#### REVIEW



# Options for Enhanced Anaerobic Digestion of Waste and Biomass—a Review

Neelam Vats<sup>1</sup> · Abid Ali Khan<sup>1</sup>  $\bullet$  · Kafeel Ahmad<sup>1</sup>

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### Abstract

Introduction Anaerobic digestion (AD) is a promising technology because it is economically feasible, environmentally friendly, and socially acceptable. Moreover, biogas generation from organic waste is considered to be the future of bio-energy.

Purpose In this paper, we review the substrates available for biogas production, different methods for improvement of AD processes (two-stage anaerobic digestion and digestate recirculation) and various pre-treatment techniques used to enhance biogas generation.

Method Two-stage digestion and co-digestion of two or more substrates appear to be promising techniques for enhanced anaerobic digestion. These techniques could help to maintain the nutrient balance in a system without the further addition of nutrients, in addition to enhancing biogas generation.

Results Pre-treatment of various substrates is mainly used to increase the hydrolysis rate and thus enhance the efficiency of AD processes.

Keywords Anaerobic digestion . Co-digestion . Substrate . Biogas . Pre-treatment

# Introduction

The energy demand for non-petroleum resources is continuously increasing due to the depletion of affordable traditional fossil energy sources (Wang et al. [2012](#page-14-0)). Dependence on traditional sources has led to global climate change, degradation of environment, and human health problems. Among the many alternative options available for the production of energy, processes using microorganisms have gained more attention because they have the potential for production of huge amounts of energy without disrupting the human environment and its activities (Bouallagui et al. [2009\)](#page-12-0). Anaerobic digestion (AD) is a promising alternative to traditional fossil energy sources and provides waste management options for the different types of wastes present in our surroundings by enabling the practice of energy recovery using biogas and digestate (Peng et al. [2016\)](#page-14-0). AD is a biological process that converts complex substrates into biogas and digestate by microbial action in the absence of oxygen. It occurs via four major processes: hydrolysis, acidogenesis, acetogenesis, and methanogenesis (Naran et al. [2016\)](#page-13-0). Biogas consists of methane  $(50-70\%)$ ,  $CO<sub>2</sub>$   $(30-40\%)$ , and very small amounts of other gases (Adinurani et al. [2015](#page-12-0)). Biogas production depends on a variety of governing factors like waste age, temperature, pH, hydraulic retention time (HRT), organic loading rate (OLR), volatile fatty acids (VFA), alkalinity, total solids (TS), volatile solids (VS), and carbon to nitrogen (C/N) ratio (Bouallagui et al. [2009\)](#page-12-0). The digestate obtained is used as a fertilizer in agricultural fields, which decreases unpleasant environmental odors arising due to decomposition of waste in the open (Kafle and Kim [2013](#page-13-0)).The wide variation of temperature has a specific role in the anaerobic process. Temperatures in the reactor shift from themesophilic temperature range (75–100 °F, 25–40 °C) to temperatures in the thermophilic range (125–140 °F, 50–0 °C). Generally, AD process efficiency is observed to be extremely high, but it is costly to maintain the reactor temperature in the optimal ranges. Anaerobic digestion is slower than aerobic digestion, typically requiring retention time of 10–30 days for mesophilic digestion.

Another important parameter that controls microbial activities in a reactor is pH. A minor fluctuation in pH or even low pH rarely hampers hydrolysis; however, methanogenic bacteria cease their activity at low pH. Biogas production depends on the quantity of VFA in the reactor, which is directly related to the pH in the digester. If pH drops lower than the optimal

 $\boxtimes$  Abid Ali Khan [akhan23@jmi.ac.in](mailto:akhan23@jmi.ac.in); [dee.abid@gmail.com](mailto:dee.abid@gmail.com)

<sup>1</sup> Department of Civil Engineering, Jamia Millia Islamia (A Central University), New Delhi, India

range  $(< 6.5)$ , the methanogenesis stage in the digester is affected (Brown and Li [2013](#page-12-0)). Haider et al. [\(2015\)](#page-13-0) observed that biogas production ceased when pH dropped when treating food waste (FW) and it started to rise again after pH adjustment with caustic soda. Zhang et al. [\(2005\)](#page-14-0) investigated increased pH solubilization in terms of total organic and (Chemical oxygen demand (COD) by 86% and 82% for twostage digestion (TSD) of kitchen waste (KW) at pH 7. Bouallagui et al. [\(2003](#page-12-0)) reported decrease in methane production when pH changed from 7.2 to 5.3 at 10% TS during AD of fruit-vegetable waste (FVW) in a tubular digester. Co-digestion of compatible wastes in suitable proportion can maintain the pH and stability of the process (Haider et al. [2015\)](#page-13-0). Previous studies reported that biogas production ceased with a drop of pH of FW and inoculum but continued as long as pH adjustment with caustic soda was used to reach neutral range.

