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Pilot scale high solids anaerobic digestion of steam autoclaved municipal solid waste (MSW) pulp

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Steam autoclaving is an efficient method for the separation and near complete recovery of organics from MSW. The material produced by the autoclave contains a high concentration of solubilized food waste absorbed onto a lignocellulosic matrix. Reported here is the operation of a 1500 gal (5677 L) high solids anaerobic digester to digest this feedstock. Total solids (TS) reductions were high, 56%, and volatile solids (VS) and biodegradable volatile solids (BVS) reductions were 63 and 79%, respectively. Gas yields were also high, producing 248 L CH_4/kg VS fed or 393 L CH_4/kg VS destroyed at a methane content of 60%. Unique design elements such as hydraulic conveyance of material, in situ classification, and in-place buffering to maintain pH stability were tested and confirmed. The digestate passed all criteria for land application of biosolids in the US, but exceeded the EU limits for Cu, Ni, and Zn.

1. Introduction

The solid waste industry has made tremendous strides in recovering materials of value from waste that would otherwise be destined to go to landfill. Paper and paperboard products are recovered at a rate of 63%, and that which can be easily and economically source-separated is largely being recovered with urban areas achieving recovery of green waste at a nationwide efficiency of 60% [1]. Many communities have also been rapidly implementing source separated food waste programs for compost and energy recovery, especially from restaurant and institutional waste streams. Residential food waste programs are in their nascent phases of implementation in many municipalities but expanding to achieve greater source separation [2].

Even with these streams heavily targeted for diversion, the reported biogenic content of MSW in the US is still >50%, largely represented by paperboard, papers and food components as well as minor waste components that may be non-categorizable [1]. Full recovery is hampered by the difficulty of economically achieving complete organics recovery by standard waste handling methods. As recoverable materials are removed the specific waste composition is shifted but still tends to remain ~50% biodegradable [3]. After the greater portion of recycling is achieved, the residual is typically wet, very heterogeneous and not easily separable. In order to achieve complete organics recovery and ultimately landfill elimination through non-incineration, new approaches must be considered.

Test markets have been established that mandate significant reduction of organics in the landfill stream. The EU allows for 35% biodegradable municipal waste to go to landfill [4], metro Vancouver currently permits 25% food waste content [5], and Massachusetts requires facilities producing 1 ton food waste per week to alternatively dispose of this material [6]. Vermont is phasing in an organics ban by 2020 [7] and California has mandated 75% recovery of the MSW for re-use in the same time span [8]. There are undoubtedly multiple pathways to these goals, but as of now no clear cut alternative has been demonstrated that economically achieves the goal of complete diversion and effective utilization of the total waste stream.

Steam autoclaving is a technology that can capture essentially all of the biodegradable organics in the MSW stream. The process was first applied to MSW in the 1980's, known as the Holloway Process and used widely to treat medical waste, and involves steam and a temperature/pressure regime to sterilize the material, pulp the paper components, and solubilize the food waste [9,10]. After size separation, 90% of the organics are contained in a moist paper pulp

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(MSW pulp) [9,11–13]. There have been several instances of application of the technology beyond medical waste applications at commercial scale, largely in the UK and Australia [14]. However, no single purveyor has demonstrated full economic utilization and disposal of the product streams recovered from the process.

The steam autoclave is efficient at recovering biogenic materials resulting in large volumes to be further utilized. For instance 1 wet ton MSW will generate on average 0.20–0.25 ton dry MSW pulp (biogenics). There are several possibilities for re-use of the MSW pulp including as secondary fiber [15–17], compost [12,18], feedstock for ethanol or advanced biofuel production [19,20], and anaerobic digestion (AD) [13,21,22]. Of recent, focus has turned to the possibility that AD is well-suited to utilize the large volumes of organics produced by the autoclave for energy production.

The USDA has been demonstrating a 2-ton per load, batch steam autoclave system that stream-pasteurizes solid waste, similar to medical waste treatment, leaving it in a relatively uniform, pulped state. This pretreatment system is located at the Crazy Horse Canyon Landfill and is used to evaluate potential end uses for the resulting MSW pulp [11,23]. Based upon the material properties of the MSW pulp and the projected large volumes of daily production, a new AD reactor design was developed by the USDA to accommodate this substrate and hold it for the required retention time for complete biodegradation. The pilot 1500 gal (5677 L) steel frame reactor was installed at the landfill to demonstrate a simple system that completely biodegrades the pulp substrate with no internal moving parts and little required supervision. This manuscript will describe the design approach and discuss the operating parameters.

2. Materials and methods

2.1. Materials

The substrate for the pilot AD study was produced via a 2 ton-per-load, batch steam autoclave system located at the Crazy Horse Canyon Landfill in Salinas, CA and described elsewhere [11,23]. The autoclave, developed and patented by CR3 (Reno, NV) consists of a horizontal rotary vessel with helical heating baffles that have been designed to externally heat the autoclave's contents, impart shear forces to the waste material, and facilitate breakdown (especially of plastic garbage bags). Briefly, the autoclave is charged with up to 1 ton of untreated curbside MSW, water is added up to ~40% total moisture, the system is sealed, and contents pressure heated at 123 °C for at least 25 min. Discharged solids are separated by trommelling (size separation), which is approximately 90% efficient in the separation of the non-recyclable paper materials and represents ~60% of the total incoming MSW [23].

Fig. 1a is a schematic outline of the autoclave which, when the autoclave cycle is completed, discharges pressure-steamed MSW into a series of rotating trommel screens. The high solids collected in the "accepts" stream for particles smaller than 0.95 cm is then sent to the anaerobic digester. This particular stream of trommel accepts is a high solids material (~40–50% solids) with a large amount of readily biodegradable soluble materials absorbed onto the pulp, which is then fed on a semi-continuous basis to the high solids digester. Four autoclave runs were performed utilizing a total of 4 ton of unsorted residential MSW obtained from the Salinas Valley Solid Waste Authority (Salinas, CA) which produced a total of 739 kg of MSW pulp (dry basis) substrate.

The inoculant for the digester was obtained from the Marina Sanitary District (Monterey, CA) and was a combination of waste activated sludge (WAS) and trickling filter sludge obtained from a primary clarifier. This source was chosen because WAS has been shown to contain a higher count of anaerobic bacteria than secondary anaerobic sludge even though it is derived from an aerobic process, with the exception being aerobic systems utilizing pure oxygen [24,25].

