Pulse shear stress for anaerobic membrane bioreactor fouling control

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ABSTRACT

Increase of shear stress at membrane surfaces is a generally applied strategy to minimize membrane fouling. It has been reported that a two-phase flow, better known as slug flow, is an effective way to increase shear stress. Hence, slug flow was introduced into an anaerobic membrane bioreactor for membrane fouling control. Anaerobic suspended sludge was cultured in an anaerobic membrane bioreactor (AMBR) operated with a side stream inside-out tubular membrane unit applying sustainable flux flow regimes. The averaged particle diameter decreased from 20 to 5 μm during operation of the AMBR. However, the COD removal efficiency did not show any significant deterioration, whereas the specific methanogenic activity (SMA) increased from 0.16 to 0.41 g COD/g VSS/day. Nevertheless, the imposed gas slug appeared to be insufficient for adequate fouling control, resulting in rapidly increasing trans membrane pressures (TMP) operating at a flux exceeding 16 L/m²/h. Addition of powdered activated carbon (PAC) enhanced the effect of slug flow on membrane fouling. However, the combined effect was still considered as not being significant. The tubular membrane was subsequently equipped with inert inserts for creating a locally increased shear stress for enhanced fouling control. Results show an increase in the membrane flux from 16 L/m²/h to 34 L/m²/h after the inserts were mounted in the membrane tube.

Key words | activity, anaerobic, fouling, membrane, shear stress, slug

INTRODUCTION

Induced surface shear is considered to be a major strategy to control membrane fouling (Cui et al. 2005). It is reported that the cake resistance is the predominant resistance, which can be minimized by increasing the membrane surface shear stress (Su et al. 2008). This finding is also evidenced by other authors (Elmaleh & Abdelmoumni 1997; Karasu et al. 2009).

Although application of a high shear stress is a widely accepted approach to alleviate membrane fouling, it is likely that the applied shear stress negatively impacts the microbial activity, resulting in a decreased performance of the membrane bioreactor (MBR). It is reported that high shear stress decreases sludge activity in aerobic MBR (Kim et al. 2001; Khan & Visvanathan 2008). In addition, the negative impact was also reported in an anaerobic MBR (Jeison 2007).

In addition, the response of cells to fluid mechanical shear stress varies with cell type and depends on their physiological characteristics, culture conditions, previous history of growth among others (Joshi et al. 1996). Thus, working with a bioreactor, different reactor operational conditions, such as impeller mixing, will have an impact on bioreactor performance (Zhang et al. 2010; Zhong 2010).

The generally observed high specific methanogenic activity of anaerobic granular sludge originates from the high density of methanogenic bacteria and the fact that micro-colonies of acetogenic bacteria are closely linked with micro-colonies of hydrogenotrophic methanogenic archaea in so-called syntrophic associations, which allows an efficient interspecies hydrogen transfer (Pol et al. 2004). A high shear stress could possibly disrupt these syntrophic aggregates, creating suspended biomass and therefore leading to a decreased anaerobic sludge activity. However, high shear, though it does harm to sludge activity, is required to minimize membrane fouling. Therefore, a gentle shear, which is high enough to provide shear, but low enough to secure sludge activity, is proposed in systems that combine biological activity with membrane separation processes.
Gas slug is perceived as a useful technique to increase shear stress near the membrane surface for achieving a high flux. However, the impact of the Taylor bubble generated shear stress on sludge activity is not yet known. Moreover, in the current research, the sustainable flux flow region was selected for the crossflow AMBR operation. Our present experiments focus on impact of pulse shear invoked by Taylor bubbles on membrane fouling and the specific methanogenic activity. Since PAC can absorb organic contaminant that may be responsible for membrane fouling and then lead to higher possible flux (Park et al. 1999; Hu & Stuckey 2007; Akram & Stuckey 2008; Vyrides & Stuckey 2009), PAC was also added into the AMBR to enhance the effect of Taylor bubble. The enhancement was based on the assumption that the Taylor bubble enhances mass transfer from the membrane surface to the sludge, while PAC reduces the concentration of contaminant. It is obvious that possible enhancement depends on the dose of PAC. However, a large dose PAC does not lead to a better effect. Unfortunately, no consensus exists on the optimal dose. It was reported that the addition of PAC at the dose of 3.4 g/L made membrane less permeable (Akram & Stuckey 2008). However, other research did not find the worsen effect while even higher dose (5 g/L) was tried (Park et al. 1999). In addition, alternative to the Taylor bubble approach, inserts were mounted in a tubular membrane to locally increase the shear stress. Although various authors confirmed the positive effect of such inserts (Mameri et al. 1999; Krsti. et al. 2002; Xu et al. 2003; Pal et al. 2008), this method has yet not been tried in anaerobic MBRs. Therefore, several experiments were performed to evaluate the potential of pulse shear invoked by fixed inserts on membrane fouling control.

