

Investigating the Effect of Commercial and Synthesized Fe⁰ particles on Methane Production Through the Anaerobic Digestion of Waste Sludge

Eljamal, Ramadan

Water and Environmental Engineering Laboratory, Interdisciplinary Graduate School of Engineering Science, Kyushu University

Maamoun, Ibrahim

Water and Environmental Engineering Laboratory, Interdisciplinary Graduate School of Engineering Science, Kyushu University

Bensaida, Khaoula

Water and Environmental Engineering Laboratory, Interdisciplinary Graduate School of Engineering Science, Kyushu University

Sugihara, Yuji

Environmental Fluid Science Laboratory, Interdisciplinary Graduate School of Engineering Science, Kyushu University

他

<https://doi.org/10.5109/4738586>

出版情報 : Proceedings of International Exchange and Innovation Conference on Engineering & Sciences (IEICES). 7, pp.184-189, 2021-10-21. Interdisciplinary Graduate School of Engineering Sciences, Kyushu University

バージョン :

権利関係 :



Investigating the Effect of Commercial and Synthesized Fe⁰ particles on Methane Production Through the Anaerobic Digestion of Waste Sludge

Ramadan Eljamal¹, Ibrahim Maamoun¹, Khoula Bensaida¹, Yuji Sugihara², Osama Eljamal^{1*}

¹Water and Environmental Engineering Laboratory, Interdisciplinary Graduate School of Engineering Science, Kyushu University, Fukuoka, Japan

²Environmental Fluid Science Laboratory, Interdisciplinary Graduate School of Engineering Science, Kyushu University, Fukuoka, Japan

*Corresponding author email: osama-eljamal@kyudai.jp

Abstract: Anaerobic digestion (AD) is a well-established technology that converts organic wastes to produce renewable energy as methane gas. However, the low conversion efficiency of organic matter to methane gas is the main challenge for the practical application of this technology. Therefore, this article aims to demonstrate the effect of the addition of commercial and synthesized Fe⁰ particles on methane gas production during the AD of waste sludge. Two set of experiments were conducted using two different waste sludges collected at different times from Fukuoka wastewater treatment plant in Japan. Different concentrations of commercial and synthesized iron particles were used in the anaerobic digesters ranged from 1 to 500 mg/L. The results should that the addition of the synthesized/Fe⁰ particles to the anaerobic digesters improved methane production by 16% and 5% at using the first and second waste sludge, respectively. On the other hand, the addition of the commercial iron particles did not improve the production of methane gas.

Keywords: Anaerobic digestion; Methane production; Synthesized iron particles; Commercial iron particles.

1. INTRODUCTION

The production of environmental organic wastes such as waste sludge, food waste, agricultural waste, and many other wastes is highly increasing, as it linked to human growth and activities. Waste sludge is one of the most discharged organic wastes to the environment. The disposal of waste sludge during the treatment of municipal wastewater costs as high as 60 % of the total operating cost of wastewater treatment plant [1]. Although different disposal routes are possible, anaerobic digestion is a promising track due to its low operating cost and its ability to transform organic matter into biogas (60–70 vol% as methane gas) [2]. Anaerobic digestion decreases the production amount of waste sludge which contributes to reduce the operating cost of wastewater treatment plant. Furthermore, when treating the municipal wastewater, anaerobic digestion significantly decreases the concentration of contaminants such as ammonia [3], chemical oxygen demand (COD) [4], sulfate [5], and phosphate [6].

Despite the numerous advantages of anaerobic digestion in producing energy and treatment wastewater, the practical application of the anaerobic digestion technology is still suffering from the low conversion efficiency of organic matter to methane gas. Therefore, many attempts and techniques have been conducted in the literature to improve the process, including bioaugmentation [7], co-digestion [8], pre-treatment of waste [9], and addition of trace elements [10]. Among of these techniques, the addition of trace elements was the most used approach to enhance the anaerobic digestion process.

The extensive use of the trace elements such as iron and bimetallic nanoparticles is attributed to the availability and cheapness of such trace elements.

Iron nanoparticles have been widely used in the environmental applications, including water treatment

and energy generation [11-16]. The low synthesis cost and the effectiveness of iron nanoparticles are the main reasons behind their extensive use in the environmental treatments.