Excess of either carbon or nitrogen stopped the AD process. The optimal range of C/N depends on the substrate chosen to balance the nutrients for maximum methane generation (Haider et al. [2015\)](#page-13-0). Biogas production is very low at higher C/N ratios due to rapid consumption of nitrogen in the digester resulting in inhibition of methanogenic activity (Yong et al. [2015](#page-14-0)). Li et al. [\(2013a,](#page-13-0) [b\)](#page-13-0) found an optimum C/N ratio 15–30 for co-digestion of chicken manure and agricultural wastes. The optimum C/N ratio for co-digestion of sisal pulp and fish wastes was in the range 15– 30 (Haider et al. [2015](#page-13-0)) and 20–30 (Kafle and Kim [2013](#page-13-0)).

Haider et al. [\(2015\)](#page-13-0) stated that the VFA/alkalinity ratio is a good indicator of digester stability. VFA/alkalinity (0.4) was found optimum for stable performance, as stated by Zuo et al. [\(2014](#page-14-0)). Higher temperature and higher OLR leads to higher production of VFA. Bouallagui et al. [\(2004\)](#page-12-0) reported reduction in VFA (1.2 g-COD/(L day)) when the pH was increased from 4.2 to 6 and increase in VFA (4.5 g-COD/ (L day)) at a pH of 5.5. The VFA/alkalinity ratio can be improved with the addition of substrates like abattoir waste water, waste activated sludge (WAS), and fish waste (Callaghan et al. [2002\)](#page-12-0). Higher TS content leads to restriction in the mobility of microbes, which affects biogas production (Adinurani et al. [2015\)](#page-12-0). Solids content should be approximately 10–15% for higher yield of biogas. Bouallagui et al. [\(2003\)](#page-12-0) reported inhibition of methanogenic activities with increase in the feed concentration from 4 to 10% TS. Aslanzadeh et al. [\(2014\)](#page-12-0) reported that increase in OLR and decrease in hydraulic retention time (HRT) does not affect process stability in terms of VS reduction, but biogas production was found to decrease at high OLR (Haider et al. [2015\)](#page-13-0). An increase in biogas production (48%) was observed when OLR increased from 1.11 to  $1.58 \text{ kg/(m}^3 \text{ day})$  in a semi-continuous two-phase reactor during AD of vegetable market waste Majhi and Jash [\(2016\)](#page-13-0).

The OLR is an important parameter because it can be used to indicate the amount of volatile solids to be fed into the digester each day (Ribeiro et al. [2017\)](#page-14-0).An increase in methane production was achieved on increasing the OLR from 1.2 to 2.4 g-COD/ (L day) and it also increased the organic matter removal

efficiency by 86%. Furthermore, increasing the OLR results in a decrease of the COD removal efficiency (74%) in the hydrothermal pre-treatment of sugarcane bagasse (SB). Karthikeyan and Visvanathan [\(2012\)](#page-13-0) observed a decrease in ammonia inhibition (19%) with OLR 7–10 kg-VS/ $(m^3$  day) in AD of green waste, FW, FVW, and paper waste. Liu et al. [\(2017\)](#page-13-0) concluded that the mesophilic digestion of FW at an OLR of 1.5 kg-VS/  $(m<sup>3</sup>$  day) was optimum for steady methane production.

OLR was mainly adjusted according to the HRT. If the concentration of organic matter in a feeding material is stable, shorter HRT and higher OLR could be achieved. Otherwise, with the same HRT, the OLR will vary with the different feeding substrates. Rapid increase in the OLR would mean that hydrolysis and acidogenesis would occur rapidly. Meanwhile, the methanogenic bacteria would be slower: they would not be able to consume the fatty acids at the same rate.

Higher production of VFA and toxic by-products often act as rate-limiting steps for the hydrolysis stage (Naran et al. [2016\)](#page-13-0). In complex substrates, hydrolytic enzyme adsorption takes place at the surface, which converts the substrates into smaller molecules for further degradation. Methanogenesis was found to be the rate-limiting step for substrates that can be biodegraded relatively simply. Several studies were focused on pre-treatment approaches to increase hydrolysis-stage activities and to optimize the AD process (Ariunbaatar et al. [2014](#page-12-0)).

In this paper, the substrates available for AD processing are summarized, along with different methods for improvement of AD processes and various pre-treatment techniques used for enhancement of biogas production.

# Variety of Substrates Investigated for Biogas Production

#### Substrates Available for Anaerobic Digestion

AD is a more appropriate technology for KW or FW than landfilling, composting, and incineration due to the high organic and moisture content present in these wastes (Bo and Pin-jing [2014;](#page-12-0) Zhang et al. [2005](#page-14-0)). FW has a lower C/N ratio due to the greater nitrogen content in the organic form (Haider et al. [2015](#page-13-0)). This causes ammonification and inhibition of methanogenesis (Yong et al. [2015\)](#page-14-0). There are a few studies that have reported successful digestion of FW due to its better biodegradability (Zhang et al. [2013\)](#page-14-0); however, most studies report low methane yield and lower stability due to VFA production (Liu et al. [2009\)](#page-13-0). The raw characteristics of various substrates are presented in Table [1](#page-2-0). FVW has a high amount of biodegradable organic matter and high moisture content; therefore, it is best used as a renewable energy source via AD (Kafle and Kim [2013](#page-13-0); Zuo et al. [2014](#page-14-0)). Co-digestion of FVW with compatible wastes can overcome its drawbacks (e.g., low solubility due to poor C/N ratio, low efficiency).

