Inter-stage thermophilic aerobic digestion may increase organic matter removal from wastewater sludge without decreasing biogas production

Sasha D. Hafner, Johan T. Madsen, Johanna M. Pedersen and Charlotte Rennuit

ABSTRACT

Combining aerobic and anaerobic digestion in a two-stage system can improve the degradation of wastewater sludge over the use of either technology alone. But use of aerobic digestion as a pre-treatment before anaerobic digestion generally reduces methane production due to loss of substrate through oxidation. An inter-stage configuration may avoid this reduction in methane production. Here, we evaluated the use of thermophilic aerobic digestion (TAD) as an inter-stage treatment for wastewater sludge using laboratory-scale semi-continuous reactors. A single anaerobic digester was compared to an inter-stage system, where a thermophilic aerobic digester (55 °C) was used between two mesophilic anaerobic digesters (37 °C). Both systems had retention times of approximately 30 days, and the comparison was based on measurements made over 97 days. Results showed that the inter-stage system provided better sludge destruction (52% volatile solids (VS) removal vs. 40% for the single-stage system, 44% chemical oxygen demand (COD) removal vs. 34%) without a decrease in total biogas production (methane yield per g VS added was 0.22–0.24 L g^{-1} for both systems).

Key words | aerobic treatment, anaerobic treatment, biogas, COD, VS, wastewater sludge

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INTRODUCTION

Thermophilic (55–60 °C) aerobic digestion (TAD) has been shown to be an effective treatment for wastewater sludge. As a stand-alone treatment, TAD can remove up to 60% of volatile solids (VS) or chemical oxygen demand (COD), and significantly reduce pathogens (Layden *et al.* 2007; Lloret *et al.* 2012). It has also been used in a two-stage configuration where TAD serves as a pre-treatment for anaerobic digestion (e.g., Ward *et al.* 1998; Jang *et al.* 2014). This combination of aerobic and anaerobic treatment may increase organic matter removal compared to either process alone, but would be expected to decrease methane production through loss of degradable substrate during pre-treatment (Ward *et al.* 1998; Dumas *et al.* 2010). A few studies report an increase in either anaerobic degradability of the organic matter remaining after TAD (Dumas *et al.* 2010) or, rarely, an increase in overall CH_4 production (Jang *et al.* 2014). However, most evidence suggests that TAD pre-treatment reduces CH_4 production through consumption of degradable organic matter that would otherwise be converted to methane, possibly combined with a decrease in its anaerobic degradability (Ward *et al.* 1998; Hasegawa *et al.* 2000; Rennuit *et al.* 2018).

Recent batch experiments have shown that an inter-stage configuration, where TAD is applied between two anaerobic stages, may be more effective than pre-treatment (Rennuit *et al.* 2018). With this configuration, the most degradable organic matter is converted to CH_4 in the first stage, preventing its oxidation and loss by TAD. Additionally, TAD treatment of the more recalcitrant organic matter in the first stage effluent seems to solubilize organic matter and increase its anaerobic degradability, resulting in a small (<3%) increase in overall CH_4 production (Rennuit *et al.* 2018).

Despite some promise, questions about inter-stage TAD treatment remain. With this configuration, the second anaerobic reactor may receive only poorly degradable material, which has previously been through two reactors (one anaerobic and one aerobic). Additionally, the feed to

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the second anaerobic reactor contains dissolved oxygen after treatment by the TAD reactor. Can the system provide stable performance under these conditions? Also, in batch trials, it is difficult to separate the contribution of inoculum from substrate. Would the same increase in CH_4 production be observed in a continuous system or would TAD decrease CH_4 production? In this study, our objective was to use semi-continuous reactor systems to compare a three-stage system with inter-stage thermophilic aerobic digestion with single-stage anaerobic digestion based on organic matter removal and CH_4 production.

METHODS

Reactor systems

Two laboratory-scale systems were used to compare single- and three-stage treatment (Figure 1). The single-stage control system (1S) consisted of one mixed 10 L (reacting volume) mesophilic anaerobic reactor. The three-stage inter-stage system (3S) included two similar anaerobic reactors (3S-1 and 3S-3) coupled with a smaller 670 mL (reacting volume) thermophilic aerobic reactor (the TAD reactor, 3S-2).

