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Methane production through anaerobic digestion: Participation and digestion characteristics of cellulose, hemicellulose and lignin

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HIGHLIGHTS

- Anaerobic digestion characteristics of lignocellulosic components are described.
- Hemicellulose was hydrolysed and acidified more quickly than cellulose.
- The biomethane potential of cellulose was higher than that of hemicellulose.
- Co-digestion of cellulose and hemicellulose had a synergistic effect on methane yield.
- Lignin caused more severe inhibition on methane yield of cellulose than hemicellulose.

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ABSTRACT

Lignocellulosic biomass is the most abundant natural resource with high biomethane potential. However, complex structure of lignocellulosic biomass has hampered the efficient utilization of this bioresource. Previous studies have investigated the overall anaerobic digestion performance of lignocellulosic biomass, but the individual participation of each lignocellulosic component during anaerobic digestion remained unclear. Thus, this study investigated the methane production characteristics of cellulose, hemicellulose, lignin and their mixtures along with the microbial communities involved in anaerobic digestion. The results showed that the biomethane potential of cellulose was higher than that of hemicellulose; however, hemicellulose was hydrolysed more quickly than cellulose, while lignin was very difficult to be digested. The higher concentrations of acetic, *n*-butyric and *n*-valeric acids hydrolysed from the hemicellulose resulted in a lower pH and more severe inhibition on methane production than that of cellulose, and the methanogenesis gradually recovered after pH adjustment. The co-digestion of cellulose and hemicellulose increased the methane yield and biodegradability compared to mono-digestions. The addition of lignin to cellulose brought more significant decrease in the methane yield of cellulose than that of hemicellulose. Substrate-related bacteria such as *Clostridium sensu stricto*, *Lutaonella*, *Cloacibacillus* and *Christensenella* showed higher relative abundance in cellulose digestate, and sugar-fermenting bacteria such as *Saccharofermentans*, *Petrimonas* and *Levilinea* were more rich in the digestate of hemicellulose. Moreover, methanogenic *Methanospirillum* and *Methanothrix* likely contributed to the methane production of cellulose, while aciduric methanogens from *Methanobrevibacter*, *Methanomassiliicoccus*, *Methanobacterium* and *Methanoculleus* contributed to that of hemicellulose. This study provides a deeper understanding of the mechanism in the bioconversion of lignocellulosic biomass during anaerobic digestion.

Abbreviations: AD, anaerobic digestion; AG, arabinogalactan; α-CE, α-cellulose; BD, biodegradability; CMY, cumulative methane yield; DMY, daily methane yield; EMY, experimental methane yield; GM, glucomannan; LI, akali lignin; MC, microcrystalline cellulose; OTUs, operational taxonomic units; RA, relative abundance; SEI, synergistic effect index; TA, total alkalinity; TAN, total ammonia nitrogen; TMY, theoretical maximum methane yield; TS, total solids; VFAs, volatile fatty acids; VS, volatile solids; XY, Xylan

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1. Introduction

Renewable energy is becoming increasingly important with growing concerns over rapid energy use and environmental pollution in the world [1]. Anaerobic digestion (AD) has been widely applied as a useful method for the bioconversion of complex substrates into renewable energy in the form of methane [2]. Lignocellulosic biomass, massively generated on earth, stands a good chance to serve as the most abundant natural resource for methane production through AD. However, most of the AD plants prefer using animal manure and food waste as feedstocks, rather than utilizing lignocellulosic biomass [2,3]. The main factor limiting the application of lignocellulosic biomass for methane production is the low hydrolysis rate caused by the complex and compact structure. Nowadays, how to improve the hydrolysis rate and methane production of lignocellulosic biomass has become a hotspot [4,5].

Lignocellulosic biomass mainly contains cellulose (15–99%), hemicellulose (0–85%) and lignin (0–40%) [4]. Since different types of lignocelulosic biomass have a different composition, therefore they respond differently during the AD process. Many previous studies have looked into the overall methane production performance and microbial community structure in the AD of lignocellulosic biomass [6–10], however, clear analysis on the individual participation of each lignocellulosic component has been scarcely reported so far. The clarity on the participation of major lignocellulosic components during AD could be used to apply physical, chemical or microbial pretreatment more directly and effectively. The clearness on methane production process of individual lignocellulosic component and their mixtures could help to develop modelling tools for the prediction of methane production, and it could also help researchers to design co-digestion strategies more efficiently by mixing biomasses with different contents of cellulose, hemicellulose and lignin. Furthermore, the exploration of the functional microbes associated with the AD of individual lignocellulosic component is significant in improving the AD performance, targeting the degradation and utilization of specific lignocellulosic component for effective bioconversion of lignocellulosic biomass to biomethane.