Up until now, the effect of iron particles on the performance of anaerobic digestion is still unclear. Therefore, the main objective of this article is to investigate the effect of the commercial and synthesized iron particles on the performance of the anaerobic digestion process. The effect of iron particles on biogas and methane generation was examined using two different waste sludges. Different concentrations of commercial and synthesized iron particles were considered to acquire the optimum concentration.

2. MATERIAL AND METHODS

2.1 sludge collection and characterization

Two waste sludges were collected from Fukuoka wastewater treatment plant at different times from the pre-stage of anaerobic digesters. The second sludge was collected two months after the first sludge collection. These waste sludges were characterized as shown in Table 1 for Chemical Oxygen Demand (COD), Total solids (TS), Total Volatile Solids (TVS), Oxidation Reduction Potential (ORP), and pH. TS, TVS, and COD were analyzed before and after the fermentation process following the standard methods [17].

2.2 Iron particles

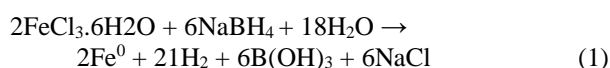
Two types of iron were used and added in the anaerobic digesters. First type is the commercial iron particles shipped from NANO IRON, s.r.o., Co., Stefanikova 116, 664 61 Rajhrad, Czech Republic.

Based on the specifications provided by the company, the commercial iron particles have an average particles size of 50 nm and a surface area of 20-25 m²/g.

Table 1 Characterization of two waste sludges collected at different times and used in the anaerobic digestion process

	pH	ORP (mV)	COD (mg/L)	TS (g/L)	TVS (g/L)	TVS/TS (%)
Sludge (1)	7.1	-38	5850	4.4	3.6	82
Sludge (2)	6.9	-24	2975	2.5	2.0	80

The second type of iron particles were prepared at water and environmental engineering laboratory, Kyushu University, Japan. The particles were synthesized following the reduction process of $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ by the addition of NaBH_4 as a strong reductant as described in Equation 1.



Briefly, the reactants of ferric chloride hexahydrate ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$, 25 g/L) and sodium borohydride (NaBH_4 , 22 g/L) were prepared using deoxygenated deionized water (DDIW) and purged with nitrogen for 10 min. Then, sodium borohydride solution was added dropwise using a peristaltic pump at a flow rate of 20 mL/min^{-1} into four neck flask reactor containing ferric chloride solution. The flask reactor was placed in a water bath set at 30°C and N_2 gas continuously bubbled during the synthesis to avoid the oxidation process of the synthesized iron particles. Besides, the reaction was implemented under continuous mixing of 400 rpm. Once sodium borohydride completely added the reaction was lasted for 5 min as an aging time. Finally, the solid particles were separated using the vacuum filtration system and then washed using deoxygenated deionized water.

2. 3 Biochemical methane potential test (BMPT)

BMPT was implemented to evaluate the production rate of methane using the waste sludge as a feedstock. The commercial and the synthesized iron particles were added separately in the biodigesters at different concentrations ranged from 1 to 500 mg/L. The BMPT was carried out using two waste sludges collected at different times with adding the same iron particles.

All BMPTs were performed using 200 mL serum vials with a working volume of 150 mL of waste sludge. After filling the bioreactors with waste sludge, they were bubbled with nitrogen for 2 min to eliminate the effect of oxygen and established the anaerobic conditions. The bioreactors were then sealed with rubber stoppers and aluminum caps and placed in the incubator for fermentation at $38 \pm 1^\circ\text{C}$ with constant agitation of 150 rpm. All groups were performed in triplicates to confirm the reproducibility of methane gas under the same conditions, and the results represent the average of three measurements. Regularly, biogas volume was determined using the water displacement method. Gas chromatograph (GC, GL Sciences Inc., Japan) equipped with a thermal conductivity detector (TCD) was used to analyze the composition of biogas. In detail, 1 mL of the biogas was taken from the headspace of bioreactor and immediately injected into the injector unit of GC for the separation process using a Hamilton gas-tight syringe (GL Sciences Inc., Japan). The temperatures of the detector, injector, and oven were 60°C , 80°C , and 60°C , respectively.

The temperature of the injector was determined to be enough for making flash vaporization for the injected sample. But the temperatures of column and detector were determined to be enough for separating the gas mixture at the column stage and avoiding any condensation for the separated sample in the detector unit, respectively. The argon gas was used as a mobile phase to push and carry the analyte sample through the column at a flow rate of 30 mL/min .

