【課程博士】

博 士 論 文 の 要 約

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学 位 論 文 題 名

Studies on pretreatment and process stabilization in anaerobic digestion

（メタン発酵における前処理とプロセス安定化に関する研究）

【Background and objectives】

Anaerobic digestion (AD) technology converts biodegradable components into methane-rich biogas, utilizing a broad spectrum of organic materials, including various types of biomass and organic wastes. This process enables simultaneous waste treatment and energy recovery. AD technologies, however, still face challenges related to conversion efficiency and process stability.

Corn stover (CS), among the wide variety of biomass, is notably recognized for its high productivity and high nitrogen uptake. The tillage system in corn cultivation, characterized by extensive mechanization and a variety of farming implements, emerges as the most energy-demanding process. As a result, fossil fuels with high CO_2 emissions need to be consumed. Sustainable CS production for renewable energy, therefore, requires addressing not only how to produce CS to maximize its use as a feedstock, but also how the tillage system contributes to a net reduction in CO_2 emissions from fossil fuels.

While AD using CS as raw material has been well studied. However, because the structure of CS is a complex network, cellulose and hemicellulose are firmly structured by lignin, which is difficult to be solubilized and biodegraded. This complex and rigid structure limits the enzymatic digestibility of lignocellulosic biomass and hinders the degradation of the entire carbohydrate structure.

A prevalent issue in AD, particularly when processing food waste and paper waste, is the pH drop that resulted from the rapid conversion of readily degradable components in food waste to volatile fatty acids (VFA) during digestion, resulting in underperformance or short-term process failure. Against this background, the main objectives of this thesis are: (1) to investigate the effect of tillage systems on the above-ground biomass yield of corn and the CO_2 emissions induced by fuel consumption of these tillage systems. (2) to develop a new approach to maintain constant digestate total solids (TS) content during AD using municipal solid waste and examine the feasibility of stabilizing the AD process by maintaining constant TS content in the digestate. (3) to investigate the effects of CO_2 addition in hydrothermal pretreatment of CS for biomethane production at lower temperature conditions.

1. Optimizing the tillage system for corn stover production

【Materials and methods】

This 2-year study was conducted in the field plots of Hokkaido University Farm No. 1, under four types of tillage systems; the field site is located in the city of Sapporo, Hokkaido, Japan (43°07′46.3″ N, 141°33′35.9″ E), which has a humid continental climate, with mean precipitation of 1,100 mm, and a mean annual temperature of 8.5 °C. The experimental field for all treatments consisted of a 300 m^2 plot, divided into four parts. Each of the tillage systems occupied 75 m^2, and planting operations for both 2019 and 2020 were carried out on May 14. The four tillage systems and their corresponding implements were as follows:

CT: Conventional tillage (plow + rotary + sowing machine + Cambridge roller)
ROT: Reduced tillage (plow + rotary + sowing machine)
CR: Reduced tillage (plow + sowing machine + Cambridge roller)
CON: Control (plow + sowing machine)

Fuel consumption rates for each tillage operation were estimated based on the typical size of tractors and trucks used in this region, alongside their average fuel and work efficiency.

The total diesel oil consumption for field operations under the four tillage systems was calculated. The calculation of CO_2 emissions was based on the tractor fuel consumption and an emission rate of 2.75 kg CO_2 per 1.0 L of diesel fuel consumed. The fuel consumption for other field operations, such as crop protection and harvesting, was not included in this study.

The study also examined the impact of the four tillage systems on soil penetration resistance (PR), measuring down to a depth of 0.3 m using a digital penetrator with a base cone diameter of 3×10^{-2} m and a cone angle of 30°. This was followed by the calculation of the seed germination rate (GR).

Plant height measurements were taken every four weeks on 25 marked plants from each row (total 100 plants per plot). The height of the plant was taken from the soil surface to the top of the most fully grown leaves. Each planet's average height was determined for each site year. On the basis of the plant height results, samples of the tallest, middle, and shortest height corn were collected from each plot to serve as samples for plant biomass measurements. These plants were manually harvested from each plot, cut 0.15 m from the ground, and taken to the laboratory for processing. The harvested plants were separated into leaves, stalks, husks, and ears, and each fraction was weighed. Volatile solids (VS) were then determined by drying samples at 105 °C for 24 h, followed by incineration at 600 °C for 3 h.

【Results and discussion】

The application of a reduced tillage (CR) system, as compared to conventional tillage (CT), can enhance total work capacity while also reducing CO_2 emissions. In terms of biomass yield, over the experimental period, both CT and CR systems yielded comparatively higher results relative to the other tillage systems tested. It was also observed that tillage systems were found to influence GR, and technical methods to improve GR are needed to achieve the full benefits of reduced tillage systems.

2. Stabilization of AD process via constant the digestate total solids content

【Materials and methods】

The Initial inoculum was obtained from a biogas plant operated by the Hokkaido University farm at mesophilic conditions and treating livestock manure. Food waste collected from the Hokkaido University central restaurant was ground using a food processor. Paper waste produced from the laboratory was shredded to give pieces measuring ~4 × 40 mm.

