Title	Anaerobic digestibility of up-concentrated organic matter obtained from direct membrane filtration of municipal wastewater
Author(s)	Hafuka, Akira; Takahashi, Taketsugu; Kimura, Katsuki
Citation	Biochemical engineering journal, 161, 107692 https://doi.org/10.1016/j.bej.2020.107692
Issue Date	2020-09-15
Doc URL	http://hdl.handle.net/2115/86151
Rights	© 2020. This manuscript version is made available under the CC-BY-NC-ND 4.0 license http://creativecommons.org/licenses/by-nc-nd/4.0/
Rights(URL)	http://creativecommons.org/licenses/by-nc-nd/4.0/
Type	article (author version)
File Information	Revised manuscript_BEJ-D-20-0038_clean.pdf

Paper submitted for publication in *Biochemical Engineering Journal* as a short communication

Anaerobic digestibility of up-concentrated organic matter obtained from direct membrane filtration of municipal wastewater

Akira Hafuka*, Taketsugu Takahashi, Katsuki Kimura

Division of Environmental Engineering, Graduate School of Engineering, Hokkaido University, Kita 13, Nishi 8, Kita-ku, Sapporo 060-8628, Japan

*Corresponding author: A. Hafuka (ahafuka@eng.hokudai.ac.jp)

Abstract: Direct membrane filtration (DMF) process is a promising technology for up-concentration and recovery of organic matter (OM) from municipal wastewater. This study investigates anaerobic digestibility of up-concentrated OM obtained from the DMF process. Effluent from a primary sedimentation basin was used as feed water. Effect of polyaluminum chloride (PACl) addition to the DMF process on the anaerobic digestibility of obtained up-concentrated OM was also investigated because PACl is sometimes used as a coagulant in the DMF process for improving effluent quality and OM recovery, and for mitigating membrane fouling. Batch anaerobic digestion experiments were conducted to achieve these objectives. Fitting the obtained data to the reaction curve model revealed that maximum biogas production based on feed volatile solids (VS_{fed}) from up-concentrated OM containing PACl reached 0.56 L-Biogas/g-VS_{fed}, which was higher than from up-concentrated OM without PACl (0.47 L-Biogas/g-VS_{fed}), waste activated sludge (0.35 L-Biogas/g-VS_{fed}), and mixed sludge (0.48 L-Biogas/g-VS_{fed}). Methane yield based on degraded VS (VS_{deg}) of up-concentrated OM containing PACl was 0.56 L-CH_4/g-VS_{deg}, which was also highest. These results indicate the up-concentrated OM obtained from the DMF process has high potential for biogas production, and the addition of PACl in the DMF process (~4.3 mg-Al/L) could enhance the biogas production.

Keywords: Biogas, Methane, Polyaluminum chloride, Batch experiment, Waste activated sludge

1. Introduction

In municipal wastewater treatment, organic matter (OM) is a main substance to be removed to maintain sanitary conditions and to protect aquatic environments. On the other hand, OM in municipal wastewater can be regarded as a resource for energy production, because biogas can be generated from up-concentrated OM via anaerobic digestion [1]. The activated sludge process is the most widespread wastewater treatment process and has been used for more than a century [2]. Although the activated sludge process provides adequate effluent quality, a large amount of energy is consumed by aeration. Mixed sludge, the mixture of primary sludge and waste activated sludge (WAS), is generated in the activated sludge process and is often treated by anaerobic digestion in large-scale wastewater treatment plants [3]. However, the methane yield of WAS is low because of its low biodegradability [4]. The high rate activated sludge (HRAS) process and the chemically enhanced primary treatment (CEPT) process have been proposed for up-concentration and recovery of OM to achieve energy neutrality in municipal wastewater treatment [1]. Although the HRAS and CEPT processes have been tested in full-scale applications, aeration is still needed.