3.RESULTS AND DISCUSSION

3.1 Characterization of iron particles

Based on the specifications provided by the NANO IRON, s.r.o., Co., the commercial iron particles have an average particles size of 50 nm and a surface area of 20-25 m^2/g . On the other hand, the size of the synthesized iron particles was analyzed using Laser Diffraction Particle Size Analyzer, SALD-2300. Both particle distribution and average particle size are depicted in Fig. 2. The results showed that the synthesized iron particles had an average particle size of 46 nm. These results were in good agreement with the obtained TEM images as shown in Fig. 1a and b.

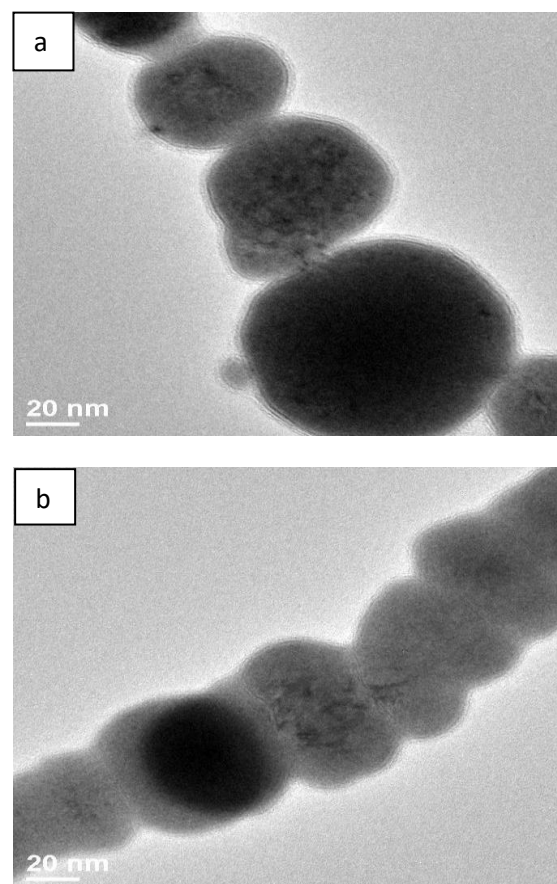


Fig. 1 a and b shows the morphological structure and size of the synthesized Fe^0 particles.

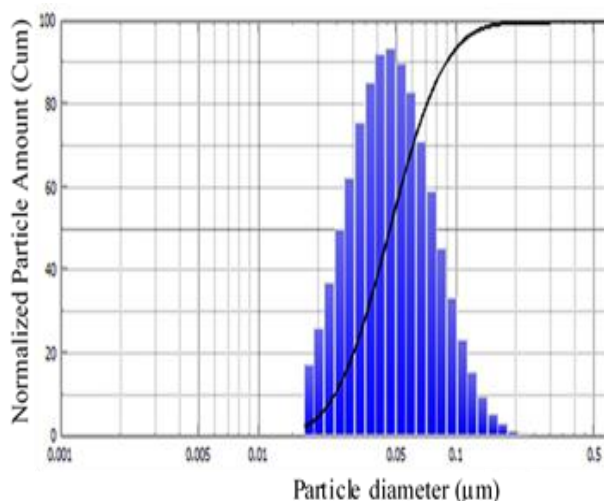


Fig.2.Size distribution and average particle size of the synthesized/ Fe^0 obtained using laser diffraction particle size analyzer.

3.2. Effect of synthesized/ Fe^0 particles on biogas and methane production using waste sludge 1

The production curves of BMPT concerning the effect of different dosages of the synthesized/ Fe^0 on the performance of anaerobic digestion using waste sludge 1 are depicted in Fig. 3. Generally, biogas and methane curves had similar trend even at using different dosages of the synthesized/ Fe^0 . However, a slight improvement in biogas and methane production was observed starting from the second day of the anaerobic digestion in the reactors of the synthesized/ Fe^0 . As the dosage of the synthesized/ Fe^0 increased from 1 to 20 mg/L, the biogas and methane production increased as well. Methane production was improved by 16 % at the dosage of 20 mg/L of the synthesized/ Fe^0 compared to the control.

This improvement in biogas and methane production could be attributed to the role of the synthesized/ Fe^0 in improving bacterial growth and optimizing the pH of the digesters [18, 19]. Furthermore, the released electrons by the corrosion reaction of the synthesized/ Fe^0 play an important role in improving bacterial activity as well as increasing methane content by further conversion of CO_2 .