The C:N:P ratio of for the 0.95 cm accepts stream was obtained in a previous detailed study [23] whereby autoclave operational parameters were correlated with the characteristics of each output stream, which included data on the C:N:P ratio for the 0.95 cm accepts stream. Due to the substrate's high carbon and low nitrogen and phosphorus ratios, YPD (yeast peptone dextrose) extract (Ohly Americas, Inc.) and trisodium phosphate (Savogran TSP) were utilized to adjust the carbon:nitrogen:phosphorus (C:N:P) ratio to a target [24] of C:N:P = 100:5:1. Additionally, small amounts of the following trace metals were added with each pulp feed to promote proper population growth [24–26]. Specifically, trace metals (with their supplier noted), were added to reach concentration in parts per million (ppm) dry weight basis of respective trace metals as follows: $FeCl_3{-}6H_2O$ (Sigma Aldrich) to 2100 ppm, $MgSO_4$ (Fisher) to 100 ppm, $CaCl_2$ (Sigma Alrich) to 2000 ppm, $ZnCl_2$ (Fisher) to 1000 ppm, $MnCl_2$ (Baker) to 80 ppm, $CoCl_2$ (Baker) to 0 ppm, $CuCl_2$ (Sigma Aldrich) to 100 ppm and $NaMoO_4$ (Mallinckrodt) to 2000 ppm.

2.2. Anaerobic system design

The pilot anaerobic digester was designed specifically to accommodate the MSW pulp produced by a steam autoclave. It addresses two significant considerations; (1) the feed, a lignocellulosic pulp matrix, can be very abrasive and is generally difficult to stir at increasing solids contents and (2) commercial scale steam autoclaving would produce a large daily volume of this lignocellulosic-rich feedstock. To minimize construction costs, a medium/high solids (20% solids) reactor basin design was utilized as outlined schematically in Fig. 1b. The digester was designed to

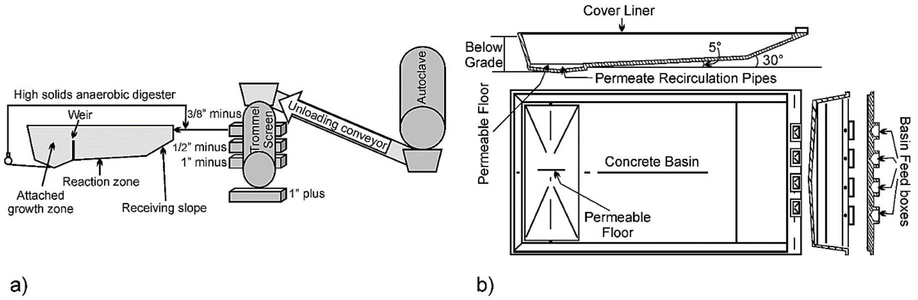


Fig. 1. a) Schematic of the 2 ton per load autoclave system for solids pretreatment, followed by solids fractionation via rotating trammel screens, and the 1500 gal high solids digester which handles the high solids from the 3/8" (~1 cm) screen. b) Schematic diagram that outlines the internal digester design with three zones, a highly-angled receiving zone, the flat-bottomed reaction zone, and the attached growth zone beyond the weir. This system is located at the Crazy Horse Landfill, Salinas, CA.

meet the following criteria:

1) Simple construction i.e., no internal moving parts to reduce building costs and avoid maintenance.
2) Able to accept multiple feedings while maintaining a hermetic seal and simultaneously fermenting the feedstock in-place.
3) Capable of inventorying large volumes of waste over long periods, even at production scale, and maintaining an extended retention time to completely reduce the biodegradable fraction.
4) Capable of operating with stability at landfill environment without requiring significant controllers or maintenance.

The pilot digester (Fig. 1) is rectangular (4 m × 1.5 m × 1.5 m), constructed of $^3/_8$" (0.95 cm) thick carbon steel, and insulated with ¼" (0.63 cm) black foam sheeting. Maximum working volume accounting for 1 ft (0.30 m) of headspace is 5500 L. The product of steam autoclaving was diluted to less than 5% solids content using recycled permeate from the digester in order to pump the slurry into the digester via a single cylinder reciprocating grout pump (Black-jack) through a duckbill check valve (Tideflex Technologies) designed to open under >6" (15.2 cm) H_2O column pressure and close again to maintain the hermetic seal.

The digester has no internal moving parts and can be considered to have three different zones based upon design characteristics (Fig. 1b). The receiving slope (~1 m length) has an angle (30°) severe enough to induce a velocity sufficient to carry the lighter weight organics in to the main reaction zone while at the same time allowing heavy debris to settle and classify separately from the substrate. The design is intended to "beach" the heavy inerts and allow for a final digestate with less contamination. The main reaction zone (~2 m length) is lightly sloped (5°) to allow for hydraulic drainage and migration of organics towards the front of the reactor. A weir separates the reaction zone from an attached growth zone (1 m length) comprised of whole oyster shells (Drakes Bay Oyster Co.) on a bed of river rock. The oyster shell (1.2 m depth) is intended to provide a medium for attachment of the biological growth and also to impart alkalinity to the reactor through leaching of the calcium carbonate under acidic conditions. The river rock (0.3 m depth) served to retain suspended solids and allow for withdrawal of permeate via an external $^1/_2$ hp Liberty centrifugal pump (Grainger) for re-circulation back to the digester either through an overhead shower system or to the feed inlet for dilution of feedstock. A stilling well at the pump inlet was utilized to measure the liquid height in the digester and estimate volume through calculations based upon reactor dimensions. Indirect heat exchange coils were embedded in the river rock to recirculate hot water from a 50 gal (189 L) tankless hot water heater at a maximum flow rate of 20 gpm (75 L/min) through the attached growth zone. Only the attached growth zone was regularly heated and this heat was partially re-distributed throughout the digester by periodic mixing as the recirculation pump was operated for a period of 15 min every 6 h. The digester was covered with a gas impermeable geotextile membrane and secured by a system of bars and clamps. Gas pressure on the cover was regulated by a back pressure valve set to 4" (10 cm) H_2O column and the gas line included a sample port for periodic gas sampling. Gas flow on-site was measured by a thermal mass flow meter (Sage Metering, Inc.) calibrated at 55% CH_4 content and was remotely monitored (SCS Engineering) once steady-state operation was achieved. For the demonstration, biogas from the digester was routed to the existing landfill flare.

2.3. Feed substrate biodegradation potential

The feed substrate delivered from the autoclave after screening, as reported previously [23], is a heterogeneous material consisting of lignocellulosic materials, glass shards, food wastes, ash, rocks dirt, and plastic shreds that pass through the screening filter. The composition was analyzed to estimate its biodegradable potential and calculate gas yields per unit of substrate available. Samples (100 g) were suspended in warm tap water (50 °C) at 4% solids with overhead mixing for a period of 30 min. The purpose of this treatment is to separate the pulp flocs, allow the curling induced by the autoclave to partially reverse, and the fibers to relax so that the solubilized organics can separate from the fibers. All soluble materials were washed free from the pulp, evaluated for their soluble COD (sCOD) content, and the dissolved organic fraction determined. The washed pulp was then subjected to component analysis including biodegradable volatile solids (BVS), determination of the acid insoluble material (lignin content) as well as concentration of monomers identified from the cellulose and hemicellulose. The hemicellulose monomers are distinct from cellulose, which are rich in six-carbon sugars, because they are generally derived from aribinoxylan polymers, which are richer in five-carbon sugars, including xylose and fructose.