MATERIALS AND METHODS

A cross flow anaerobic MBR was operated using a microfiltration (MF) membrane module in gas-lift mode. A tubular PVDF membrane (Norit, the Netherlands) was employed (30 nm pore size). The inner diameter and length were 5.2 mm and 0.74 m, respectively. A cylindrical vessel was used as anaerobic bioreactor with an effective volume and inner diameter of 4.5 L and 10 cm, respectively. Temperature of the bioreactor was kept at 35 °C by means of a water jacket surrounding the bioreactor. A pump (Watson Marlow 323D) was also used for pumping supernatant into the tubular membrane in which, during some experiments, inserts were placed. Other three pumps (Watson Marlow 323D) were used for mixing the reactor (gas mixing), permeation extraction, and substrate supply. A schematic graph of the experimental setup is shown in Figure 1. Gas lift operation mode lasted during most of the experiments. Inserts were only included to test its effect on controlling membrane fouling and involved only after the gas-lift operation mode was finished. Permeate from the membrane was returned to a plastic pipe that served to adjust the water level in bioreactor. By the use of the plastic pipe, the flux of the membrane could be changed within a wide range. Meanwhile, the inflow and the outflow of the overall reactor was always the same and no dilution of the influent took place. The critical flux was based on a flux step method (Le Clech et al. 2005). In this experiment, a flux was maintained within 10 min followed by a backflush for 6 s. After the backflush, the flux was increased by 4 L/m²/h. TMP variation was recorded accordingly.

PAC (SAE2, Norit, the Netherlands) was rinsed with demineralized water carefully to remove ash from the PAC surface. The PAC was stored in an oven at a temperature.
of 100 °C for one day to remove water. Finally, a sieve (diameter 0.105 mm) was employed to select PAC with larger diameters. Selected PAC was added into the bioreactor.

Inserts were placed in the tubular membrane to test its effect on membrane fouling control. The number of the inserts was up to 20. The distance between each two inserts was 3 cm. They were tied together and kept in place by a plastic wire of 1 mm diameter. **Figure 2** shows dimension of an insert placed in tubular membrane.

Granular sludge was taken from a plant treating wastewater containing lactose (Purac Biochem, Gorinchem, The Netherlands). The sludge was crushed into small particles and inoculated in the bioreactor. The substrate is prepared with gelatin:acetate:propionate and butyrate to obtain a COD ratio 2:1:1:1. The substrate was diluted with demineralized water until a total COD concentration of 20 g/L. The imposed organic sludge load was 0.3 g COD/g VSS/d. Samples taken from the membrane permeate was injected into the reagent and the temperature was kept at 148 °C for 2 h. Then COD concentration was measured by means of a spectrophotometer (NOVA60). Specific methanogenic activity was measured following pressure increase using the method of Zandvoort (Zandvoort et al. 2002). The linear part of the curve describing methane generation versus time was used to calculate the methanogenic activity. Particle size distribution was measured by means of a particle size counter (Model 3000, Pacific Scientific Instruments).

