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Research article

Combining microwave irradiation with sodium citrate addition improves the pre-treatment on anaerobic digestion of excess sewage sludge

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This study investigated the synergistic effect of sodium citrate (SC; $Na_3C_3H_5O(COO)_3$) and microwave (MW) treatment on the efficiency of the anaerobic digestion of excess sewage sludge. In terms of the methane yield, an increase of the digestion's efficiency was observed. Taking into account the cost for the MW energy supplied to the system, the optimum treatment conditions were a MW energy input of 20 MJ/kg TS and a SC concentration of 0.11 g/g TS, obtaining a methane yield of 218.88 ml/g VS, i.e., an increase of 147.7% compared to the control. MW treatment was found to break the sludge structure, thereby improving the release of extracellular polymeric substances (EPS) and volatile fatty acids (VFAs). The treatment of sodium citrate further strengthened the breakage of loosely bound extracellular polymeric substances (LB-EPS) and tightly bound extracellular polymeric substances (TB-EPS). The increased VFA content stressed the improved digestion by this pretreatment. Furthermore, the preliminary economic analysis showed that at this point in the research, only operational but no financial gains were achieved.

1. Introduction

The disposal of excess sewage sludge from wastewater treatment plants (WWTP) is of increasing importance, since (i) the amount produced on yearly basis is steadily growing, and (ii) its final disposal possibilities are limited. The stringent legislation regarding the presence of heavy metals and toxic non-biodegradable organics leads to the fact that only a small amount of this sludge can be used in agriculture as fertilizer (Hanć, A. et al., 2009). For final disposal, large amounts of sludge must be combusted in dedicated sludge incinerators, co-digested with other types of waste, or dried and utilized as a secondary fuel in cement kilns or coal fired power stations. Due to the high water content and poor mechanical dewatering properties of the excess sludge, with typically only 20–25% DS-content achievable, these final disposal routes are only feasible after an energy consuming pre-drying step (Appels et al., 2008). It is reported that sludge disposal represents over 60% of total treatment plant operating expense (Appels et al., 2008; Neyens and Baeyens, 2003), which is a major drawback for waste water treatment.

As a treatment process of waste sludge, anaerobic digestion (AD) is generally recognized as a stable, and energy-yielding process with the production of biogas containing 55–70% CH_4 (Appels et al., 2008). AD can moreover (i) decrease the total sludge volume for disposal, (ii) improve the sludge dewaterability and (iii) stabilize the sludge (Appels et al., 2011). There are four major steps in anaerobic digestion: hydrolysis, acidogenesis, acetogenesis and methanogenesis. The hydrolysis phase is often regarded as the rate-limiting step in the whole process (Appels et al., 2010; Abelleira et al., 2012). However, pretreatment of waste activated sludge can accelerate this step. In this regard, a great number of studies investigated pre-treatment methods to disintegrate the sludge and to release organic matter into the aqueous sludge phase, aiming to accelerate the hydrolytic stage and produce more biogas. Various treatment methods have been conducted for sludge digestion on pilot-scale and lab-scale. They comprise ultrasound (Pham et al., 2009; Bougrier et al., 2005), mechanical disintegration, (Zhang

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et al., 2012), chemical action (Dewil et al., 2007), ozone oxidation (Ahn et al., 2002), Fenton peroxidation (Kaynak and Filibeli, 2008), bio-hydrolysis (Gopi Kumar et al., 2012), thermal treatment (Val del Río et al., 2011), alkaline pre-treatment (Lin et al., 2007) and thermo-alkaline pre-treatment (Uma Rani et al., 2012), and combinations thereof Most results not only showed an improvement of biodegradability and sludge dewatering with accelerated hydrolysis, but also revealed a reduction of pathogens and foaming.

Applying microwaves as sludge pretreatment process is quite novel and promising. Microwaves are electromagnetic waves (oscillating frequency about 0.3–300 GHz) that can promote structure alteration of micro-organisms and organic matter in the sludge, and further generate heat (Climent et al., 2007). In addition to their thermal effect, microwaves also cause an additional a-thermal effect by altering the dipole orientation in the polarized side chains of the cell membrane molecules. The a-thermal effect results in the cleavage of the hydrogen bonds, so that the floc matrix of the micro-organisms disintegrates and the structure of the present proteins break (Park et al., 2004, 2010; Tang et al., 2010; Eskicioglu et al., 2007a). The effect of microwave radiation is influenced by four parameters, i.e. radiation time, strength (frequency), penetration depth and concentration (Park et al., 2010). Microwave technology has been widely used in several aspects like synthesis, extraction, digestion and stabilization (Huang et al., 2015), for many advantages of microwave such as: 1) reduction of energy consumption, 2) reduction of reaction time, 3) environmentally friendly heating (Huang et al., 2015; Wang et al., 2015).