For our purposes, the biodegradable volatile solids were defined as follows:

$$\text{BVS}\ (\%) = \text{cellulose}\ (\%) + \text{hemicellulose}\ (\%) + sCOD\ (\%) \qquad (1)$$

This can be seen as an estimate since it relies only on adding up quantifiable components, not direct measurement. Small debris such as plastics will characterize as volatile but not be biodegradable; the same is true for lignin. There will also be biodegradable organics that escape characterization. COD is reported in units of mg O_2 required to fully oxidize substrate per unit volume (typically mg O_2/L) and is not a direct weight of soluble organics but has been used adequately as a surrogate to estimate the biodegradable potential. The average for this study was 0.25 kg sCOD per kg dry MSW pulp material and this value is consistent with previous analysis of this waste stream.

2.4. Field testing

Water additions and removals were monitored by flow meter and recorded. Digester volume was measured by height and input. Temperature, pH, total dissolved solids (TDS), conductivity, and oxidation reduction potential (ORP, or redox potential) were recorded in the field using a handheld YSI PRO1030 Professional Plus Multiparameter Instrument (Model # PRO1030). Carbon dioxide content in biogas was determined by fyrite analysis (Bacharach).

2.5. Laboratory analysis

Total solids (TS) was determined by drying overnight at 105 °C, and volatile solids (VS) and ash (NVS, non-volatile solids) were determined by firing the dried residue in a muffle furnace at 550 °C for 6 h, all in accordance with standard methods [27].

Volatile fatty acids (VFAs) content was determined with an Agilent Technologies 1260 Infinity series HPLC equipped with a refractive index (RI) detector (Model #G1362A) and a Biorad 300 × 7.8 mm organic acid column (Aminex HPX-87H). The mobile phase (10 mM H_2SO_4) was delivered by quaternary pump (Model #G1311) at a flow rate of 0.5 mL/min. The column was maintained at 85 °C and the RID at 55 °C. Calibration was by the external standards method (C_1–C_6 acids).

Standard TAPPI methods were performed to determine the cellulose, hemicellulose, and acid insoluble lignin contents of the pulp feeds and the residue from AD as follows: TAPPI T203 − om9 for cellulose, TAPPI T223-083 for pentosan (hemicellulose), and

TAPPI T222 om-02/88 for insoluble lignin. Monomers from carbohydrate polymers were quantified with an identical HPLC system as described above but equipped with an Aminex HPX-87P column with water as the mobile phase and calibrated with external cell wall standards (glucose, xylose, galactose, arabinose, and mannose) [28].

Total alkalinity measured as mg/L as $CaCO_3$ (Method 8203) was determined using a HACH drop count titration kit. Soluble COD was performed with a Chemetrics COD kit using the mid-range tubes and heated with a HACH meter (model # CR2200). COD was measured using a UV–Vis spectrophotometer (Molecular Devices M2) and calibrated using 1000 and 10,000 ppm COD standards (A-7301 and A-7310, respectively, from Chemetrics) [28].

Fyrite analysis was supported by periodic sampling of digester biogas for analysis on a 6890N Agilent GC. Biogas was collected in a Tedlar gas bag (Sigma Aldrich) and immediately transferred to an evacuated 150 mL serum bottle (Fisher) with a butyl rubber stopper (Geo-microbial Technologies, Inc.) and a crimp seal (Fisher). 200 mL of biogas were transferred using a syringe and 20-gauge needle so that the bottle became pressurized after sampling to ensure exclusion of air. The GC was outfitted with a purged packed inlet, an Alltech Carbosphere 80/100 packed column and thermal conductivity detector. 300 μl of gas were sampled from the serum bottles with a glass syringe (Hamilton) and injected onto the column. The following conditions were used to separate CO_2 and CH_4: oven isothermal (240 °C); carrier gas (He) flow rate 40 mL/min; injector temperature (250 °C), column temperature (240 °C), detector temperature (250 °C).

2.6. *Elemental and micrometals analysis*

Ultimate analysis, total organic carbon (TOC), total Kjeldahl nitrogen (TKN), total phosphorus, and micrometals analyses were all performed by ALS Global (Tucson, AZ).

3. Results and discussion

3.1. *Start-up procedures*

The digester was sealed and pressure-checked to ensure no leaks were present at the cover, and 300 gal (1135 L) of well water were introduced to prime the system. The water was warmed to 37 °C by indirect contact in the attached growth zone for one week to stabilize the temperature of the reactor at which time the inoculant (1146 L) was introduced. Once again, the reactor was acclimated over a one-week period prior to the introduction of substrate. At this point the pH was 7.38 and the temperature was 36 °C. The inoculant contained 3.45% total solids (TS) and 72.8% volatile solids (VS) on a TS basis. As a result, the applied inoculant was 40 dry kg and consisted of 29 kg VS and 11 kg of non-volatile solids (NVS). Micrometal analysis was performed and incorporated into the system mass balance.

3.2. *Feed introduction*

MSW pulp substrate was introduced to the digester by 4 weekly plug feeds directly following pulp production by the steam autoclave. Ideally, the digester would be fed daily or multiple times daily; in this situation due to manual feed operation all pulp was introduced in a single weekly feed. The feed introduction was accomplished using a grout pump and the pulp was slurried by recirculating permeate from within the digester to dilute to ~5% solids. The action of the grout pump created sufficient pressure within the inlet feed pipe to open a check valve and allow the pulp to be introduced to the digester without exposing the digester to atmosphere or risking biogas escape. In theory at this point, the slurried organics would flow from the receiving slope (30° angle) to the main reaction zone while the heavy debris would “beach” on the receiving slope providing in situ classification and allow for a less contaminated end product. The pulp substrate dewaters to its water holding capacity as the permeate drains to the attached growth zone for re-circulation within the digester.

The four feeds comprised a total of 739 dry kg of MSW pulp at an average solids content of 34.8%. The total volatile solids added to the reactor were 550 kg (74.5% on TS) with the remainder being non-volatile solids (NVS). An additional total of 367 gal (1389 L) of water were also added with the pulp and 160 gal free water were added to achieve the target concentration of 20% TS. Over the feed period, the total organic loading rate (OLR) was 4.75 kg/m^3 d.

The VS component of the MSW pulp was determined to be 29.3% cellulose, 8.2% hemicellulose, and 33.4% soluble COD for a total of 70.9% biodegradable volatile solids (BVS) on a volatile solids basis, or 52.8% BVS on a total solids basis. For mass closure, the VS had a lignin content of 12.4% with 16.6% unidentified composition, which may or may not be convertible to biogas. All pulp feeds were analyzed for heavy metal contents and compared against threshold levels established for both the US and the EU (Table 1). In all cases on a concentration basis (mg/kg dry pulp) the MSW pulp had heavy metal contents well below those for the EPA 503 biosolids ceiling concentrations [29] and also below the PAS-100 PTE (potentially toxic element) limits for compost [30].