Gas-lift mode was employed for membrane operation. A peristaltic pump (Watson Marlow 323D) was employed for biogas extraction from the head space of the bioreactor and to inject the gas into the tubular membrane. Sludge moved into the tubular membrane automatically due to the gas motion. Gas and liquid superficial velocity were 0.74 m/s and 0.34 m/s, respectively. To determine the liquid flow pattern resulting from the applied gas rate, the method of Dziubinski (Dziubinski et al. 2004) was used. Based on Dziubinski’s equations, in order to characterize the flow pattern, the parameters x and y are introduced and used as ordinates in an x-y graph describing the flow pattern (**Figure 3**). The coordinates x and y are determined by the following two equations:

\[
x = \frac{\nu_G}{\nu_L} \sqrt{\frac{\rho_G \rho_{\text{H}_2\text{O}}}{\rho_{\text{air}} \rho_L}} \quad (1)
\]

\[
y = \frac{\nu_L}{\nu_G} \sqrt{\frac{\rho_L}{\rho_{\text{H}_2\text{O}}}} \quad (2)
\]

where $\nu_G$ is superficial gas velocity (m/s); $\nu_L$: superficial liquid velocity (m/s); $\rho_G$: gas density (kg/m³); $\rho_{\text{air}}$: air density (kg/m³); $\rho_{\text{water}}$: water density (kg/m³); $\rho_L$: liquid density (kg/m³).

The two boundary lines are used to discriminate the different flow patterns:

**Bubble-slug**:

\[
y = 0.0685x^{-1.03} \quad (3)
\]

**Slug-churn**

\[
y = 0.556x^{-0.92} \quad (4)
\]

**Figure 2** | The shape and size (mm) of inserts placed in tubular membrane.

**Figure 3** | Map for determining flow pattern in membrane.
By measuring gas and sludge velocity in the tubular membrane, three lines were derived and plotted in Figure 3, describing the flow pattern inside the membrane. Figure 3 shows two intersections, which are used as critical points indicating flow pattern transition. The two critical points correspond to gas velocities of 0.1 m/s and 0.74 m/s, respectively. Hence, it was concluded that the gas velocity should be between 0.1 m/s and 0.74 m/s in order to maintain slug flow. Outside these boundaries, the flow pattern shifts to either bubble flow or churn flow, which are both not desired. Hence, during all the experiments in which gas-lift operation mode was used, the gas superficial velocity was maintained at 0.74 m/s.

RESULTS

Effect on permeate quality, particle size distribution and SMA

The influence of shear stress generated by Taylor bubbles on the effluent COD concentration and the COD removal efficiency is shown in Figure 4. At the beginning of the experiments, effluent COD concentrations were recorded around 6.4 g/L, likely caused by poor bioreactor mixing. Enhanced mixing immediately resulted in much lower effluent COD values for the next 4–5 weeks. Air intrusion via a broken gas pipe at day 50 resulted in a sharp increase in the effluent COD (Figure 4). Full recovery was only observed after 20 days of continuous operation. Results presented in Figure 4 indicate that long term operation is feasible and the sludge activity, in terms of COD removal efficiency, is not negatively influenced by the shear stress generated in membrane module, which is supported by an increase in SMA (see the paragraph below).

The impact of shear stress on sludge morphology was investigated by analyzing the particle size distribution (PSD) during the course of the continuous flow experiment. In addition to the high superficial liquid velocity of 0.34 m/s, the wake zone, which is a secondary flow zone, is generally regarded as high turbulent zone, causing increased shear (Cui et al. 2003). As clearly illustrated in Figure 5, the percentage of small particles increased during the experimental run time. Apparently, the anaerobic sludge particles could not resist the shear stress generated by Taylor bubbles in the membrane module. The mean diameter gradually shifted from 20 μm to 5 μm. The decrease in particle size was also discovered in an submerged anaerobic membrane bioreactor in which coarse bubbles were involved (Jeison 2007). SMA experiments conducted at the beginning and at the end of experiment indeed showed an increase from 0.16 g CH4/g VSS/d at day 1 to 0.41 g CH4/g VSS/d at day 70. However, the decrease of particle size, though might reduce substrate mass transfer, did not well explain the SMA increase. The development of microbial population may have caused the SMA increase.