Direct membrane filtration (DMF) of municipal wastewater using microfiltration or ultrafiltration membranes is another option to up-concentrate the OM [5]. In the DMF process, membranes provide solid-liquid separation based on their pore sizes. Therefore, OM with a size larger than that of membrane pore size cannot pass through the membrane and then it is concentrated. As DMF is a membrane-based process, it does not require intensive aeration. In addition, a high recovery efficiency of OM (70-90%) based on chemical oxygen demand (COD) was reported for the DMF process [6-9], as membranes can provide high solid-liquid separation efficiency. The reported COD concentrations of up-concentrated OM are in the range of 8,000 to 23,000 mg/L [6-9], which can be supplied for anaerobic digestion process. The main concern of DMF process is membrane fouling, which decreases filtration flux and increases energy consumption. Therefore, some studies focusing on control and mitigation of membrane fouling in the DMF process have been reported [6-9]. Polyaluminum chloride (PACl) is sometimes used as a coagulant in the DMF process to increase OM recovery and to mitigate membrane fouling [7-9]. It has been reported that use of PACl also improved effluent quality by removing OM and nutrient (i.e., phosphorus) [7-9]. On the other hand, inhibitory effects of PACl on anaerobic digestion of WAS has been reported [10]. So far, anaerobic digestibility of up-concentrated OM obtained from the DMF process has not been evaluated in detail. Investigation of the anaerobic digestibility of up-concentrated OM is important to estimate the energy efficiency of the whole DMF process. The aim of this study is to first investigate the anaerobic digestibility of up-concentrated OM and compare it with that of mixed sludge and WAS. The second aim is to investigate the effect of PACl on the digestibility of up-concentrated OM. Batch anaerobic digestion experiments were used to achieve these objectives. The maximum biogas production and maximum biogas production rate were estimated by fitting the experimental data to the reaction curve (RC) model.

2. Material and Methods

2.1. Up-concentration of OM in DMF system

Up-concentrated OM was obtained from two laboratory-scale DMF reactors which were installed at an existing full-scale municipal wastewater treatment plant (Soseigawa wastewater treatment plant, Sapporo, Japan). Conventional activated sludge process is used in the plant and the treatment capacity is 144,000 m^3/d. Effluent (i.e., not primary sludge but wastewater) from the primary sedimentation basin in the plant was used as the feed water. The average concentration of COD in the feed water was 194 ± 5 mg/L. The DMF reactors were set up according to a previously reported configuration with some modifications [6]. Each DMF reactors was composed of two filtration tanks sequentially connected (working volume: 4.6 L each). Flat-sheet ceramic membranes (total effective area: 0.147 m^2) with a nominal pore size of 0.1 μm were immersed in each tank. The feed water was filtered through the membranes with dead-end filtration using suction pumps. Up-concentrated OM was continuously obtained from the second tank. The operational condition of each DMF reactors is shown in Table S1. Intermittent filtration and intermittent aeration were conducted throughout the operation. Mechanical scoring with granules and backwashing with tap water and/or sodium hypochlorite solution (500 mg/L) were conducted to avoid membrane fouling. Granules (BCN, Nisshinbo Chemical Inc., Tokyo, Japan) were added in each tank and were fluidized mechanically with stirrers for physical membrane cleaning. The granules were made from polyethylene glycol and they were cylindrical (diameter: 4 mm; height: 4 mm). One DMF reactor was operated with PACl addition and the other one was operated without PACl. The PACl addition was set at 4.3 mg-Al/L, which was the same addition in a previous study [9]. PACl stock solution (7 mL) was directly added to the first tank of the DMF reactor every 15 minutes. The hydraulic retention time of the first tank was 1.1 h, which ensured sufficient contact time.