An attempt was made to target a carbon:nitrogen:phosphorus (CNP) ratio of 100:5:1.5 utilizing YPD extract at the time of solids introduction. However, the actual CNP (100:4.2:0.06) content of the pulp was not determined until after adjustments were made, and, as a result the nitrogen content, was slightly over the target value, while phosphorus content was under the targeted value. The former did not pose a problem, but low P could have resulted in lowered population growth especially during the early stages of operation. To assess population health, the food-to-microorganism ratio (F:M) was calculated regularly on a VS basis. F:M was calculated based on (1) influent flow; i.e., CBOD (mg/l) concentration, and (2) mixed volatile suspended solids concentration (mg/l). Thus, F:M measured the amount of incoming food (CBOD) divided by the density of microorganisms, and was measured regularly to ascertain the “health” of the microbe population, Early on in the operation the food-to-microorganism ratio (F:M) on a VS basis was 3.75:1 after the initial feed, but it rose to 19:1 after all feed introduction (assuming no population growth). A large biological deficit such as this requires bacterial population growth and results in a lower utilization rate. Additionally, lignocellulosic feeds require large populations of hydrolytic organisms [31] and thus the culture needed a period of time to acclimate to substrate.

Table 1
Heavy metals and potentially toxic element contents of each pulp feed and the MSW digestate compared to the EPA 40 CFR 503B and BSi PAS-100:2005 and PAS-110:2014 limits for land application.

	mg/kg, dry basis							
	As	Cd	Cr	Cu	Pb	Hg	Ni	Zn
Pulp feeds								
1	1.5	0.65	12.7	22.5	8.5	0.38	14.4	189
2	1.4	0.24	8.7	20	11.8	0.41	6.8	177
3	1.5	0.52	31.3	53.3	49	0.07	14.7	220
4	0.7	0.45	9.4	16.9	17.1	0.2	6.9	144
PAS-100		1.5	100	200	200	1	50	400
MSW digestate	5	1	37	217	40	0	233	636
PAS-110		1.2	80	160	160		40	320
EPA 503 CCL	41	39	1200	1500	300	17	420	2800

3.3. Acclimation period

The MSW pulp contains two very distinct substrates for anaerobic digestion, the lignocellulosic pulp matrix and the sCOD fraction derived from the solubilized food waste and other dissolved organic materials. As discussed above, the sCOD represents on average 25% by weight of the dry pulp feed and is readily convertible to volatile fatty acids and biogas. The lignocellulosic fraction is more slowly degraded and provides the longer term biogas yield. During the acclimation phase, in particular at high solids with very high F:M ratio, the concentration of VFAs can attain very high levels due to the limitations in dilution and the lack of sufficient methanogens to convert the VFAs. Increasing levels of VFAs derived from the sCOD will result in a depression of pH and can lead to inhibition of methanogenesis and a "sour" digester [32]. Through experience with the substrate it has been discovered that the addition of a solid alkaline media provides the early stage buffering capacity necessary to counteract the pH depression effects associated with high VFA levels. Leaching of the solid media at lower pH restores the carbonic acid equilibrium at neutral or slightly alkaline levels even in the presence of elevated VFAs. This is achieved preferably by a divalent counterion such as calcium that will bind the VFAs in their salt form, making the VFA readily available for transfer across the cell wall of the bacteria while also shifting the carbonic acid equilibrium [33]. The maintenance of neutral pH allows the bacterial population to replicate and acclimate without pH inhibition, and eventually the VFA concentration is reduced and the digester achieves steady operation.

Fig. 2 illustrates the operational parameters that were monitored throughout digester operation. At the initial feed, pH was 7.38. By the third pulp addition, the pH had dipped to 6.5 under the presence of elevated VFA levels (briefly >20,000 mg/L). It should be noted that the pH remained sufficiently high to maintain methanogenesis. At the point of lowest pH depression, the microbial population had established sufficiently high numbers so as to convert the VFAs at a rapid enough rate to begin diminishing the overall concentrations. In addition, the alkalinity had risen to levels ~5000 mg/L and remained constant throughout operation. The VFA levels continually decreased and by Day 30 of overall operation, there was <5000 mg/L VFA in solution as they were being converted as quickly as they were produced. In addition, during this period the pH also rebounded and stabilized ~7.6 for the entirety of operation. Soluble COD followed a similar trend, rising with elevated VFA and then decreased until a background level of ~6000 mg/L was established, likely representing soluble but non-convertible compounds.

3.4. Operational stability

As mentioned above, the addition of the oyster shells provided both surface area and buffering, allowing the pH to be maintained so that methanogenesis could continue unabated. The pH dipped briefly to 6.4 for a period of one week while biogas production continued and VFA levels peaked; by then the population adjusted and reduced VFA levels and in turn the pH rose. Beyond this point pH was stable ~7.6. Alkalinity was initially low but rose steadily with pulp addition and VFA production to a constant level of 5000 mg/L indicating that the digester had achieved a sufficient buffer capacity to support stable bacterial growth and operation. All other operational parameters (sCOD, TDS, conductivity) followed the same trend once steady-state was established within the digester. At operational stability monitoring was performed remotely with weekly site visits for sampling. A simple to operate system that is suitable for inclusion into a solid waste handling operation was thus demonstrated.

3.5. Biogas production and rates

Fig. 3a shows the total and daily biogas production curves. In total, the digester produced 265,666 L of biogas at a 60% CH_4 content, or 312 L biogas per kg TS in the digester. Throughout, the digester produced continuously ~2000 L of biogas per day which is equivalent to a rate of 2 L CH_4/kg VS/d. The digester was operated for a period of 105 days after final feed and produced 246 L CH_4/kg $VS_{applied}$ over the course of fermentation. The rate of conversion was lower than was anticipated which can be attributed to a number of factors observed throughout operation, specifically: (1) the operating F/M ratio (19:1) was much lower than is desirable and typically reported in the literature to affect rate [34]; (2) the phosphorus contents were low and possibly hindered sufficient population growth to achieve the desired gas production rates; and (3) temperature was likely a factor in the low rates of gas production. Mean daily ambient temperature in Monterey County during the summer months is ~65 °F (18 °C) and even though the reactor was insulated, the daily temperature fluctuations likely had an impact on rate. Overnight lows were typically 50–60 °F (10–16 °C) and observed biogas production was diurnal. In addition, only the attached growth zone was heated to mesophilic temperatures 37 °C (98 °F) with periodic mixing (15 min recirculation @ 75 L/m every 6 h) serving to provide heat to the rest of the reactor. As a result, sub-mesophilic temperatures in the reaction zone were observed throughout operation.

