멤브레인(Membrane Journal)
Vol. 21, No. 4 December, 2011, 360-366

음식물 침출수를 처리하는 막결합 고온혐기성 소화시스템에서 교차여과와 막간압력이 파울링에 미치는 영향

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(2011년 12월 1일 접수, 2011년 12월 26일 수정, 2011년 12월 26일 채택)

Effect of Cross-flow Velocity and TMP on Membrane Fouling in Thermophilic Anaerobic Membrane Bioreactor Treating Food Waste Leachate

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(Received December 1, 2011, Revised December 26, 2011, Accepted December 26, 2011)

요 약: 음식물 폐기물 침출수를 처리하는 분리막 결합 고온 혐기성소화공정(생물학적 반응조) (Anaeorobic Membrane Bioreactor, AnMBR)의 파일럿 운전에서 분리막의 교차여과 속도와 막간압력이 파울링에 미치는 영향을 관찰하였다. 연구결과 정압여과 하에서 교차여과 속도가 증가할수록 파울링의 속도는 현격히 감소되었다. 그러나 이와 같은 영향은 낮은 막간압력에서 더욱 효과적이었다. 막간압력이 증가할수록 여과대상 물질의 압축성으로 인해 투과성이 상대적으로 낮은 파울링층(혹은 케익층)이 분리막 표면에 형성된 것에 기인된 듯하다. 여과대상 시료의 입도분석을 해 본 결과 입자크기는 약 10∼100 μm 범위에서 분포하였고 이에 따라 브라운확산에 의한 역수송보다 분리막 표면에서 교차여과에 의해 발생하는 전단력이 입자의 역수송에 더욱 기여하고 있음을 예측할 수 있었으며 이는 AnMBR의 연속운전을 통해 재확인할 수 있었다. 운전 후 막 부검을 실시한 결과 유기 및 무기 파울링이 모두 관찰되었으나 어느 것이 지배적인 파울링 기작을 나타내는지는 앞으로 더욱 연구가 필요하다. 무기 파울링의 경우 대부분 분리막 표면에서 스케일링 형성이 지배적이었으며, 따라서 분리막의 공극 막힘에 주로 기여하는 작은 콜로이드성 유기물질의 경우 분리막 표면에서 전단력에 의한 역수송 효과는 그다지 크지 않을 것으로 사료된다.

Abstract: The effect of cross-flow velocity and transmembrane pressure (TMP) on membrane fouling was observed from pilot-scale operation of thermophilic anaerobic membrane bioreactor (AnMBR) treating food waste leachate. It was found that fouling rate was reduced significantly as cross-flow velocity increased at constant TMP mode of operation while this effectiveness was more pronounced at lower TMP. Higher TMP resulted in less permeable fouling layer possibly due to compressibility of foulant material on membrane surface. Particle sizes of membrane concentrate ranged from 10 to 100 μm, implying that shear-induced diffusion enhance back transport of these particle sizes away from the membrane effectively. From the continuous operation of AnMBR, it was confirmed that shear rate played an important role in the reduction of membrane fouling. Membrane autopsy works at the end of operation of AnMBR showed clearly that both organic and inorganic fouling were significant on membrane surface. Surface shear by cross-flow velocity was expected to be less effective to remove irreversible fouling which can be mainly caused by the adsorption of organic colloidal materials into membrane pores.

Keywords: *cross-flow velocity, TMP, membrane, anaerobic bioreactor, food waste leachate*

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1. Introduction

Food waste has been generated by various high concentration points such as restaurants, residential areas and markets etc. In South Korea, over 9,000 tons of food waste leachate per day is generated mainly from composting or other recycling procedure for the treatment of food waste [1]. Majority of food waste leachate has been treated by ocean dumping, however, this will be prohibited strictly from the year of 2012.

The use of anaerobic treatment for the food waste leachate should be attractive because the leachate contains high concentration of biodegradable organic material. Most of all, the anaerobic treatment is considered as an energy-producing rather than energy-consuming because it can produce useful energy in the form of methane [2]. Although there has been upsurge of interests in the anaerobic treatment for the high-strength wastewater which is generally higher than 1,500 mg/L as soluble COD, the anaerobic treatment has still difficulty to maintain high amount of active biomass in the bioreactor since the growth rate of anaerobes in anaerobic reactor is very slow. Biomass retention is necessary feature for the successful application of anaerobic digestion for wastewater treatment. Biofilms and granule formation are the traditional way to achieve such retention, enabling reactor operation at high biomass concentrations, and therefore at high organic loading rates [3,4].