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OFMSW organic fraction of municipal solid waste, MSW municipal solid waste, TOC total organic carbon, HRT hydraulic retention time, d days

Lignocellulosic biomass (energy crops, SB, Corn Stover (CS), straw, etc.) is less resistant to microbial attack due to the complex relation between lignin, cellulose, and hemicellulose that does not allow proper microbial action to yield methane gas (Bruni et al. [2010a](#page-12-0); Zhong et al. [2011](#page-14-0); Vats et al. [2019a\)](#page-14-0). Sugarcane bagasse (SB) is found mostly in tropical countries and is typically utilized for electricity production by combustion, as feedstock for animals, and as fuel in various boilers. Oil palm mesocarp fiber (OPMF) is not easily degradable due to its high lignocellulose content, so mono-digestion of it is not preferred. As suggested by Saidu et al. [\(2014\)](#page-14-0), biological pre-treatmentis required for any lignocellulose biomass to be used as a substrate in AD processes. This pre-treatment decrease the lignin and cellulose content in OPMF to 9% and 10.6% respectively by making lignin, cellulose, and hemicellulose available to microbial degradation. Rice husk used as energy crop is slowly biodegradable due to its higher nitrogen ratio and lignin content. Wheat straw (WS) is found in agricultural waste and is generally used for mulching or as fodder. The remaining WS is burnt or left unused (Bolado-Rodriguez et al. [2016](#page-12-0)). Straws have high C/N ratios due to nitrogen deficiency and the cellulose present in the straws is responsible for its prolonged digestion time (Yong et al. [2015](#page-14-0)). The biogas produced from different substrates is shown in Table [2.](#page-5-0)

WAS produced in biological units has high buffering capacity and low C/N ratio; therefore, it is mostly co-digested with easily biodegradable organic matter to produce maximum methane from the sludge. WAS is rich in nutrients but is not used as a fertilizer in agricultural fields (Gaur and Suthar [2017](#page-13-0); Gaur et al. [2017](#page-13-0)). The AD of WAS increases its potential for land application because this improves its dewaterability and stabilizes the sludge produced in the treatment plants. This produces high-quality biosolids for land application and a carbon source to promote denitrification (Bougrier et al. [2007\)](#page-12-0).

Livestock manure like poultry waste (PM) and other such wastes have high buffering capacity and acts as a potential source for energy production using AD. High organic matter content in livestock waste causes the accumulation of VFA, ammonia, and ammonium ions. Poultry droppings comprise more easily degradable organic materials, but also contain lignin (40–50% of the TS). This makes AD difficult without the addition of other substrates (Wang et al. [2012\)](#page-14-0). Poultry waste is around 20% TS so it is diluted to improve the TS proportion and, thereby, the AD performance (Callaghan et al. [2002](#page-12-0)). PM mainly consists of carbohydrate (53% TS), lignin, and starch (Kaparaju and Rintala [2005](#page-13-0)). PM has high ammonium content which inhibits the AD process. When co-digested with potato waste, PM buffers the potato waste, which has low nitrogen.

#### Improving Biogas Yield Through Co-digestion

AD of mono feedstock is inhibited due to poor macro and micro nutrients, high nitrogen and heavy metal concentrations, low organic matter content, and accumulation of VFA

which results in an optimized reactor (Haider et al. [2015](#page-13-0); Peng et al. [2016\)](#page-14-0). The AD efficiency can be increased by addition of a compatible waste substrate able to establish nutrient balance and buffering capacity of the system to obtain maximum output from both wastes (Bouallagui et al. [2009](#page-12-0)). Digestion of multiple substrates in a single reactor is known as co-digestion. This process is feasible, economic, and ecologically beneficial, with technological benefits as well. According to Haider et al. [\(2015\)](#page-13-0) the microorganism concentration determines the process stability, biodegradation rate, and lag time. Chen et al. ([2010](#page-13-0)) observed that an appropriate second substrate reduces the need of adding other alkaline or acid sources to maintain the pH, which is required in single-stage digestion (SSD) to provide a proper working environment for the microbes. Co-digestion decreases the free ammonia accumulation, maintains a pH in the reactor optimal for maximum production of methane or biogas, and creates an appropriate balance of carbon and nitrogen in the system (Haider et al. [2015;](#page-13-0) Vats et al. [2019b](#page-14-0)). Callaghan et al. [\(2002\)](#page-12-0) also observed more stable performance of animal manure and FVW with low and high C/N ratios than of mono-digestion of animal manure. Kalamaras and Kotsopoulos [\(2014\)](#page-13-0) reported a 60–70% increase in biogas generation with cattle manure and cardoon silage, sorghum silage, maize silage, and milk thistle in the first 15 days of AD. Kafle and Kim [\(2013\)](#page-13-0) investigated the decreased risk of ammonia inhibition in swine manure (SM) and the acidification problem in apple waste (AW) during co-digestion of AW and SM. A ratio of 33:67 (VS basis) was recommended for AW and SM for maximum biogas production because the reactor performance depends on the proportion of each substrate. Callaghan et al. ([2002](#page-12-0)) determined that the biogas yield was increased from 0.23 to  $0.45 \text{ m}^3/\text{kg-VS}$  by increasing the FVW content from 20 to 50% during co-digestion with cattle manure. Li et al. ([2009](#page-13-0)) suggested 75% (VS basis) KW for optimal codigestion activity with cow manure. Bouallagui et al. [\(2003\)](#page-12-0) observed an increase in acidogenic activities and decrease in methanogenic activities at short HRT and high loading rate. It can be concluded that the AD process gives good results with optimized ratios of the feedstocks used (Wang et al. [2012\)](#page-14-0).The addition of cow dung to Spartina alterniflora (an estuarine cordgrass) resulted in improved C/N ratio of 14.19 with different proportions of feedstock substrate (Chen et al. [2010\)](#page-13-0). The methane yield was increased by 7.09–44.26%. It was concluded that the addition of cow dung or other nutrient rich material to lignocellulose biomass increases the process efficiency. An improvement in biogas yield was observed with 15– 20% of potato waste with PM from 0.13-0.15  $m^3/kg$ VSadded waste to 0.30–0.33m<sup>3</sup> /kg-VSadded waste at OLR 2 kg-VS/ $(m^3$  day) (Kaparaju and Rintala [2005\)](#page-13-0), thereby creating a positive environment in the reactor (Mata-Alvarez et al. [2000\)](#page-13-0).

