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The anaerobic digestion of biologically and physicochemically pretreated oily wastewater

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HIGHLIGHTS

- The oil-degrading bacteria enhance the oil degradation in the oily wastewater.
- The ultrasonic pre-treatment improves the initial methane production.
- The citric acid maintains the methane production during the whole digestion stage.
- The oil-degrading bacteria, ultrasonic and citric acid improves the digestion.

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ABSTRACT

To enhance the degradation of oily wastewater and its biogas production, a biological–physicochemical pretreatment was introduced prior to the anaerobic digestion system. The digestion thereafter proceeded more efficiently due to the inoculation by oil degrading bacteria (*Bacillus*). A 2-stage pre-mixing is more effective than directly mixing. The effects on the methane production were also investigated by pre-treatment with ultrasonic (US) treatment, combined with citric acid (CA) addition. US pre-treatment was found to improve the initial methane production, and CA pre-treatment could maintain this improvement during the whole digestion stage. Pre-mixing *Bacillus* at 9 wt.% inoculation, combined with US for 10 min and a CA concentration of 500 mg/L provided the optimum conditions. The most effective enhancement of methane yield was 1100.46 ml/g VS, exceeding that of the control by 280%. The change of coenobium shape and fatty acid content further proved that such pretreatment of oily wastewater can facilitate digestion.

1. Introduction

Restaurant organic waste is the main source of oily wastewater. Such as the daily food waste (FW) yield exceeded 20 million ton in China in 2010 (Wang et al., 2013). Lipids are the major organic component in wastewaters from many industries including dairies, edible oil refineries, slaughterhouses, and wool scouring factories. Food waste, to a large extent, and domestic wastewater to a lesser extent, contain significant proportions (about 5–38%) of oil (Labatut et al., 2011). But neither landfill nor incineration of FW offer an effective solution to their disposal, the former for reasons of the lack of available land, rising landfill fees and environmental pollution, the latter for reasons of higher water content of FW which could cause large amount energy consumption. Since both disposal methods are environmentally unfriendly, the use of anaerobic digestion (AD) to treat the semi-liquid FW is gaining interest (Appels et al., 2011), encouraged by the energy-gains for producing biogas (Appels et al., 2008a; Li et al., 2010; Zhang et al., 2011), providing an economically attractive application (Dewil et al., 2006).

AD is an extensively adopted means for reducing bio-solids' volume and producing biogas (Appels et al., 2008b; Wan et al., 2011). The technology has been given great attention recently for its important role in clean energy production and pollution reduction (Li et al., 2009). With the high content of moisture, oil and salt, FW is perishable, as well as biodegradable. Through anaerobic bacteria, lipids can be degraded by hydrolysis and β-oxidation to acetate and hydrogen, which in turn are converted to methane. A large amount of methane can be produced from lipids because they have a high carbon content. Li et al. (2011) reported that oil exhibits higher biogas production than kitchen waste as co-substrate. Theoretically, 1 g of glycerol trioleate, a common lipid in nature, can be converted to 1.08 L of methane at standard temperature and pressure (STP) by biodigestion, whereas 1 g of glucose can be converted to only 0.37 L. Although the methane production is considerable and the high oil content can be considered a good substrate for the anaerobic digestion process (Palatsi et al., 2011), the anaerobic digestion of oily wastewater is still problematic (Resch et al., 2011),

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due to the low solubility of oil, poor biodegradability, and surface action whereby biomass flocs are shielded and do not participate in the biochemical reactions (Chu et al., 2002; Pereira et al., 2005).

In view of the above problems, oil removal should proceed the further wastewater treatment. A moderate removal of oil can be achieved by dissolved air flotation. Previous studies have reported that adding enzymes like lipase and cellulase into anaerobic digesters treating food-processing wastewater resulted in an improved digestion (Romano et al., 2009; Valladão et al., 2007). Pretreatments such as physical, chemical and biological treatment were previously demonstrated to increase the biogas yield, with 90% of methane being produced in the initial stage (Romano et al., 2009), thus proving the applicability of the treatment. Gaur et al. (2010) applied coarse sand and gravel/coarse sand bioreactors to the pretreatment of simulated turkey fat-containing wastewater and draw a conclusion that the reactors facilitates the conversion of slowly degradable organic matter. It is also found that the additional pea gravel cap could act as a sieve to entrap organic matter including fat globules from the wastewater and slightly improved the relative removal of COD. This is a combination of physical and biological treatment applied to digestion of oily wastewater. Cavaleiro et al. (2013) applied combinations of different temperatures, NaOH and lipase from Candida rugosa to promote wastes hydrolysis and obtained a higher COD solubilisation (66% for greaves; 55% for rinds). The biochemical methane potential of raw greaves and rinds was 707 ± 46 and 756 ± 56 L CH_4 kg^{-1} VS, respectively. This is a combination of chemical and biological treatment applied to digestion of oily wastewater. Valladão et al. (2007) used a pool of hydrolysis enzymes with 21.4 U/g lipase activity, in the solid-state fermentation by Penicillium restrictum of waste from the Orbignya oleifera (Babassu) oil processing: 0.1% of the enzymatic pool was used in the pre-hydrolysis stage for a feed with 1200 mg oil and grease, present at a ratio of about 1:1. The COD removal efficiency was 85%, with a biogas yield of 175 ml after 4 days. These papers have provided an effective way to treat oily wastewater by AD, but the low solubility of oil is still unsolved.

