

平成 30 年 6 月 20 日現在

機関番号：12102

研究種目：基盤研究(C) (一般)

研究期間：2015～2017

課題番号：15K00599

研究課題名 (和文) Formation of Granules with High Phosphorus Content to Realize Efficient Phosphorus Recovery from Waste Activated Sludge by Using Anaerobic Digestion Followed by Aerobic Granulation Technology

研究課題名 (英文) Formation of Granules with High Phosphorus Content to Realize Efficient Phosphorus Recovery from Waste Activated Sludge by Using Anaerobic Digestion Followed by Aerobic Granulation Technology

研究代表者

雷 中方 (LEI, Zhongfang)

筑波大学・生命環境系・准教授

研究者番号：30634505

交付決定額 (研究期間全体) : (直接経費) 3,700,000 円

研究成果の概要 (和文) : リンは多く分野に重要な役割を果たし、再生できない資源の一つである。日本は世界の8番目のリンの消費国であり、すべてのリンを海外から輸入に依存している。本研究は熱水前処理と嫌気性消化と好気性グラニュール化のコンビネーションによって廃棄活性汚泥からリンの高含有グラニュール生産を実現する可能性等を検討した。熱水処理条件の最適化により、汚泥からのメタンガス生産性が改善された結果、エネルギー収支の面からも、前処理法は効率的な方法であると明らかになった。また、合成した消化液或いは廃水を用いてグラニュール化した結果、6-10%のリンを含有し、そのうちリンの生物学的利用性は95%に高いグラニュールを得ました。

研究成果の概要 (英文) : Phosphorus (P) as a non-renewable resource plays important roles in agricultural and industrial activities. Japan is the 8th largest P consumption nation with all the P resources being imported from other countries. This project investigated the feasibility of P-rich granules production from waste activated sludge by combining hydrothermal pretreatment, anaerobic digestion and aerobic granulation. Results show that hydrothermal treatment can be a cost-effective process when the treatment temperature and duration are optimized according to the energy balance of enhanced methane production from the pretreated waste activated sludge. P-rich granules (6-10%) with P bioavailability up to 95% could be successfully cultivated using synthetic digestate or wastewater.

研究分野：持続環境学

キーワード：リン回収 好気性グラニュール化 廃棄活性汚泥 熱水前処理 嫌気性消化

1. 研究開始当初の背景

(1) Wastewater treatment plants (WWTPs) contribute a lot to the sustainable utilization of water in our society, in which activated sludge process is applied worldwide. Waste activated sludge (WAS), a major by-product from WWTPs, is difficult and costly to handle with mainly due to its huge production amount. In Japan, WAS production is about one-fourth of the total waste biomass, 79 million tones wet and 2 million tones dried mass, respectively. After being thickened, anaerobically digested, chemically adjusted, and then mechanically dewatered, the treated WAS can be incinerated and the resultant ash is always landfilled. The treatment cost of these processes amounts to 40-60% of the total operation and maintenance costs of the WWTP. In fact, because of being rich in organics and mineral nutrients, WAS can be recycled and reused after being appropriately treated. The main bottlenecks are associated with how to quickly separate the treated WAS from the mainstream treatment units and keep the bioavailability of the nutrients in the WAS after being treated.

(2) Phosphorus (P) is a non-renewable resource and plays a major role in agricultural and industrial activities. The reserve of P mineral is estimated to deplete within 50-100 years. More importantly, Japan is the 8th largest P consumption nation in the world and all the P resources are imported from other countries. Meanwhile, our preliminary results of WAS samples from several WWTPs nearby indicate that the P content in WAS is about 2-5% of the total solids (dry weight). The successful recovery of P from WAS can greatly ameliorate the pending crisis of P resource, thus reducing the dependence on P import from other countries.

2. 研究の目的

(1) The major objective of this project was to cultivate P-rich aerobic granules by using the liquid digestate of WAS. To achieve this target, the operation strategies, characterization of WAS digestate and additional additives were considered to make clear which factor impacts greatly on the effective P accumulation into the granules, and which factor could largely enhance P bioavailability.