2.2. Batch anaerobic digestion experiments

The mixed sludge and WAS were also obtained from the Soseigawa wastewater treatment plant. The mixed sludge was thickened by centrifuge to reduce the sludge volume. The digested sludge was obtained from a mesophilic sludge digester of another full-scale municipal wastewater treatment plant (Ebetsu wastewater treatment plant, Ebetsu, Japan) and was used as seed sludge. All sludge samples were screened through a 1 mm mesh before use. The batch anaerobic digestion experiments were conducted in 200 mL glass vials at 35 °C according to previously reported methods [11]. This kind of small-scale batch experiment is useful to evaluate biogas production potential of substrates [11-13]. Each vial was filled with 0.66 g-VS of seed sludge (80 mL) and 0.22 g-VS of substrate sludge (60-120 mL). The produced biogas volume was normalized to standard condition of 101.3 kPa and 0 °C. All samples were prepared in duplicate, and control samples filled with only seed sludge were also prepared. The maximum biogas production and maximum biogas production rate based on the feed volatile solids (VS_{fed}) were estimated by fitting the experimental data to the reaction curve (RC) model which is described in Eq. (1) [14]:

$$B = P\left(1 - exp\left(-\frac{R_m(t-\lambda)}{P}\right)\right), (1)$$

where *B* is the cumulative biogas production (L/g-VS_{fed}), *P* is the maximum biogas production (L/g-VS_{fed}), R_m is maximum biogas production rate (L/(g-VS_{fed}·d)), t is digestion time (d), and λ is lag time (d). We obtained these values by using Origin Pro 9.65 software. The RC model was reported as a simplified model for anaerobic digestibility test and the equation has been widely applied for various substrates including sewage sludge [15].

2.3. Analytical methods

Selected physical and chemical properties of sludge samples were analyzed before and after the batch digestion experiments. The pH of the sludge samples was measured using a pH meter (D-73, Horiba, Ltd., Kyoto, Japan). Concentrations of total solids (TS) and VS were determined according to standard methods [16]. Concentrations of total nitrogen (T-N), ammonium (NH_4^+-N), total phosphorus (T-P), and orthophosphate (PO_3^{4-}-P) were determined according to previously reported protocols using Hach reagent kits [17]. Following appropriate sample dilution, T-N and T-P concentrations were measured according to a Hach methods (Methods 10127 and 10072, respectively) with a spectrophotometer (DR 3900, Hach Co., Loveland, USA). Concentrations of NH_4^+-N and PO_3^{4-}-P concentrations in the samples were also determined using Hach methods (Method 10031 and 8114, respectively) after centrifugation and filtration through 0.45 μm-pore size membranes (25HP045AN; Toyo Roshi Kaisya, Ltd., Tokyo, Japan). Microwave digestion was performed before determination of total aluminum (T-Al) concentrations. Each sludge sample (1 mL), concentrated HNO_3 (2 mL), and concentrated HCl (6 mL) were added into digestion vials and samples were then digested on a microwave digestion system (ETHOS TOUCH CONTROL; Milestone General K.K., Kawasaki, Japan). Concentrations of T-Al were determined by an inductively coupled plasma atomic emission spectrometer (ICPE-9000; Shimadzu Corporation, Kyoto, Japan). The methane (CH_4) and carbon dioxide (CO_2) content of the biogas were determined using a gas chromatograph (GC-14B; Shimadzu Corporation, Kyoto, Japan) equipped with a thermal conductivity detector and a 6.0 m × 3.0 mm stainless-steel-packed column (Shincarbon St, Shinwa Chemical Industries, Ltd., Kyoto, Japan). The capillary suction time (CST) of the digested sludge samples was measured using a CST apparatus (Type 304B CST; Triton Electronics Ltd., Essex, England).

3. Results and Discussion

Physical and chemical properties of sludge were shown in Table 1. The TS concentrations of the sludge varied from 2.3 to 4.9 mg/L because of differences between the operational conditions of the wastewater treatment plant and laboratory-scale DMF reactors. The VS/TS ratio of the up-concentrated OM containing PACl was 73%, which was low compared with another substrate sludge due to PACl addition. Compared with other substrate sludge, the T-P and T-Al concentrations of the up-concentrated OM containing PACl were high because of the added PACl in the DMF process. In the DMF reactor without PACl addition, the trans-membrane pressure (TMP) which reflects membrane fouling reached 20 kPa after 100 hours of operation. On the other hand, TMP was below 10 kPa in the DMF reactor with PACl addition (data not shown). This result indicates the PACl addition was effective for mitigating membrane fouling.