The anaerobic reactors (1S, 3S-1, 3S-3) were cylindrical, with an internal diameter of 280 mm and a height of 360 mm (22 L total volume). Continuous mixing was provided by two impellers, each with four flat blades, and was sufficient for preventing any surface crust or mat formation. Three electric heating elements fixed to a stainless steel base plate maintained a temperature of 37 °C. The TAD reactor (3S-2) was a 3 L fermenter, and was aerated with compressed air through perforated stainless steel tubing at a constant rate of

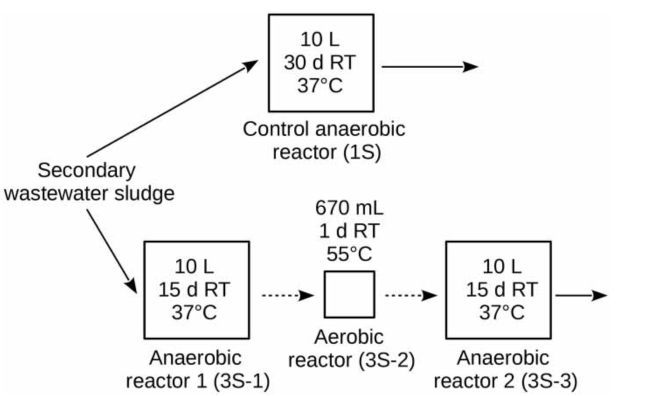

Figure 1 | The two laboratory-scale reactor systems compared in this work: control single-stage (1S) (top) and three-stage inter-stage system (3S) (bottom). Volumes are reacting volumes. Transfers shown with a dashed line were done using pumps, while all others were manually carried out three times per week.

0.25 L kg^{-1} min^{-1} (L of air standardized to 1 atm and 0 °C per kg reacting mass). This aeration rate was selected based on previous studies. Hasegawa *et al.* (2000) used aeration rates of 0.08 and 0.4 L L^{-1} min^{-1}, while Jang *et al.* (2014) used much higher rates of 10–40 L L^{-1} min^{-1}. The TAD reactor was insulated and heated to 55 °C by an electric hotplate. Mixing was by three flat blade impellers (1,100 rpm).

Target retention times (RT) were 30 days for 1S and 31 days for the 3S system (15 days for 3S-1 and 3S-3, and 1 day for 3S-2), but were generally lower (see Results and discussion). Feed for both systems was the same secondary wastewater sludge (waste activated sludge) collected from a municipal wastewater treatment plant (Ejby Mølle, Odense, Denmark). Sludge was collected every 4 weeks or more frequently and stored below 5 °C between feedings. Manual feeding of 1S, feeding of 3S-1, and wasting from 1S and 3S-3 was done three times per week (every Monday, Wednesday, and Friday). Because of the short RT of the TAD reactor 3S-2, manual feeding was not practical. Instead, effluent was transferred from 3S-1 to 3S-2 automatically using a peristaltic pump (Qdos, Watson-Marlow, Ringsted, Denmark) every three hours. An identical pump was used to transfer sludge from 3S-2 to the final anaerobic reactor 3S-3. Wasting from 3S-2 was always done 10 min before feeding. Pumping rates were set based on mass.

Anaerobic reactors 1S and 3S-1 were started with a mixture of digestate and raw secondary sludge from a wastewater treatment plant (Ejby Mølle, Odense, Denmark) at a wet mass ratio of 1.4:1 (inoculum-to-substrate ratio on VS basis approximately 0.7:1). The TAD reactor 3S-2 was started with raw secondary sludge and no inoculum.

