



Enhancing anaerobic digestion performance and degradation of lignocellulosic components of rice straw by combined biological and chemical pretreatment

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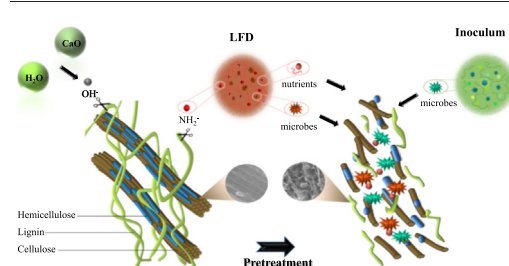
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HIGHLIGHTS

- The different combination bio-chemical pretreatment methods were investigated.
- CaO-LFD bio-chemical pretreatment achieved the best effect.
- Physico-chemical structure and AD performance of rice straw were improved.
- Methane yield of CaO-LFD was $274.65 \text{ mL} \cdot \text{gVS}^{-1}$, 57.56% more than that of the control.

GRAPHICAL ABSTRACT



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ABSTRACT

In order to determine eco-friendly pretreatment method, the combination of different pretreatment reagents such as: CaO, ammonia solution (AS), liquid fraction of digestate (LFD), CaO-AS and CaO-LFD were used in this study. The features of physico-chemical structures and anaerobic digestion (AD) performance of rice straw were investigated using different combined biological and chemical pretreatment methods. The results showed that CaO-LFD bio-chemical pretreatment achieved the best effect among different pretreatment conditions. The removal rate of lignocellulosic components from CaO-LFD pretreated rice straw was 20.73% higher than that of the control sample. The ether and ester bonds between lignin and hemicellulose were ruptured during pretreatment. Moreover, the methane yield from CaO-LFD pretreated rice straw was $274.65 \text{ mL} \cdot \text{gVS}^{-1}$, which was 57.56% more than the control. Compared with the untreated rice straw, T_{80} decreased by 42.86%. CaO-LFD combined pretreatment has advantages as both biological and chemical pretreatment, which complement each other to improve the degradation of the rice straw. Meantime, AD performance was improved and excellent economic viability was achieved. Therefore, this study provides sustainable insight for exploring efficient pretreatment strategy to stabilize and enhance AD performance for further application.

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1. Introduction

China is one of the world's largest energy consuming countries which accounted for 23% of global energy consumption and contributing 27% to global energy demand growth in 2016, according to the BP Statistical Review of World Energy (2017) (BP, 2017). To reduce the

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reliance on unsustainable fossil fuels and to get relieved from the concern over global climate change, countries have to look for sustainable and renewable energy resources to enable partial or full replacement of using fossil fuels (Ren et al., 2016). Lignocellulosic biomass is a carbon-neutral source for renewable energy generation, which became an increasingly important renewable raw material for biogas production in anaerobic digestion technology (Khor et al., 2015). China is one of the largest agricultural countries in the world, producing averagely 180 to 270 million tons of rice straw in every year (Dai et al., 2017). Even though there are several methods available for reutilization of rice straw, the rate of utilization has been still less than 50% (Yun et al., 2014). The remaining amount of straw often dumped or burned in open environment, causing tremendous environmental problems (Zhang et al., 2016). Anaerobic digestion (AD) of rice straw has been considered as one of the environmentally friendly ways while treating organic wastes to produce renewable energy in the form of biogas and control greenhouse gas emissions. Rice straw is one of the most abundant lignocelluloses (LCH) crop residues, which exhibit great potential to produce biogas with high methane content through AD. It has been confirmed that AD is an attractive technology for simultaneous clean bioenergy production and waste treatment (Li et al., 2017).

However, the inherent characteristics of LCH made it resistant to enzymatic degradation by anaerobic microbes (Zheng et al., 2014). So that pretreatment of rice straw prior to AD has been proven to be necessary to improve biodegradability and biogas production (Wei et al., 2015). Chemical pretreatment is believed to be the promising method to improve the bioconversion of cellulose and destruct the rigid lignin structures in various physical, chemical, biological and combined pretreatment methods (Paudel et al., 2017; Romero-Güiza et al., 2017). Though NaOH was the most common pretreatment reagent used in alkaline pretreatment, the high concentration of Na^+ in reactors may cause cation toxicity which can inhibit the activity of methanogens (Geng et al., 2016). In addition, sodium ions in anaerobic discharges might be environmentally harmful as they can lead to negative impacts by increasing soil salinity (Zheng et al., 2014). In contrast, Ca^{2+} have less inhibiting effect than Na^+ , environmentally friendly and economically feasible, which make CaO a better choice to alkaline pretreatment agent (Zhang and Jahng, 2010). It has been reported that using CaO (4% and 10%) for the pretreatment of microalgae was taken place at different temperatures (25 °C, 55 °C and 72 °C) enhanced proteins and carbohydrates solubilization by 31.4%–32.4% and increased the biochemical methane potential by 25% (Solé Bundó et al., 2017). The carbon to nitrogen ratio (C:N) in straw is mostly in the range of 53:1–78:1, which is believed to be not suitable for anaerobic digestion where the optimum working condition is in between the C:N ratio of 20:1–30:1 (Li et al., 2015). However, using pretreatment reagents like ammonia solution (AS) could be used to enhance biodegradability and increase the content of nitrogen which can decrease the C/N ratio to its optimum level (20:1–30:1). Besides, in aqueous form, AS causes swelling of the rice straw which brings significant morphological changes (Kim et al., 2016). It was reported that 4% AS pretreated corn stover produced 26.7% higher total biogas yield compared to the untreated corn stover and consumed 80.6% of cellulose and 68.52% of hemicelluloses (Yuan et al., 2015).