(2) The aerobic granulation conditions would be

optimized to achieve high bioavailability ($\geq 80\%$) of P in the granules, efficient removal of pollutants, easy control of the operation and less addition of additives. The mechanisms involved in the granulation with enhanced P recovery and P bioavailability would be further elucidated in the context of P recovery from the liquid WAS digestate.

3. 研究の方法

Three sets of experiments were carefully designed and conducted to fulfill the targets of this research.

(1) Enhanced anaerobic digestion of waste activated sludge (WAS) was realized by using hydrothermal (HT) pretreatment and the optimal conditions were finalized according to the performance of methane production and P release.

(2) N recovery as ammonia was achieved from liquid WAS digestate by using air stripping.

(3) P-rich granules with high bioavailability were then cultivated by using aerobic granulation through treating liquid WAS digestate, which provides a new promising process for fast P recovery and easy post-treatment of WAS digestate.

In the above three sets of experiments, the crucial influencing factors were also determined and optimized, and the mechanisms involved were explored.

4. 研究成果

(1) Results show that hydrothermal treatment can be a cost-effective process when HT temperature and duration are optimized according to the energy balance of enhanced methane production from the pretreated WAS. P-rich granules (6-10%) with P bioavailability up to 95% could be successfully cultivated by using synthetic digestate or wastewater. Through the proposed processes, the released ammonia can also be recovered simultaneously with P when coupling with magnesium ammonium phosphate (MAP) precipitation after WAS being HT pretreated. Besides excellent organics and N removal, a constant 5 mg/L Fe^{2+} addition to the influent of aerobic granulation system can achieve stably

efficient P removal (92%) and recovery from wastewater to produce P-rich granules with high P bioavailability. In addition, a comparative study has been conducted between the lab-scale aerobic granular sludge system and Johkasou system with respect to their pollutants removal, energy consumption, and investment and operation costs, indicating the greater application potential of the aerobic granular sludge process.

(2) The results from this project suggest that the combination of HT pretreatment, anaerobic digestion and aerobic granulation processes might be the most prospective solution to manage organic solid wastes or high-strength organic wastewater, targeting efficient pollutants removal and energy/resources recovery. In the case of P recovery from WAS, not only organics can be decomposed and recovered as biogas (mainly methane) but also P-rich granules are produced, which can be easily re-utilized for multipurpose due to its high P bioavailability. Based on our experimental results, about 40,000 - 100, 000 tons of P could be annually recycled and reused in Japan if these processes could be applied.

(3) In addition to the pollutants removal and resources recovery efficiencies, the mechanisms involved have been explored, especially on P accumulation and its distribution in different kind of aerobic granules. The contributions of the main factors have been identified, including operation strategy, influent characteristics, additional additives, extracellular polymeric substances (EPS), etc. Furthermore, through this project we proposed some new and promising processes such as ammonia stripping and algal-bacterial granular sludge processes which possess great potentials for nutrients recovery from organic waste or wastewater.

(4) During the three years' investigation, the major results from this project have been published in 16 papers in international journals and 22 conference presentations as well, which provided good opportunities for us to communicate with the peer researchers in the similar research fields. Moreover, we extended the proposed processes to treat not only WAS but also other organic solid wastes like manure waste, reflecting their high efficiencies in simultaneous organics, N and P removal or recovery when their

optimal conditions are applied.

5. 主な発表論文等

[雑誌論文] (計 16 件)

