High Rate Methane Fermentation of Food Waste and Sewage Sludge by an Anaerobic Membrane Bioreactor

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論 文 内 容 要 旨

This dissertation was accomplished under the background of the urgent necessity for renewable energy to replace traditional fossil fuels and the demand for organic solid waste treatment. With the development of society and the expansion of population, the increasing utilization of materials and energy cause unignorable environmental issues such as global warming and resources shortage. Large quantity of waste, both industrial and municipal waste, are generated at the same time. Under such situation, the residual value inside the waste attracts more and more attention and the concepts of waste-to-resources and waste-to-energy were proposed. Especially for organic solid waste with the characteristics of high moisture and organic content, it may cause solid, air, water pollution with improper treatment while it also can become treasure for energy and material production with suitable treatments. Anaerobic digestion, which can transfer organic matters into bioenergy via microorganisms activities, is considered as the most promising technology to realize the gorgeous transformation of organic solid waste. However, though the technology has been applied for several decades, efforts are still attributing to improve the process efficiency. Facing the shortcoming in system imbalance caused by the shortage of necessary elements and nutrients in the traditional single-substrate digestion, the application of co-digestion, which digests two or more kind of substrates simultaneously, presents a more stable performance. On the other hand, the enhancement in the microorganisms density in the digester via digester design optimization is regarded as another hint to obtain a better digestion efficiency. Anaerobic membrane bioreactor (AnMBR) is such kind of novel technology combining traditional anaerobic digester and membrane technology to intercept microorganisms via membrane filtration. It has been more commonly used in wastewater treatment rather than the solid waste digestion while the promotion is on the way. One of the biggest challenges in the application of AnMBR in the solid waste treatment is the high solid concentration in both substrate and digester which bring difficulties in long-term

stable operation and maintenance. What's more, there is very little research on the application of high-solid AnMBR for the co-digestion of organic solid waste. Therefore, this dissertation described a systematic research on this content.

The first study in this dissertation, as shown in Chapter 3, investigated the long-term performance of food waste and sewage sludge in a thermophilic AnMBR. The AnMBR used in this study contained a 13 L CSTR and a 2 L membrane unit inserted a 0.1 m^2 hollow fiber membrane with average pore size of 0.1 μm. The reactor was originally fed with pure food waste at a HRT of 15 d. Sewage sludge, collected from local wastewater treatment plant, replaced food waste at TS ratios of 25%, 50% and 100%. According to the different combination of substrate, the digester went through the mono-digestion of food waste, the co-digestion of food waste and sewage sludge (75%: 25%; 50%: 50%) and the mono-digestion of sewage sludge periods. The system was fed with constant TS loading and HRT. The long-term stable operation showed the feasibility of the utilization of high solid thermophilic AnMBR for food waste and sewage sludge treatment. It should be noted that the mono-digestion of these two substrate failed in CSTR reported by other researchers. The addition of sewage sludge restricted the biogas generation due to the less biodegradable of sewage sludge compared to food waste. While for a sludge substitution with a 25% TS-based substrate, the biogas yield of 0.812 L/g-VS_{fed} was at 91% and 158% of that of the mono-digestion of food waste and sewage sludge, respectively. Furthermore, with the assistance of membrane, excellent permeate quality was obtained compared with that from traditional CSTR which was another merit of AnMBR. The function of membrane in the anaerobic digestion was also analyzed. Except the interception of microorganisms and suspended solid, the rejection of soluble organic matters were also observed. The results showed that the membrane filtration rejected about 70% of COD, 80% of soluble carbohydrates and 17% of soluble proteins while the concentration of soluble carbohydrates were at a far lower level than that of soluble proteins. It may due to the higher digestion efficiency and the larger and complex structure of carbohydrates than for proteins. Except digestion performance, the membrane performance was also important component in the operation. The long-term operation results indicated that with the application of filtration-relaxation mode, the AnMBR can operated stably at a high flux of 5 LMH under the high solid (~27 g/L) condition which was much higher than the working condition reported in the literature for the wastewater treatments.