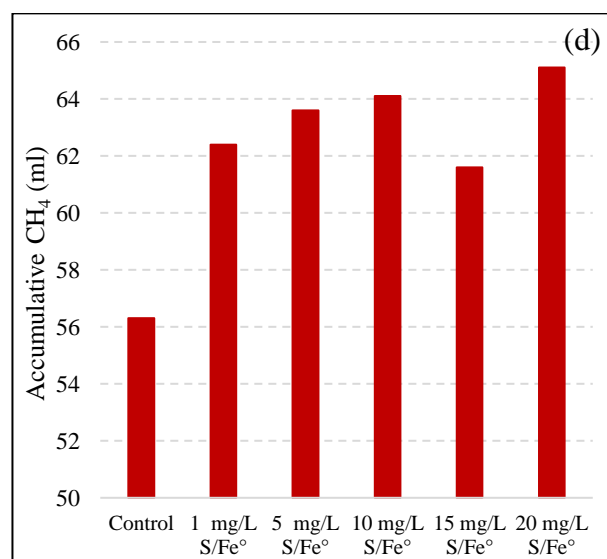
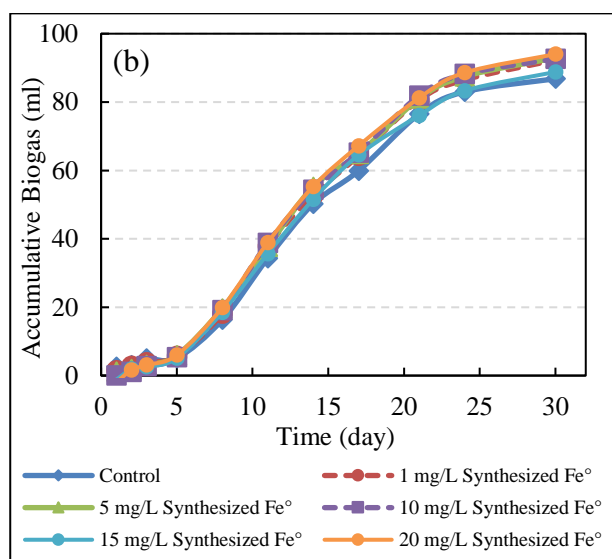
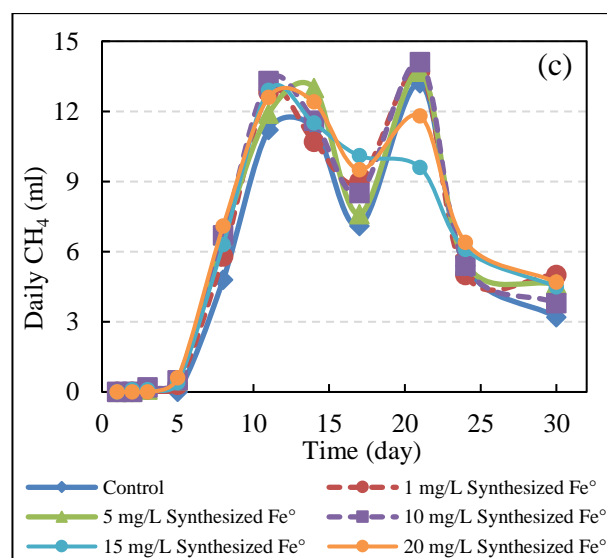
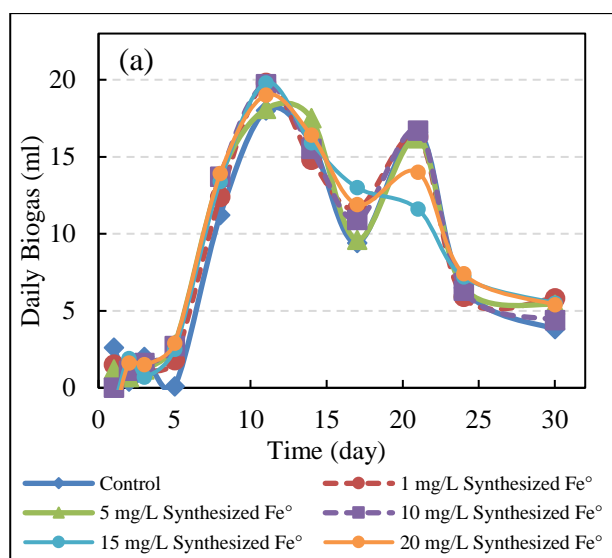


Fig.3. BMPT curves showing the effect of different dosages of the synthesized/ Fe^0 (S/ Fe^0) on the performance of anaerobic digestion using waste sludge 1 ; a) daily biogas production, b) accumulative biogas, c) daily methane production, d) accumulative methane.