- (1) Cai W., Zhao Z., Lei Z., Zhang Z., Adachi Y., Lee D.-J., 2018. Influence of ferrous iron dosing strategy on aerobic granulation of activated sludge and bioavailability of phosphorus accumulated in granules. *Bioresource Technology Reports*, 2, 7-14 (peer reviewed)
- (2) Chen K., Zhao Z., Yang X., Lei Z., Zhang Z., Zhang S., 2018. Desorption trials and granular stability of chromium loaded aerobic granular sludge from synthetic domestic wastewater treatment. *Bioresource Technology Reports*, 1, 9-15 (peer reviewed)
- (3) Yuan T., Cheng Y., Huang W., Zhang Z., Lei Z., Shimizu K., Utsumi M., 2018. Fertilizer potential of liquid product from hydrothermal treatment of swine manure. *Waste Management*, 77, 166-171 (peer reviewed)
- (4) Huang W., Zhao Z., Yuan T., Yu Y., Huang W., Lei Z., Zhang Z., 2018. Enhanced dry anaerobic digestion of swine excreta after organic nitrogen being recovered as soluble proteins and amino acids using hydrothermal technology. *Biomass and Bioenergy*, 108, 120-125 (peer reviewed)
- (5) Ahmad J. S. M., Cai W., Zhao Z., Zhang Z., Shimizu K., Lei Z., Lee D.-J., 2017. Stability of algal-bacterial granules in continuous-flow reactors to treat varying strength domestic wastewater. *Bioresource Technology*, 244, 225-233 (peer reviewed)
- (6) Yu Y., Lei Z., Yuan T., Jiang Y., Chen N., Feng C., Shimizu K., Zhang Z., 2017. Simultaneous phosphorus and nitrogen recovery from anaerobically digested sludge using a hybrid system coupling hydrothermal pretreatment with MAP precipitation. *Bioresource Technology*, 243, 634-640 (peer reviewed)
- (7) Yuan T., Huang W., Lei Z., Zhao Z., Zhang Z., 2017. Effects of different alkalis on hydrolysis of swine manure during dry anaerobic digestion and resultant nutrients availability. *International Biodeterioration & Biodegradation*, 123, 138-145 (peer reviewed)

- (8) Huang W., Zhao Z., Yuan T., Huang W., Lei Z., Zhang Z., 2017. Low-temperature hydrothermal pretreatment followed by dry anaerobic digestion: A sustainable strategy for manure waste management regarding energy recovery and nutrients availability. *Waste Management*, 70, 255-262 (peer reviewed)
- (9) Cai W., Huang W., Li H., Sun B., Xiao H., Zhang Z., Lei Z., 2016. Acetate favors more phosphorus accumulation into aerobic granular sludge than propionate during the treatment of synthetic fermentation liquor. *Bioresource Technology*, 214, 596-603 (peer reviewed)
- (10) Huang W., Zhao Z., Yuan T., Lei Z., Cai W., Li H., Zhang Z., 2016. Effective ammonia recovery from swine excreta through dry anaerobic digestion followed by ammonia stripping at high total solids content. *Biomass and Bioenergy*, 90, 139-147 (peer reviewed)
