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| Author(s) | Hafuka, Akira; Mimura, Kazuhisa; Ding, Qing; Yamamura, Hiroshi; Satoh, Hisashi; Watanabe, Yoshimasa |
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**Performance of anaerobic membrane bioreactor during digestion and thickening
of aerobic membrane bioreactor excess sludge**

Akira Hafuka^{a,*}, Kazuhisa Mimura^b, Qing Ding^c, Hiroshi Yamamura^a, Hisashi Satoh^d,

Yoshimasa Watanabe^e

^a*Department of Integrated Science and Engineering for Sustainable Society, Faculty of*

Science and Engineering, Chuo University, 1-13-27 Kasuga, Bunkyo-ku, Tokyo

112-8551, Japan

^b*Technical Research & Development Institute, Sanki Engineering Co., Ltd., 1742-7*

Shimotsuruma, Yamato-shi, Kanagawa 242-0001, Japan

^c*Division of Civil and Environmental Engineering, Graduate School of Science and*

Engineering, Chuo University, 1-13-27 Kasuga, Bunkyo-ku, Tokyo 112-8551, Japan

^d*Division of Environmental Engineering, Graduate School of Engineering, Hokkaido*

University, North-13, West-8, Sapporo 060-8628, Japan

^e*Research and Development Initiatives, Chuo University, 1-13-27 Kasuga, Bunkyo-ku,*

Tokyo 112-8551, Japan

E-mail address:

A. Hafuka: hafuka.14p@g.chuo-u.ac.jp

K. Mimura: kazuhisa_mimura@eng.sanki.co.jp

Q. Ding: dingqing1988@gmail.com

H. Yamamura: yamamura.10x@g.chuo-u.ac.jp

H. Satoh: qsatoh@eng.hokudai.ac.jp

Y. Watanabe: yoshiw@tamacc.chuo-u.ac.jp

*Corresponding author. Tel./fax: +81-3-3817-7283.

E-mail address: hafuka.14p@g.chuo-u.ac.jp (A. Hafuka).

ABSTRACT

In this study, we evaluated the performance of an anaerobic membrane bioreactor in terms of digestion and thickening of excess sludge from an aerobic membrane bioreactor. A digestion reactor equipped with an external polytetrafluoroethylene tubular microfiltration membrane module was operated in semi-batch mode. Solids were concentrated by repeated membrane filtration and sludge feeding, and their concentration reached 25 400 mg/L after 92 d. A high chemical oxygen demand (COD) removal efficiency, i.e., 98%, was achieved during operation. A hydraulic retention time of 34 d and a pulse organic loading rate of 2200 mg-COD/(L-reactor) gave a biogas production rate and biogas yield of 1.33 L/(reactor d) and 0.08 L/g-COD_{input}, respectively. The external membrane unit worked well without membrane cleaning for

90 d. The transmembrane pressure reached 25 kPa and the filtration flux decreased by 80% because of membrane fouling after operation for 90 d.

Keywords: Anaerobic digestion, Membrane filtration, Excess sludge, Microfiltration, Tubular PTFE membrane module

1. Introduction

The use of aerobic membrane bioreactors (MBRs) is a promising technique for municipal and industrial wastewater treatments because they produce higher quality effluents and have smaller footprints compared with conventional treatment processes (Judd, 2007). Although the amount of excess sludge produced by an MBR is smaller than that produced in conventional activated sludge processes (Wei et al., 2003), anaerobic digestion of MBR excess sludge is necessary along with the widespread use of MBRs. Anaerobic digestion requires no aeration, reduces the biomass yield, and generates methane-containing biogas. It is therefore used to treat sludge as well as wastewaters and food wastes (Lettinga et al., 2001; Khalid et al., 2011; Zhang et al., 2007). However, the growth rates of anaerobic microorganisms are low and the effluent quality is not sufficient. To overcome these disadvantages of anaerobic treatment, anaerobic membrane bioreactors (AnMBRs) have attracted much attention recently