Sample collection and analysis

Feed dry matter (DM) and VS were determined weekly or every time a new batch of raw sludge was collected from the wastewater treatment plant, based on drying at 105 °C and combustion at 550 °C (APHA, AWWA, WEF 1998). Effluent from all three anaerobic reactors (1S, 3S-1, and 3S-3) was collected weekly and analyzed for DM and VS using the same method. Effluent pH was measured using an Orion ROSS electrode (Thermo Orion, MA, USA). COD of feed and effluent samples was measured in triplicate using Hach COD vials (Hach, Loveland, CO, USA).

Cumulative biogas production was measured using a peristaltic pump and pressure sensor system (Andtec, Ski, Norway). Biogas samples were collected weekly and analyzed for CH_4 and CO_2 using a gas chromatograph with a thermal conductivity detector (Agilent 7890A, column:

J&W 113-4332GS GASPRO, oven temperature 250 °C). Dissolved oxygen in the TAD reactor was measured using a Knick SE 706 Memosens electrode (Knick International, Berlin, Germany). Data collection commenced 2 weeks after starting the reactor systems.

Data analysis

All data analysis was done using R (R Core Team 2017). Biogas and CH_4 production were determined from measured volumes and composition using the cumBg function in the biogas package (Hafner *et al.* 2015) and expressed as dry volume at 0 °C and 101.325 kPa. Methane concentration was normalized for calculations (Richards *et al.* 1991).

Loading rates were determined based on mass. To estimate RT and calculate VS loading, an averaging interval of 1 week was used. Removal rates of VS and COD were calculated from point estimates of concentrations matched by day. Because feed composition changed significantly over time, apparent variability in removal efficiency will be inflated with this approach, but average values over long periods are expected to be accurate. A single set of DM and VS observations thought to be incorrect from day 39 were dropped because of unusually high variability and an implausible large drop in DM and VS in 1S effluent.

Paired *t*-tests were used to compare VS and COD removal between the two systems using the t.test function in the stats package (R Core Team 2017), with observations matched by day. Biogas composition (CH_4 concentration) was compared among the three anaerobic reactors using a single-factor analysis of variance (ANOVA) carried out with the aov function, followed by Tukey's HSD test done using the Tukey.HSD function in the stats package (R Core Team 2017). Methane yield (per kg fed VS) was calculated in two periods based on total VS loading and total CH_4 production during these periods. Period 2 was from 30 to 60 days, and period 3 60 to 97 days after starting the reactors. Because a different biogas meter was used for each anaerobic reactor, and small systematic biases are likely, we made no attempt to compare biogas production using a statistical model.

RESULTS AND DISCUSSION

System performance

The three-stage system (3S) had consistently greater organic matter removal, as quantified by both VS ($P = 2 \times 10^{-8}$, $n = 10$) and COD ($P = 0.001$, $n = 10$). Based on point estimates, VS removal (mean ± standard deviation) was 52 ± 12% for 3S and 40 ± 12% for 1S. Removal of COD was 44 ± 17% for 3S and 34 ± 17% for 1S. This difference may be due to preferential removal of more oxidized components during TAD or error in VS measurement due to loss of volatile fatty acids (VFAs) or other volatile compounds. The first anaerobic stage 3S-1 accounted for more than half of total VS removed (60 ± 12%) in the 3S system, with the remainder removed by the following aerobic (3S-2) and anaerobic (3S-3) reactors. The increase in overall organic matter destruction suggests that aerobic and anaerobic communities utilize different components of sludge (Rennuit *et al.* 2018).

Methane production from 3S-1 was nearly twice as high as from 1S due to a higher loading rate, while production from 3S-3 was much lower (about 12% of the total 3S production; Figure 2). Total CH_4 yield from the 3S system was indistinguishable from the yield from 1S; both were about 0.23 L g^{-1} (per g VS; Table 1). However, the average CH_4 concentration in biogas from 3S was higher than it was for 1S: 85 ± 6.7% for 3S-1 (mean ± standard deviation) and 85 ± 4.5% for 3S-3 versus 81 ± 7.7% ($P = 0.001$ for 1S vs. 3S-1 and $P = 0.005$ for 1S vs. 3S-3 based on Tukey's HSD test). The high CH_4 concentration in 3S-3 could be due to three factors: removal of inorganic carbon by stripping in the TAD reactor, low biogas yield, and a slightly elevated pH compared to the 1S reactor. The last two processes increase the relative fraction of inorganic carbon that remains in the aqueous phase (Hafner & Rennuit 2017).