Anaerobic membrane bioreactor (AnMBR) combines anaerobic treatment with membrane filtration. The success of anaerobic wastewater treatment can be attributed to an efficient uncoupling of the solids retention time from the hydraulic retention time through biomass retention [5]. An advantage of membrane is that it can prevent washout of microorganisms from the bioreactor due to complete physical retention of suspended solids [6]. Since biomass is physically retained inside the reactor, there is no risk of washout of cells and the solid retention is not dependent on the formation of biofilms or granules. Therefore, long solid retention time can be achieved while operating short hydraulic detention times as required for reducing reactor size and cost [7]. Membranes can be installed in hard-shell encased vessels as side-stream with anaerobic bioreactor or they can be submerged directly into the reactor to reduce footprint.

Membrane fouling caused by the deposition of foulant material on membrane surface and/or within membrane pores is main drawback in the operation of AnMBR [4-6]. Membrane fouling can increase both capital and operational cost by increasing transmembrane pressure at constant flux mode of operation or decreasing permeate flux at constant pressure mode of it. Membrane fouling increases frequency of chemical cleaning and requires replacement with new membrane eventually. Thus, reducing membrane fouling is very critical for the sustainable operation of AnMBR. Several studies have been conducted with the AnMBR to reduce membrane fouling. Introduction of large portion of biogas produced in submerged AnMBR was recycled to control membrane fouling by providing physical scouring effect with gas sparging along membrane surface [4]. It was also reported that application of activated carbon such as powdered activated carbon (PAC) along the membrane surface reduced membrane fouling by both adsorption and scouring effects [8-10]. These fouling mitigation methods were studied with submerged AnMBR where the membranes are immersed directly into anaerobic bioreactor [9,10]. In the case of hard-shell encased membrane module as side-stream for anaerobic bioreactor, the applications of these methods may be limited because membranes are encased in pressurized vessel. Instead, both cross-flow velocity and transmembrane pressure (TMP) should be main operating parameters feasible to control membrane fouling. Nevertheless, little information on the combined effects of both parameters on the reduction of membrane fouling in AnMBR are available particularly for the treatment of high strength wastewater such as food waste leachate. The objective in this study was to observe combined effects of cross-flow velocities and TMP on the reduction of membrane fouling from the pilot-scale AnMBR in order to better

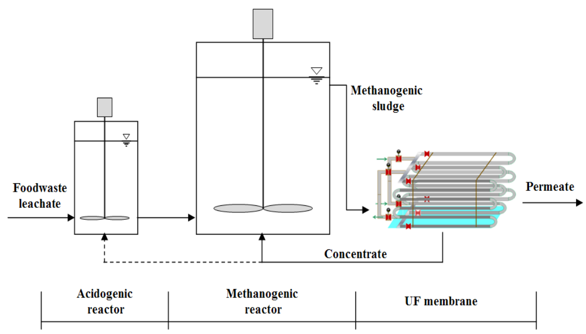

Fig. 1. Schematic diagram of AnMBR system treating food waste leachate.

understand fouling behavior in the pilot-scale AnMBR treating food waste leachate.

2. Experimental

A pilot-scale AnMBR system was installed within the municipal food waste treatment plant (Incheon, Korea) managed by the Environmental Cooperation of Incheon. Fig. 1 shows schematics of pilot-scale design of AnMBR treating food waste leachate. The main system of the AnMBR consisted of thermophilic acidogenic reactor, methanogenic reactor and one-channel tubular ultrafiltration (UF) membrane (Jinshui Membrane, China). The membrane was composed of PVDF and the pore size of the membrane used was 0.01 µm. The effluent from the methanogenic reactor was pressurized into the hard-shell encased tubular UF membrane module. Membrane concentrate was recycled back into the methanogenic reactor to prevent wash-out of anaerobes and maintain high concentration of biomass in the reactor.

The feed flow rate and organic loading rate (OLR) of food waste leachate for the methanogenic reactor were 3 m^3/day and 5.0 kg COD_{Cr}/m^3/day, respectively. The total HRT was 20 days and operating temperature of the anaerobic reactor was 55 ± 1°C. The average COD_{Cr} and suspended solids concentration of the methanogenic sludge were about 26,600 mg/L and 15 g/L, respectively. A PVDF one-channel tubular UF membrane module was installed as a side stream to treat the effluent from thermophilic methanogenic reactor. Seven tubular membranes were encased in one module. The membrane module was operated under cross-flow filtration at constant TMP and total recycle mode of operation. The diameter of tubular one-channel UF membrane was 11 mm and total membrane surface area in this study was 13.1 m^2. The operational conditions of cross-flow velocity and TMP ranged from 1 to 3 m/s, and from 1 to 3 bar, respectively. Membrane permeate effluent was further treated by ammonia stripping for the nitrogen removal followed by aerobic MBR for the treatment of remaining organics to meet effluent quality standards. At the end of operation, membrane surface was observed by energy-dispersive X-ray analysis (SEM-EDX, Hitachi, S-4200) in order to quantify inorganic components in foulant materials. Fourier Transform Infrared (FTIR) analysis (Bruker, Vertex 80V) was also performed to analyze organic fraction of foulant layer.

