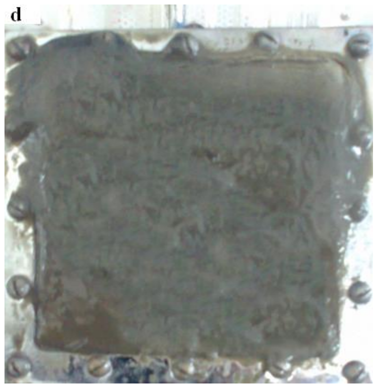
(a) MLSS of 6±0.4 g/L



(b) MLSS of 8±0.5 g/L



(c) MLSS of 10±0.5 g/L



(d) MLSS of 14±0.3 g/L

Fig. 3 The photography pictures of SFDMs at several concentrations (Resolution of 8 Megapixel)

the images, increasing the MLSS concentration leads to an upward trend in deposition rate of particles on the mesh filter surface at agitation stage which results in developing dynamic membrane thickness. The compressibility index, i.e., the index of SS/cake volume, indicates the compaction of SFDM; this is evaluated at four concentrations. This factor is a criterion for analyzing the TMP trend. So that the more compressibility index rises, the more pressure drop occurs.

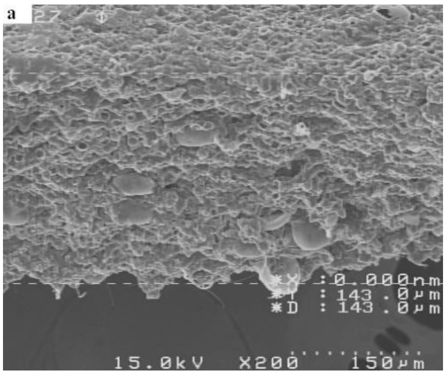
The thickness and compressibility index of dynamic membranes are depicted in Fig. 5. As can be seen, the compressibility index remains approximately constant for both MLSS of 6±0.4 and 8±0.5 g/L, while their thicknesses increase. Such a trend proves that the colloidal particles only settle on each other with a uniform rate and there is no cake layer compression. Hence, the TMP trend shows negligible increase in both agitation and aeration stages.

The membrane thickness increases along with increasing the deposition rate of suspended solids. Colloidal particles penetrate membrane pores and bring about their blockage, due to drag force of peristaltic pump. Hence, the membrane compression and the reduction trend of SFDM porosity increase by increasing the MLSS concentration from 8±0.5 g/L to 14±0.3 g/L, which result in a drastic increase of TMP trends at both MLSS. The compression of layer and membrane pores blocking reach the maximum value for the MLSS of 14±0.3 g/L, in which the membrane thickness increases up to 814 μm (Fig 4(b)) and pressure drop reaches the maximum value of 3.2 kPa (Fig. 6(b)).

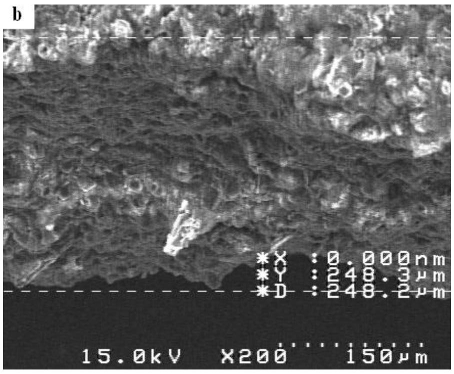
3.4 Evaluation of formation stage

In the agitation stage, the formation process of SFDM with distinct concentrations of 6±0.4, 8±0.5, 10±0.3 and 14±0.3 g/L is examined. During the SFDM formation process, the values of turbidity and TMP for different MLSSs are depicted in Figs. 6(a) and 6(b) in which the formation time of each SFDM is presented in dashed line. According to Fig. 6(a), the maximum and minimum formation times are 17 and 11 min for MLSSs of 6±0.4 and 14±0.3 g/L, respectively. In contrast, the maximum and minimum effluent turbidity are 74 and 43 NTU for MLSSs of 8±0.5 g/L and 14±0.3 g/L, respectively.