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Table 2 Biogas/methane yield at optimum conditions by co-digestion of different substrates Table 2 Biogas/methane yield at optimum conditions by co-digestion of different substrates CS corn straw, AW apple waste, SM swine manure, VS volatile solids, TKN total Kjeldahl nitrogen, FW food waste, VS volatile solids, VFA volatile fatty acids, TA total alkalinity, d days

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#### Improving Biogas Yield with Digestate Recirculation

Digestate recirculation in the form of liquid or solids improves biogas production for energy crops (WS, CS) due to the availability of nutrients and appropriate bacteria in the digestate. Peng et al. [\(2016\)](#page-14-0) found that WS cannot be digested alone due to inadequate nutrients and the high C/N ratio (Passos et al. [2016\)](#page-14-0) needed for sufficient methane production. In this study, digestate liquor was recycled to maintain the nutrients during the AD process. An improvement of 21% in methane production was observed with recirculation of microbes and nutrients. Moreover, the methane content in the biogas declined to less than 50% after a 50-day digestion period, whereas with recirculation of nutrients and microbes, it was more than 50%. The rate of hydrolysis also improves with digestate recirculation. Lee et al. [\(2010](#page-13-0)) explored digestate sludge recirculation to help in balancing alkalinity, VFA content, and methanogens. In the study, different OLR  $(4.16, 8.4, and 11.8 g-COD/ (L day) was used in a two-stage$ methanogenic reactor producing biogas (15.7, 58.6, and 67.4 L/ day, respectively). The methane content decreased when the OLR was > 58.5 g-COD/ (L day) in the first reactor.

#### Improving Biogas Yield with Two-Stage Digestion

Two-stage digestion utilizes two reactors (i.e., for acidogenesis and methanogenesis) that operate separately, where organic biomass digestion takes place to upgrade the productivity of each process separately (Massanet-Nicolau et al. [2015](#page-13-0)). The processes of solubilization, hydrolysis, and acidogenesis take place in acidogenesis reactor where the hydrogen and VFA are produced. The hydrolyzed biomass is used by hydrogen producing bacteria in this reactor. In the second stage, the VFA produced are converted to methane (Majhi and Jash [2016](#page-13-0)). According to Bouallagui et al. [\(2004\)](#page-12-0) in TSD, buffering of OLR takes place in the first stage and consistent feeding is attained in the second stage (Bouallagui et al. [2004](#page-12-0)). However, TSD reactors are well known for system failure, design complexity, and operating difficulties despite their advantages of higher energy production than with SSD. A number of studies have been focused on TSD, some mainly on varying the OLR and HRT. Different reactor configurations such as combined continuous stirred tank reactor (CSTR) and upflow anaerobic sludge blanket reactor (UASB) (Aslanzadeh et al. [2014](#page-12-0)), and coupled anaerobic sequential batch reactor (SBR) (Bouallagui et al. [2004\)](#page-12-0) were also investigated. Whereas, others looked at solid-liquid phase separation during acidogenesis and subsequent transfer of liquid to the methanogenesis reactor (Majhi and Jash [2016\)](#page-13-0) and the use of a tubular reactor (Bouallagui et al. [2003\)](#page-12-0). The two processes (acidogenesis and methanogenesis) differed regarding the separation of liquid in the acidogenic phase and that in the methanogenic phase, and in the way the microorganisms were retained in the reactors.