System stability

Composition of the feed varied over time: DM ranged from 33 to 63 g kg^{-1}, VS from 24 to 46 g kg^{-1}, and COD from 39 to 71 g kg^{-1}. (VS was generally close to 73% of DM, and the COD:VS ratio was generally close to 1.6.) RT dropped below target values for all three anaerobic reactors, apparently due to spills, water vapor loss from the TAD reactor (3S-2), and increased feeding rates to provide effluent for methanogen activity tests (results not reported). However, despite a gradual drop in RT (below 10 days for 3S-1 for some weeks), neither biogas production nor the pH of the anaerobic reactors dropped significantly over time (Figure 2, Table 1).

The pH of the anaerobic reactors ranged from 7.3 to 8.5, while the pH of the TAD was almost always between 8.5 and 9.0 (Figure 2). VFAs were not measured, but it is possible that depletion of high VFA concentrations in the anaerobic reactors may have contributed to increases in pH following

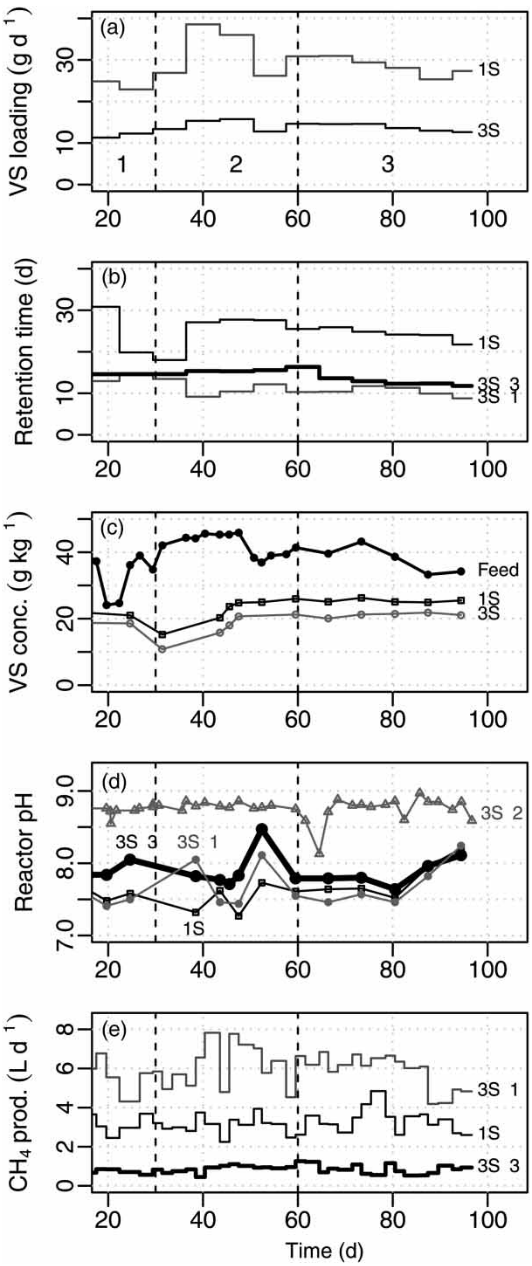

Figure 2 | Operating conditions and performance of single-stage (1S) and inter-stage (3S) wastewater sludge digestion systems. (a) VS loading, (b) retention time, (c) feed and effluent VS concentrations, (d), reactor pH, and (e) CH_4 production. Vertical dashed lines and the numeric labels in part (a) show the start-up period (1) and the two evaluation periods (2 and 3).

reductions in VS loading rate around day 50 and after day 80. Conversely, the aerobic reactor 3S-2 showed an unexplained drop in pH around day 65, which could have been caused by production of VFAs. Dissolved oxygen in the TAD reactor was usually between 3.5 and 4.5 mg L^{-1} (70% to 95% of saturation), but was, unfortunately, not measured during the pH drop. Biogas always had a high concentration of CH_4: never below 67% for 1S, 70% for 3S-1, and 75% for 3S-3, providing no evidence of inhibition or low methanogen activity. Results show that 3S-3 was capable of stable performance at a low RT despite receiving aerated feed with low degradability.