3.3 Effect of synthesized/ Fe^0 particles on biogas and methane production using waste sludge 2

The effect of the synthesized/ Fe^0 on biogas and methane production was investigated using waste sludge 2, which had different characteristics than waste sludge 1 as shown in Table 1. This BMPT was implemented to confirm the positive effect of the synthesized/ Fe^0 on biogas and methane production as observed using waste sludge 1. In this BMPS, different dosages of the synthesized/ Fe^0 were used ranged from 10 to 500 mg/L, to clearly acquire the optimum dosage based on the highest biogas and methane production. The results of biogas and methane production curves are presented in Fig. 4. In all cases, biogas and methane production curves had similar trend as the control. However, a slight enhancement in biogas and methane production was observed from the second day of operation. In this experiment of waste sludge 2, methane production enhanced by 5% at the optimum dosage of 50 mg/L of the synthesized/ Fe^0 , which is much lower that enhancement obtained from waste sludge 1. From these investigations on the effect of the synthesized/ Fe^0 on methane production using two different waste sludges

collected at different times and had different characteristics, several important points can be highlighted concerning the effect of the synthesized/ Fe^0 on methane production. First, despite the low enhancement rate of methane production by the addition of the synthesized/ Fe^0 using two different waste sludge, however the positive effect of the synthesized/ Fe^0 on the performance of the anaerobic digestion could be confirmed using two waste sludges. Second, the effect of the synthesized/ Fe^0 on biogas and methane production is largely governed by the organic content in the reactors as well as the abundance of the fermentative bacteria. Based on the results obtained from this work, the high organic matter (TS and TVS) content in anaerobic digesters would increase the positive effect of the synthesized/ Fe^0 on methane gas production. Therefore, some important measures must be taken in the account when applying the synthesized/ Fe^0 in the practical application of anaerobic digestion. Among these measures, the addition of the synthesized/ Fe^0 in the anaerobic digesters must be based on the organic matter content (TS and TVS), and the abundance of the fermentative bacteria in the raw sludge