(Smith et al., 2012, 2014). AnMBR can control the hydraulic retention time (HRT) and solid retention time (SRT) separately using membrane filtration. High solid concentrations can therefore be attained because anaerobic microorganisms are not washed out from the reactor. In addition, the effluent water quality is high because suspended solids (SS) are removed. Many researchers have studied the use of AnMBRs in the treatment of synthetic or actual wastewaters; for example, microbial community shifts, the membrane fouling properties of extracellular polymeric substances, and the removal of trace organic chemicals in synthetic wastewaters have recently been reported (Ding et al., 2015; Wijekoon et al., 2015; Win et al., 2016). Performance evaluation and optimization of the operational parameters of AnMBRs have been performed using actual wastewaters or kitchen waste slurries (Ng et al., 2015; Xiao et al., 2015; Yu et al., 2016). However, few studies have focused on excess sludge treatment using an AnMBR (Meabe et al., 2013). In general, primary and secondary excess sludges are transferred to a thickener, which generates a total solids (TS) concentration of 3–6%, before anaerobic digestion (Gerardi, 2003). Use of an AnMBR for sludge treatment could reduce or avoid sludge-thickening processes because solids accumulate in an AnMBR during membrane filtration (Pierkiel et al., 2005). The objective of this study was therefore to evaluate the performance of an AnMBR for digestion and thickening of an

actual MBR excess sludge. The results will help to expand the use of AnMBRs in sludge treatment.

2. Materials and methods

2.1. AnMBR setup and operating conditions

The AnMBR consisted of a digestion reactor (DR) with a working volume of 20.0 L, and an external membrane unit (MU). The DR was agitated continuously with a stirrer. The DR was covered with a heating jacket to maintain a temperature of 35 °C (mesophilic condition). The MU was a polytetrafluoroethylene (PTFE) tubular microfiltration (MF) membrane module (POREFLON LPM-X240, Sumitomo Electric Industries, Ltd., Osaka, Japan). The total effective area of the membrane module was 0.06 m² and the inner diameter of each membrane tube was about 5.1 mm. The AnMBR was operated in semi-batch mode without sludge discharge, except for sampling. Membrane filtration and MBR excess sludge feeding were performed twice a week. The DR was inoculated with 1.0 L of homogenized upflow anaerobic sludge blanket (UASB) granular sludge containing 84 300 mg/L TS and 52 000 mg/L volatile solids (VS), obtained from a full-scale UASB reactor used to treat food manufacturing wastewater in the plant. Membrane filtration of the digested sludge was performed at a

constant cross-flow velocity of 0.7 m/s using a mono pump (NY 40, Heishin Ltd., Kobe, Japan). The transmembrane pressure (TMP) was monitored using manometers (GC61-174, Nagano Keiki Co., Ltd., Tokyo, Japan) during membrane filtration. The MBR excess sludge feed was obtained from a full-scale MBR reactor treating miscellaneous drainage waste in the plant. The raw MBR excess sludge was screened (1 mm) and stored at 4 °C before feeding into the DR. Digested sludge samples (0.1 L) were collected once a week before membrane filtration and after sludge feeding. In theory, the SRT in an AnMBR is infinite, but it was set at 700 d because of sludge sampling. The HRT was controlled by changing the membrane filtrate volume. The HRT was set at 67 d in the first 43 d (phase 1) and was changed to 34 d after 43 d (Phase 2). The volume of biogas produced from the DR was measured using a wet gas meter (W-NK-0.5A, Shinagawa Co., Tokyo, Japan). The pH and oxidation–reduction potential (ORP) in the reactor were measured using a pH meter (D-74, Horiba, Ltd., Kyoto, Japan) and ORP meter (TRX-999, Tokyo Chemical Laboratories Co., Ltd., Tokyo, Japan), respectively. Membrane cleaning was not performed during the operation.

2.2. Analytical methods

The MBR excess sludge feed, membrane filtrate, and digested sludge were sampled

once a week. The TS and VS concentrations in the sludge were determined using standard methods (APHA, 2012). The chemical oxygen demand with potassium dichromate (COD_{Cr}), total nitrogen (T-N), and total phosphorus (T-P) concentrations in the sludge and membrane filtrate were determined by Hach methods (Methods 8000, 10127, and 10072, respectively), using a spectrophotometer (DR 3900, Hach Co., Loveland, USA), after appropriate sample dilution. The ammonium (NH_4^+ -N) and orthophosphate (PO_4^{3-} -P) concentrations in the samples were also determined using Hach methods (Method 10031 and 8114, respectively) after centrifugation to obtain sample supernatants. The biogas was collected using an aluminum gas bag and the methane content was determined using gas chromatography (GC-14B, Shimadzu Co., Kyoto, Japan). The solubilization rate from T-N (or T-P) to NH_4^+ -N (or PO_4^{3-} -P) was calculated using Eq. (1):

Solubilization rate

$$= \frac{\text{NH}_4^+ - \text{N (or PO}_4^{3-} - \text{P)} \text{mass}_{\text{increased}}}{\text{T - N (T - P)} \text{mass}_{\text{input}}} \quad (1)$$

The biogas yield was calculated using Eq. (2):

$$\text{Biogas yield} = \frac{\text{Biogas volume}_{\text{produced}}}{\text{COD mass}_{\text{input}}} \quad (2)$$

3. Results and discussion

Table 1 shows the characteristics of the MBR excess sludge fed to the DR. The average COD concentration was 21 200 mg/L and the VS/TS ratio was constant at 84%. Fig. 1(a) shows the changes in pH and ORP in the DR. The initial pH was 6.4 and it gradually increased to 7.0 over 92 d. The average pH values in the reactor were 6.6 and 6.8 in phase 1 and phase 2, respectively. The ORP immediately decreased to around -400 mV after starting the operation and was stable during operation, confirming anaerobic digestion conditions in the DR.