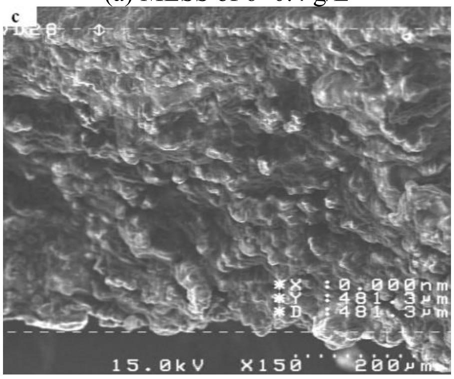
According to Fig. 3(a), the SFDM of 6±0.4 g/L posses a visible break which is clear evidence for lack of enough adhesive force between colloidal particles on the membrane surface, which is consistent with previous studies (Le-Clech *et al*. 2003, Mori *et al*. 2006). This phenomenon leads to passing particles from the break and increasing its effluent turbidity rather than the MLSS of 8±0.5 g/L during the SFDM formation process (Fig. 6(a)). The obtained results can be confirmed by previous studies which state the MLSS concentrations have a direct impact on viscosity rather than adhering force (Rosenberger *et al*. 2002, Hasar *et al*. 2004). Fig. 3(b) depicts a uniform surface of SFDM at the concentration of 8±0.5 g/L. As can be seen from Figs. 3(a)-(d), the uniformity and permeability of SFDM decrease with increasing the concentration of sludge and compressibility index. As mentioned above, the quality of



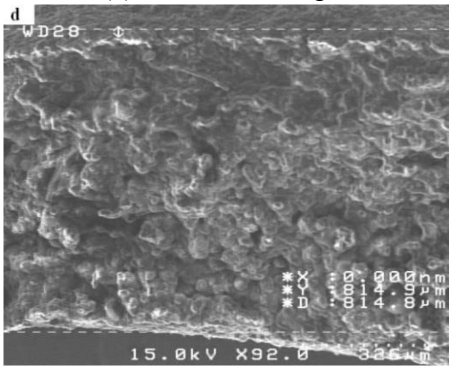
(a) MLSS of 6±0.4 g/L



(b) MLSS of 8±0.5 g/L



(c) MLSS of 10±0.5 g/L



(d) MLSS of 14±0.3 g/L

Fig. 4 The cross sectional SEM images of SFDM (Magnification 2000x)

permeate decreases as the SFDM thickness increases and the SFDM uniformity reduces at the MLSSs of 10±0.3 and 14±0.3 g/L. This is because the permeate flow cannot pass from the blocked SFDMs and the drag force imposed by peristaltic pump can just compress the available dynamic membranes. Hence, the colloidal particles penetrate into the membrane pores which result in increasing the biomass and the suspended solids concentrations inside of the formed dynamic membrane and effluent turbidity, respectively. This phenomenon leads to settling more particles on the mesh filter and thickening the SFDM, thereby increasing the TMP trend and turbidity in the effluent (Ye *et al*. 2008). Hence, the higher TMP and the higher effluent turbidity for the MLSS concentrations of 10±0.5 and 14±0.3 g/L rather than others can be related to the drag force of peristaltic pump and high concentration of MLSS inside the SFDMs which result in increasing the thickness and density of settled colloidal particles on mesh filter.

According to the obtained results, the effluent turbidity increases by increasing MLSS which is consistent with previous studies (Katayon *et al*. 2006). Several authors reported that the increasing MLSS results in higher fouling (Lukáš *et al*. 2011, Meng *et al*. 2007), while it is not clear which parameter can determine exactly the permeate flux. Some authors reported that TMP and permeate quality are not under effect of MLSS variations (Chang and Kim 2005), however others stated that in higher MLSS concentrations, TMP can be reduced considerably (Durante *et al*. 2006). In this study, the best results for the batch stage are 14 min of formation time and 43 NTU of effluent turbidity for the MLSS of 8±0.5g/L.