Fig. 1 shows the temporal change in the cumulative biogas production from each type of sludge. Each data point represents the average value of two replicates. The RC model was fitted to the experimental data and the solid lines in Fig. 1 were obtained. The RC model was well fitted to the experimental data and the obtained parameters are summarized in Table 2. A long lag time was not observed because of the good acclimatization of the seed sludge to the substrate sludge. The biogas production from up-concentrated OM containing PACl was the highest among the four types of sludge, and the maximum biogas production reached 0.56 L/g-VS_{fed}. The biogas production from up-concentrated OM was comparable to that from mixed sludge. The maximum biogas production from WAS was the lowest among all sludge types because of the sufficient stabilization with long solid retention time in the plant. The maximum biogas production rates from up-concentrated OM, up-concentrated OM containing PACl, and mixed sludge were comparable, between 0.076 and 0.079 L/(g-VS$_{fed}$·d). These results indicate that the up-concentrated OM containing PACl has the highest potential for biogas production. In this study, the mixed sludge contained primary sludge, but the OM obtained from the DMF process did not contain the primary sludge. Therefore, further increase in biogas production could be expected by anaerobic digestion of primary sludge and OM obtained in primary sedimentation basin-DMF process.

Although Chen *et al*. [10] reported that PACl has inhibitory effects on the anaerobic digestion of WAS, no inhibitory effects were observed in this study because of the low PACl dosage (4.3 mg-Al/L) in the DMF reactor. In previous studies about DMF, the range of PACl dosage was between 4 to 15 mg-Al/L [7-9]. In fact, the T-Al concentration (190 mg/L) of up-concentrated OM containing PACl was almost equal to that of the seed sludge (120 mg/L; Table 1). On the contrary, biogas production of the up-concentrated OM was enhanced by PACl addition in the DMF process. Gong *et al*.

[7] also reported the bio-methane potential of OM obtained from membrane-based pre-concentration (i.e., DMF) and reviewed the bio-methane potential of WAS from other studies. However, the anaerobic digestibility of sludge is largely affected by the characteristics of raw wastewater, and there are significant differences in the anaerobic digestibility of sludge among treatment plants. In our study, we successfully conducted a direct comparison of the biogas production from up-concentrated OM, mixed sludge, and WAS obtained from the same full-scale municipal wastewater treatment plant.

There was no significant difference in the methane content in the biogas produced from each sludge (69–73%; Table 3). The VS degradation rate of mixed sludge was the highest (80%) because it contained primary sludge. The VS degradation ratios of up-concentrated OM with and without PACl were 72% and 62%, respectively. The methane yields of up-concentrated OM were higher than those of mixed sludge and WAS, which means that up-concentrated OM obtained from DMF of municipal wastewater has high potential for methane production. The difference in the methane yields was probably come from the difference of constituents (i.e., carbohydrates, protein, and lipid) in the substrates [18]. Further research is needed to confirm this hypothesis. Considering the maximum biogas production and the methane content, the maximum methane production of up-concentrated OM containing PACl was 407 mL-CH_4/g-VS_{fed}, which is slightly larger than previously reported value (367 mL-CH_4/g-VS_{fed}) [7]. Finally, we investigated the dewaterability of digested sludge (i.e., degree of ease in dewatering of sludge) by measuring the CST/TS value which is one of the index parameters of dewaterability of sludge [19]. Improved dewaterability of sludge is one of the advantages, as actual digested sludge is usually dewatered after anaerobic digestion. Although we expected better dewaterability of up-concentrated OM containing PACl because it contained a coagulant, there is no significant decrease in the CST/TS value. This result also suggested that the PACl content was low and the dewaterability was not improved. In a future study, we will scale up a reactor and conduct continuous operation of the reactor treating OM obtained from the DMF process. Investigation of energy efficiency and economic feasibility in an OM recovery process containing the DMF is also needed considering costs for membrane filtration and PACl use.