- (11) Huang W., Huang W., Yuan T., Zhao Z., Cai W., Zhang Z., Lei Z., Feng C., 2016. Volatile fatty acids (VFAs) production from swine manure through short-term dry anaerobic digestion and its separation from nitrogen and phosphorus resources in the digestate. *Water Research*, 90, 344-353 (peer reviewed)
- (12) Huang W., Yuan T., Zhao Z., Yang X., Huang W., Zhang Z., Lei Z., 2016. Coupling hydrothermal treatment with stripping technology for fast ammonia release and effective nitrogen recovery from chicken manure. *ACS Sustainable Chemistry and Engineering*, 4(7), 3704-3711 (peer reviewed)
- (13) Huang W., Huang W., Li H., Lei Z., Zhang Z., Tay J.H., Lee D.-J., 2015. Species and distribution of inorganic and organic phosphorus in enhanced phosphorus removal aerobic granular sludge. *Bioresource Technology*, 193, 549-552 (peer reviewed)
- (14) Li B., Huang W., Zhang C., Feng S., Zhang Z., Lei Z., Sugiura N., 2015. Effect of TiO₂ nanoparticles on aerobic granulation of algal-bacterial symbiosis system and nutrients removal from synthetic wastewater. *Bioresource Technology*, 187, 214-220 (peer reviewed)
- (15) Huang W., Li B., Zhang C., Zhang Z., Lei Z., Lu B., Zhou B., 2015. Effect of algae growth on aerobic granulation and nutrients removal from synthetic wastewater by using sequencing batch reactors. *Bioresource Technology*, 179, 187-192 (peer reviewed)
- (16) Huang W., Cai W., Huang H., Lei Z., Zhang Z., Tay J.H., Lee D.-J., 2015. Identification of inorganic and organic species of phosphorus and its bio-availability in nitrifying aerobic granular sludge. *Water Research*, 68, 423-431 (peer reviewed)
- [学会発表] (計 22 件)
- (1) Tian Yuan, Yanfei Cheng, Wei Huang, Zhenya Zhang, Zhongfang Lei, Kasuya Shimizu: Fertilizer potential of liquid product from hydrothermal treatment of swine manure. 日本水処理生物学会第 54 回大会 11 月 8 ~ 10 日, 大阪 (2017)
- (2) Zhongfang Lei and Zhenya Zhang: Hydrothermal pretreatment and its effects on resources and energy recovery from organic solid wastes. The 15th IWA World Conference on Anaerobic Digestion, Side Event III - Workshop on Anaerobic Fermentation Biotechnology for Organic Solid Waste, 14-16 October, 2017, Wuxi, China
- (3) Zhongfang Lei and Zhenya Zhang: Energy and Resources Recovery from Waste and Wastewater Treatment, International Workshop on Environmental Science Technology and Sustainability Assessment, 3-6 August, 2017, Zhengzhou, China
- (4) 小倉舜, 稲森隆平, 稲森悠平, 趙子文, 張振亜, 雷中方: 曝気量の違いによる共存藻類等の有無による好気性グラニューール法の処理性能の比較. 第 51 回日本水環境学会年会, 3 月 15 ~ 17 日, 熊本 (2017)
- (5) Johan Syafri Mahathir Ahmad, Wei Cai, Meishan Jin, Minh Tu Nguyen, Zhongfang Lei, Zhenya Zhang: Study on Stability of Aerobic Algal-bacterial Granular Sludge in A Continuous Flow Reactor. 日本水処理生物学会第 53 回大会, 11 月 10 ~ 12 日, 千葉, 日本 (2016)
- (6) Boaiqi Zhang, Zhongfang Lei, Zhenya Zhang: Effect of Direct Current Electric Field on Mature Aerobic Granular Sludge during Synthetic Ammonia Wastewater