Application

Use of a full-scale inter-stage TAD reactor requires energy for aeration and possibly heating. The requirement for oxygen transfer to sludge can be estimated from COD destruction in the TAD reactor. Although it was not directly measured here, this value can be estimated as 10% of feed COD (with significant uncertainty), based on the difference in COD removal in the two complete systems (recognizing that CH_4 production did not differ). Given a feed COD concentration of 60 g kg^{-1}, oxygen transfer must be at least 6 kg m^{-3} (sludge volume basis). Assuming an oxygen transfer efficiency of 1 kg kWh^{-1} (0.28 kg MJ^{-1}) (Environmental Dynamics International 2017), the required energy supply for aeration would be 21 MJ m^{-3}. The high aeration rate used in the laboratory-scale reactor here would consume even more energy, and is probably not feasible for full-scale application. In comparison, energy recovered in biogas would be 360 MJ m^{-3} (based on values in Table 1). Although the metabolic heat produced from TAD can be sufficient for heating sludge to thermophilic conditions (i.e., autothermal TAD, or ATAD) (Layden *et al.* 2007), the short RT and low degradation in the TAD in this inter-stage system would limit this effect. The maximum energy required for heating (increase from 37 °C to 55 °C, with no heat generation or recovery) would be 75 MJ m^{-3}. These estimates are smaller than the energy that can be recovered through biogas, but still significant. A better understanding of the effect of aeration intensity on CH_4 production by the second anaerobic reactor (3S-3) would be useful.

Increases in RT or a switch to thermophilic anaerobic digestion are alternative biological approaches for increasing organic matter destruction. However, they are unlikely to provide as large an improvement as inter-stage TAD. Improvements due to RT increase are generally only high with large increases from a low RT, which are possible only with a reduction in capacity or construction of additional digesters. For example, Moen *et al.* (2003) found an increase from 56% to 66% VS destruction with a doubling of RT from 10 to 20 days for mesophilic anaerobic digestion. Nges & Liu (2010) reported an increase of 45% to 54% VS destruction with a change from 15 to 25 days' RT. Thermophilic anaerobic digesters may perform significantly better

Table 1 | Reactor feeding and biogas production during two evaluation periods[a]

		Sludge fed (kg)		VS fed[b] (g)		CH_4 (L)		CH_4 yield[c] (L g^{-1})	
Period	**Duration (d)**	**1S**	**3S**	**1S**	**3S**	**1S**	**3S**	**1S**	**3S**
2	30	10.1	22.9	431	978	93	213	0.216	0.218
3	37	13.8	28.0	528	1,074	124	249	0.235	0.232

[a]Period 2 was from 30 to 60 days after starting the reactors, and period 3 was from 60 to 97 days.
[b]Sludge volatile solids fed to each system.
[c]Methane production per g VS fed based on totals over each period.

than mesophilic digesters at short RTs, but for wastewater sludge, differences for RTs $\geq$ 15 days are small, if present at all (de la Rubia *et al.* 2002; Moen *et al.* 2003; Nges & Liu 2010).

CONCLUSIONS

Inter-stage thermophilic aerobic digestion combined with anaerobic digestion in a three-stage system can substantially improve organic matter removal from sludge while matching single-stage methane production. Even with low RTs, first- and second-stage anaerobic reactors provided relatively stable performance in this study. More work is needed to understand differences between aerobic and anaerobic sludge degradation, and the source of complementarity apparent in a combined system. Future work should focus on how RT and aeration intensity in the aerobic reactor affect organic matter destruction and changes in anaerobic degradability, and whether it is possible to significantly increase methane yield and the overall energy balance by using inter-stage thermophilic aerobic digestion.

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