4. Conclusions

This study investigated the anaerobic digestibility of up-concentrated OM obtained from DMF of municipal wastewater. We compared the biogas production from up-concentrated OM, mixed sludge, and WAS obtained from the same source of municipal wastewater. The maximum biogas production and the methane yield of up-concentrated OM containing PACl were higher than those of up-concentrated OM without PACl, mixed sludge, and WAS. The presence of PACl in the up-concentrated OM did not show an inhibitory effect on the anaerobic digestibility, but instead enhanced the biogas production from up-concentrated OM. These results indicated that the up-concentrated OM obtained from DMF of municipal wastewater has high potential for biogas production. The obtained values in the batch anaerobic digestion experiments will be useful for estimation of energy efficiency and economic feasibility of the DMF process.

Acknowledgements

This research was partly supported by a Grant-in-Aid for Scientific Research (KAKENHI Grant No. 18K13860 and No. 18H05333) from the Japan Society for the Promotion of Science.

References

1. H. Guven, R.K. Dereli, H. Ozgun, M.E. Ersahin, I. Ozturk, Towards sustainable and energy efficient municipal wastewater treatment by up-concentration of organics, Prog. Energy Combust. Sci. 70 (2019) 145-168.

https://doi.org/10.1016/j.pecs.2018.10.002.

2. L. Appels, J. Baeyens, J. Degreve, R. Dewil, Principles and potential of the anaerobic digestion of waste-activated sludge, Prog. Energy Combust. Sci. 34 (2008) 755-781. https://doi.org/10.1016/j.pecs.2008.06.002
3. D. Jenkins, J. Wanner, Activated Sludge - 100 Years and Counting, IWA Publishing, London, 2014.
4. A. Gonzalez, Atwm. Hendriks, J.B. van Lier, M. de Kreuk, Pre-treatments to enhance the biodegradability of waste activated sludge: Elucidating the rate limiting step, Biotechnol. Adv. 36 (2018) 1434-1469. https://doi.org/10.1016/j.biotechadv.2018.06.001
5. S. Hube, M. Eskafi, K.F. Hrafnkelsdottir, B. Bjarnadottir, M.A. Bjarnadottir, S. Axelsdottir, B. Wu, 2020. Direct membrane filtration for wastewater treatment and resource recovery: A review. Sci. Total Environ. 710, 136375. https://doi.org/10.1016/j.scitotenv.2019.136375
6. K. Kimura, D. Honoki, T. Sato, Effective physical cleaning and adequate membrane flux for direct membrane filtration (DMF) of municipal wastewater: Up-concentration of organic matter for efficient energy recovery, Sep. Purif. Technol. 181 (2017) 37-43. https://doi.org/10.1016/j.seppur.2017.03.005.

7. H. Gong, Z.Y. Jin, H. Xu, Q. Yuan, J.E. Zuo, J. Wu, K.J. Wang, Enhanced membrane-based pre-concentration improves wastewater organic matter recovery: Pilot-scale performance and membrane fouling, J. Clean Prod. 206 (2019) 307-314. https://doi.org/10.1016/j.jclepro.2018.09.209.

8. Y.X. Zhao, P. Li, R.H. Li, X.Y. Li, Direct filtration for the treatment of the coagulated domestic sewage using flat-sheet ceramic membranes, Chemosphere. 223 (2019) 383-390. https://doi.org/10.1016/j.chemosphere.2019.02.055.

9. Z.Y. Jin, H. Gong, H. Temmink, H.F. Nie, J. Wu, J.E. Zuo, K.J. Wang, Efficient sewage pre-concentration with combined coagulation microfiltration for organic matter recovery, Chem. Eng. J. 292 (2016) 130-138. https://doi.org/10.1016/j.cej.2016.02.024.