Using TSD for vegetable waste improves the biogas and methane production. The use of TSD along with recirculation helps maintain the buffering capacity, alkalinity, and the VFA content in a two-stage acidogenesis reactor. Zuo et al. [\(2014](#page-14-0)) studied two-stage systems using vegetable waste along with recirculation from a methanogenic to anacidogenic reactor. This combination increased the biogas yield from 0.50 to 0.66 L/g. Massanet-Nicolau et al. [\(2015](#page-13-0)) found a 13.4% higher methane yield with TSD than with SSD with 20-day HRT digesting pelletized grass. Moreover, digestion could be performed at high OLR without impacting the stability of the methanogenic process. Bo and Pin-jing [\(2014](#page-12-0)) concluded for TSD of KW that with increase in the COD loading rate, the efficiency decreased by 44% and no increase in methanogenesis was observed. Aslanzadeh et al. ([2014\)](#page-12-0) investigated a combination of CSTR and UASB reactors for TSD. The supernatant from the CSTR reactor was used as feed in the UASB reactor for methane production. Higher OLR with shorter HRT was achieved using this type of TSD for treatment of FW. A lower reactor volume was required for TSD of FW (by 26%) and municipal waste (by 65%). Bouallagui et al. [\(2004\)](#page-12-0) reported higher stability in the TSD of FVW in coupled anaerobic SBR. Majhi and Jash [\(2016](#page-13-0)) developed a robust methanogenic stage reactor for solid-liquid separation to increase the performance of the reactor. Higher biogas production (19–21%) was observed than with SSD. Nasr et al. ([2012\)](#page-13-0) stated that TSD gives better stability with shorter HRT than SSD, which led to process improvement. A total of 18.5% improvement in energy recovery in TSD of thin stillage (compared to SSD reactor performance) was observed. It has been concluded from various studies that good phase separation results in higher system stability at higher OLR Bo and Pin-jing [2014;](#page-12-0) Bouallagui et al. [2004\)](#page-12-0). Zuo et al. [\(2014](#page-14-0)) investigated TSD of high moisture content vegetable waste and concluded that TSD was better than SSD because the recirculation of acidic effluent from the second stage to the first stage creates a more favorable environment for methane generation.

It can be concluded that SSD generally operates at low OLR and the TSD acidogenic process can operate at higher OLR, specifically more than two-times higher than with SSD, and with shorter HRT (Bouallagui et al. [2004](#page-12-0)).

# Pre-treatment Methods

Pre-treatment methods may be used at large scale to enhance the biomethane production and these help in accelerating the digestion process (Carlsson et al. [2015](#page-12-0)). Several pre-treatment methods have been proposed by various researchers for treating the organic waste. Pre-treatment methods that have been studied so far include physical, chemical, thermal, and biological methods, as well as combinations of them. Ultrasonic treatment and ozonation are also well-studied methods for improving process performance (methane production) and stability of the

process (Ekpeni et al. [2014](#page-13-0)). Ariunbaatar et al. ([2014](#page-12-0)) reported that pre-treatment helps in degradation of complex organic substrate molecules and recalcitrant particles. Wet organic waste can also be used effectively for energy production if an appropriate pre-treatment method is employed. This has been proven successful for lignocellulose biomass due to its abundance in nature. Because it is composed of lignin, cellulose, and hemicellulose, hydrolysis of this biomass is problematic and adequate penetration of microorganisms past the surface does not occur. The degrade performance and results in much less production of energy and biogas. Lignin and cellulose are difficult to degrade due to the complex relation between them that limits both hydrolysis and the surface areas available for microbial action. Various studies have been done on chemical, thermal, physical, and biological pre-treatment in which significant improvement in the digestion process has been observed. Pre-treatment approaches modify the physical properties of the organic materials, thereby improving filterability, dewaterability, and solubilization. This, in turn, improves biogas generation and process performance (Pei et al. [2016](#page-14-0)). Dewaterability and filterability are not improved by the form of pre-treatment method applied; rather, they depend on the organic matter released during the pre-treatment method. Carrere et al. ([2009\)](#page-12-0) suggested that biogas production from PM can be improved with thermal and thermo-chemical pre-treatment, which improve biodegradability. The methane yield of 170 mL-CH<sub>4</sub>/g-COD was achieved at 190 °C, which is higher by 1.21 times than the control for the liquid-fraction of manure. It was also observed that at temperatures of 25, 135, and 150 °C and  $pH \sim 12$ , the biodegradability of the liquid and solid fraction of PM also decreased as compared with the control.

# Mechanical Pre-treatment

In one case, mechanical treatment increased the surface availability for enzymatic action, thus increasing the methane yield by 25% (Bruni et al. [2010b\)](#page-12-0). Mechanical methods used at large scale include French presses, bead mills, sonicators, homogenizers, and micro fluidizers. High-pressure homogenization is one of the best techniques being used to enhance the AD process. It works by changing the microbial contact area by disruption of cell walls (Ekpeni et al. [2014\)](#page-13-0). Abouelenien et al. ([2014\)](#page-12-0) observed that pre-treatment of CM using the ammonia stripping technique (CM has too much ammonia that inhibits the AD process) achieved the required C/N ratio in the reactor. This pre-treatment increased the stability of the reactor, improving ammonia inhibition and VFA degradation in ways that enhanced the methane yield of the system. Methane yield of 695 mL/g-VS was achieved (42% of the control at thermophilic temperature). Mechanical pre-treatment of the waste accelerates the digestion for lignocellulose biomass and helps in decreasing the crystallinity of the structure so that enzymatic action is properly achieved. A 25% increase in methane production of lignocellulose substrate has been achieved at full-scale biogas

plants using a mechanical milling pre-treatment technique. At laboratory and full scales, digestion of Baker's yeast in a highpressure homogenizer has been proven satisfactory for cell wall disruption (Ekpeni et al. [2014](#page-13-0)).

