Pretreatment methods to enhance anaerobic digestion of organic solid waste

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Highlights

- Pretreating organic solid wastes leads to an enhanced anaerobic digestion process.
- Pretreatments may also reduce the cost for post treatment of digestates.
- Efficiency of pretreatment methods depends on the substrates' characteristics.
- Only few pretreatment methods are successfully applied at full-scale to date.

Abstract

This paper reviews pretreatment techniques to enhance the anaerobic digestion of organic solid waste, including mechanical, thermal, chemical and biological methods. The effects of various pretreatment methods are discussed independently and in combination. Pretreatment methods are compared in terms of their efficiency, energy balance, environmental sustainability as well as capital, operational and maintenance costs. Based on the comparison, thermal pretreatment at low (<110 °C) temperatures and two-stage anaerobic digestion methods result in a more cost-effective process performance as compared to other pretreatment methods.

Keywords:
Anaerobic digestion
Pretreatment
Organic solid waste
Sustainability
Economic cost

1. Introduction

2. Mechanical pretreatment

2.1. Process description and mode of action

2.2. Mechanical pretreatment of OFMSW

2.3. Mechanical pretreatment of miscellaneous OSW

3. Thermal pretreatment

3.1. Process description and mode of action

3.2. Thermal pretreatment at lower temperatures (<110 °C)

3.3. Thermal pretreatment at higher temperature (>110 °C)

Abbreviations: AD, anaerobic digestion; COD, chemical oxygen demand; CSTR, continuously stirred reactor; FW, food waste; HHW, household waste; HMF, hydroxymethylfurfural; HPH, high pressure homogenizer; HRT, hydraulic retention time; MSW, municipal solid waste; OFMSW, organic fraction of municipal solid waste; OLR, organic loading rate; OM, operational and maintenance; OSW, organic solid waste; PEF, pulsed electric field; SRT, solid retention time; SS, sewage sludge; THP, thermal hydrolysis process; TPAD, temperature phased anaerobic digestion; WWTP, wastewater treatment plant; TS, total solid; UASB, upflow anaerobic sludge blanket; VS, volatile solid; VFA, volatile fatty acid; WAS, waste activated sludge.

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Anaerobic digestion (AD) is one of the oldest and well-studied technologies for stabilizing organic wastes [1]. Among the treatment technologies available for treating organic solid wastes (OSW), AD is very suitable because of its limited environmental impacts [2–5] and high potential for energy recovery [2,3,6]. Such positive aspects coupled with the recent concerns on rapid population growth, increasing energy demand, and global warming have promoted further research on the AD process development and improvement in order to enhance biogas production, achieve faster degradation rates and reduce the amount of final residue to be disposed [3,4,7].

AD is a biological process that converts complex substrates into biogas and digestate by microbial action in the absence of oxygen through four main steps, namely hydrolysis, acidogenesis, acetogenesis and methanogenesis. Most researchers report that the rate-limiting step for complex organic substrates is the hydrolysis step [8–26], due to the formation of toxic by-products (complex heterocyclic compounds) or non desirable volatile fatty acids (VFA) formed during the hydrolysis step [27,28]; whereas methanogenesis is the rate-limiting step for easily biodegradable substrates [24,27,29,30]. Extensive research has been conducted on pretreatment methods to accelerate the hydrolysis step [31,32] and to obtain suitable by-products from this step [28], as well as to improve the quality of useful components like nitrogen and phosphorus to be recycled [33].

According to European Union Regulation EC1772/2002, substrates such as municipal solid waste (MSW), food waste (FW), and slaughterhouse wastes need to be pasteurized or sterilized before and/or after AD. Taking this regulation into account, pretreatment methods could be applied, thus obtaining a higher energy recovery and eliminating the extra cost for pasteurization and/or sterilization [34,35]. Pretreatment methods could nevertheless be unsustainable in terms of environmental footprints, even if they enhance the AD process performance [36]. The effects of various pretreatment methods are highly different depending on the characteristics of the substrates and the pretreatment type. Hence, it is difficult to compare and systematically assess the applicability and sustainability of such methods at a full scale.

In the recent past a number of reviews have been published with a common aim to assess the pretreatment effects. Table 1 shows that most of the research on pretreatment methods has been conducted on wastewater treatment plant (WWTP) sludge and/or lignocellulosic substrates; whereas there is a limited number of reviews on the recently growing interest of pretreatment methods to enhance AD of OSW, specifically the organic fraction of municipal solid waste (OFMSW). Therefore, this paper aims to review the most recently studied pretreatment methods including mechanical, thermal, chemical and biological methods to enhance AD of OSW, with an emphasis on OFMSW. The pretreatment methods will be compared in terms of efficiency, energy balance, cost and process sustainability.