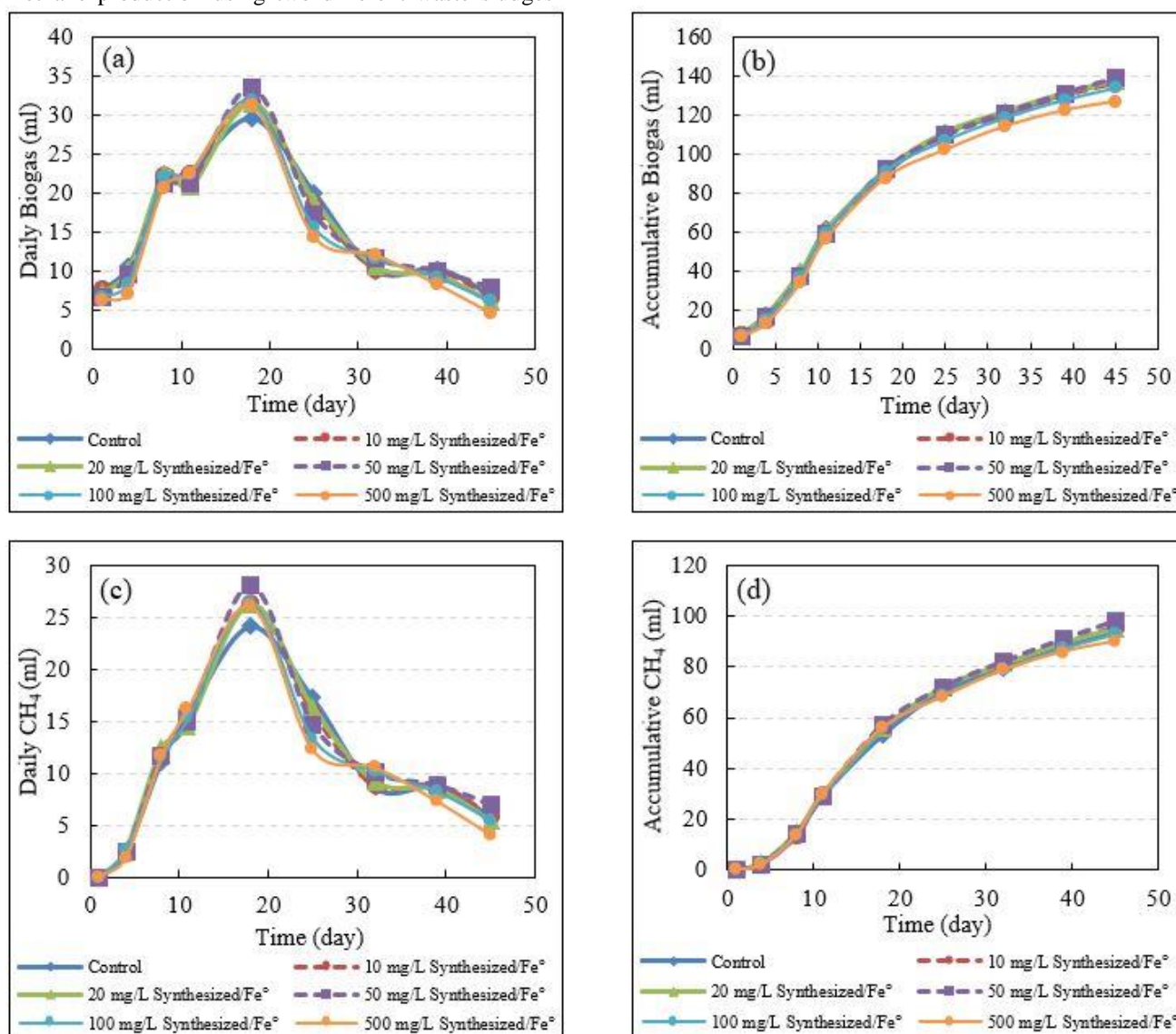


Fig. 4. BMPT curves showing the effect of different dosages of the synthesized/ Fe^0 (S/Fe^0) on the performance of anaerobic digestion using waste sludge 2 ; a) daily biogas production, b) accumulative biogas, c) daily methane production, and d) accumulative methane.

3.4 Effect of commercial/ Fe^0 particles on biogas and methane production using waste sludge 1

In this research, the effect of the commercial/ Fe^0 towards the performance of the anaerobic digestion process was investigated as well. The BMPT was performed using waste sludge 1 at different dosages of the commercial/ Fe^0 ranged from 1 to 20 mg/L. The results of biogas and methane production curves are depicted in Fig. 5. The results showed that the addition of the commercial/ Fe^0 did not improve biogas and methane production even at various concentrations of commercial/ Fe^0 . In comparison, the performance of the synthesized/ Fe^0 in improving methane production was higher than that the performance of the commercial/ Fe^0 in this study. This could be attributed to the freshness and

the high surface energy of the freshly synthesized/ Fe^0 , generating more electrons in the system to stimulate bacterial activity and increase the conversion of CO_2 to methane gas during the process. In the literature, the effect of the commercial/ Fe^0 on methane production varied from study to study. For instance, the results of this study are totally contradicted with that results obtained by Yinghong Feng et al. [18]. The difference in the results could be attributed to the specifications of the commercial/ Fe^0 used in each study as well as the characteristics of the used waste sludge, including organic matter content and the abundance of fermentative bacteria in each sludge.