As a result, the digester had to be operated for a longer period of

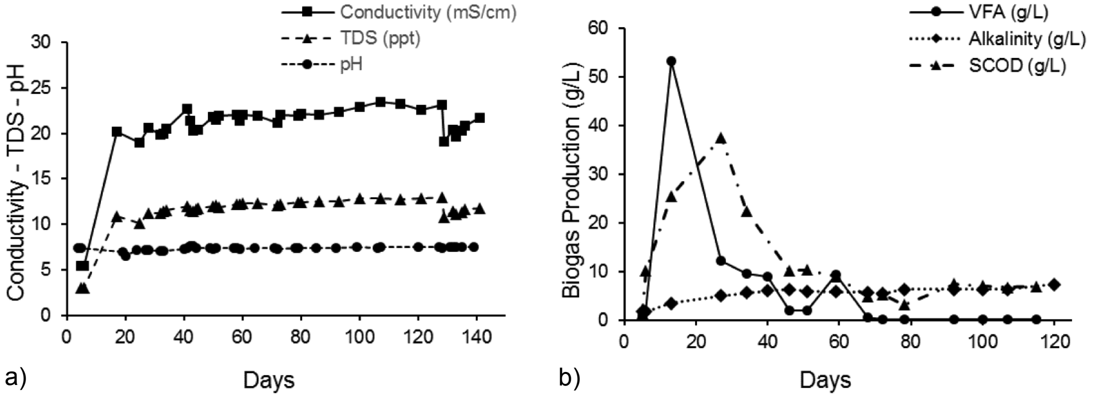


Fig. 2. The operational parameters of the digester over the 120 day run with a) conductivity, TDS and pH, and b.) VFA, alkalinity and SCOD as a function of time. Once the parameters reached steady-state after ~25 days, the digester was monitored remotely and required little attention.

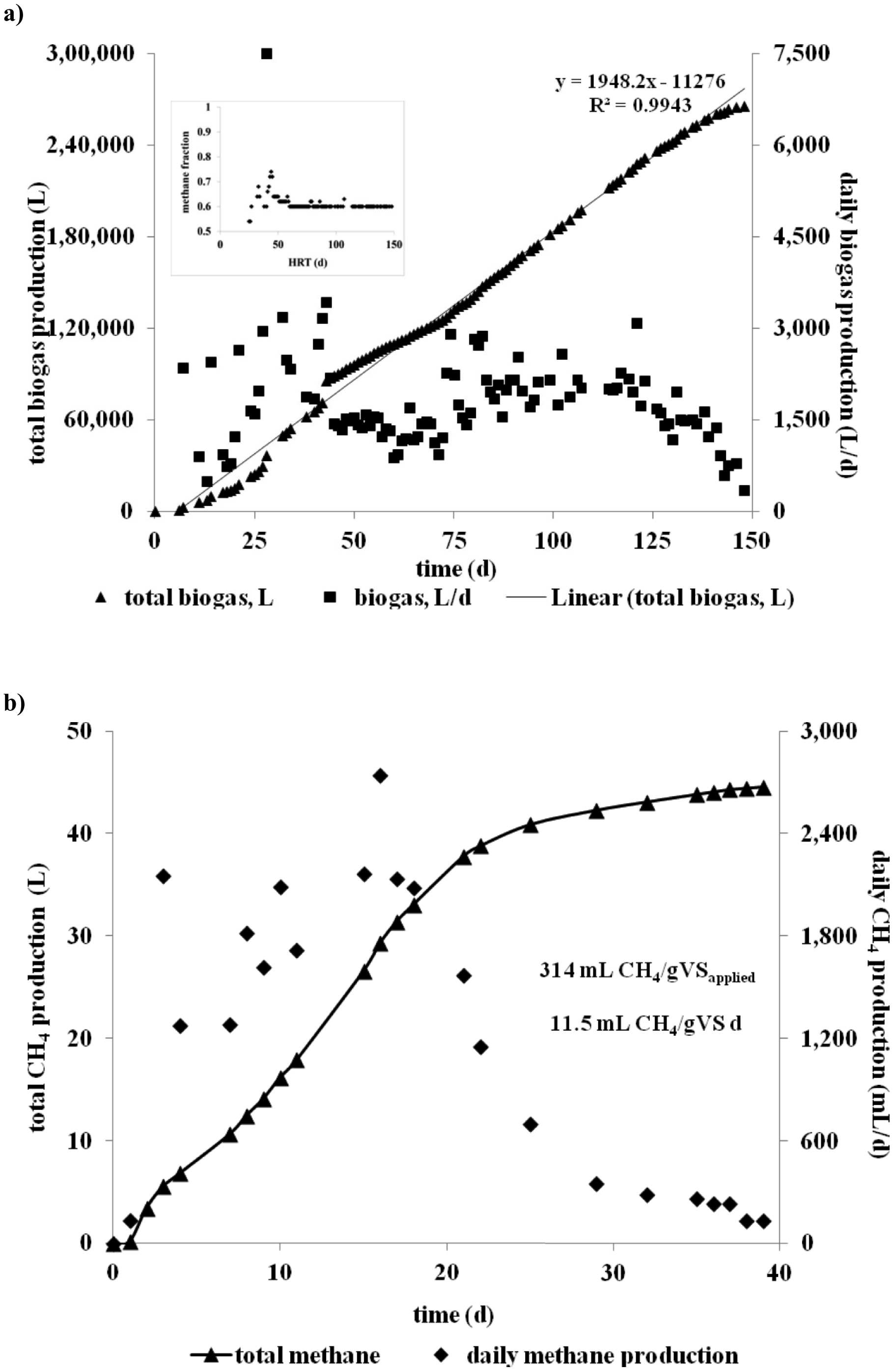


Fig. 3. a) Daily and total biogas production from pilot anaerobic digester. (Inset: methane fraction); b) biomethane potential test on digester feedstock performed in the laboratory under ideal conditions.

time than was anticipated to achieve near complete conversion of the biodegradable material. Although this pilot-scale system used here is an above-ground, steel vessel, it has been designed with the anticipation that at commercial scale it would be an in-ground, concrete-lined basin with a similar, scaled geometry. An in-ground digester would be expected to have an enhanced rate due to better temperature maintenance throughout the reactor.

In comparison (Fig. 3b) the same substrate was returned to the laboratory and digested under controlled conditions (2:1 F/M ratio, 37 °C, nutrient and micrometal concentrations controlled). 314 mL CH_4 were produced per g VS applied to the laboratory flask over a 40-d biomethane potential experiment, indicating that although the total HRT in the field was too long, near complete conversion of the biodegradable fraction was achieved at pilot scale. The laboratory control flask produced CH_4 at a rate of 11.5 mL/gVS d, which is a rate nearly 6 times faster than that observed under somewhat less than ideal conditions at the landfill. Applying the theoretical biogas production rate to the landfill operation (ideally controlled conditions) indicates that it is feasible to complete conversion in 30 d.

