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Integrated anaerobic digestion and algae cultivation for energy recovery and nutrient supply from post-hydrothermal liquefaction wastewater

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ABSTRACT

Post-hydrothermal liquefaction wastewater (PHWW), which contains approximately 80% of original feedstock resources, shows great potential to achieve sustainable development of an environment-enhancing energy system. A combination of anaerobic digestion and algae cultivation was proposed for methane recovery and nutrient supply from PHWW. Granular activated carbon (GAC) and ozone were used to enhance energy recovery from the PHWW. The results indicated that with GAC addition, the maximum methane yield increased by 67.7%–228 mL/g $COD_{removal}$. In addition, *Chlorella vulgaris* displayed optimal growth in a 5-fold diluted digestate with a 2.32 g/L maximum biomass content and 180 mg/(L·d) biomass production rate. The total energy yield was 565 kJ/g COD, which was 27.4 times higher than that without GAC. Integration of anaerobic digestion and algae cultivation, particularly with GAC addition during fermentation, is a feasible and advantageous process for energy recovery from PHWW.

1. Introduction

Biomass, which is currently the world's largest source of renewable energy, makes up more than 10% of the global energy supply (Van Doren et al., 2017). For example, in China, the annual output of agricultural straw can reach 740 million tons, which is equivalent to 317 million tons of standard coal in calories (Cao et al., 2017). Hence, development and utilization of biomass resources can greatly alleviate the depletion of non-renewable fossil fuels. Hydrothermal liquefaction (HTL) is a promising technology for converting wet bio-waste or biomass into crude oil (Biller and Roth, 2018; Cheng et al., 2018). However, a large quantity of aqueous phase, namely post-hydrothermal liquefaction wastewater (PHWW), is produced as a by-product of HTL. This high-strength wastewater accumulates most of the feedstock nutrients (approximately 80%) and a substantial amount of organics (up to 40%), which provides a significant opportunity for carbon and nutrient recovery (Nie and Bi, 2018; Zhang et al., 2018; Zhou et al., 2013).

Due to rich nutrient content of PHWW, algae cultivation using PHWW has been extensively studied. Jena et al. (2011) highlighted the possibility of using nutrient-rich PHWW as a growth media for the cultivation of microalgae. By adding PHWW to deionized water at a 0.2% v/v concentration, 0.52 g/L *C. minutissima* was obtained, this established the proof of concept for combing algae cultivation with PHWW for nutrients recycling. Biller et al. (2012) demonstrated the feasibility of using microalgae for treating PHWW. Edmundson et al. (2017) also quantified the maximum specific growth rates of algae on recycled nutrients derived from HTL processing by-products, which provided evidence for substantial improvement of large-scale algae biomass production. Additionally, a paradigm, referred to as the environment -enhancing energy (E^2 – Energy) system, which can capture carbon, recycle nutrient, and clean wastewater, was proposed by Yu et al. (2011). In this system, the PHWW treatment was integrated with microalgae cultivation to support multiple stages of biomass production and biofuel conversion, it was regarded as the key step to achieving such a large potential in bioenergy production. However, a high dilution ratio (10–500 times) was required to reduce the negative effects from inhibiting substances in PHWW, such as fatty acids, *N*-heterocyclic compounds and ammonia (Pham et al., 2013; Zheng et al., 2017). The

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high dilution ratio resulted in the requirement of abundant fresh water and a larger reactor. Moreover, the biomass concentration and productivity obtained in the diluted PHWW were lower.

Compared to microalgae strains, anaerobic microbes seem to be more tolerant to the mixture of chemical compounds in PHWW (Xu et al., 2018; Zhou, 2010). Moreover, gas can be produced during the anaerobic digestion process. These advantages make it a feasible and advantageous process for energy recovery from PHWW. A successful anaerobic digestion occurred at a 6.70% concentration of PHWW in the research of Zhou et al. (2015), which resulted in a biogas yield of 500 mL/g $COD_{removal}$ and 53.0% energy recovery efficiency (Zhou et al., 2015). Additionally, Fernandez et al. (2018) operated a semi-continuous anaerobic digestion reactor to degrade the PHWW using *Tetraselmis* and *Chlorella* algae, 327 mL/g $VS_{Tetraselmis}$ and 263 mL/g $VS_{Chlorella}$ of methane were produced under 20–30% (v/v) PHWW with clarified manure. Nitrogen and phosphorus in PHWW would be left in the digestate because anaerobic digestion is known to reduce negligible amounts of nutrients (Wang et al., 2010). Thus, further treatment, such as microalgae cultivation, is required to enhance nutrients recovery from PHWW.