Fig. 1(b) shows the temporal changes in the TS and VS concentrations of the digested sludge after sludge feeding. In previous studies, AnMBRs with 20 000–30 000 mg-TS/L of digested sludge were operated using waste activated sludge or kitchen waste slurry (Dagnew et al., 2012; Xiao et al., 2015). We therefore aimed to achieve more than 25 000 mg-TS/L during operation. TS and VS both accumulated during operation because no sludge was removed. The TS and VS concentrations had increased from 11 500 mg/L and 8900 mg/L, respectively, to 25 400 mg/L and 20 200 mg/L, respectively, after 92 d. The average TS and VS concentrations in the feed MBR sludge were 15 000 mg/L and 12 700 mg/L, respectively (Table 1), indicating that the feed sludge accumulated 1.7-fold during operation for 92 d.

Fig. 2(a) shows the changes in the COD concentrations of the digested sludge and membrane filtrate, and COD removal efficiencies, based on the COD mass. The COD concentration in the digested sludge gradually increased from 11 100 mg/L to 37 800 mg/L during 92 d. The COD removal efficiency decreased slightly during operation but a high COD removal efficiency, i.e., 98%, was achieved because of SS removal by membrane filtration. This high removal efficiency is comparable to those obtained in previous studies using starch wastewater or kitchen waste slurry (Xiao et al., 2015; Yu et al., 2016). Fig. 2(b) shows the changes in the pulse organic loading rate (OLR) to the DR and biogas production rate from the reactor. The average OLR in phase 1 was 1300 ± 300 mg-COD/(L-reactor) and was increased to 2200 ± 200 mg-COD/(L-reactor) in phase 2. Significant biogas production was first observed on day 13 from start-up. The biogas production rate increased from phase 1 to phase 2 with increasing OLR. Within one cycle (1 week), biogas was mainly produced 1–2 d after sludge feeding.

Table 2 shows the HRTs, SRTs, biogas yields, and methane contents in phase 1 and phase 2. The biogas yield increased from 0.03 L/g-COD_{input} in phase 1 to 0.08 L/g-COD_{input} in phase 2. As shown in Fig. 1(a), pH increased to appropriate range of anaerobic digestion in phase 2 (Gerardi, 2003). In addition, microbial biomass might also increase in phase 2 due to solid thickening. Therefore, we thought that the biogas

yield increased in phase 2. Yu et al. (2012) also reported that the digestibility of MBR excess sludge was lower than that of sludge produced using an activated sludge process. The methane content of the biogas was around 65% in both phase 1 and phase 2. This value was slightly higher than that from general waste activated sludge (Appels et al., 2008).

Fig. 3(a) shows the temporal changes in the nitrogen concentrations in the digested sludge and membrane filtrate. NH_4^+ -N in the digested sludge and T-N in the membrane filtrate were also measured but these values were equal to NH_4^+ -N in the membrane filtrate, indicating that organic nitrogen was completely removed by membrane filtration, whereas NH_4^+ -N passed through the membrane. The T-N and NH_4^+ -N concentrations increased from 1080 mg-N/L and 72 mg-N/L, respectively, to 2600 mg-N/L and 383 mg-N/L, respectively, during 92 d. The NH_4^+ -N/T-N ratio was 8% on day 8 and it increased to 15% by day 92. The T-N to NH_4^+ -N solubilization rate inside the DR was calculated from the mass balance; it was around 30% in both phase 1 and phase 2. The NH_4^+ -N concentration reached 383 mg-N/L after 92 d but was still under 1500 mg- NH_4^+ /L, which is still below the inhibition level for anaerobic digestion (Rajagopal et al., 2013). This could be the effect of membrane filtration, which can reduce the amount of NH_4^+ -N in the DR. Fig. 3(b) shows the temporal changes in the