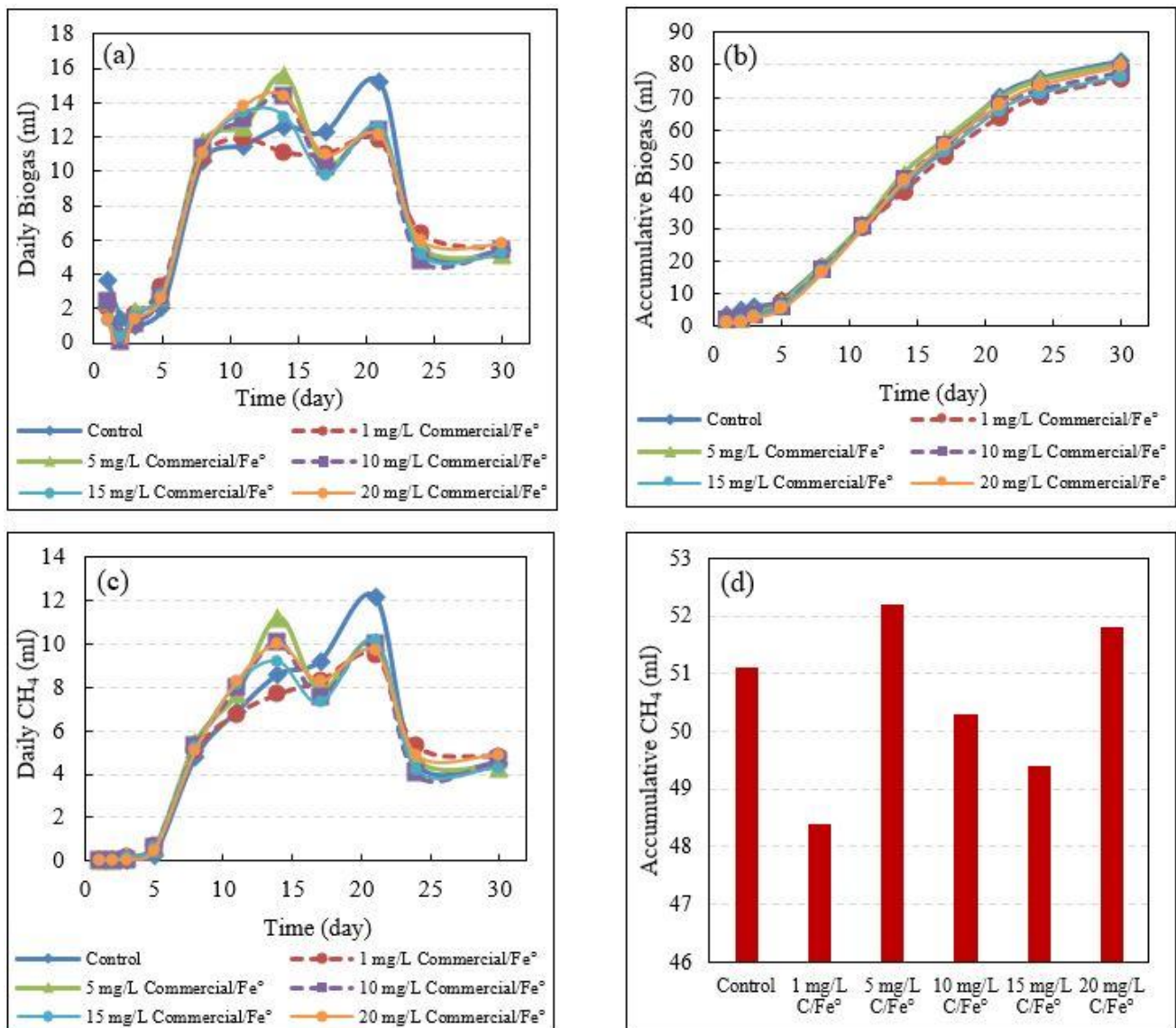


Fig. 5. BMPT curves showing the effect of different dosages of the commercial/ Fe^0 (C/Fe^0) on the performance of anaerobic digestion using waste sludge 1 ; a) daily biogas production, b) accumulative biogas, c) daily methane production, and d) accumulative methane.

4. CONCLUSIONS

In this research, the effect of the addition of commercial and synthesized/Fe⁰ particles on biogas and methane production during the anaerobic digestion of waste sludge was investigated. Two set of experiments were conducted using two different waste sludges collected at different times from Fukuoka wastewater treatment plant in Japan. Different concentrations of commercial and synthesized iron particles were used in the anaerobic digesters ranged from 1 to 500 mg/L. The results should that the addition of the synthesized/Fe⁰ particles to the anaerobic digesters improved methane production by 16% and 5% at using the first and second waste sludge, respectively. Despite the low enhancement rate of methane production by the addition of the synthesized/Fe⁰ using two different waste sludges, the positive effect of the synthesized/Fe⁰ on the performance of the anaerobic digestion could be confirmed using two waste sludges. The effect of the synthesized/Fe⁰ on biogas and methane production is largely governed by the organic matter content in the reactors. On the other hand, the addition of the commercial iron particles did not improve the production of methane gas.

ACKNOWLEDGMENTS

This work was supported by JSPS KAKENHI Grant Number JP21K12311.