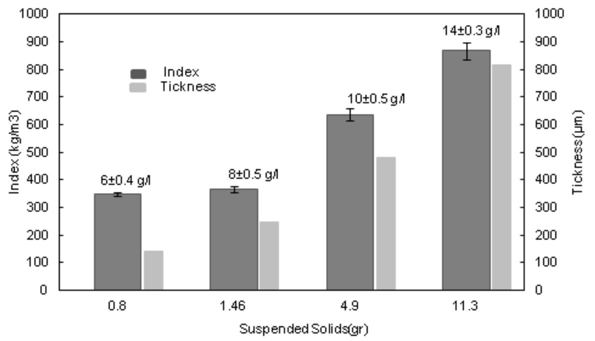
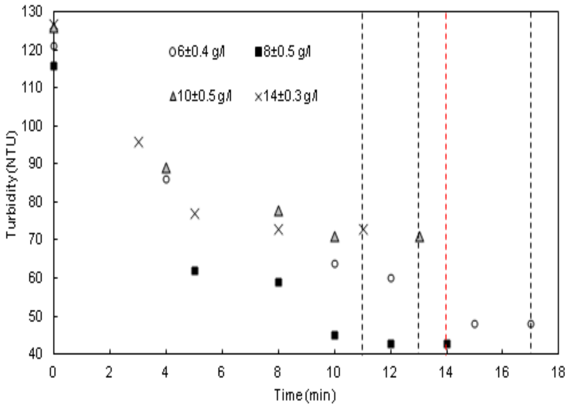


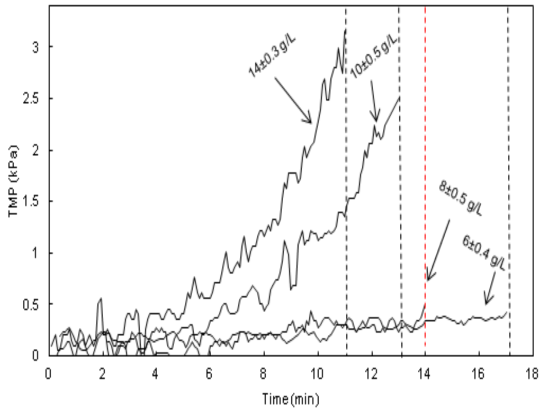
Fig. 5 Thickness and compressibility index of SFDM versus suspended solids during formation

3.5 Operational performance of SFDM

To investigate the performance of SFDM in the bioreactor, a 72-hour operation of treatment is performed for all MLSSs. The operating flux is kept at 30 LMH during the aeration stage. Average amounts of turbidity, COD and their removal efficiency are depicted in Figs. 7(a)-7(c). As can be seen from the charts, the COD concentration and effluent turbidity at MLSS of 8±0.5 g/L are lower than other concentrations, so that the best performance for wastewater treatment is observed in this concentration. So that, the above-mentioned reasons can be applied also for this phenomenon. According to Fig. 7(c), the maximum COD removal efficiency is 97% which is related to the MLSS of 8±0.5 g/L. As can be seen from the Fig. 8, there is a negligible increase in the TMP trends for the concentrations of 6±0.4 and 8±0.5 g/L. It means that the compression

