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研究課題名（和文）Innovative Nano-biotechnology for Hydrogen and Methane Co-Production with Zero CO2 Emission through a Novel Design of Three-Stage Continuous Flow Anaerobic Digestion System

研究課題名（英文）Innovative Nano-biotechnology for Hydrogen and Methane Co-Production with Zero CO2 Emission through a Novel Design of Three-Stage Continuous Flow Anaerobic Digestion System

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交付決定額（研究期間全体）：（直接経費）　　3,100,000円

研究成果の概要（和文）：業績がRenewable and Sustainable Energy Reviews（インパクトファクター15.9）に掲載されました： 水酸化マグネシウムでコーティングした鉄ナノ粒子を用いて、廃棄物汚泥からのメタン生成を改善する新しい方法。
2本目の論文のタイトルは 廃棄物汚泥の嫌気性消化によるバイオガス生産に対する鉄ナノ粒子とアロエベラバイオマス添加剤の卓越した貢献がEnergy誌に受理された（インパクトファクター9.0）。

研究成果の学術的意義や社会的意義
Scientific: understanding of applying iron-based bimetallic nanoparticles to a continuous flow Anaerobic Digestion system has been clarified.
Social: H2 and CH4 co-production and utilizing CO2 have a double beneficial to both of environment and economy by increasing the efficiency of energy recovery

研究成果の概要（英文）：Achievements have been published in Renewable and Sustainable Energy Reviews (impact factor of 15.9), title: A novel method to improve methane generation from waste sludge using iron nanoparticles coated with magnesium hydroxide.
The second paper, title: Exceptional contribution of iron nanoparticle and Aloe vera biomass additives to biogas production from anaerobic digestion of waste sludge has been accepted by Energy (impact factor of 9.0)

研究分野：Environmental

キーワード：Anaerobic Digestion　Biogas production　Methane generation　Coated nanoparticles　Waste sludge　Nano-biotechnology　bimetallic nanoparticles　zero CO2 emission

Innovative Nano-biotechnology for Hydrogen and Methane Co-Production with Zero CO_2 Emission through a Novel Design of Three-Stage Continuous Flow Anaerobic Digestion System

１．研究開始当初の背景

Anaerobic Digestion (AD): The AD process consists of four main steps: (i) Hydrolysis step (ii) Acidogenesis step (iii) Acetogenesis step (iv) Methanogenesis step. The major challenges that the AD process is facing is that each step requires different conditions to achieve optimal operation. Therefore, the process is separated into two stages (R1 & R2) as shown in **Figure 1**; the first for hydrolysis/acidogenesis and the second for acetogenesis/methanogenesis. This will lead to a solid control and optimal operation. A novel third stage (R3) is added as a microalgal/photobioreactor in order to utilize the CO_2 stream from the previous stages in biomass recirculation.

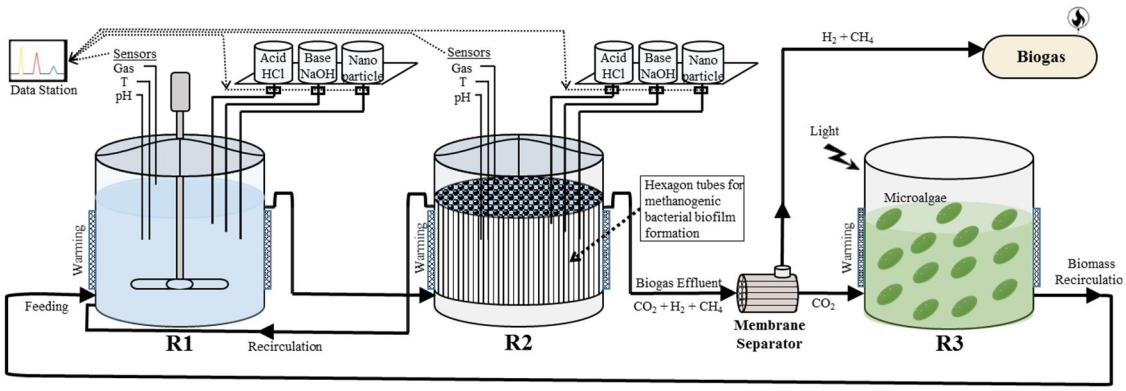

Figure 1*. Design of three-stage anaerobic digestion system, Reactor 1 (R1): Hydrolysis/Acidogenesis reactor, Reactor 2 (R2): Acetogenesis/Methanogenesis reactor, and Reactor 3 (R3): Microalgal/Photobioreactor.*