2. Mechanical pretreatment

2.1. Process description and mode of action

Mechanical pretreatment disintegrates and/or grinds solid particles of the substrates, thus releasing cell compounds and increasing the specific surface area. An increased surface area provides better contact between substrate and anaerobic bacteria, thus enhancing the AD process [3,24,25]. Esposito et al. suggested that a larger particle radius results in lower chemical oxygen demand (COD) degradation and a lower methane production rate [37]. Likewise, Kim et al. showed that particle size is inversely proportional to the maximum substrate utilization rate of the anaerobic microbes [38]. Therefore, mechanical pretreatments such as sonication, lysis-centrifuge, liquid shear, collision, high-pressure homogenizer, maceration, and liquefaction are conducted in order to reduce the substrate particle size.

In addition to size reduction, some methods result in other effects depending on the pretreatment. Hartmann et al. reported that the effect of maceration is more due to shearing than cutting of the fibers [39]. Sonication pretreatment generated by a vibrating probe mechanically disrupts the cell structure and floc matrix [40]. The main effect of ultrasonic pretreatment is particle size reduction at low frequency (20–40 kHz) sound waves [41]. High-frequency sound waves also cause the formation of radicals such as OH−, HO2−, H+, which results in oxidation of solid substances [42].
A high pressure homogenizer (HPH) increases the pressure up to several hundred bar, then homogenizes substrates under strong depressurization [43]. The formed cavitation induces internal energy, which disrupts the cell membranes [44]. These pretreatment methods are not common for OFMSW, but they are more popular with other substrates such as lignocellulosic materials, manure and WWTP sludge. Size reduction through beads mill, electroporation and liquefaction pretreatments of OFMSW has been studied at lab scale, whereas rotary drum, screw press, disc screen shredder, FW disposer and piston press treatment are successfully applied at full scale. Both electroporation and liquefaction pretreatments cause cellular structure damage, thus the effect on the AD process is similar to maceration [45,46].

The advantages of mechanical pretreatment include no odour generation, an easy implementation, better dewaterability of the final anaerobic residue and a moderate energy consumption. Disadvantages include no significant effect on pathogen removal and the possibility of equipment clogging or scaling [47,48].

### 2.2 Mechanical pretreatment of OFMSW

Mechanical pretreatments such as rotary drum were used as an effective technology for OFMSW separation and pretreatment prior to AD, which could enhance the biogas production by 18–36% [49,50]. Davidson et al. found small variations in both methane yields per gVS (gram volatile solids) and content of methane in biogas while studying the biomethane potential of source-sorted OFMSW pretreated with different mechanical methods including screw press, disc screen shredder, FW disposer and piston press [51]. Similarly, Zhang and Banks found no significant enhancement with such pretreatment methods [52]. Hansen et al. studied the effects of the same pre-treatment technologies on the quantity and quality of source-sorted OFMSW. They found that screw press pretreatment resulted in a smaller substrate particle size, while a shredder with magnetic separation yielded a higher (5.6–13.8% as compared to the other methods) methane production [53]. In contrast, Bernstad et al. reported that the screw press pretreatment method also result in a loss of biodegradable materials and nutrients, even though it enhances the biogas production in general [54].

Izumi et al. studied the effect of the particle size on FW biomethanation. Size reduction through a beads mill resulted in a 40% higher COD solubilization, which led to a 28% higher biogas production yield. However, excess size reduction to particles smaller than 0.7 mm caused an accumulation of VFA [8]. As the methanogens are sensitive to acidic intermediates [55], excessive size reduction may result in a decreased AD process performance. Few research on electroporation, liquefaction, and high frequency sonication pretreatment methods to enhance OFMSW has been conducted. Electroporation pretreatment of OFMSW resulted in 20–40% higher biogas production [46], and liquefaction resulted in 15–26% higher biogas production [3], whereas sonication resulted in 16% higher cumulative biogas production as compared to untreated substrates [56]. The higher biogas production was mostly due to the more extensive solubilization of the particulates.

#### 2.3 Mechanical pretreatment of miscellaneous OSW

Maceration, sonication and HPH are the simplest mechanical pretreatments for OSW such as WWTP sludge and lignocellulosic substrates. Size reduction of lignocellulosic substrates results in a 5–25% increased hydrolysis yield, depending on the mechanical methods used [34], whereas for WWTP sludge and manure, the effects of pretreatments significantly differ. Generally, applying maceration pretreatment enhances biogas production by 10–60% [3]. For instance, maceration of fibers in manure up to 2 mm resulted in a 16% increase of the biogas production, while size reduction up to 0.35 mm resulted in a 20% increase, and no significant difference was observed with further size reduction [57].