MW pre-treatment can contribute to an augmented biogas yield, as shown by several studies. The summary of these studies have been listed in Table 1.

Other previous research has investigated how microwave treatment influences other characteristics of sludge, like pathogen destruction (Kuglarz et al., 2013), energy efficiency (Tang et al., 2010), and solubilization (Chang et al., 2011).

It is proven that MW causes the breakage of the sludge structure and the release of organics while improving its dewaterability. However, the cell structure of microorganisms could be excessively broken when too much radiation power or too long radiation time is used. This excessive breakage will lead to the over-leakage of intracellular organics. Hence, a combination treatment like microwaves with sodium citrate could avoid these drawbacks, since less energy needs to be applied to the sludge in order to reach the same effect.

Sodium citrate is generally used as anticoagulant of blood in pharmaceuticals and as acidity regulator in food. In this study it can be used as a cation-binding agent for sludge treatment. It mainly affects the mineral components of EPS (Ca^{2+}, Mg^{2+}, Fe^{3+}) by complexation, according to the following reaction:

$$EPS - M^{n+} + RCOONa \rightarrow EPS - Na^{+} + (RCOO)_n M$$

This loosens the EPS structure and makes it prone to a more direct disrupture when MW is applied. (Ebenezer et al., 2015b) used sodium citrate as deflocculant to remove the EPS of sludge and reduced the irradiation energy of microwave by 1.4 times. This provided a way for sodium citrate to pretreat the excess sludge.

The goal of this study is to improve the efficiency of MW pretreatment for subsequent AD. The addition of sodium citrate prior to MW treatment disrupts the sludge biomass structure and releases COD and EPS into the aqueous phase. The outcome of the study is assessed through biomethane potential tests, COD and VFA measurements and characterization of the EPS components.

2. Materials and methods

2.1. Sludge sampling and characterization

The excess sludge was obtained from the buffer tank of a municipal WWTP located in Mechelen, Belgium. After sampling, the sludge was immediately stored at 4 °C prior to further treatment and analysis. The characteristics of the raw sludge are shown in Table 2.

2.2. Sodium citrate pre-treatment

Dosage optimization for sodium citrate was conducted in 60 conical flasks with a volume of 250 mL, filled with 100 g sludge. The range of SC addition was 0.05–0.14 g/g TS, according to (Ebenezer et al., 2015b). The sludge and sodium citrate were continuously mixed for 2 h at 70 rpm. After the reaction was terminated, the microwave pre-treatment was applied, as described further. Soluble COD (SCOD) concentrations were determined before and after the pre-treatment. The experiments were conducted in triplicate.

2.3. Microwave pre-treatment

A microwave synthesis reactor (Anton Paar, Monowave 300, max power of 850 W) was used to treat the sludge; the treatment was conducted in a glass vial filled with 30 mL of sludge and different energy levels were applied. The power was kept constant at 850 W. The irradiation duration was varied between 3 and 30 s, thus leading to a specific energy (SE) input ranging from 10 to 40 MJ/kg TS, calculated according to the formula below.

Table 2
Characterization of the raw sludge.

Parameter	Value
pH	7.56 ± 0.02
Total Solids (TS) (g TS/kg sludge)	48.5 ± 0.2
Volatile Solids (VS) (g VS/kg sludge)	33.3 ± 0.1
Soluble COD (SCOD) (mg O_2/L)	2519 ± 122
Total COD (TCOD) (mg O_2/L)	84,263 ± 1043
Soluble proteins (mg/L)	17,379 ± 1060
Soluble carbohydrates (mg Glu-eq/L)	3066 ± 159

Table 1
Summary and comparison of the previous studies on MW treatment.

Reference	Treatment method	Results
Beszédes et al. (2011)	MW pretreatment (5 W/g and 30 min)	Methane content increased up to 60%, COD solubility enhanced to a maximum of 57%.
Zhou et al. (2010)	MW treatment (700 W and 9min)	SCOD concentration increased 1.8–4.0-fold.
Yu et al. (2009)	MW treatment (900 W and 60s)	EPS concentration (1500–2000 mg/L) and disintegration (1.5–2%) accompanied.
Yu et al. (2010)	MW treatment (900 W and 60s)	Settle ablity increased from 39.58 mm/h to 45.08 mm/h.
Ebenezer et al. (2015a)	MW pretreatment (14,000 kJ/kg TS)	COD solubilization increased 28%, and reduction in SS increased 38%.
Zhang et al. (2016)	MW pretreatment (600 W–100 °C)	The methane production was 338.44 mL CH_4/gVS.