#### Ultrasonic Pre-treatment

Ultrasonic (US) pre-treatment is another viable option for organic matter solubilization and degradation. It disrupts the physical, chemical, and biological properties. Moreover, the degree of disintegration depends on the sonication parameters and on the feedstock characteristics and therefore, the optimum range for sonication varies as well. Cesaro et al. [\(2012\)](#page-12-0) reported increased biodegradability with US treatment of solid waste that increased the biogas obtained (42% higher than the untreated waste) after 45 days. Sludge degradation at lower frequencies is more efficient, and increases particle solubilization for enhanced stability of the reactor (Bougrier et al. [2006\)](#page-12-0). Naran et al. ([2016\)](#page-13-0) investigated the ultrasonic effect on biogas production for digestion of FW and WAS. The highest amount of organic matter removal was 11.1% and 39.5% (for FW and WAS, respectively) over the removal by mono-digestion of FWand WAS. Li et al. [\(2013a](#page-13-0), [b](#page-13-0)) investigated US and thermos-chemical pre-treatment effects on methane production from fat, oil, and grease (FOG) and synthetic KW. US treatment does not efficiently increase biogas production in any of the co-digestions studied and it caused inhibitory effects on FOG co-digestion. It also leads to a longer lag phase in the digestion of FOG due to inhibition of methanogenesis. With US treatment, 60% COD solubilization was obtained, which increased the biogas yield by 24% more than from untreated substrate (Cesaro et al. [2012\)](#page-12-0).

#### Chemical Pre-treatment

Bruni et al. [\(2010a](#page-12-0)) found that chemical treatment methods could increase methane production by 66% compared with other methods (biological, mechanical, combination of steam and biological method, and thermal treatment) for digestion of biofibers. Steam explosion of plant biomass is one of the oldest methods and requires less chemicals and energy. This method involves the injection of high-pressure steam into a reactor filled with biomass. Macromolecular explosion occurs when the steam is injected into the biomass where rapid release of the substrates takes place, causing disruption of the structure (Han et al. [2010\)](#page-13-0). The disadvantage of steam explosion is that it generates a number of toxic compounds in the reactor that need to be removed from the system before proceeding to biogas generation. As determined by Harun et al. [\(2011](#page-13-0)), chemical pre-treatment requires lower operating cost if compared with enzymatic pretreatment. Chemical pre-treatment includes both acid and alkali treatments and several studies have been performed on such treatments that cause significant changes in the biomass structure and helps improve the reactor performance.

#### Alkali Hydrolysis (AH) Pre-treatment

Alkali pre-treatment is generally used for wastes with a high concentration of protein, lignin, cellulose, and hemicellulose (Harun et al. [2011](#page-13-0)). Most of the studies found used pretreatment with NaOH (among other alkali hydrolysis chemicals) (Zheng et al. [2009\)](#page-14-0). Researchers suggest that pre-treatment with NaOH and calcium hydroxides are effective in increasing methane production of crops by improving their structure and lignin solubilization (Ferreira et al. [2013](#page-13-0)). Phenol and organic acids are among the most easily degradable compounds during alkali and acid pre-treatment. Chandra et al. [\(2012](#page-13-0)) observed that alkali pretreatment (4% NaOH) resulted in a 1.12-times increase of methane, compared with that from untreated substrate. If lignin degradation does not take place, then cellulose and hemicellulose degradation also stops due to the inhibition of microbial action. Ferreira et al. [\(2013\)](#page-13-0) noted that AH with bases solubilizes the lignin component and modifies the structure of lignin. It was observed that the use of an oxidizing compound like  $H_2O_2$  in combination with alkali treatment improves the digestibility of crop residues in comparison to alkaline pre-treatment only. Bruni et al. [\(2010a](#page-12-0)) observed lower methane yield at a 10% CaO w/w ratio due to the formation of complexes of calcium and lignin. Chandra et al. [\(2012\)](#page-13-0) reported the result on pre-treatment of WS using chemical and hydrothermal pre-treatment techniques and observed significant biogas enhancement by 87.5% with 4% sodium hydroxide, as compared with untreated substrate.

#### Acid Pre-treatment

Acid pre-treatment helps in removal of lignin and cellulose from the substrates. Harun et al. ([2011\)](#page-13-0) observed that acid pretreatment method is mostly used for substrates with an excess of carbohydrates. Rafique et al. ([2010](#page-14-0)) reported disadvantages like generation of toxic compounds, unrecyclable reagents, and high energy demand. A variety of chemical acids including sulfuric, hydrochloric, phosphoric, nitric, and maleic have been used in acid pre-treatment processes. Inhibitors like furfural and HMF have been obtained in acid hydrolysis with 1.5% HCL of WS and SB (Bolado-Redriguez et al. [2016](#page-12-0)). Reduction of lignin, cellulose, and hemicellulose have been reported at molecular level with acid hydrolysis of SB. Reduction of hemicellulose content by up to 92.78% was observed, along with improved efficiency of the process using a sulfuric acid pre-treatment (Chandel et al. [2014](#page-12-0)).