3.6. Dissolved counterion content

High but stable conductivity and TDS levels were observed throughout operation. It was a concern that the calcium carbonate in the oyster shell in the growth zone would impart elevated counterion concentrations and lead to an inhibition of microbial activities [33]. With this in mind, the permeate from the digester was sampled periodically for Ca, Na, and K levels and those results are presented in Fig. 4. It is known that counterion concentrations can have varying effects on degradation rates; however at higher concentrations than were observed in this demonstration. Calcium is only slightly soluble at neutral pH in pure water and molar concentrations remained throughout below 0.01 M, much too low for an inhibitory effect. Interestingly both Na and K were also much lower than anticipated despite being highly soluble in water at neutral pH, with levels stable at 0.05 M and 0.025 M, respectively.

3.7. Solids reductions, gas yields, and conversion efficiencies

Gas yields are determined on a methane basis and are quite high for this substrate. A total of 265,666 L biogas at 60% CH_4 content were produced, yielding 157,803 L CH_4. Table 2 lists the gas yields based upon TS, VS, and BVS fed and also the weight of each constituent destroyed. For instance, 187 L CH_4 were produced per kg TS fed. The total methane production per kg TS destroyed is 337 L/kg and corresponds well with theoretical equations (0.35 L CH_4/kg $COD_{destroyed}$ @ STP) [35]. Likewise, 388 and 396 L CH_4/kg VS and BVS destroyed, respectively, were produced during operation of this digester corresponding to the stoichiometric calculation of gas yield based upon carbohydrate polymer composition (i.e., $C_6H_{10}O_5$).

At the end of testing, the basin was gradually dewatered and once the bed was fully drained, the residuals were carefully characterized for volume and composition. The final volume of dewatered solids was 1.3 m^3 and represented a volume reduction on a cubic yard basis of 54% as determined by the following equation:

Table 2
Overall mass balance and gas yields.

	TS	VS	NVS	BVS	sCOD	paper
in (kg)	842	643	200	482	184	206
out (kg)	374	236	138	83	24	59
diff (kg)	468	407	61	399	160	147C
% red	56%	63%	31%	83%	87%	71%
L/kg_{fed}	187	246		327		
$L/kg_{destroyed}$	337	388		396		

$$\text{Vol. reduction } (\%) = (\text{Feed}_{\text{in}} - \text{Resid}_{\text{out}})/Feed_{in} \times 100\,\% \tag{2}$$

The total weight of residual obtained (Fig. 5) was 374 kg for a total solids (TS) reduction of 56%, corresponding well with the volume reduction. Considering that the total estimated biodegradable volatile solids in the total feed to the digester was only 57%, the TS reduction indicates that the available feed had been nearly completely degraded.

The residual also had a 63% VS content corresponding to a 63% volatile solids reduction following the equation:

$$\text{red., } \% = ((\text{Feed} \times \%VS_{\text{Feed}}) - (\text{Resid} \times \%VS_{\text{resid}}))/(\text{Feed} \times \%VS_{\text{Feed}}) \times 100\% \tag{3}$$

where, the feed and residue are in units of weight.

Although the conversion efficiency on a VS basis seems low, it should be considered that there are significant components, i.e., lignin and plastics that will be volatile but not biodegradable. In addition, production of biomass necessary to degrade the substrate contributes to the VS of the residual and lowers the apparent effectiveness of treatment.

Biodegradable volatile solids reduction is defined as:

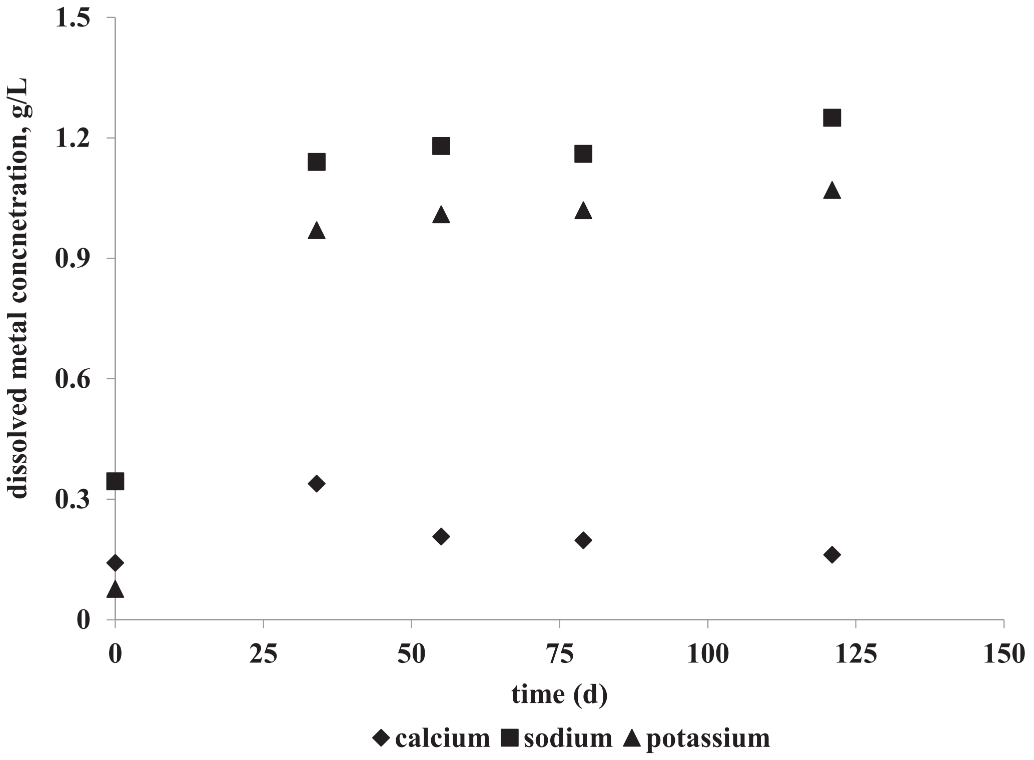
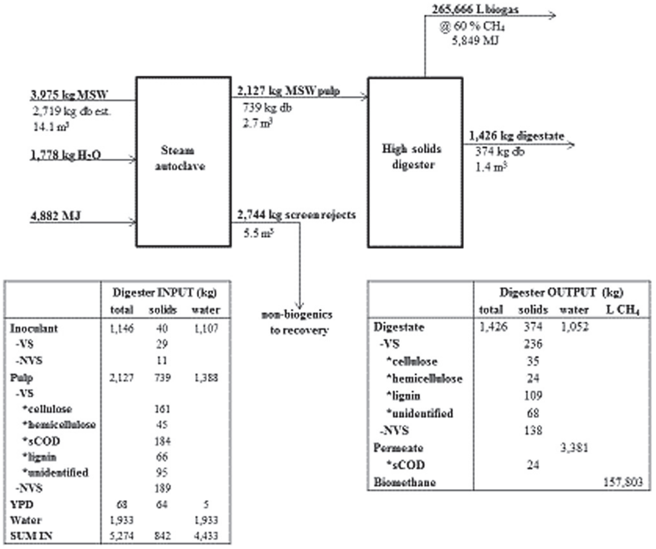


Fig. 4. Soluble counterion concentrations in high solids digester permeate.