The integration of anaerobic digestion and algae cultivation for energy recovery and nutrient supply from PHWW was proposed in this study. Compared to previous research, anaerobic treatment would potentially function as a detoxification step by using PHWW for algae cultivation (Zheng et al., 2017). Organic pollutants, such as phenol and benzene, which are known to be toxic to algal growth, can be degraded during digestion, thus, a lower dilution ratio of PHWW can be used for algae cultivation. Besides, except for the nitrogen and phosphorus in PHWW can compensate for the cost for algae cultivation, organic carbon in PHWW can also be recovered as methane-rich biogas. To mitigate the effects of inhibitors from the PHWW and to enhance energy and nutrients recovery efficiency (Lee et al., 2009; Pham et al., 2013; Younker and Walsh, 2015), a detoxification method using granular activated carbon (GAC) and ozone was employed to treat PHWW in this study. Fig. 1 shows the schematic of the process. The objective of this study was 1) to illustrate the effect of GAC and ozone on the anaerobic digestion and algae cultivation from PHWW, 2) to reveal the energy recovery and nutrient supply efficiency from PHWW by integrating anaerobic digestion and algae cultivation, and 3) to maximize the economic value of the PHWW to contribute to the sustainable development of the E^2 -Energy system.

2. Materials and methods

2.1. Post-hydrothermal liquefaction wastewater

The original PHWW was collected from a pilot-scale HTL experiment. The HTL reactor was operated using swine manure at a reaction temperature of 270 ± 10 °C and with a solids content of 13%. The retention time of the HTL reaction was 1 h. The PHWW was stored in a cold chamber at 4 °C and filtered using a 0.45 μm filter before use. The characteristics of the PHWW have been reported in previous research (Yang et al., 2018).

2.2. Anaerobic digestion

2.2.1. Inoculated sludge

A mesophilic anaerobic inoculum was collected from a full-scale anaerobic digester operating at the Urbana-Champaign Sanitary District (Urbana, Illinois, USA), and was enriched using the methods reported in previous research (Si et al., 2016). The inoculum was washed by DI water three times before use.

2.2.2. Fermentation of post-hydrothermal liquefaction wastewater

Assays were performed in 160 mL serum bottles that were hermetically sealed with stoppers. The operating conditions for anaerobic digestion of PHWW are shown in Table 1. At the beginning of anaerobic treatment, 20 mL of sludge were added to the bottles, and 0.5 g $NaHCO_3$/g COD was added as buffer. The concentration of the fermentation was controlled at 10 g COD/L. Anaerobic digestion of raw PHWW (RP) was conducted as the control group (C). Twenty grams per liter of GAC (Calgon F-400) were added to the bottles to ferment with the PHWW as the GAC-treated PHWW group (AC). The concentration of GAC was chosen according to previous research (Zhou et al., 2015). PHWW treated with 2.1 mg O_3/mL was used for the anaerobic digestion as the ozone-treated PHWW group (O). The ozone dosage was applied based on our previous research (Yang et al., 2018), and was produced by a portable ozone producer (SATA 03601).

All bottles were placed in a water bath at 37 °C and were processed for 50 days. The gas volume was measured daily using a glass syringe, and gas content was measured using a gas chromatograph. Every seven days, a 1.5 mL liquid sample was extracted for chemical analysis. All of the experiments were performed in triplicate. At the end of the experiment, the digested liquid was collected for microalgae cultivation.

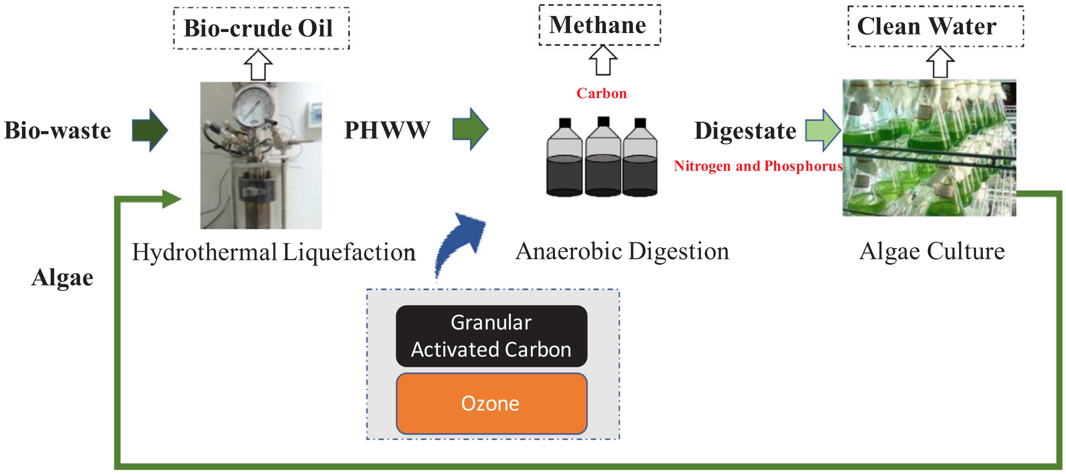

Fig. 1. The schematic of the process. HTL: Hydrothermal liquefaction; PHWW: Post-hydrothermal liquefaction wastewater.