#### Oxidation (Ozone) Pre-treatment

Ozone is a strong oxidant that reacts directly or indirectly by breaking into radicals. The direct reaction causes loss in biomethane production due to destruction of easily fermentable sugar by ozone radicals, whereas indirect reaction with ozone causes the degradation of complex organic compounds. High ozone doses  $(0.034 - 0.202 \text{ g} - O_3/\text{g} - T\text{S})$  are not effective for FW pre-treatment due to loss of fermentable sugars (Ariunbaatar et al. [2014\)](#page-12-0). Ozone treatment of larger molecules breaks them into smaller one and makes them more easily accessible to microorganisms. Oxidation of organic matter has a positive impact on COD solubilization and VS reduction, which improves the rate of biogas generation. This improves retention time in the digester and reduces the initial lag phase of sludge digestion. Ozone pre-treatment has been found highly effective for sewage sludge AD as stated by Bougrier et al. [\(2006](#page-12-0)). Ozone pretreatment has been successfully applied to pharmaceutical and municipal sludge, and increases the hydrolysis rate and decreases antibiotic resistant genes. Ozone increased the solubilization rate of sludge by 15.75–25.09% (Pei et al. [2016](#page-14-0)). Beszedes et al. [\(2009\)](#page-12-0) reported combined ozonation-acid pretreatment was a less time-consuming process for getting effective results than ozone treatment only. Enhancement of biogas production by up to 10 times was observed when ozonation and microwave pre-treatment methods were combined.

### Biological Pre-treatment

Biological pre-treatment is an effective method to increase the biogas or methane yield. In this method enzymes work to degrade cell walls rather than with violent disruption as in mechanical and steam methods. Biological methods generally work under milder conditions. The enzymes selected for digestion of microalgae depends on the composition of cell walls of the target microalgae. The cell wall molecules tend to convert into some usable products (Gonzalez-Fernandez et al. [2012](#page-13-0)). Saidu et al. ([2014](#page-14-0)) reported reduction of 10.91, 8.96, and 10.63% in hemicellulose, cellulose, and lignin content (respectively) during biological pre-treatment of OPMF with oyster mushrooms (Pleurotus florida, the fungal enzyme source). OPMF, cattle manure, and palm oil mill effluent produced the most biogas with OLR 1.62 g-VS/(L day) at 10-day HRT. This production was higher than for other reactors with varying OLR. Zhong et al. ([2011\)](#page-14-0) reported an increase in biogas (33%) and methane (76%) yields for CS using the oyster mushroom P. florida. CS is a lignocellulosic biomass and its components, namely, lignin, cellulose, and hemicellulose, were degraded significantly during biological treatment with P. florida. Very few studies have been done on enzymatic hydrolysis and their effect on oxidation of lignin in AD (Zhong et al. [2011](#page-14-0)). Passos et al. [\(2016\)](#page-14-0) observed improvement in methane yield (8 and 15%) for microalgae with a 1% cellulase and enzyme mix, respectively, as the biological enzymes within 6 h. An increase in the methane yield from digestion of filamentous algae by 17% and 4% was obtained with cellulase and xylanase enzymes, respectively. Bruni et al. [\(2010b\)](#page-12-0) biologically pre-treated biofibers with enzymatic and partial aerobic microbial conversion. Enzymatic pre-treatment results were less desirable due to lower methane production (than steam explosion with phosphoric acid). Biological pretreatment requires low energy, mild environmental conditions, and no requirement for chemicals, which makes it quite easy to handle and operate.

### Thermal Pre-treatment

The thermal pre-treatment is considered to be an eco-friendly and green processing technology (Carlsson et al. [2015](#page-12-0); Gaur et al. [2017\)](#page-13-0). Thermal treatment at low temperature has gained considerable amount of attention due to its lower consumption of energy as compared with thermal treatment at higher temperatures (> 100 °C). High-pressure treatment also often consumes more energy (Wang et al. [2012](#page-14-0); Vats et al. [2019c](#page-14-0)). The advantages of thermal pre-treatment include the prevention of corrosion problems, no formation of toxic compounds, fewer requirements of chemicals for the neutralization of the hydrolysates produced, and production of less waste (Ferreira et al. [2013\)](#page-13-0). Inhibitors like furfural and hydro methyl are produced in excess or at higher temperatures in the AD process, which does not allow the microorganisms to function properly for methanogenesis (Bolado-Redriguez et al. [2016\)](#page-12-0).Solubilization and dewaterability are increased during the thermal pre-treatment, which in turn increases the process performance. These lead to economic savings and improvement of the biomass hydrolysis step (Ma et al. [2011;](#page-13-0) Cano et al. [2014\)](#page-12-0). Thermal pre-treatment methods involve temperature ranges from 60 to 270 °C, and can extend from 15 min to several hours. Kalamaras and Kotsopoulos [\(2014\)](#page-13-0) reported that although thermal pre-treatment does not increase the soluble fraction or alter the fiber composition, it still helps improve biogas production. FW produced more biogas when pre-treated thermally at 80 °C and relatively less biogas was obtained at  $> 100$  °C. The biomethane produced with thermal pre-treatment at 80 °C for 1.5 h was 52% higher than for untreated biomass. Moreover, if thermal treatment was extended, positive results were obtained, but the energy requirement also increased with increasing temperature. Bolado-Redriguez et al. ([2016\)](#page-12-0) reported increase in methane gas production (29%) by thermal pre-treatment whereas alkaline pre-treatment of the solid fraction increased biodegradability and methane production by 30% for WS and SB. Biological sludge exhibited the highest methane gas production (50% higher) compared with other forms of waste studied. This was due to cell lysis that took place during steam explosion for thermal hydrolysis (Cano et al. [2014](#page-12-0)). Carlsson et al. ([2015\)](#page-12-0) reported that low hydrolysis was mainly due to two components in microbial cells (as in WAS) and another component (lignocellulose in plants), but that these can be overcome by thermal pre-treatment. Methane yield was increased at 70– 190 °C for the manure soluble fraction and a temperature of at least 135 °C was required for the total soluble fraction of PM. Thermal treatment led to the increase of pH from 7.6 to 8.2, as proven by the solubilization of proteins (Carrere et al. [2009\)](#page-12-0).