The AD experiment was carried out in thermophilic conditions (53 ± 2 °C) using a semi-continuous, horizontal cylindrical reactor with a mechanical mixer (235 L working volume) for 64 days. In this study, the C/N ratio was 40, and three levels of OLR were used. The reactor was equipped with a side port for material charging, a top port for gas collection, and a bottom port for digestate sampling. Pre-treated raw materials were added four times each week (on Monday, Tuesday, Thursday, and Friday). Anaerobic conditions were maintained by supplying nitrogen gas to the reactor after material was added. Generated biogas was collected in a gas bag for desulfurization before further utilization, and digestates were collected for component and stability analyses.

The entire experiment was divided into four phases. Phase 1 (weeks 1–4) was the period when the steady condition (TS content of 4% w.b.) was adopted, and the OLR was gradually increased from 1 to 3. The effects of the steady conditions were assessed. Phase 2 (weeks 5, 6) acted as the control condition, corresponding to the period without steady TS control, and the OLR was fixed to 3. Raw material was added without adding tap water in week 5, and liquid fraction from the disposed digestate was then added instead of raw material in week 6. Phase 3 (week 7) was the period without feeding, designed to stabilize the digestate solids content and VFA concentration under the influence of raw material charging in the previous week. The constant condition (6% w.b.) was adopted again in Phase 4 (weeks 8, 9) with a fixed OLR of 3 to further assess the effectiveness of the stable conditions.

【Results and discussion】

AD using FW and PW was carried out under thermophilic conditions in a semi continuous type of reactor to examine the feasibility of stabilizing the AD process by maintaining constant TS content in the digestate. Two levels of controlled conditions (desired values of 4% w.b. and 6% w.b.) were compared with conventional control over an experimental period of 64 days. The results identified that by applying stable digestate conditions newly developed using a mass balance equation during the AD process, the TS content of the digestate was maintained within 1% w.b. of the desired level. Considering the increase of digestate TS and the system imbalance caused by VFA accumulation during the conventional control phase, it was apparent that the digestate TS content during AD can be controlled and digestion process can be stabilized by adopting steady conditions.

In addition, the biogas production and methane concentration were increased over the levels for conventional control by 10.2% and 13.5%, respectively, with steady control in Phase 1, and by 16.6% and 21.2%, respectively, when using steady control in Phase 4. Therefore, we consider that AD performance can be improved by controlling the digestate TS to a steady level during AD. In future work, considerably more work will need to be done to determine the behavior of microbial communities when adopting steady conditions.

3. CO_2-assisted hydrothermal pretreatment of corn stover for enhanced biomethane production

【Materials and methods】

The initial inoculum (TS: 4.76 ± 0.06, VS: 3.72 ± 0.03) was sourced from a biogas plant operated by the Hokkaido University farm at mesophilic conditions treating livestock manure. CS (TS: 94.6 ± 0.04, VS: 84.5 ± 0.17) harvested from a farm owned by Hokkaido University was used. This dried CS was then pulverized in a laboratory-scale grinder to a 60-mesh size.

The pretreatment reactor was connected to the liquid CO_2 tank and liquid CO_2 flowed into the reactor without a pump after passing through the heater, due to a higher CO_2 cylinder pressure. After reaching a desired pressure (3 or 5 MPa), the reactor will be placed in a water bath for heating. CO_2 free conditions was obtained by supplying N_2 (0.5 MPa) to the reactor. 9 grams of milled, air-dry CS will mix with 90 g of distilled water solution pleased in temperature-controlled batch reactor with 190 mL volume. The pretreatment temperature was 50, 70 and 90 °C with or without CO_2, pretreatment time was fixed at 1 h to examine the effect of pretreatment temperature and time on final product. Liquid and solid phases of the reaction solution were then separated by vacuum filtration for further use. The solid, liquid fractions or whole slurry after pretreatment was exposed to AD under thermophilic conditions (53 ± 2 °C) in a 200 mL triangular beaker to produce biogas under the inoculum to feedstock ratio of 2:1 (VS based) for 30 days. Anaerobic conditions were maintained by supplying nitrogen gas to the reactor after feedstock was added.

【Results and discussion】

The effects of CO_2 addition in hydrothermal pretreatment (HTP) of CS for biomethane production at lower temperature conditions were investigated. The HTP of CS was assessed at temperatures ranging from 50 °C to 90 °C with CO_2 pressures of 3 and 5 MPa, along with a control group (CO_2 free). The findings revealed that CO_2-assisted HTP could effectively remove hemicellulose in CS. This removal, along with an increase in pore area and porosity of the pretreated sample, is likely to enhance the adsorption capabilities for microorganisms and enzymes on the lignocellulosic substrate, thereby improving the bioconversion process of carbohydrates present in CS.

The highest Substrate-specific methane yield (SMY) (n-m^3/kg VS) was achieved with the liquid fraction at a pretreatment condition of 90 °C under 5 MPa. Nonetheless, for AD purposes, utilizing the entire slurry is more advantageous, considering the overall mass balance.

This study has shown that implementing reduced tillage practices can lead to a decrease in CO_2 emissions. Furthermore, within the newly established AD steady conditions, it is feasible to manage the TS content of the digestate throughout the AD process. This control helps to stabilize the digestion process, resulting in high levels of methane production and organic matter degradation. Additionally, the study highlighted the positive impact of CO_2-assisted hydrothermal pretreatment on improving methane yield and the organic matter degradation in the AD process using CS. This study not only enhances our understanding of biomass-to-energy conversion processes but also provides valuable insights that benefit the bio-ecosystem, thereby contributing to sustainable bioenergy production.