10. Y.N. Chen, Y.X. Wu, D.B. Wang, H.L. Li, Q.L. Wang, Y.W. Liu, L. Peng, Q. Yang, X.M. Li, G.M. Zeng, Y.R. Chen, Understanding the mechanisms of how poly aluminium chloride inhibits short-chain fatty acids production from anaerobic fermentation of waste activated sludge, Chem. Eng. J. 334 (2018) 1351-1360. https://doi.org/10.1016/j.cej.2017.11.064.

11. Z.Y. Yu, X.H. Wen, M.L. Xu, M. Qi, X. Huang, Anaerobic digestibility of the waste activated sludge discharged from large-scale membrane bioreactors, Bioresour.

Technol. 126 (2012) 358-361. https://doi.org/10.1016/j.biortech.2012.09.024.

12. N. Sahu, A. Sharma, P. Mishra, B. Chandrashekhar, G. Sharma, A. Kapley, R.A. Pandey, Evaluation of biogas production potential of kitchen waste in the presence of spices, Waste Manage. 70 (2017) 236-246. https://doi.org/10.1016/j.wasman.2017.08.045.
13. V. Wyman, A. Serrano, F.G. Fermoso, D.K.V. Gomez, Trace elements effect on hydrolytic stage towards biogas production of model lignocellulosic substrates, J. Environ. Manage. 234 (2019) 320-325. https://doi.org/10.1016/j.jenvman.2019.01.015.
14. A. Donoso-Bravo, S.I. Perez-Elvira, F. Fdz-Polanco, Application of simplified models for anaerobic biodegradability tests. Evaluation of pre-treatment processes, Chem. Eng. J. 160 (2010) 607-614. https://doi.org/10.1016/j.cej.2010.03.082.
15. A.Y. Cetinkaya, K. Yetilmezsoy, Evaluation of anaerobic biodegradability potential and comparative kinetics of different agro-industrial substrates using a new hybrid computational coding scheme, J. Clean Prod. 238 (2019) 117921. https://doi.org/10.1016/j.jclepro.2019.117921.
16. APHA, AWWA, WEF, Standard Methods for the Examination of Water and Wastewater, 23rd ed., American Public Health Association, Washington, DC. 2017.

17. A. Hafuka, R. Mashiko, R. Odashima, H. Yamamura, H. Satoh, Y. Watanabe, Digestion performance and contributions of organic and inorganic fouling in an anaerobic membrane bioreactor treating waste activated sludge, Bioresour. Technol. 272 (2019) 63-69. https://doi.org/10.1016/j.biortech.2018.09.147.

18. C.P.C. Bong, L.Y. Lim, C.T. Lee, J.J. Klemes, C.S. Ho, W.S. Ho, The characterisation and treatment of food waste for improvement of biogas production during anaerobic digestion - A review, J. Clean Prod. 172 (2018) 1545-1558. https://doi.org/10.1016/j.jclepro.2017.10.199.

19. X.Y. Li, Y.Z. Peng, Y.L. He, S.Y. Wang, S.Y. Guo, L.K. Li, Anaerobic stabilization of waste activated sludge at different temperatures and solid retention times: Evaluation by sludge reduction, soluble chemical oxygen demand release and dehydration capability, Bioresour. Technol. 227 (2017) 398-403. https://doi.org/10.1016/j.biortech.2016.12.069

Table and figure captions

Table 1. Characteristics of sludge used for the batch anaerobic digestion experiments.

Table 2. Parameters obtained with the RC model.

Table 3. Methane content, VS degradation ratio, methane yield, and CST/TS value after

30 days digestion in the batch experiments.

Fig. 1. Cumulative biogas yield from each type of sludge in the batch experiments. The solid lines are predictions calculated using the RC model.