Problem Statement: The limitation of fossil fuel, as one of the main energy sources, includes unstable geopolitical situation in the Middle East, and carbon emission that causes the global warming and climate change. On the other hand, the low efficiency of the conventional methods that convert organic waste into biogas energy requires more efforts to achieve higher efficiency.

Solution: Alternative technologies to fossil fuels such as clean and renewable energy are being under consideration by several researchers worldwide. One of the available technologies is anaerobic digestion (AD) which is a biological process that converts the organic waste into biogas energy.

２．研究の目的

The purpose of this research proposal is to develop an innovative nano-biotechnology for hydrogen (H_2) and methane (CH_4) co-production, with zero CO_2 emission, from organic waste through a new design of the three-stage continuous flow AD system enhanced by bimetallic nanoparticles.

３．研究の方法

The following methodology applied to this research:

1. *Reactor experiments*: The setup of the three-stage continuous flow AD system will be

implemented as shown in **Figure 1** with 3000-mL Plexiglas reactors for R1, R2 and R3. The system will be operated by a programmable controller operating system which allows to control all conditions continuously along the running time of experiments, that helps in adjusting and optimizing the operation variables and conditions for high energy recovery from the organic waste. Liquid and gas samples will be taken periodically for more detailed analysis to evaluate the system performance.

2. *Synthesis of NZVI-based bimetallic nanoparticles*: The nanoparticles will be synthesized in accordance with Eljamal et al. [6] based on chemical reduction of $FeCl_3 \cdot 6H_2O$ by $NaBH_4$. The second metal (Pd, Zn, Cr, Cu, Co, Cd, Ag, Mn and Ni) also will be added into the solution of NZVI to form different types of bimetallic nanoparticles.
3. *Characterization of nanoparticles*: Synthesized nanoparticles will be characterized before and after use in reactors by Transmission and Scanning Electron Microscopies (TEM & SEM) coupled with energy dispersive X-ray spectroscopy for morphological investigation, X-Ray Diffractometer (XRD) for crystallinity examination, and laser analyzer for particle size measurements.
4. *Organic waste*: 3 different organic wastes will be used separately or combined in co-digestion, **I.** Activated sludge: taken from Mikasagawa wastewater treatment center in Fukuoka, **II.** Food waste: taken from canteen of Kyushu university and **III.** Recirculated algae biomass.
5. *Analytical methods*: The chemical element concentrations will be analyzed by spectrophotometer and/or inductively coupled plasma mass spectrometry techniques, gas samples will be analyzed by gas chromatography, bacterial growth rate will be estimated by colony forming unit (CFU).

４．研究成果

Biochemical methane potential (BMP) tests have been conducted to optimize the conditions of biogas and methane production and applied to a semicontinuous operation system.

The results showed that under the optimum conditions, the methane production increased by 46.6% in the batch tests and 120% in the semicontinuous operation system compared to the control reactor. The improved methane production originated from the synergetic effect of combining Fe0 and Mg(OH)2. Adding coated/Fe0 stimulated bacterial growth increased methane content and maintained the pH within the optimum range in the bioreactors. The dosing time of coated/Fe0 was investigated during the four stages of the anaerobic digestion process, and the best dosing time was found in the methanogenic stage (on Day 4). Overall, based on the experimental and predicted methane production, the coated/Fe0 has great potential for the practical applications of anaerobic digestion. moreover, the investigation also extended to evaluate the co-digestion by using aloe vera biomass. significant increase in the energy recovery from the organic waste by enhancing the production of H2 using nanoparticles and CH4, shorter retention time of treatment by accelerating the anaerobic digestion process

The main Achievements have been published in Renewable and Sustainable Energy Reviews with an impact factor of 15.9 under the title A novel method to improve methane generation from waste sludge using iron nanoparticles coated with magnesium hydroxide.