Many studies have found that pre-treatment of sewage sludge increases the methane yield but an extremely limited numbers of studies were found about manure pre-treatment with thermal and thermo-chemical methods. Steam pre-treatment with NaOH as catalyst produced a maximum methane yield of 49 mL-CH4/g-VS for digested biofibers of manure. Combined steam pre-treatment with  $H_3PO_4$  and NaOH, and with biological treatment resulted in increased methane yield by  $2 \pm 0.5$  and  $1.7 \pm 0.4$ , respectively. This method is also effective for substrates with higher volatile solid content (Naran et al. [2016](#page-13-0)).

# Thermo-Chemical Pre-treatment

Thermo-chemical pre-treatment is a key method to enhance biogas generation. It is an effective and economical approach to extract maximum resources from waste (Rafique et al. [2010\)](#page-14-0). Kalamaras et al. [\(2014\)](#page-13-0) reported an increment in biogas production with thermo-chemical pre-treatment with NaOH for lignin materials. Milk thistle crop was investigated for biogas production and fiber composition using mechanical, thermo-chemical, and thermal pre-treatment. The thermochemical experiment was carried out in an autoclave at 120 °C for 20 min with 2% w/v NaOH. The methane yield of 271 L-CH4/kg-VS was obtained using pre-treated substrate. Thermal pre-treatment did not show any increment in the soluble fraction of the milk thistle crop, whereas the soluble fraction was increased with thermo-chemical treatment from 31 to 55.1%. Ma et al. [\(2011](#page-13-0)) reported the highest solubilization (32%) of KW by a thermo-chemical method (comparable to other pre-treatment methods), but the higher solubilization due to pre-treatment did not result in the highest biogas production. This trade-off between solubilization and biodegradability may be due to the formation of inhibitory materials during pre-treatment. Chandra et al. [\(2012\)](#page-13-0) reported a drop in pH in the pre-hydrolysis step due to oxidation of sulfur and phosphorus content when performing the hydrothermal pre-treatment. Sodium hydroxide maintained the pH. For digester stability, along with pH and buffering capacity, a 10% TS concentration of the biomass must be maintained. Naran et al. ([2016](#page-13-0)) studied and found that pre-treated FW and WAS using alkali thermal treatment obtained the highest removal of total suspended solids (12.8% and 12.9% respectively).

Alkali thermal pre-treatment was found more effective for samples with higher total suspended solids due to its rapid hydrolysis and mineralization process, while alkali thermal pre-treatment does not seem to be effective for substrates with higher VSS content. Li et al. ([2013a,](#page-13-0) [b\)](#page-13-0) found that pH 10 at 55 °C and pH 8 at 55 °C were optimal conditions for generation of methane production  $(9.9 \pm 1.5\%$  and  $9.5 \pm 0.4\%$ , respectively). Table [3](#page-10-0) summarizes the different studies carried out on pre-treatment techniques and biogas production of various organic wastes.

<span id="page-10-0"></span>





FW food waste, HMF hydroxymethylfurfural, WAS waste activated sludge, TSS total suspended solids, VS volatile solids

# <span id="page-12-0"></span>Conclusions

In this paper, we reviewed a variety of different options for improved performance AD processes using various substrates and their potential for enhanced biogas generation. The codigestion of multiple substrates with inoculum can be effective for enhanced biogas generation. The use of multiple substrates helps in balancing the C/N ratio or nutrients in AD processes. Various pre-treatment techniques can also be used to increase the biodegradability and solubilization of the biomass, which ultimately increases the process performance and improves biogas production. The pre-treatment of energy crops such as sugarcane tend to cause the loss of lignin, cellulose, and hemicellulose content. This results in better microbial access to the available surface and increases the biodegradability of the process. Based on this review, it was observed that AD with multiple organic fractions appears a very promising option for enhancing biogas generation.

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# Compliance with Ethical Standards

Conflict of Interest The authors declare that there is no conflict of interest.

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