Type of sludge	pH (-)	TS (g/L)	VS (g/L)	VS/TS (%)	T-N (mg-N/L)	NH_4^+-N (mg-N/L)	T-P (mg-P/L)	PO_4^{3-}-P (mg-P/L)	T-Al (mg/L)
Seed sludge	7.4	11.4 ± 0.1	8.3 ± 0.0	73 ± 0	1700 ± 0.0	1030.0 ± 5.0	410 ± 13	290 ± 15	120 ± 0.65
Up-concentrated OM	7.0	3.4 ± 0.1	2.8 ± 0.1	84 ± 0	240 ± 5.0	23.5 ± 0.0	58 ± 4.0	5.1 ± 1.9	7.3 ± 0.06
Up-concentrated OM containing PACl	6.9	3.7 ± 0.2	2.7 ± 0.1	73 ± 0	210 ± 13	18.5 ± 0.5	96 ± 7.5	3.3 ± 1.7	190 ± 0.91
Mixed sludge	6.9	2.3 ± 0.0	1.9 ± 0.0	84 ± 0	150 ± 2.5	23.0 ± 0.0	34 ± 0.8	7.4 ± 1.6	8.8 ± 0.36
WAS	6.7	4.9 ± 0.0	3.9 ± 0.0	80 ± 0	330 ± 30	5.0 ± 3.0	88 ± 4.0	16.3 ± 7.7	22 ± 0.67

Note: TS = total solids; VS = volatile solids; T-N = total nitrogen; NH_4^+-N = ammonium; T-P = total phosphorus; PO_4^{3-}—P = orthophosphate; T-Al = total aluminum.

Type of sludge	P (L-Biogas/g-VS$_{fed}$)	R_m (L-Biogas/(g-VS$_{fed}$·d))	λ (d)	r^2 (-)	RSS (-)
Up-concentrated OM	0.47	0.079	0.2	0.998	8.62×10^{-4}
Up-concentrated OM containing PACl	0.56	0.076	0.2	0.999	8.73×10^{-4}
Mixed sludge	0.48	0.077	0.2	0.997	1.41×10^{-3}
WAS	0.35	0.062	0	0.991	1.97×10^{-3}

Note: P is the maximum biogas production. R_m is maximum biogas production rate. λ is lag time. RSS is residual sum of squares.

Type of sludge	Methane content (%)	VS degradation ratio (%)	Methane yield ($L\text{-}CH_4/g\text{-}VS_{deg}$)	CST/TS ((s·L)/g)
Up-concentrated OM	71.2 ± 0.1	62 ± 5	0.54 ± 0.05	17 ± 1
Up-concentrated OM containing PACl	72.7 ± 0.2	72 ± 7	0.56 ± 0.06	18 ± 0
Mixed sludge	71.9 ± 0.4	80 ± 3	0.43 ± 0.01	13 ± 0
WAS	69.0 ± 0.0	53 ± 12	0.50 ± 0.13	21 ± 3