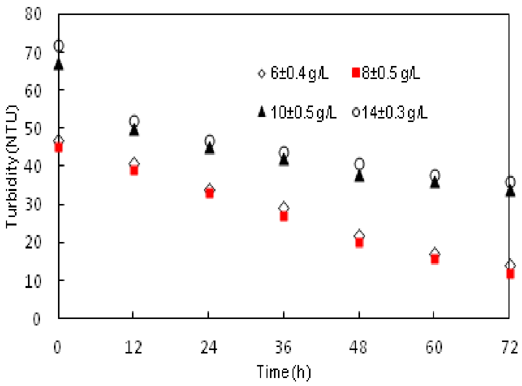


(a) Turbidity variations with time at four concentrations of MLSS

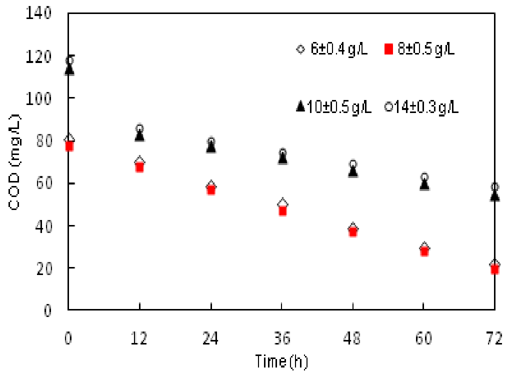


(b) TMP trend with time at four concentrations of MLSS

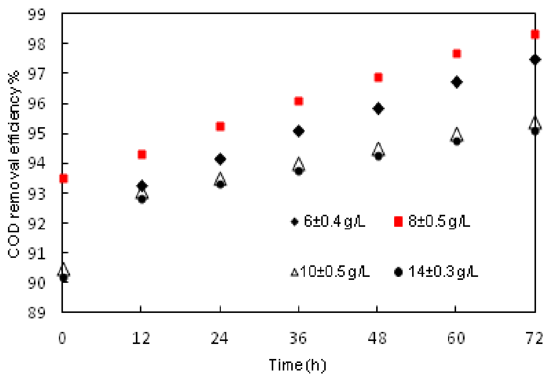
Fig. 6 Turbidity and TPM versus time in the formation stage



(a) Effluent turbidity variations with time at four concentrations of MLSS



(b) Effluent COD variations with time at four concentrations of MLSS



(c) COD removal with time at several concentrations of MLSS

Fig. 7 Effluent turbidity, COD and COD removal versus time in the aeration stage

of the cake layers for the MLSSs of 6±0.4 and 8±0.5 g/L is lower than other MLSSs, due to regular steeling particles on the surface of each other rather than the pore blocking by the soluble particles (Rezvani *et al*. 2014). Due to the dynamic membrane compressibility and the pore blocking resulted from the drag force and the MLSS concentration sharp increase in the TMP trend are observed in Fig. 8. So that, for the MLSS of 14±0.3 g/L, there is no visible permeate flow and the TMP and the compressibility index reach the maximum values of 3.2 kPa and of 850 kg/m^3, respectively.

Due to the fact that turbidity reduction is indicative of microfiltration process (Poostchi *et al*. 2012), a significant turbidity reduction indicates that the dynamic membrane is increment, a drastic reduction in membrane porosity and a operated as a microfiltration process for the MLSS of 8±0.5 g/L. This shows the benefit of using mesh filter instead of conventional membrane.

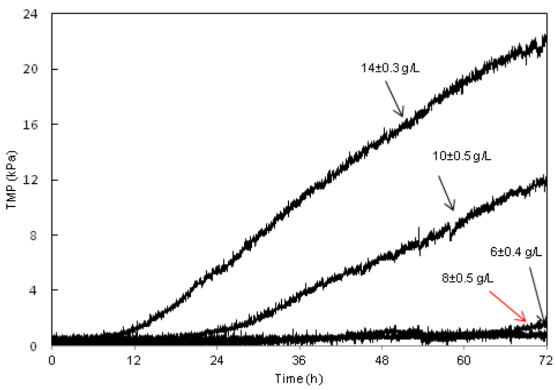


Fig. 8 TMP variations with time during 72-hour operation for different MLSSs

4. Conclusions

In this study, four concentrations of activated sludge were compared in terms of formation and performance of SFDM which was formed within the two distinct stages of batch and continuous. The performances of formed SFDMs were evaluated to determine the best MLSS concentration for treating municipal wastewater treatment. The advantage of the addition of agitation stage was to expedite the SFDM formation as uniformly and rapidly as possible. Critical fluxes for all concentrations were acquired to determine the subcritical flux of 30 LMH as operational flux for aeration stage. A drastic reduction of membrane porosity and a sharp increase in TMP were observed due to increasing the dynamic membrane compressibility and the pore blocking resulted from the drag force and increasing the MLSS. Compressibility index, as a criterion for analyzing the pressure drop, was consistent with TMP trend. So that the more compressibility index increased, the more pressure drop was observed. According to the obtained results from the effluent quality and the TMP profile, the optimum MLSS concentration of 8±0.5 g/L was considered for the dynamic membrane formation on the mesh filter at both the agitation and aeration stages. Results showed that the SFDM formed by 8±0.5 g/L was able to improve particle removal efficiency of mesh filter in term of turbidity reduction as well as a microfiltration membrane does.

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