Summary: In response to the low efficiency of the anaerobic digestion (AD) process in generating methane gas, we apply for the first time the use of coated/Fe0 with Mg(OH)2 to enhance the production rate of methane gas from the degradation of waste sludge. A series of batch tests investigated several operations factors followed by a semi-continuous operation system examined the long-term production of methane gas in the presence of the coated/Fe0 were performed. The coating ratio of Mg(OH)2/Fe0 and the dosage of coated/Fe0 were optimized to acquire the highest production rate of methane as 0.5% and 25mg/gVS, respectively. Under these optimum conditions, the methane production increased by 46.6% in the batch tests and 120% in the semi-continuous operation system compared to the control reactor. The results revealed that both Fe0 and Mg(OH)2 did not significantly improve the production of methane when each one was used alone at different dosages, and the improved methane production originated from the synergetic effect of combining these two materials. The crucial role of Mg(OH)2 coating layer was associated with the controlled reactivity release of Fe0, which was indicated by the slow release of Fe2+ and Fe3+ in the bioreactors. Furthermore, the addition of coated/Fe0 stimulated bacterial growth, increased methane content, and maintained the pH within the optimum range in the bioreactors. The dosing time of coated/Fe0 was investigated during the four stages of AD process, and the best dosing time was found in the methanogenic stage (on Day 4). Overall, based on the experimental and predicted methane production, the coated/Fe0 has a great potential for the practical applications of AD.

Currently, the second paper under the title Exceptional contribution of iron nanoparticle and Aloe vera biomass additives to biogas production from anaerobic digestion of waste sludge has been accepted by Energy with an impact factor of 9.0

Summary: Aloe vera is a medicinal plant cultivated for numerous applications in the medical field, health care products, and many others. After the extraction of Aloe vera gel from leaves, a considerable amount of Aloe vera waste (AVW) is produced. Therefore, this study aimed to investigate the improvement of the anaerobic digestion (AD) performance of waste sludge (WS) by utilizing AVW and Fe0 nanoparticles. Accordingly, a series of biogas generation experiments were conducted at different mixing ratios of AVW and WS (AVW/WS: 0, 1, 2, and 3), to find out the optimum ratio for maximizing biogas generation. To gain insights into the improved biogas generation by the addition of AVW, pH value was monitored during the fermentation process and adjusted to different initial values (3, 7, and 12). Moreover, to further enhance the performance of

AD, the effect of different concentrations (10 and 50 mg/L) and dosing times (0, 6, 12, 24, 48, and 96 hrs.) of Fe0 nanoparticles on the digestion of the optimized ratio of AVW and WS (AVW/WS:2) were investigated. Results showed that the utilization of AVW in the anaerobic digesters at the optimum ratio (AVW/WS:2) resulted in improving biogas and methane generation by 62.5 % and 96.0 %, respectively, which is an excellent improvement rate of biogas and methane generation compared with the previously reported values in the literature. The addition of Fe0 at the optimum concentration (10 mg/L) and dosing time (12 hrs.) to the optimized ratio of AVW and WS (AVW/WS:2) resulted in enhancing methane generation by 146.9 %. Overall, based on the experimental and predicted methane data, the AVW has great potential for the enhancement of CH4 production in practical applications.

５．主な発表論文等

〔雑誌論文〕　計7件（うち査読付論文　7件／うち国際共著　7件／うちオープンアクセス　3件）

１．著者名 Ramadan Eljamal, Ibrahim Maamoun, Khaoula Bensaida, Gulsum Yilmaz, Yuij Sugihara, Osama Eljamal	４．巻 158
２．論文標題 A novel method to improve methane generation from waste sludge using iron nanoparticles coated with magnesium hydroxide	５．発行年 2022年
３．雑誌名 Renewable and Sustainable Energy Reviews	６．最初と最後の頁 1 - 13
掲載論文のDOI（デジタルオブジェクト識別子） 10.1016/j.rser.2022.112192	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