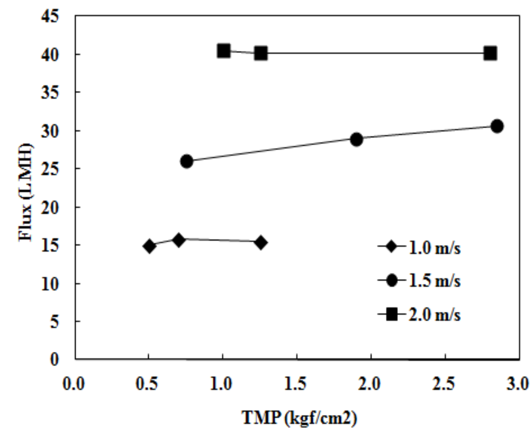

Fig. 2. Combined effect of cross-flow velocity and TMP on permeate flux.

3. Results and Discussion

3.1. Combined Effect of TMP and Cross-flow Velocity on Permeate Flux

Fig. 2 shows the combined effects of TMP and cross-flow velocity on permeate flux in AnMBR. Result indicates that permeate flux increase as cross-flow velocity increases at fixed TMP, suggesting that cross-

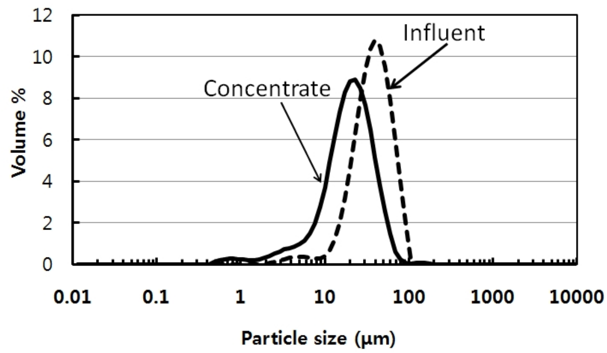

Fig. 3. Particle size distribution of feed suspension and membrane concentrate.

flow velocity influence fouling mitigations. Fig. 3 indicates that particle sizes of feed solution in AnMBR range from 10 to 100 μm. However, the particle sizes were shifted to be smaller in membrane concentrate possibly due to break-up of microbial flocs under high shear rate along membrane surface [11]. It was expected that higher shear rate at higher cross-flow velocity should improve back transport of these particle sizes away from membrane surface. However, the permeate flux was relatively independent upon TMP. This can be explained by the fact that increasing resistance and/or thickness of fouling layer (or cake layer) on membrane surface should be countered by increasing TMP, thus the permeate flux is approached to limiting flux.

The particle back diffusivity as a function of particle size can be estimated from using equations (1) and (2). The Brownian back diffusion coefficient (D_B) for small colloids and macromolecules can be calculated from equation (1) of the Stokes-Einstein equation. Assuming that volume fraction of the cake on membrane has maximum packing density, shear-induced diffusivity (D_s) can be calculated as a function of shear rate which is estimated by equation (2).

$$D = D_B + D_S = \frac{k_B T}{6\pi\mu r_p} + 0.03 r_p^2 \gamma \quad (1)$$

$$\gamma = 8U/d \quad (2)$$

where k_B is Boltzmann constant, T is absolute temperature, r_p is particle radius, γ is shear rate, U is cross-flow velocity and d is diameter of tubular membrane. Results in Fig. 4 showed that the back transport of particles bigger than about 0.2 μm of particle size was dominated by shear-induced back diffusion (D_S) while Brownian back-diffusion (D_B) was more important with particles smaller than 0.2 μm. This result shows that shear rate controlled by cross-flow velocity can be an important way to increase back diffusion of foulant away from membrane in the AnMBR treating food waste leachate. Since effluent from anaerobic bioreactor contains wide range of particles, however, interaction between particles of different sizes can also affect the back-transport of each while the mechanisms by which such interactions still need to be explored.