	Digester INPUT (kg)		
	total	solids	water
Inoculant	1,146	40	1,107
-VS		29	
-NVS		11	
Pulp	2,127	739	1,388
-VS			
*cellulose		161	
*hemicellulose		45	
*sCOD		184	
*lignin		66	
*unidentified		95	
-NVS		189	
YPD	68	64	5
Water	1,933		1,933
SUM IN	5,274	842	4,433

	Digester OUTPUT (kg)			
	total	solids	water	L CH_4
Digestate	1,426	374	1,052	
-VS		236		
*cellulose		35		
*hemicellulose		24		
*lignin		109		
*unidentified		68		
-NVS		138		
Permeate			3,381	
*sCOD		24		
Biomethane				157,803

Fig. 5. Heat and mass balance for the autoclave/anaerobic digester system.

$$\text{BVS } red., \% = \sum_{in}(\text{cell.} + \text{hemi.} + sCOD) - \sum_{out}(\text{cell.} + \text{hemi.} + sCOD) \Big/ \sum_{in}(\text{cell.} + \text{hemi.} + sCOD) \times 100\% \tag{4}$$

BVS reduction was 83%, the lignocellulosic fraction was reduced 71%, and the sCOD was degraded 87%. It should be noted that the digester was still producing gas when demonstration was ceased, so a residual component was anticipated. Analogous digestion of the material in the laboratory netted no detectable carbohydrate residue and demonstrates that the biodegradable fraction of the material is fully degradable under favorable conditions and rates (Fig. 3b).

In total 842 kg of material were added to the digester including feed, inoculant, and supplements; 374 kg of digestate were recovered for further analysis. Chemical analysis of the residual (digestate) indicated that it had a 29% lignin content defined as organic-derived acid insoluble residue after residual dissolution in strong acid and volatilization at 550 °C. The residue had a remaining carbohydrate content of 15.9% and 37% NVS, leaving approximately 17% of the composition unidentified.

3.8. Overall mass balance

The overall mass balance (including the autoclave and the digester) (Fig. 5) indicates that 3975 kg of unsorted residential MSW were processed to produce 739 kg (dry basis) of MSW pulp. 2744 kg of trommel screen rejects (wet basis), largely non-biogenic in nature, were also separated and can be sorted for recovery of materials of value. The autoclave alone reduced volume 40%. The anaerobic process reduced the solids to 374 kg digestate residue. Processing the MSW in the autoclave required 849 kJ/kg material in place and at the pilot scale No. 2 fuel oil was utilized. At large scale, the biogas produced by the digester can heat the autoclave and the excess can be used otherwise. The digester produced 5700 MJ which was 20% more than required to heat the autoclave. It is possible to reduce the energy requirements at larger scale with multiple autoclaves thereby increasing the excess biogas yield for other purposes [11].

3.9. Digestate quality

The digestate was carefully sampled by dividing the reactor into 4 zones for separate examination. The digester was hydraulically designed to convey organics into the reaction zone (Zones 3 and 4) while allowing heavier debris to segregate in Zone 1 and allow for a higher quality digestate to be obtained from the main reaction bed. Qualitatively, visual observation upon removing the cover indicated that the scheme for separating non-organics was successful; aggregation of inorganics like dirt, rocks, metal debris, and textile fibers was apparent. The material in the reaction zone was of a dark brown color reminiscent of organic material with little visual contamination. The ash content though high is mostly acid soluble materials indicating that it is mainly salts, not debris. Zones 1 and 2 conversely have higher acid insoluble ash indicating higher debris content. Based upon these results and observations, it can be concluded that the design elements related to sloping the receiving zone for conveyance and beaching of heavy debris was successful.

Heavy metals content in the digestate is a direct result of the metals in the feed substrate and inoculant and the micrometal addition. The concentrations can be expected to double based upon solids destruction and be enriched in the digestate product. At neutral pH it is anticipated that most of the metals of interest will be limited in solubility and be contained in the solid phase. Table 1 compares the concentrations of 8 pollutants in the digestate as defined by the US EPA code 40 CFR 503 B for land application of biosolids as well as the British Standards Institute PAS-110 potentially toxic elements standards for digestate [29,36]. Heavy metal contents in the digestate were in all cases below the standards set for the 503 biosolids regulations suggesting that the residue is applicable for land application in the US despite its original source. Additionally, given the relatively high N content, lower amounts per acre would be required to supply the annual requirements for nitrogen. Potential land application would thus be regulated by annual application rates and cumulative metal application. While all heavy metals content in the MSW pulp feed were lower than the PAS-100 standard, the MSW digestate exceeded that of the PAS-110 standard for Cu, Ni, and Zn, similar to that reported in literature for analogous conditions [12].

Finally, the autoclave process subjects the waste materials to temperatures sufficient for sterilization and is a major advantage to the technology. The digestate product even after open air-drying contained very little/no pathogen level, well below all acceptable levels for land application [29,36]. The properties of the digestate will be discussed more fully in an upcoming publication.

4. Conclusions

Unsorted residential MSW was efficiently segregated into its organic fraction by steam autoclaving and, using a specially-designed 1500 gal (5677 L) high solids anaerobic digester, converted to biogas, whereby total solids (TS) reductions were 55%, and volatile solids (VS) and biodegradable volatile solids (BVS) reductions were 63 and 79%, respectively. Gas yields were 248 L CH4/kg VS fed or 393 L CH4/kg VS destroyed at a methane content of 60%. Conversion rates were lower than anticipated but that may have been due to the complex nature of the feedstock derived from the autoclaving process. Specifically, the material produced by the autoclave contains a high concentration of solubilized food waste absorbed onto a lignocellulosic matrix, making it a complex, high solids feedstock. Environmental controls were identified for improvement that could enhance conversion.

Several unique design elements were introduced into the system that may facilitate its use in a landfill environment, where ease of construction and ease of operation are important considerations.

Hydraulic conveyance of the high-solid material from the “loading zone” into the deeper active/reactive zone via simple gravity-feed helps maintain anaerobic conditions for a semi-continuous feeding, without complicated feed controllers. Thus semi-continuous, controlled feeding was realized with no moving parts in the conveyance system. *In situ* classification, and in-place buffering via simple additives (coral shells, for example to maintain pH stability were tested and confirmed, and may eliminate the need for complex pH control systems. The digestate passed all criteria for land application of biosolids in the US.