In this study, cellulose, hemicellulose, lignin and their mixtures are used as substrates for AD, wherein daily methane yield (DMY), cumulative methane yield (CMY), volatile fatty acids (VFAs) accumulation and process stability were investigated, by which the participation and methane production characteristics of cellulose, hemicellulose and lignin during AD have been clarified. In addition, bacterial and archaeal communities involved in the AD of cellulose, hemicellulose and their mixtures have been analysed to elucidate the functions of microorganisms. Insights gained from this research would provide the foundation for future studies on the effective bioconversion of lignocellulosic biomass to renewable energy.

2. Materials and methods

2.1. Substrates and inoculum

Microcrystalline cellulose (68005882, Shanghai Urchem, China) and α-cellulose (C104841, Aladdin, China) were both sourced from cotton. Xylan (HWG25024, HWRK, China), glucomannan (S30903, Yuanye, China) and arabinogalactan (S30898, Yuanye, China) were used as representatives of hemicellulose. Alkali lignin (370959, Sigma-Aldrich, USA) was also used as a single substrate and co-substrate for AD to act as one of the lignocellulosic components. Inoculum was the anaerobic sludge from the Beijing Donghuashan Biogas Plant, which operates at large scale using continuous anaerobic digester treating swine manure. The inoculum was degassed for 20 days at room temperature (25–28 °C) to reduce its endogenous methane production [10].

2.2. Analytical methods

Total solids (TS) and volatile solids (VS) of the substrates and inoculum were analysed by standard methods [11]. The elemental compositions, including C, H, N, O and S, were tested by an elemental analyser (Vario EL cube, Elementar, Germany). A pH meter (Mettler Toledo, USA) was used to measure the pH value. By using a barometer (WAL Mess-und Regelsysteme GmbH, Germany), daily biogas production was calculated from the difference in the gas pressure inside the digesters. The biogas composition was measured daily at a constant time by using a gas chromatograph (GC) system (7890B, Agilent, USA), equipped with an analytical column (Agilent Hayesep Q) and a thermal conductivity detector. Operation temperatures of 60 and 220 °C were set for the column oven and the detector, respectively. The carrier gas used was helium at a constant pressure of 34.5 kPa. Digestate was centrifuged at 12,000 rpm for 5 min and the supernatant was filtered sequentially by a micro-filtration membrane of 0.45 and 0.22 μm to prepare the samples for the analysis of the VFAs and ethanol. Then the samples were quantified by a GC system (7890A, Agilent, USA) equipped with a DB-WAXETR capillary column (30 m × 530 μm × 1.0 μm) and a flame ionization detector. The operation temperatures of the column and detector were 50 and 250 °C, respectively, and high purity nitrogen was used as the carrier gas at a flow rate of 10 mL/min. The concentrations of total ammonia nitrogen (TAN) and total alkalinity (TA) were measured according to the instructions of the HACH test kits (HACH, USA) [10].

2.3. Experimental design

The total initial organic loading of mono-digestions and co-digestions was 15 g-VS/L. Cellulose, hemicellulose and lignin were digested in separate bottles to compare the AD characteristics of different lignocellulosic components. Microcrystalline cellulose (MC), α-cellulose (α-CE) and alkali lignin (LI) were water-insoluble, while xylan (XY), glucomannan (GM) and arabinogalactan (AG) were water-soluble. However, all these substrates at a high concentration of 15 g-VS/L in water formed suspensions. To investigate the interactions between lignocellulosic components, co-digestions of cellulose with hemicellulose, cellulose with LI and hemicellulose with LI at a mixing ratio of 1:1 based on VS content were performed, as well as a co-digestion of MC, XY and LI (1:1:1). Each digester was inoculated with the inoculum at a substrate-to-inoculum ratio of 1, which was also calculated on the basis of VS content [12]. In order to mix the substrates, inoculum and water, the digesters were shaken manually for 2 minutes at the time of installing the AD experiment. Hydrochloric acid (0.6 mol/L) was used to adjust the initial pH of the fermentation matrix to 7.0 ± 0.1. Every digester had a total volume of 1 L and was filled with deionized water to reach a working volume of 0.4 L. Then, each digester was purged with high purity nitrogen gas for 2 min to eliminate O_2, and the digester was sealed with rubber stopper and covered with perforated lid. Four blank digesters only containing 15 g-VS/L of inoculum with the same working volume were used as control group, and the DMY of substrates was calculated by subtracting the methane production of control group from test group. All AD tests were conducted in quadruplicate at 37 °C for 50 days, and the digesters were shaken at a speed of 130 rpm every day for 1 min before testing the biogas pressure. When the AD process was complete, the digestate was sampled to analyse the bacterial and archaeal communities.

2.4. Calculation of biogas production

The biogas pressure was quickly tested before and after releasing biogas from anaerobic digesters under a constant temperature of 37 °C (310.15 K). The biogas yield was calculated based on the pressure difference and was adjusted to the standard conditions (0°C, 101 kPa) by using ideal gas law shown below [13]:

$$V_{biogas} = \frac{P \times V_{head} \times C}{R \times T}, \quad (1)$$

where V_{biogas} represents the daily biogas yield (L), P stands for the