The liquid fraction of digestate (LFD) pretreatment is a sustainable biological pretreatment to take cooperative action on structurally inhibiting lignocellulosic materials. The good thing while using LFD is the availability of abundant specific lignocellulose-degrading microbes, inorganic anions and cations (NH_4^+ -N and NO_3^- -N) and organic substance (amino acids, protein, sugar and so on) (Akhiar et al., 2017). As a result, recirculating LFD for pretreatment in AD process increases the rate of bioconversion of the substrate, reduces the amount of LFD discharged to the environment, and also promote anaerobic biogasification efficiency of lignocellulose. Compared with the chemical pretreatment (ammonia solution and NaOH), LFD

pretreatment achieved the same effect in the co-digestion of cattle manure and corn stover, which achieved 25.40%–30.12% higher cumulative methane production and 14.48%–16.84% higher volatile solid removal rate than that of untreated corn stover (Wei et al., 2015). Similarly, it had been found that the total lignin, cellulose and hemicellulose contents after LFD pretreatment were reduced by 8.10%–19.40%. Moreover, during AD of corn stover, 70.4% more biogas production and 41.7% shorter technical digestion time were achieved than the untreated stover (Hu et al., 2015). On the other hand, large amount of digestate was generated from the anaerobic digesters at the end of the process, causing serious environmental pollution. The re-using of LFD for pretreatment is a good option, which can reduce the discharge of LFD, avoid possible pollution and minimizes the cost of pretreatment.

Rate of biodegradability of biomasses could be affected by several factors, as a result using only single pretreatment method could not provide efficient results due to its limited functioning mode (e.g. NaOH primarily targets lignin, but not hemicellulose) and intrinsic disadvantages (Haghighi Mood et al., 2013). Therefore, none of pretreatment methods (physical, chemical, or biological) can be declared as a “winner”. In recent years, applying combined pretreatment incorporating two or more single pretreatment techniques are getting common (Zheng et al., 2014). Some studies have found that combined pretreatment approaches like alkaline-ultrasonic, bacterial treatment-NaOH and ensiling-fungal could significantly improve the degradation of biomass (Dai et al., 2015; Thomsen et al., 2016; Zhou, 2015). However, few researches have focused on the combined effects and economic feasibility of biological (LFD) and chemical (CaO) pretreatment.

To enhance further methane production, it is necessary to research physicochemical characterization of rice straw with the aforementioned pretreated methods. Therefore, the objectives of this study were: (1) to analyze the composition and structural changes of lignocellulose during different pretreatments; (2) to investigate the effect of combined biological and chemical pretreatment on the AD performance of rice straw; (3) to establish a sustainable pretreatment process for enhancing anaerobic digestion of rice straw.

2. Materials and methods

2.1. Raw material and inoculum

The rice straw used in this study was collected from Tianjin District, China. The rice straw was air-dried in open field and then ground to the size of 20-mesh by a knife mill (YSW-180, Zhengde, China). The inoculum for AD process was taken from a biogas station in the Shunyi District (Beijing, China). LFD for pretreatment was obtained from an anaerobic reactor which has been operated continuously for more than 1 year, where pig manure was used as feedstock. LFD referred as the filtrate which passed through a 20-mesh sieve. The characteristics of rice straw, inoculum and LFD are listed in Table 1.

Table 1
Characteristics of rice straw, inoculum and LFD*.

| Items | Rice straw | Inoculum | LFD |
|---|--------------|--------------|--------------|
| TS (%) ^a | 93.29 ± 0.09 | 7.01 ± 0.04 | 2.35 ± 0.16 |
| VS (%) ^a | 83.78 ± 0.02 | 3.89 ± 0.02 | 1.22 ± 0.04 |
| TC (%) ^b | 40.83 ± 0.21 | 28.36 ± 0.79 | 29.18 ± 0.76 |
| TN (%) ^b | 0.80 ± 0.02 | 2.38 ± 0.08 | 2.09 ± 0.03 |
| C/N | 50.83 | 11.90 | 13.94 |
| Cellulose (%) ^b | 35.32 ± 0.56 | – | – |
| Hemicellulose (%) ^b | 28.92 ± 0.01 | – | – |
| Lignin (%) ^b | 7.88 ± 0.20 | – | – |
| NH_4^+ -N (mg·L ⁻¹) ^a | – | 536 ± 42 | 1400 ± 33 |

* Values are means ± SD (n = 3).

^a Content of fresh matter.

^b Content of dry matter.

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