- Treatment. 日本水処理生物学会第 53 回大会, 11 月 10 ~ 12 日, 千葉, 日本 (2016)
- (7) 小倉舜, 雷中方, 稲森隆平, 稲森悠平, 張振亜, 類家翔, Nguyen Tu Minh, 徐開欽: 好気性グラニユール法における共存藻類等の有無による人工排水を基質とした処理性能の比較. 日本水処理生物学会第 53 回大会, 11 月 10 ~ 12 日, 千葉, 日本 (2016)
- (8) Meishan Jin, Wei Cai, Johan Syafri Mahathir Ahmad, Zhongfang Lei, Zhenya Zhang: Study on Algal Granules' Growth and Stability under Controlled Artificial Sunlight. 日本水処理生物学会第 53 回大会, 11 月 10 ~ 12 日, 千葉, 日本 (2016)
- (9) Zhongfang Lei, Wenli Huang, and Zhenya Zhang: Functional Granular Sludge: Achievements and Perspectives (Invited Presentation). International Symposium on Environmental Education and Sustainability Sciences, May 31 - June 1, Wuxi, China (2016)
- (10) Zhenya Zhang and Zhongfang Lei: Anaerobic Fermentation Systems: Advantages and International Strategies. (Invited Presentation). International Symposium on Environmental Education and Sustainability Sciences, May 31 - June 1, Wuxi, China (2016)
- (11) Weiwei Huang, Zhongfang Lei, and Zhenya Zhang: Short-term Dry Anaerobic Digestion for Ammonia and Volatile Fatty Acids Accumulation in Swine Manure and Their Subsequent Separation from Phosphorus Resource. International Symposium on Environmental Education and Sustainability Sciences, May 31 - June 1, Wuxi, China (2016)
- (12) 黄 薇薇, 張 振亜, 雷 中方: 「短期間乾式豚糞の嫌気性消化における揮発性脂肪酸及びアンモニアの生産とそれらの分離」, 東京大学・天津市政府共同研究プロジェクト「都市と農村の融合に基づく持続的発展」2016 年度第 1 回日中共同セミナー, 4 月 1 2 日, 東京 (2016)
- (13) 雷 中方, 張 振亜: 「中国畜産廃棄物を湿式及び乾式嫌気性消化処理する二つの事例」, 東京大学・天津市政府共同研究プロジェクト「都市と農村の融合に基づく持続的発展」2016 年度第 1 回日中共同セミナー, 4 月 1 2 日, 東京 (2016)
- (14) Huseini Muhammad Reza, Zhongfang Lei, Zhenya Zhang: Adsorbent development from primary sludge treated by hydrothermal technology for ammonia recovery from wastewater. 日本水処理生物学会第 52 回大会, 11 月 11 ~ 13 日, 北九州, 日本 (2015)
- (15) Dapeng Zhang, Wei Cai, Zhongfang Lei, Zhenya Zhang: Study on biosorption of chromium(VI) onto aerobic granules and its influence factors. 日本水処理生物学会第 52 回大会, 11 月 11 ~ 13 日, 北九州, 日本 (2015)
- (16) Wenli Huang, Wei Cai, Weiwei Huang, Zhenya Zhang, Zhongfang Lei: Phosphorus species, distribution and bio-availability in aerobic granular sludge, サマースクール「土・水・生命環境とコロイド界面現象 - コロイドの凝集と沈降」, 7 月 31 日, 筑波, 日本 (2015)
- (17) Weiwei Huang, Ziwen Zhao, Tian Yuan, Zhongfang Lei, Wei Cai, Zhenya Zhang: Effective ammonia recovery from swine manure by dry anaerobic digestion and air stripping at high total solids content. The 67th Annual Meeting of the Society for Biotechnology, Japan, 26-28 October, Shiroyama, Japan (2015)
- (18) Wei Cai, Weiwei Huang, Danni Li, Junjun Li, Zhongfang Lei, Zhenya Zhang: Effect of hydrothermal pretreatment on solubilization and subsequent volatile fatty acids fermentation of waste activated sludge. The 67th Annual Meeting of the Society for Biotechnology, Japan, 26-28 October, Shiroyama, Japan (2015)
- (19) Junjun Li, Wei Cai, Danni Li, Zhongfang Lei, Zhenya Zhang: Volatile fatty acids (VFAs) accumulation from waste activate sludge under alkaline anaerobic fermentation. The 67th Annual Meeting of the Society for Biotechnology, Japan, 26-28 October, Shiroyama, Japan (2015)
- (20) Danni Li, Wei Cai, Junjun Li, Zhongfang Lei, Zhenya Zhang: Hydrothermal pretreatment of primary sludge and its impact on subsequent anaerobic VFAs accumulation and biogas production. The 67th Annual Meeting of the Society for Biotechnology, Japan, 26-28 October, Shiroyama, Japan (2015)
- (21) Ziwen Zhao, Weiwei Huang, Tian Yuan, Wei Cai, Zhongfang Lei, Zhenya Zhang: Effect of hydrothermal pretreatment on VFAs

production from rice straw through anaerobic fermentation. The 67th Annual Meeting of the Society for Biotechnology, Japan, 26-28 October, Shiroyama, Japan (2015)

- (22) Tian Yuan, Weiwei Huang, Ziwen Zhao, Zhongfang Lei, Zhenya Zhang: Accumulation of volatile fatty acids and change in phosphorus fractions during alkaline dry anaerobic fermentation of swine manure: Effect of different alkalis. The 67th Annual Meeting of the Society for Biotechnology, Japan, 26-28 October, Shiroyama, Japan (2015)

[その他]

ホームページ等

Researchgate

https://www.researchgate.net/profile/Zhongfang_Lei;

ResearcherID

<http://www.researcherid.com/rid/B-3611-2014>

Researchmap

https://researchmap.jp/Lei_Zhongfang/

筑波大学研究者総覧

<http://www.trios.tsukuba.ac.jp/researcher/0000003223>

6. 研究組織

(1) 研究代表者

雷 中方 (LEI, Zhongfang)

筑波大学・生命環境系・准教授

研究者番号：30634505

(2) 研究分担者

張 振亜 (ZHANG, Zhenya)

筑波大学・生命環境系・教授

研究者番号：20272156