phosphorus concentrations in the digested sludge and membrane filtrate. $\text{PO}_4^{3-}\text{-P}$ in the digested sludge and T-P in the membrane filtrate were determined, but the values were equal to $\text{PO}_4^{3-}\text{-P}$ in the membrane filtrate. The T-P and $\text{PO}_4^{3-}\text{-P}$ concentrations gradually increased from 320 mg-P/L and 37 mg-P/L, respectively, to 635 mg-P/L and 345 mg-P/L, respectively, during operation. The $\text{PO}_4^{3-}\text{-P}$ /T-P ratio inside the reactor was 16% on day 8 and it increased to 26% by day 85. In general, $\text{PO}_4^{3-}\text{-P}$ solubilization occurs in anaerobic digestion (Latif et al., 2015). However, $\text{PO}_4^{3-}\text{-P}$ solubilization was not observed in phase 1. The T-P to $\text{PO}_4^{3-}\text{-P}$ solubilization rate in phase 2 was estimated to be 32%.

Membrane fouling is the main problem in MBRs (Dagnew et al., 2012). The TMP and filtration flux were therefore measured (Fig. 4). We were able to operate the AnMBR system for 90 d without membrane cleaning. There was a trade-off between TMP and filtration flux, as expected. We performed 22 membrane filtrations and filtered a total of 42 L of digested sludge during all operations. The initial flux was about 0.6 m/d, but it declined to 0.1 m/d after 92 d, i.e., an approximately 80% decline in the filtration flux. In contrast, the TMP gradually increased and it reached 25 kPa after 92 d. Although an MF membrane might be sufficient for sludge filtration in an AnMBR, because of its high COD removal efficiency (Fig. 2a), membrane cleaning is needed for

longer operation. In previous studies, ultrafiltration membranes were often used in AnMBRs for the treatment of activated sludge, kitchen waste slurry, and dairy manure (Meabe et al., 2013; Xiao et al., 2015; Zitomer et al., 2005). To the best of our knowledge, this study is the first to use a PTFE MF membrane in an AnMBR for sludge treatment. Cross flow filtration of digested sludge in a polytetrafluoroethylene tubular microfiltration membrane which has a large inner diameter of 5.1 mm might prevent severe membrane fouling.

4. Conclusions

We evaluated the performance of an AnMBR with an external PTFE tubular MF membrane module during digestion and thickening of MBR excess sludge. A TS concentration of 25 400 mg/L after 92 d and high COD removal efficiency, 98%, were achieved. An HRT of 34 d with a pulse OLR of 2200 mg-COD/(L-reactor) gave a biogas production rate and yield of 1.33 L/(reactor d) and 0.08 L/g-COD_{input}. The TMP reached 25 kPa and the filtration flux decreased by 80% after 92 d without membrane cleaning. We plan to shorten the HRT and perform long-term AnMBR operation with membrane cleaning.

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Table and figure captions

Table 1

Characteristics of MBR excess sludge fed to DR ($n = 12$).

Table 2

HRTs, SRTs, biogas yields, and methane contents ($n = 6$) in phase 1 and phase 2.

Fig. 1. (a) pH and ORP in DR and (b) TS and VS concentrations of digested sludge.

Times of membrane filtration and sludge addition are indicated by vertical lines.

Fig. 2. (a) COD concentrations of digested sludge and membrane filtrate and COD

removal efficiencies based on COD mass and (b) OLR to DR and biogas production rate

from DR in each cycle.

Fig. 3. (a) Nitrogen concentrations and (b) phosphorus concentrations in digested sludge

and membrane filtrate.

Fig. 4. TMP and filtration flux of MU.

Table 1

Characteristics of MBR excess sludge fed to DR ($n = 12$).

| Parameter | Unit | Value |
|--------------------|--------|--------------------|
| TS | mg/L | $15\,000 \pm 1200$ |
| VS | mg/L | $12\,700 \pm 1000$ |
| VS/TS | % | 84 ± 0 |
| COD | mg/L | $21\,200 \pm 1300$ |
| T-P | mg-P/L | 414 ± 23 |
| PO ₄ -P | mg-P/L | 158 ± 54 |
| T-N | mg-N/L | 1270 ± 230 |
| NH ₃ -N | mg-N/L | 32 ± 16 |
| pH | - | 6.6 ± 0.1 |

Table 2

HRTs, SRTs, biogas yields, and methane contents ($n = 6$) in phase 1 and phase 2.

| Phase | HRT (d) | SRT (d) | Biogas yield (L/g-COD _{input}) | Methane content (%) |
|-------|---------|---------|--|---------------------|
| 1 | 67 | 700 | 0.03 | 68.5 ± 0.1 |
| 2 | 34 | 700 | 0.08 | 64.5 ± 1.7 |