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References

[1] US EPA, Advancing Sustainable Materials Management: Facts and Figures 2013, Assessing Trends in Material Generation, Recycling and Disposal in the United States, United States Environmental Protection Agency, Office of Resource Conservation and Recovery, 2015.
[2] R. Yepsen, Biocycle nationwide survey: residential food waste collection in the U.S, BioCycle 53 (1) (2012) 23–33.
[3] M. Kayhanian, Biodegradability of the organic fraction of municipal solid waste in a high-solids anaerobic digester, Waste Manag. Res. 13 (2) (1995) 123–136.
[4] D. Watson, Municipal Waste Management in the United Kingdom, 2013. ETC/SCP EEA.
[5] G. Moore, Organics Disposal Ban Explainer, 2014 (Accessed January 2016), http://www.metrovancouver.org/media-room/video-gallery/issues/3897434104001.
[6] Recycling Works Massachusetts, Comply with the Organics Waste Ban, 2014 (Accessed January 2016), http://recyclingworksma.com/commercial-organics-waste-ban.
[7] R. Spencer, Rolling out a statewide organics ban, BioCycle 55 (3) (2014) 82–84.
[8] California AB-341, Solid Waste: Diversion, Mandatory Commercial Recycling, Sacramento, 2011. Chapter 476, California AB-341, Sacramento.
[9] M.H. Eley, C.C. Holloway, Treatment of municipal solid wastes by steam classification for recycling and biomass utilization - scientific note, Appl. Biochem. Biotechnol. 17 (1–3) (1988) 125–135.
[10] C.C. Holloway, Method for separation, recovery, and recycling of plastics from municipal solid waste, United States Patent #4,844,351 (1989).
[11] K.M. Holtman, D.V. Bozzi, D. Franqui-Villanueva, R.D. Offeman, W.J. Orts, A pilot-scale steam autoclave system for treating municipal solid waste for recovery of renewable organic content: operational results and energy usage, Waste Manag. Res. 34 (5) (2016) 457–464.
[12] E.K. Papadimitriou, J.R. Barton, E.I. Stentiford, Sources and levels of potentially toxic elements in the biodegradable fraction of autoclaved non-segregated household waste and its compost/digestate, Waste Manag. Res. 26 (5) (2008) 419–430.
[13] E.I. Stentiford, P.G. Hobbis, J.R. Barton, Z. Wang, C.J. Banks, Evaluating the Effect of Autoclaving on the Rate of Bioprocessing of Waste, Report to Defra, University of Leeds, UK Waste Implementation Programme, 2010.
[14] Mechanical Heat Treatment of Municipal Solid Waste, Food and Rural Affairs (Defra), UK, Department for the Environment, 2013, pp. 1–50.
[15] C.R. Ashley, K.T. Hodgson, Papermaking properties and morphology of cellulose fiber recovered from municipal solid waste, Tappi J. 2 (10) (2003) 19–22.
[16] C.R. Ashley, K.T. Hodgson, Papermaking properties of cellulose fiber recovered from municipal solid waste, Prog. Pap. Recycl. 13 (4) (2004) 5–11.
[17] C.R. Ashley, K.T. Hodgson, Morphological properties of cellulose fibre recovered from municipal solid waste, Appita J. 57 (3) (2004) 210–213.
[18] M. Farrell, D.L. Jones, Critical evaluation of municipal solid waste composting and potential compost markets, Bioresour. Technol. 100 (19) (2009) 4301–4310.
[19] A. Li, B. Antizar-Ladislao, M. Khraisheh, Bioconversion of municipal solid waste to glucose for bio-ethanol production, Bioprocess Biosyst. Eng. 30 (3) (2007) 189–196.
[20] A.D. Li, M. Khraisheh, Bioenergy II: bio-ethanol from municipal solid waste (MSW): the UK potential and implication for sustainable energy and waste management, Int. J. Chem. React. Eng. 7 (2009) A78.
[21] A. García, C. Maulini, J.M. Torrente, A. Sánchez, R. Barrena, X. Font, Biological treatment of the organic fibre from the autoclaving of municipal solid wastes; preliminary results, Biosyst. Eng. 112 (4) (2012) 335–343.
[22] C.C. Glass, E.M.N. Chirwa, D. Bozzi, Biogas production from steam-treated municipal solid waste wastewater, Environ. Eng. Sci. 22 (4) (2005) 510–524.
[23] K.M. Holtman, A. Kodama, A.P. Klamczynski, A. Flynn, D.V. Bozzi, L. Torres, D. Franqui-Villanueva, J. Mao, G.M. Glenn, W.J. Orts, Thermal properties of poly(ethylene terephthalate) recovered from municipal solid waste by steam autoclaving, J. Appl. Polym. Sci. 126 (5) (2012) 1698–1708.
[24] M. Kim, R.E. Speece, Aerobic waste activated sludge (WAS) for start-up seed of mesophilic and thermophilic anaerobic digestion, Water Res. 36 (15) (2002) 3860–3866.
[25] R.E. Speece, Anaerobic Biotechnology and Odor/corrosion Control for Municipalities and Industries, Archae Press, Nashville, TN, 2008.
[26] W.F. Owen, D.C. Stuckey, J.B. Healy, L.Y. Young, P.L. McCarty, Bioassay for monitoring biochemical methane potential and anaerobic toxicity, Water Res. 13 (6) (1979) 485–492.
[27] APHA, Standard Methods for the Examination of Water and Wastewater, sixteenth ed., American Public Health Association, Washington, D.C., USA, 1985.
[28] K.M. Holtman, R.D. Offeman, D. Franqui-Villanueva, A.K. Bayati, W.J. Orts, Countercurrent extraction of soluble sugars from almond hulls and assessment of the bioenergy potential, J. Agric. Food Chem. 63 (9) (2015) 2490–2498.
[29] US EPA, Title 40 code of federal regulations, Part 503, Fed. Regist. 58 (32) (1993) 9248–9415.
[30] BSI, PAS, 100: Specification for Composted Materials, British Standards Institution, Milton Keynes, UK, 2005.
[31] P.L. McCarty, Anaerobic waste treatment and fundamentals. Part one: chemistry and microbiology, Public Works 95 (9) (1964) 107–112.
[32] P.L. McCarty, Anaerobic waste treatment and fundamentals. Part two: environmental requirements and control, Public Works 95 (10) (1964) 123–126.
[33] I.J. Kugelman, K.K. Chin, Toxicity, Synergism, and Antagonism in Anaerobic Waste Treatment Processes, Anaerobic Biological Treatment Processes, Advances in Chemistry, American Chemical Society, Washington D.C., USA, 1971, pp. 55–90.
[34] A.G. Hashimoto, Effect of inoculum/substrate ratio on methane yield and production rate from straw, Biol. Wastes 28 (4) (1989) 247–255.
[35] Metcalf & Eddy, Inc, Wastewater Engineering: Treatment and Reuse, fourth ed., McGraw-Hill, New York, NY, 1989, p. 993.
[36] BSI, PAS, 110:2014 Specification for Whole Digestate, Separated Liquor and Separated Fibre Derived from the Anaerobic Digestion of Source-segregated Biodegradable Materials, British Standards Institution, Milton Keynes, UK, 2014.