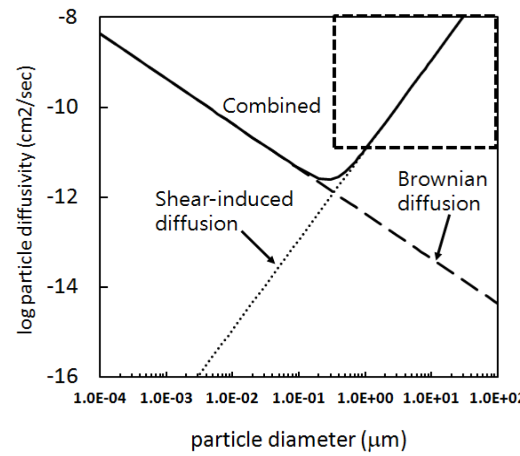

Fig. 4. Back transport diffusivity as a function of particle size (cross-flow velocity = 1.5 m/s).

3.2. Combined Effect of TMP and Cross-flow Velocity on Fouling Resistance

Total fouling resistance was measured using permeate flux (J) data at different TMPs and cross-flow velocities using Darcy's equation described below. From Equation (3), the permeate flux is proportional to TMP but inversely proportional to the total resistance (R_{tot}) and viscosity of feed suspension (μ). Total resistance was assumed to be summation of cake resistance on membrane surface (R_c) and membrane resistance

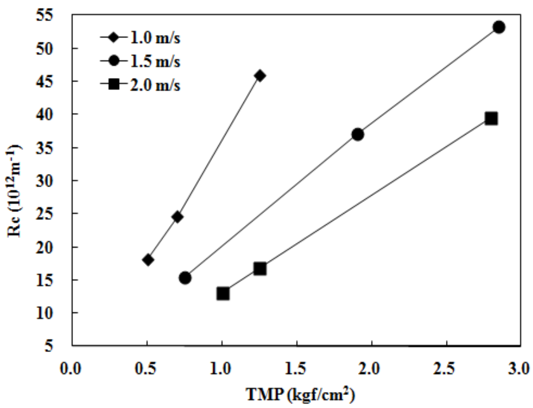

Fig. 5. Effect of TMP on fouling resistance at different cross-flow velocities.

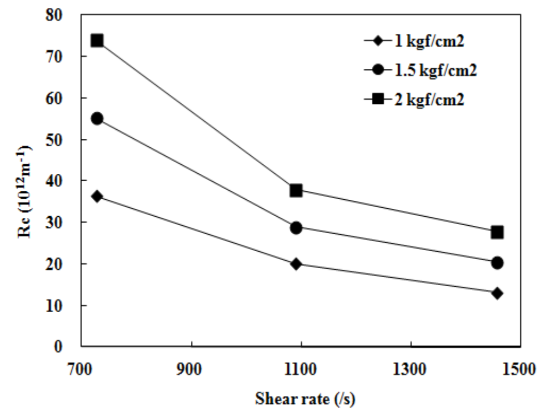

Fig. 6. Effect of cross-flow velocity on fouling resistance at different TMPs.

(R_m). It is true that irreversible fouling resistance which is mainly caused by adsorption of foulant into membrane pores can be included in total resistance. Since particle size distribution in feed solution to the membrane ranged from 1 to 100 μm (Figure 3) and membrane pore size was about 0.01 μm, however, it was assumed that cake resistance on membrane surface should be dominant fouling resistance.

$$J = \frac{TMP}{\mu R_{total}} \tag{3}$$

$$R_C = \frac{TMP}{\mu J} - R_m \tag{4}$$

It was found that cake resistance increased with increasing TMP while the extent of the increase was more pronounced at lower cross-flow velocity (Fig. 5). Since biologically generated particles found in AnMBR are deformable, the fouling layer should be characterized compressible. The compressibility can lead to higher fouling resistance at higher TMP since it is related to the TMP using a power-law equation below.

$$\alpha = \alpha_0 \Delta P^n \tag{5}$$

where α and n are average specific cake resistance of entire cake depth on membrane and compressibility, respectively. The specific cake resistance (α, m/kg) can be estimated by using equation (6).

$$\frac{t}{(V/A)} = \frac{\mu\alpha C_0}{2\Delta P}\left(\frac{V}{A}\right) + \frac{\mu R_m}{\Delta P} \tag{6}$$

where A is the membrane surface area, V is the permeate volume per membrane area accumulated in time, t. C_0 is solids concentration in bulk suspension and R_m is membrane resistance. Using equation (6), the specific cake resistance of α can be estimated from the slope of linear relationship between t/(V/A) and (V/A). In fact, the effluent from anaerobic bioreactor was characterized as highly compressible material having compressibility near 1.0. This value is within the range for microbial suspensions which is from 0.3 to 1.0. Increasing TMP compresses structure of cake layer and it can produce more compact structure of cake layer due to increasing in cake resistance.