Effect of Taylor bubble on fouling

The impact of the Taylor bubble approach on membrane fouling was evaluated by assessing the TMP increase during step flux experiments applying a sustainable flux flow regime. Results are depicted in Figure 6. Notably, a rapid increase in TMP is observed when applying a liquid flux exceeding 16 L/m²/h. In order to enhance the effect of Taylor bubble on membrane fouling control, PAC was added to the bioreactor. The effect of PAC additions was minimal or even negligible when the imposed flux was lower than 16 L/m²/h. However, a clear effect was observed at fluxes exceeding 16 L/m²/h. Addition of PAC in two different doses, i.e. 1.6 g/L and 10 g/L, did not show difference in terms of TMP build-up during filtration at elevated fluxes. Although PAC clearly shows its positive effect on lowering the TMP, its impact is
likely insufficient to cause a breakthrough for enhanced AMBR operation. It is deduced that PAC can adsorb but not all foulants. Too much PAC cannot give better effect than an optimized dose or even give worse effect, as discovered by other researchers (Park et al. 1999; Akram & Stuckey 2008). The optimized PAC varied in these literatures, which may attribute to raw material and manufacture process of the PAC as well as pretreatment of the PAC before dosing and amount of adsorbable foulant. It is found that a type of PAC (screened by a 100 μm sieve and stored in an oven at 105°C, Norit, UK) gave positive effect at 1.67 g/L but negative effect at 3.4 g/L (Akram & Stuckey 2008). Alternatively, the other type of PAC (an average size of 100 μm, sieved through 120×200 meshes and rinsed several times with ultrapure water to remove inorganic ashes completely, then dried overnight in an oven at 105°C, and always stored in a desiccator before use, Norit SA4, US) did not show negative effect while PAC dose increased from 1 to 5 g/L (Park et al. 1999). The PAC used in this experiment was undergone certain pretreatment and negative effect was also not found while high dose (10 g/L) was used.

Increasing flux by fixed insert

In the next set of experiments a cascade of inserts was placed in the inner space of the tubular membrane as an alternative to the Taylor bubble operation. Experiments were performed using reactor supernatant for the filtration tests. Compared to anaerobic sludge, the supernatant has similar fouling capacity as indicated by Jeison and van Lier (Jeison & van Lier 2007). The supernatant was obtained by settling sludge for one night.

Results in Figure 7 clearly show the positive effect of the mounted inserts on supernatant filterability, applying a flux of 34 L/m²/h. In fact, when the inserts were absent, frequent back flush was necessary to lower the TMP. Back flush could be significantly lowered when the inserts were present, whereas the TMP level was lowered by almost one order of magnitude. Very interestingly, Figure 7 shows that the use of inserts significantly decreased the TMP increasing rate under the same flow conditions. As can be seen from Figure 7(b), TMP was maintained at low level even while flux was kept high for 7 h, which was similar to the data in Figure 7(a), which were collected in short term experiment.

The significantly increased flux can be explained by the increase of mass transfer at membrane surface. Mass transfer coefficient at membrane surface is given by (Taha & Cui 2002):

\[ k = 1.62 \left( \frac{d_i D}{d_h L} \right)^{0.33} \]

where \( k \): mass transfer coefficient (m/s); \( d_i \): diameter of tubular membrane (m); \( d_h \): equivalent hydraulic diameter (m); \( \gamma \): shear rate (l/s); \( D \): mass diffusivity, m²/s; \( L \): length of tubular membrane (m).

For the sections of flow channel taken up by inserts, the inserts significantly reduced the equivalent hydraulic diameter of the tubular membrane, which also increased shear rate in the resulting narrow flow channel while flow rate was kept the same. By this way, the use of the inserts increased
mass transfer coefficient at membrane surface, which made foulant easier to shift from membrane surface to the bulk solution. Therefore, pore blocking and cake layer formation did not easily occur, which made higher flux possible.

CONCLUSIONS

The Taylor bubble approach did not offer a satisfactory effect on fouling control while the membrane module was operated in a gas lift mode under anaerobic condition. The addition of PAC, though showed positive effect, did not make a breakthrough in controlling the membrane fouling.

The application of inert inserts could significantly improve achievable flux to as high as 34 L/m²/h.

REFERENCES


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