The second study in this dissertation, as shown in Chapter 4, focused on the change in the functional bacterial and archaea community during thermophilic anaerobic digestion of food waste and sewage sludge in a membrane bioreactor. Furthermore, the reaction kinetics of anaerobic digestion was also explained connected with microbial analysis results. The anaerobic activated sludge samples were taken from the stable period of a thermophilic AnMBR for food waste and sewage sludge digestion. The samples were analyzed via high-throughput sequencing technology. Among the four steps, hydrolysis, acidogenesis, acetogenesis, methanogenesis, of anaerobic digestion, hydrolysis was determined as the rate-limiting step in both mono- and co-digestion periods according to the calculation results. Meanwhile, the reaction ratios decreased with the addition of sewage sludge. Correspondingly, an increasing trend in the microorganism diversity was also observed. The microbial genera were divided into several functional groups based on their roles played in the anaerobic digestion. Generally, the bacterial community working on the decomposition of organic matters from large complex molecules to small simple molecules while the archeal community generate methane. The change of functional groups in bacterial community related with the composition of substrate. The relative abundance of carbohydrates-degrading-bacteria decreased with the addition of sewage sludge which contained less carbohydrates. While the change in proteins and peptides-degrading-bacteria did not change in one direction with the increase of sludge. The matched phenomenon was also observed as the accumulation of soluble proteins in the digester. It was due to the change of archeal community which had a negative influence on the co-cultured proteins and peptides-degrading-bacteria. The shift in the functional bacterial community reflected the shift in main types of organic matters consumed and further influenced methane generation pathway. In the mono-digestion of food waste and the co-digestion of 75% food waste and 25% sewage sludge, carbohydrates, together with proteins were used as the main substrate and methane was generated via hydrogenotrophic pathway. In the co-digestion of 50% food waste and 50% sewage sludge, proteins were consumed mostly and methane generation by both hydrogenotrophic and acetoclastic methanogens. While in the mono-digestion of sewage sludge, with the increasing diversity of microorganisms, carbohydrates, proteins and organic acids were all consumed, and acetate was the absolute dominant substrate used for methane production.

The third study in this dissertation, as shown in Chapter 5, investigated the effect of temperature on the mono- and co-digestion of food waste and sewage sludge in AnMBRs from the perspective of methane generation performance, membrane filtration performance, membrane fouling and microbial community. The experiments conducted by two identical AnMBR systems operated under mesophilic and thermophilic conditions, respectively. The results shown that the temperature had little influence on the methane generation capacity of AnMBR in both mono- and co-digestion conditions that may due to the high microorganisms concentration in the membrane bioreactor. While the phenomenon was different with that reported to traditional CSTRs where the advantage of temperature was obvious. However, thermophilic AnMBR presented faster specific reaction rates as its advantage. Though the reduction ratios of organic matters were similar under mesophilic and thermophilic conditions, the permeate quality was better from the mesophilic digester, especially for in proteins. The phenomenon were consistent with the results of membrane rejection. Both AnMBRs had a higher rejection to soluble carbohydrates than soluble proteins while the thermophilic digester had a less rejection to soluble proteins than the mesophilic one. These may because the higher temperature presented a better decomposition capacity on organic matters and carbohydrates were easier to be consumed than proteins. In the membrane foulants analysis, the thermophilic AnMBR suffered a severe membrane fouling than the mesophilic one while the main foulants in both reactors was cake layer. However, due to a fast growth of cake layer under the thermophilic condition, 80% of the total resistance on the membrane surface caused by foulants can be removed via physical cleaning protocols. While that value for mesophilic condition was nearly 60%. A big difference observed in the microbial community structure under different temperature. The mesophilic AnMBR had a higher microbial diversity than the thermophilic one due to the more suitable environment for general microorganisms growth under a mild conditions. Furthermore, the mesophilic system was observed to be more stable under different feeding conditions than the thermophilic one. Unlike the changeable metabolic pathway in the mono- and co-digestion in the thermophilic condition, methane generated in mesophilic AnMBR was always via acetoclastic methanogens. When it was come to the influence of sewage sludge addition in the substrate, the microbial diversity under both temperature showed increasing trends. What's more, the microbial diversity in the co-digestion periods were higher than the superimposed values obtained from the mono-digestion periods showed the advantage in enriching species diversity of co-digestion.

Last but not the least, the fourth study in this dissertation, as shown in Chapter 6, conducted a technical evaluation on a high solid external submerged thermophilic AnMBR used for mono- and co-digestion of food waste and sewage sludge from the perspective of anaerobic digestion efficiency, waste-to-energy efficiency and system energy balance. The evaluation connected the relation between the experiment results and calculation results and the relation between the theoretical performance and piratical performance. The results showed that the system can reached methane recovery rates over 50% compared to the calculation results from chemical analysis of substrate. The energy recovery rates of the substrate via anaerobic digestion in AnMBR range from 40% to 90% based on different calculation methods under all test conditions. In the system energy balance analysis, the blower was found to consume over 85% of electricity among all power-consuming devices under the experimental scale. For the piratical application in a large scale, a system energy analysis was conducted at a processing capacity of 100 m^3/d, and found that the thermophilic AnMBR system can achieve positive energy production in the mono- and co-digestion of food waste and sewage sludge.