As shown in Fig. 6, increasing shear rate decreases fouling resistance for all TMPs applied in this study. As mentioned, fouling layer becomes less compressible at lower TMP and thus effectiveness of shear rate to remove fouling layer may be more pronounced. However, it was observed that further increase in shear rate above 1,100 s^{-1} was not very effective to reduce fouling resistance, suggesting that optimum shear rate to mitigate membrane fouling should exist. Since col-

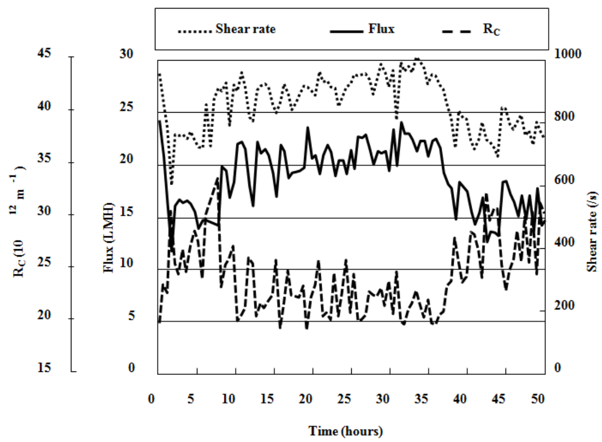

Fig. 7. Behavior of fouling resistance and flux as a function of shear rate (TMP = 0.8 kgf/m^2) in longer-term operation.

loidal materials which is smaller than 0.2 μm may not be transported easily away from the membrane surface by shear-induced back diffusion (Fig. 4), contribution of these small colloids to membrane fouling can not be overlooked.

3.3. Fouling Behavior in Continuous Operation

Continuous operation was conducted to observe the fouling behavior at different shear rates in AnMBR under 0.8 bar of TMP. As shown in Fig. 7, the shear rate below 1,000 s^{-1} has close relationship with fouling behavior in AnMBR. The transient behavior of permeate flux has similar pattern with shear rate. That is, as shear rate increased, permeate flux increased while cake resistance decreased. After increasing shear rate from 700 to 900 s^{-1}at 3 hr operation, permeate flux increased to about 20 L/m^2/hr from 10 L/m^2/hr. However, sharp decrease in permeate flux was observed at 40 hr operation at lower shear rate. This continuous observations confirm strongly that shear rate can be an important parameter to control membrane fouling in AnMBR treating food waste leachate.

3.4. Microscopic Observations of Membrane Fouling

Energy Dispersive X-ray (EDX) analysis of the foulant accumulated on membrane surface at the end of operation showed that large portion of Ca^{2+} was detected. It appeared that the formation of inorganic fouling such as calcium carbonate scaling should be formed on membrane surface because of the production of alkalinity in the AnMBR. It was observed that higher than 90% of Ca^{2+} was removed through the membrane while only 10 and 40% of removal was achieved for $Mg^{2+}$and Na^{+}, respectively. Considering pore size of the membrane tested in this study, 90% removal of Ca^{+2} cannot be achieved. High calcium removal may be caused by inorganic precipitate on membrane surface due to high calcium concentration (> 800 mg/L) in the food waste leachate and increase in the alkalinity from methanogenic sludge (> 10,000 mg/L) [12]. Our FTIR analysis also showed that the foulant material on membrane surface consisted mainly of polysaccharides and carboxylic groups (data are now shown), suggesting that microbial agent such as soluble microbial products (SMPs) should be one of main sources of organic fouling in AnMBR. Further works, however, are needed to know dominant species of inorganic scaling and how they can interact with organic fouling in AnMBR treating food waste leachate.

4. Conclusions

Membrane fouling could be reduced by increasing shear rate along membrane in AnMBR treating food waste leachate but the effectiveness was more pronounced at lower TMP under which less compressible fouling layer could be formed. Increasing shear rate along the membrane surface reduced membrane fouling, but optimum value above which no significant reduction of membrane fouling was observed. Continuous operation of AnMBR at different cross-flow velocities confirmed that the shear rate should affect reduction of membrane fouling significantly. Membrane autopsies at the end of operation indicated both organic and inorganic fouling on membrane surface while further works are needed to identify which one is more responsible under cross-flow velocity of the AnMBR treating food waste leachate.

Acknowledgement

This research is financially supported by a research project (10ECOS01) of Hyundai Engineering & Construction Co., Ltd. and Korea Ministry of Environment (MOE) as “Human Resource Development Project for Waste to Energy”.

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