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Mesophilic anaerobic digestion of pulp and paper industry biosludge–long-term reactor performance and effects of thermal pretreatment

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ABSTRACT

The pulp and paper industry wastewater treatment processes produce large volumes of biosludge. Limited anaerobic degradation of lignocellulose has hindered the utilization of biosludge, but the processing of biosludge using anaerobic digestion has recently regained interest. In this study, biosludge was used as a sole substrate in long-term (400 d) mesophilic laboratory reactor trials. Nine biosludge batches collected evenly over a period of one year from a pulp and paper industry wastewater treatment plant had different solid and nutrient (nitrogen, phosphorus, trace elements) characteristics. Nutrient characteristics may vary by a factor of 2–11, while biomethane potentials (BMPs) ranged from 89 to 102 NL CH_4 kg^{-1} VS between batches. The BMPs were enhanced by 39–88% with thermal pretreatments at 105–134 °C. Despite varying biosludge properties, stable operation was achieved in reactor trials with a hydraulic retention time (HRT) of 14 d. Hydrolysis was the process limiting step, ceasing gas production when the HRT was shortened to 10 days. However, digestion with an HRT of 10 days was feasible after thermal pretreatment of the biosludge (20 min at 121 °C) due to enhanced hydrolysis. The methane yield was 78 NL CH_4 kg^{-1} VS for untreated biosludge and was increased by 77% (138 NL CH_4 kg^{-1} VS) after pretreatment.

1. Introduction

Biorefinery concepts, where wastes and/or by-products are used as a resource, have attracted interest in recent years. In many cases, the pulp and paper industry has resolved its water pollution issues by treating its wastewater with an aerobic activated sludge process (Stoica et al., 2009; Meyer and Edwards, 2014). However, wastewater treatment produces large volumes of waste-activated sludge (referred as biosludge). Stoica et al. (2009) reported a yearly biosludge production of about 2900–4000 t/mill as total solids (TS) for three Swedish pulp and paper mills. As a consequence, sludge management is one of the major costs (up to 50–60%) of the wastewater treatment process (Mahmood and Elliott, 2006; Meyer and Edwards, 2014). At present, biosludge is often dewatered and incinerated, which is not energetically favorable due to the high energy consumption of dewatering. Further, the solid content of the dewatered biosludge remains low (18–50% TS), decreasing the heating value (Stoica et al., 2009). Incineration may not be the best utilization option if nutrient recovery from biosludge is desired, as nitrogen is lost in the incineration process and phosphorus recovery from ashes has proven challenging, mainly due to impurities (Reijinders, 2014). Recently, pulp and paper biosludge has been suggested for use in algae cultivation after anaerobic digestion (AD) in microalgae-utilizing biorefinery concepts (Kouhia et al., 2015).

In addition to methane production, the advantage of AD for sludge treatment is that the costly dewatering step is not necessarily required. AD is a conventional technology used around the world to stabilize municipal wastewater treatment plant sludges but has rarely been applied for pulp and paper industry biosludge. Pulp and paper industry biosludge has very different characteristics compared with municipal biosludge, particularly its high content of lignocellulosic material, which hinders the anaerobic degradability of the former. The lignin and cellulose contents of pulp and paper biosludge have been reported to be 36–50% and 19–27% TS, respectively, while these contents are usually <1% TS in municipal biosludge (Meyer and Edwards, 2014). The characteristics of wastewater and biosludge may vary between mills (e.g., according

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to the pulp and paper processes, raw materials, and wastewater treatment procedures used on site) (Bayr and Rintala, 2012; Ekstrand et al., 2013). However, the raw material mixture and operational parameters both in pulp and paper mill processes and wastewater treatment (e.g., nutrient dosing in wastewater treatment) are also likely to change over a longer period of time. The literature data are scarce, but it is possible that the characteristics of biosludge even from the same mill may vary over time and may potentially affect the AD if applied for sludge treatment.

Because of its low degradability, methane yields from biosludge with a high lignocellulosic content are usually low. Meyer and Edwards (2014) reviewed the biomethane potentials (BMP) reported for pulp and paper industry biosludges, and only two out of thirteen BMPs exceeded 100 L CH_4 kg^{-1} VS. Karlsson et al. (2011) studied biosludges from six Swedish pulp and paper mills, and the BMPs were 43–155 NL CH_4 kg^{-1} VS after 20 days incubation. In comparison, BMPs for municipal biosludge are often around two times higher, 200–250 L CH_4 kg^{-1} VS (Girault et al., 2012; Wang et al., 2014). The low methane yield and slow degradability requiring a long hydraulic retention time (HRT) have hindered the economical sustainability of pulp and paper industry biosludge AD (Mahmood and Elliott, 2006).