REFERENCES

- [1] R. Eljamal, I. Kahraman, O. Eljamal, I.P. Thompson, I. Maamoun, G. Yilmaz, Impact of nZVI on the formation of aerobic granules, bacterial growth and nutrient removal using aerobic sequencing batch reactor, *Environmental Technology & Innovation* 19 (2020) 100911.
- [2] L. Appels, J. Baeyens, J. Degreè, R. Dewil, Principles and potential of the anaerobic digestion of waste-activated sludge, *Progress in energy and combustion science* 34 (2008) 755-781.
- [3] A. Palakodeti, S. Azman, B. Rossi, R. Dewil, L. Appels, A critical review of ammonia recovery from anaerobic digestate of organic wastes via stripping, *Renewable and Sustainable Energy Reviews* 143 (2021) 110903.
- [4] T.W. Amen, O. Eljamal, A.M. Khalil, Y. Sugihara, N. Matsunaga, Methane yield enhancement by the addition of new novel of iron and copper-iron bimetallic nanoparticles, *Chemical Engineering and Processing-Process Intensification* 130 (2018) 253-261.
- [5] T.W. Amen, O. Eljamal, A.M. Khalil, N. Matsunaga, Evaluation of sulfate-containing sludge stabilization and the alleviation of methanogenesis inhibition at mesophilic temperature, *Journal of Water Process Engineering* 25 (2018) 212-221.
- [6] C. Keating, J.P. Chin, D. Hughes, P. Manesiotis, D. Cysneiros, T. Mahony, C.J. Smith, J.W. McGrath, V. O'Flaherty, Biological phosphorus removal during high-rate, low-temperature, anaerobic digestion of wastewater, *Frontiers in microbiology* 7 (2016) 226.
- [7] J. Jiang, L. Li, Y. Li, Y. He, C. Wang, Y. Sun, Bioaugmentation to enhance anaerobic digestion of food waste: Dosage, frequency and economic analysis, *Bioresource technology* 307 (2020) 123256.
- [8] X. Fonoll, S. Astals, J. Dosta, J. Mata-Alvarez, Anaerobic co-digestion of sewage sludge and fruit wastes: evaluation of the transitory states when the co-substrate is changed, *Chemical Engineering Journal* 262 (2015) 1268-1274.
- [9] J. Ariunbaatar, A. Panico, G. Esposito, F. Pirozzi, P.N. Lens, Pretreatment methods to enhance anaerobic digestion of organic solid waste, *Applied energy* 123 (2014) 143-156.
- [10] Y.Y. Choong, I. Norli, A.Z. Abdullah, M.F. Yhaya, Impacts of trace element supplementation on the performance of anaerobic digestion process: A critical review, *Bioresource technology* 209 (2016) 369-379.
- [11] R. Eljamal, O. Eljamal, I. Maamoun, G. Yilmaz, Y. Sugihara, Enhancing the characteristics and reactivity of nZVI: Polymers effect and mechanisms, *Journal of Molecular Liquids* 315 (2020) 113714.
- [12] S. Takami, O. Eljamal, A.M. Khalil, R. Eljamal, N. Matsunaga, Development of continuous system based on nanoscale zero valent iron particles for phosphorus removal, *Journal of JSCE* 7 (2019) 30-42.
- [13] A. Khalil, Treatment and regeneration of nano-scale zero-valent iron spent in water remediation, 2017.
- [14] K. Bensaida, R. Eljamal, Y. Sugihara, O. Eljamal, The impact of iron bimetallic nanoparticles on bulk microbial growth in wastewater, *Journal of Water Process Engineering* 40 (2021) 101825.
- [15] R. Eljamal, Upgrading of Aerobic Sequencing Batch Reactor System with Adding Nanoscale Zero Valent Iron for Wastewater Treatment, *School of Engineering Sciences, Kyushu University*, 2019.
- [16] O. Eljamal, A.M. Khalil, N. Matsunaga, Experimental and modeling column study of phosphorus removal by permeable reactive materials, *International Journal of Environmental and Agriculture Research (IJOEAR)* 3 (2017) 62-70.
- [17] A. American Public Health, A. American Water Works, F. Water Environment, Standard methods for the examination of water and wastewater, APHA-AWWA-WEF, Washington, D.C., 1998.
- [18] Y. Feng, Y. Zhang, X. Quan, S. Chen, Enhanced anaerobic digestion of waste activated sludge digestion by the addition of zero valent iron, *Water research* 52 (2014) 242-250.
- [19] J. Zhou, X. You, B. Niu, X. Yang, L. Gong, Y. Zhou, J. Wang, H. Zhang, Enhancement of methanogenic activity in anaerobic digestion of high solids sludge by nano zero-valent iron, *Science of the Total Environment* 703 (2020) 135532.