As possible solution to the poor solubility by traditional means, the present study investigates the use of pre-processing stages such as ultrasonic and thermo-chemical pretreatments to enhance the oil degradation (Apul and Sanin, 2010; Elbeshbishy et al., 2011; Kvesitadze et al., 2012; Saha et al., 2011): the pretreatment of the oily feedstock can break the long chain or network structure, and transform the structure of complex organic materials into small molecules. This process leads to the increase of the feedstock surface area, improves the contact of micro-organisms with the feedstock, and enhances the solubility of oil and grease resulting in more readily biodegradation, thereby improving its efficiency. Salsabil et al. (2009) treated the waste sludge by sonication at 108 MJ kg TS^{-1} and observed an increase of the instantaneous specific soluble COD uptake rate and the biogas yield. They concluded that combining sonication and anaerobic digestion provided a sludge reduction of about 80%, leading to an increase of biogas production due to the increase of available soluble COD. But this improvement of biogas is fairly limited by the poor solubility of oil. Neither simple biological pretreatment nor simple physicochemical pretreatment could achieve a relatively high effect of oil degradation. Despite the moderate biodegradable nature of the wastewater, its biological treatment by traditional means has so far not been successful, mostly hampered by the presence of grease-type feedstock.

On this basis, further references to oil degradation are very scarce. The present research will provide possible solutions of improved treatment by two combined actions: (i) the introduction of oil degrading bacteria (*Bacillus*) in the pretreatment stage; (ii) the use of US and CA as the emulsification method on the basis of (i).

2. Methods

2.1. Characteristics of inoculum and feeds

Diluted activated sludge (~20,000 mgSS/L) was collected from the wastewater treatment plant of Shunyi District, Beijing, China. After settling in the secondary clarifier, the activated sludge was thickened using a thickening table to reach a moisture content of 85–90%. The concentrated sludge was air dried for 20 days at ambient temperature, collected in sealed plastics bags and stored at −20 °C for later experiments. Then it was acclimatized to a synthetic high-oil containing feedstock during a period of 6 months. The artificial oily wastewater used in this study contained 6 g-VS waste oil per L water. The waste oil was separated from the raw wastewater by decanting. The raw wastewater was collected from the canteen of the Beijing University of Chemical Technology. After the separation, the waste oil was stored at 4 °C for later experiments. Waste oil was the only organic matter in the artificial oily wastewater. The characteristics of waste oil were shown in Table 1.

2.2. The oil degrading bacteria

The oil degrading bacteria were separated from the activated sludge taken from the anaerobic digester by the oil separation medium. The oil in the separation medium was from the oily wastewater. Basic tests had proved that the bacteria are kind of bacillus which can use oil as a carbon source. The other specific characteristics need to be further tested.

2.3. Experimental setup and procedure

2.3.1. Experimental setup

The oily wastewater was treated in 3 parallel experiments, each comprising 2 glass cylinder digesters, immersed in a thermostatic water bath at 35 ± 1 °C. The working volume of each digester was 0.8 L. Each digester was sealed with a rubber lid and two sampling lines were installed: one line under the liquid surface for the collection of liquid samples; another was located above the surface in the headspace and attached to a gas-tight bag for biogas collection. In each digester, anaerobically treated activated sludge in the amount of 30 ± 1 gTS was used as inoculum. The digesters were incubated at 35 ± 1 °C for 10 d, during which the yield and composition of biogas, the pH value were measured, and the contents of oil and fatty acids were detected by gas chromatography. To assure anaerobic conditions, the head space was purged with inert gas (300 ml-N_2/min) for 5 min.

2.3.2. Anaerobic digestion experiments

Table 2 shows the experimental conditions of the batch anaerobic digestion. The oil concentration throughout the experiments was 6 g-VS/L, typical of a restaurant food waste. The corresponding HRT was shown in Table 2. The mix type was designed as follows: no mix, direct-mix and pre-mix, to determine the optimum mix type. Annotation "Direct-mix" represents experiments where *Bacillus* were mixed with the activated sludge and together

Table 1
Characteristics of waste oil (mean value ± standard deviation).

Parameter	Cavaleiro et al. (2013)	This study
Total solids (%)	88 ± 0	96 ± 0
Volatile solids (%)	86 ± 0	95 ± 0
Oleic acid (C18:1) (%)	41 ± 2	30 ± 5
Palmitic acid (C16:0) (%)	29 ± 2	7 ± 1
Stearic acid (C18:0) (%)	30 ± 4	4 ± 1
Linoleic acid (C18:2) (%)	–	57 ± 6
Linolenic acid (C18:3) (%)	–	2 ± 1