Various pretreatment methods have been screened to enhance the methane production of lignocellulosic biosludge, with thermal pretreatments being among the most promising technologies (Wood et al., 2009; Saha et al., 2011; Bayr et al., 2013). Thermal pretreatments for pulp and paper industry biosludges have been suggested to require temperatures above 150 °C, which particularly improve the hydrolysis of hemicellulose (Hendriks and Zeeman, 2009; Fernandez-Cegri et al., 2012). Lignin solubilization begins at 160 °C, but inhibitive phenolic compounds are also formed from lignin (Hendriks and Zeeman, 2009). Enhanced methane production from pulp and paper industry biosludge has been demonstrated at 150 °C (Bayr et al., 2013) and with microwave pretreatment at 75–175 °C (Saha et al., 2011). Several studies with municipal biosludge show that thermal pretreatment at temperatures <150 °C (e.g., at 121 °C) can also improve methane production (Bougrier et al., 2008; Carrére et al., 2010), but lower pretreatment temperatures are rarely studied for pulp and paper industry biosludge. A lower pretreatment temperature would reduce the input energy and prevent the formation of phenolic compounds from lignin.

Most previous AD studies with pulp and paper industry biosludges have been conducted with batch BMP assays. Only a few continuous studies have reported on the AD of pulp and paper industry biosludges without dewatering (Meyer and Edwards, 2014), although at least one full-scale reactor has been treating dewatered pulp and paper mill biosludge in Norway (Kepp et al., 2000). Compared to batch assays, continuous studies offer better understanding of the effect of HRT and the organic loading rate (OLR) on the AD process with possibly varying biosludge properties. Long-term studies could reveal the effect of varying biosludge properties on AD and the potential adaptation of microorganisms to inhibitive substances (Chen et al., 2008).

In this study, which was conducted using a mesophilic AD process and pulp and paper industry biosludge, the following three objectives were set: 1. Determine the long-term performance of mesophilic AD of biosludge in relation to varying substrate characteristics and different HRTs; 2. Find out how thermal pretreatment temperature affects methane production potential; and 3. Specify how implementation of thermally pretreated biosludge affect the performance of continuous AD. Finally, the filtered digestate produced in this study was used for algae cultivation as reported elsewhere (Polishchuk et al., 2015).

2. Materials and methods

2.1. Biosludge and inocula

Biosludge originated from a wastewater treatment plant that treats pulp and paper industry wastewater. Incoming wastewater at this treatment plant included a minor fraction (<10% of volume) of municipal wastewater. During the 400-day study period, a new biosludge batch (~70 L in a 100-L container) was obtained every second month, for a total number of nine batches. The solid concentration of feed sludge was increased from the second biosludge batch onwards to achieve higher organic loading in reactor trials. The solid concentration was increased by settling; the biosludge was kept in a 100-L container for 24 h, after which the liquid fraction (ca. 50% of the volume) was removed. The biosludge was stored at 7 °C before use.

The inoculum for the reactor trials and pretreatment screening BMP assay was mesophilically digested municipal sewage sludge from the Viinikanlahti Wastewater Treatment Plant (Tampere, Finland). All other BMP assays were conducted using digestate from the present experimental reactors (collected on days 136, 205, 289, and 400) as the inoculum.

2.2. Pretreatments

Thermal (at 80 °C, 105 °C, 121 °C, and 134 °C) pretreatments were screened to improve the degradability of biosludge. In the low thermal pretreatment condition, 500 ml of sludge in a loosely (not gas tight) closed 1-L glass bottle was warmed to 80 °C in a water bath (7 min) and subsequently kept at 80 °C in an incubator for two hours. The sludge was allowed to cool at room temperature (two hours). In the thermal pretreatment group, 500 ml of biosludge were autoclaved (KSG Sterilisatoren GmbH) in a loosely closed 1-L glass bottle at three different temperatures. The temperature was increased to 105 °C, 121 °C, and 134 °C after 36, 45, and 50 min, respectively, and kept there for 20 min before cooling to room temperature (about three hours). The pressure increased to 2.2 bars (gauge pressure) in 30 min with all treatments (back-up pressure with air and self-generated steam pressure) and was slowly released within 30 min while the sludge cooled.

2.3. BMP assays

BMP assays were conducted in 120-ml serum-bottles with a working volume of 60 ml at 35 °C as described by Kinnunen et al. (2014). A $VS_{substrate}$:$VS_{inoculum}$ ratio of 0.5 was used. The BMPs were determined for four of the nine biosludge batches (batches 3, 5, 7, and 9) using adapted inoculum from semi-continuous experimental reactors used in this study. In the pretreatment screening assays, the BMPs were determined for biosludge after thermal pretreatments using inoculum from a municipal wastewater treatment plant. In addition, a comparative BMP assay was conducted using digestate from the reactor trials as the inoculum (adapted inoculum).

2.4. Semi-continuous reactor trials

Three parallel 6-L, semi-continuous completely stirred tank reactors (CSTRs) (with a working volume of 5 L) were run for 400 days at 35 °C. A mechanical timed mixer (30 rpm) was on for 15 min and then off for 15 min during days 0–325; after that, the mixing was changed to continuous to avoid observed sludge flotation and gas tube clog up. The reactors were fed five days per week through a tube on the top of the reactor. Prior to feeding, the digestate was removed from the bottom of the reactor (7–10% less than the