論文審査結果の要旨

生ごみは発生量が多く、有機物の割合が高いので、代表的廃棄物系バイオマス資源の一つである。また、都市下水処理に伴い発生する下水汚泥も安定的なバイオマス資源の一つである。メタン発酵は、廃棄物系バイオマスからエネルギーを生産するための重要な方法であり、その効率化と応用拡大が望まれている。近年、嫌気性膜バイオリアクター（AnMBR）は革新的排水処理技術として提案され、その応用範囲の拡張が期待されている。本研究は嫌気性膜バイオリアクターによる生ごみと下水汚泥のメタン発酵の効率化を目指して、生ごみと下水汚泥の単独発酵と混合発酵に関する連続実験を行い、原料組成と運転温度の変化による AnMBR 性能への影響について検討を行った。本論文は以下の通り 7 章から構成されている。

第 1 章では、本研究の背景と研究目的を説明し、本論文の構成について紹介した。

第 2 章では、生ごみと下水汚泥の処理方法を紹介し、メタン発酵の既存技術と効率改善方法をまとめ、混合発酵と嫌気性膜分離技術を用いた都市系廃棄物のメタン発酵の研究課題を抽出した。

第 3 章では、嫌気性膜バイオリアクターによる生ごみと下水汚泥の TS 比を 100:0、75:25、50:50、0:100 に変化させて高温メタン発酵連続実験を行い、それぞれの条件における有機物の分解率、膜ろ過やバイオガス生成のパフォーマンスを把握した。汚泥比率 25%の混合発酵条件では、混合発酵による相乗効果も見られた。また、TS 濃度が 2.7-3.7%の高い条件においても中空糸膜は 5LMH の高いフラックスで安定した運転ができることが示された。

第 4 章では、嫌気性膜バイオリアクターによる生ごみと下水汚泥の高温混合メタン発酵における微生物群集の構造を検討した。メタン発酵反応を 4 段階に分けてそれぞれの段階の反応速度を計算して比較した結果、加水分解は、発酵全体の律速段階であることを明らかにした。炭水化物分解細菌、タンパク質分解細菌および各種酸生成細菌などの機能性細菌群集の構造は主に有機物の種類変化に影響され、またメタン生成古細菌がさらに影響を受けた。メタン生成経路は生ごみの単独発酵では水素経路が優勢であったが、下水汚泥の単独発酵では酢酸経路が優勢となった。

第 5 章では、生ごみと下水汚泥の高濃度混合メタン発酵に及ぼす温度の影響を検討するために、高温と中温嫌気性膜バイオリアクターにおける有機物の分解、バイオガス生成、膜性能、微生物群集を比較した。中温と高温の条件下では、バイオガス生成に大きな差が見られなかったが、高温の条件下での反応速度がより高かった。また高温条件下では、膜ファウリングはより深刻に発生した。また中温条件における微生物の多様性と安定性が高かった。

第 6 章では、本研究の実験結果に基づき、嫌気性膜バイオリアクターによる生ごみと下水汚泥の混合メタン発酵に関するエネルギー収支を評価した。本研究の高濃度 AnMBR システムは、生ごみと下水汚泥の単独発酵および混合発酵のいずれでもエネルギー生産を実現できることを解明した。また、同システムにおける電力消費の 85%は膜洗浄ブロワーに利用されていることを示唆した。

第 7 章では本研究で得られた主な成果を総括し、応用に関する今後の展望について述べている。

以上のように、本論文は高濃度嫌気性膜バイオリアクターの応用拡大を目指して、生ごみと下水汚泥を対象とした連続実験を行い、プロセスの安定性とメタン発酵性能を検討し、微生物群集の変化とバイオエネルギー回収に関する重要な知見が得られている。その成果は都市廃棄物系バイオマスのエネルギー利用や循環型社会の構築に貢献することができ、環境工学の発展に寄与するところは少なくない。

よって、本論文は博士(工学)の学位論文として合格と認める。