１．著者名 Ramadan Eljamal, Ibrahim Maamoun, Mohd Faizul Idham, Osama Eljamal	４．巻 8
２．論文標題 The role of coated nanoscale zero-valent iron with magnesium hydroxide in improving methane production during the anaerobic digestion of waste sludge	５．発行年 2022年
３．雑誌名 Proceedings of International Exchange and Innovation Conference on Engineering & Sciences (IEICES)	６．最初と最後の頁 291-296
掲載論文のDOI（デジタルオブジェクト識別子） 10.5109/5909106	査読の有無 有
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 該当する

１．著者名 Osama Eljamal	４．巻 8
２．論文標題 Opportunities to exploit stored energy in wastewater	５．発行年 2022年
３．雑誌名 Proceedings of International Exchange and Innovation Conference on Engineering & Sciences (IEICES)	６．最初と最後の頁 23-26
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１．著者名 Khaoula Bensaid, Ramadan Eljamal, kareman Eljamal, Yuji Sughihara, Osama Eljamal	４．巻 40
２．論文標題 The impact of iron bimetallic nanoparticles on bulk microbial growth in wastewater	５．発行年 2021年
３．雑誌名 Journal of Water Process Engineering	６．最初と最後の頁 1 -10
掲載論文のDOI（デジタルオブジェクト識別子） 10.1016/j.jwpe.2020.101825	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

１．著者名 Ramadan Eljamal, Ibrahim Maamoun, Khaoula Bensaida, Yuji Sugihara, Osama Eljamal	４．巻 7
２．論文標題 Investigating the Effect of Commercial and Synthesized Fe^0 particles on Methane Production Through the Anaerobic Digestion of Waste Sludge	５．発行年 2021年
３．雑誌名 Proceeding of International Exchange and Innovation Conference on Engineering & Sciences (IEICES)	６．最初と最後の頁 184 - 189
掲載論文のDOI（デジタルオブジェクト識別子） 10.5109/4738586	査読の有無 有
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 該当する

１．著者名 Eljamal Osama、Eljamal Ramadan、Falyouna Omar、Maamoun Ibrahim、Thompson Ian P.	４．巻 303
２．論文標題 Exceptional contribution of iron nanoparticle and Aloe vera biomass additives to biogas production from anaerobic digestion of waste sludge	５．発行年 2024年
３．雑誌名 Energy	６．最初と最後の頁 131761～131761
掲載論文のDOI（デジタルオブジェクト識別子） 10.1016/j.energy.2024.131761	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

１．著者名 JIANNING DING、Eljamal Osama	４．巻 9
２．論文標題 Hydrogen production of anaerobic digestion: A review	５．発行年 2023年
３．雑誌名 Proceedings of International Exchange and Innovation Conference on Engineering & Sciences (IEICES)	６．最初と最後の頁 247～252
掲載論文のDOI（デジタルオブジェクト識別子） 10.5109/7157979	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

〔学会発表〕　計5件（うち招待講演　5件／うち国際学会　0件）

１．発表者名 Osama Eljamal
２．発表標題 Potential for Energy Recovery from Wastewater
３．学会等名 CAMPUS Asia 2022 Summer School, Pusan, Korea（招待講演）
４．発表年 2022年

１．発表者名
Osama Eljamal

２．発表標題
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３．学会等名
J-MENA, Academic Seminar（招待講演）

４．発表年
2022年

１．発表者名
Osama Eljamal

２．発表標題
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３．学会等名
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１．発表者名
Osama Eljamal

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３．学会等名
International Conference on Emerging Contaminants and Environmental Health; Today and Future（招待講演）

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2021年

１．発表者名
Osama Eljamal

２．発表標題
Opportunities to exploit stored energy in wastewater

３．学会等名
The 8th International Exchange and Innovation Conference on Engineering & Sciences（招待講演）

４．発表年
2022年

〔図書〕　計0件

〔産業財産権〕

〔その他〕

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６．研究組織

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （機関番号）	備考

７．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

８．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関