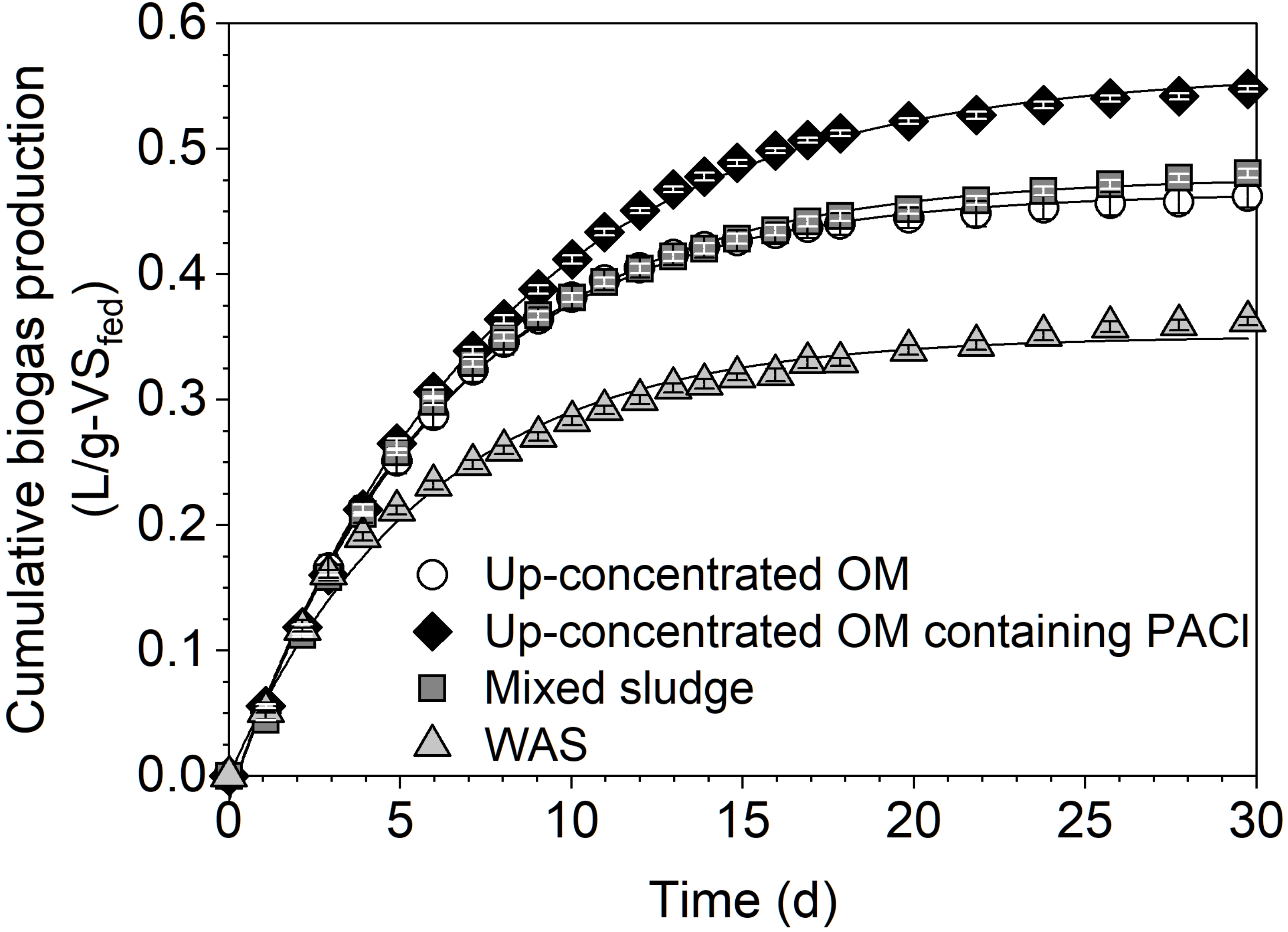
Cumulative biogas production
(L/g-VS_{fed})
0.0
0.1
0.2
0.3
0.4
0.5
0.6
0
5
10
15
20
25
30
Time (d)
○ Up-concentrated OM
◆ Up-concentrated OM containing PACl
■ Mixed sludge
△ WAS

Table S1. Operational condition of each DMF reactor.

	DMF reactor without PACl addition		DMF reactor with PACl addition	
	First tank	Second tank	First tank	Second tank
Working volume (L)	4.6	4.6	4.6	4.6
HRT (h)	1.5	4.9	1.1	4.2
PACl addition (mg-Al/L)	-	-	4.3	-
Filtration flux (LMH)	15.6	8.8	24.1	9.7
Intermittent filtration (filtration (min)/pause (min))	4/1	4/1	12/3	12/3
Intermittent aeration (aeration (min)/pause (min))	1/4	1/4	1/14	1/14
Granule volume (vol%)	30	30	30	30
Frequency of backwashing with tap water (backwashing (s)/operating time (h))	60/1.0	50/0.5	50/0.5	50/0.5
Frequency of backwashing with sodium hypochlorite (backwashing (s)/operating time (d))	120/2	120/2	-	-