Barjebruch and Kopplow treated surplus sludge with HPH at 600 bar, and showed that the filaments were completely disintegrated [44]. Engelhart et al. studied the effect of HPH on the AD of sewage sludge (SS), and achieved a 25% increased VS reduction [58]. This improvement was induced by the increased soluble protein, lipid, and carbohydrate concentration. The HPH of WWTP sludge has been applied at full scale, resulting in a 30% biogas enhancement, thus the working volume of digesters could be decreased by 23% [3].

Sonication prior to the AD process resulted in an enhancement of the biogas production of 24–140% in batch systems, and 10–45% in continuous or semi-continuous systems [3]. However, not all studies confirm the enhancement of VS destruction or higher biogas production. Sandino et al. studied sonication of waste activated sludge (WAS) and obtained only a negligible increase in both VS destruction and mesophilic methane production [59].

### Table 1

Reviews on different pretreatment methods to enhance AD using various substrates.

<table>
<thead>
<tr>
<th>Substrate</th>
<th>Pretreatment methods</th>
<th>Important findings</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>OFMSW</td>
<td>All pretreatment methods</td>
<td>- Physical pretreatments are widely applied for OFMSW, whereas other methods are not spread at industrial level</td>
<td>[61]</td>
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<tr>
<td></td>
<td></td>
<td>- Further research on pretreatment should focus more on the modelling as well as mass and energy balance of the pretreatment effect and the whole AD process</td>
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<tr>
<td>All organic</td>
<td></td>
<td>- The most popular pretreatment methods are thermal and ultrasonic for WWTP sludge, chemical for lignocellulosic substrates, and mechanical for OFMSW</td>
<td>[39]</td>
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<tr>
<td>substrates</td>
<td></td>
<td>- Systematic studies on energy balance and economic feasibility are necessary</td>
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<td>- Further development of descriptive and predictive variables is required</td>
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<tr>
<td>Lignocellulosic</td>
<td>Thermal, thermo-chemical, chemical</td>
<td>- Pretreatments could improve the digestibility of lignocellulosic substrates</td>
<td>[73]</td>
</tr>
<tr>
<td>substrates</td>
<td>Thermal, thermo-chemical, and chemical</td>
<td>- Pretreatments could result in more efficient process as compared to the conventional process</td>
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<td></td>
<td></td>
<td>- Thermal pretreatments as well as lyme and ammonia based chemical methods are more effective in improving the digestibility of lignocellulosic substrates</td>
<td>[34]</td>
</tr>
<tr>
<td>Pulp &amp; paper sludge</td>
<td>Thermal, thermo-chemical, chemical</td>
<td>- Pretreatments could result in reduced HRT, increased methane production, and reduced sludge size</td>
<td>[40]</td>
</tr>
<tr>
<td>WWTP sludge</td>
<td>Ultrasound, chemical, thermal, and microwave</td>
<td>- The effect of pretreatment methods depends on the characteristics of sludge and the intensity of the method</td>
<td>[3]</td>
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<tr>
<td></td>
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<td>- Pretreatments result in enhanced biogas production (30–50%)</td>
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<tr>
<td>WWTP sludge</td>
<td>Thermal, thermo-chemical, and chemical</td>
<td>- Thermal pretreatment at high temperature (&gt;175 °C) as well as thermo-chemical methods are more effective in improving sludge dewaterability</td>
<td>[60]</td>
</tr>
<tr>
<td>WWTP sludge</td>
<td>Thermal and thermo-chemical</td>
<td>- Pretreatments result in enhanced biogas production (30–50%)</td>
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<td>- Comprehensive model for evaluating the economic feasibility was developed</td>
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<td></td>
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<td>- Pretreatments could yield a better digestate with high recoverable nutrients</td>
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</table>
3. Thermal pretreatment

3.1. Process description and mode of action

Thermal treatment is one of the most studied pretreatment methods, and has been successfully applied at industrial scale [3,31,61]. Thermal pretreatment also leads to pathogen removal, improves dewatering performance and reduces viscosity of the digestate, with subsequent enhancement of digestate handling [2,31,32,62]. Various temperatures (50–250 °C) to enhance the AD of different OSW (mainly WWTP sludge and lignocellulosic substrates) have been studied. However, to the best of our knowledge, no systematic research on various temperature and treatment times to enhance AD of OFMSW has been conducted.

The main effect of thermal pre-treatment is the disintegration of cell membranes, thus resulting in solubilization of organic compounds [17,63–65]. COD solubilization and temperature have a direct correlation. Higher solubilization can also be achieved with lower temperatures, but longer treatment times. Mottet et al. compared different thermal pretreatment methods and found no significant difference between steam and electric heating, whereas microwave heating solubilized more biopolymers [66]. The higher rate of solubilization with microwave pretreatment can be caused by the polarization of macromolecules [47,63]. Concerning the lignocellulosic substrates, temperatures exceeding 160 °C cause not only the solubilization of hemicellulose but also solubilization of lignin. The released compounds are mostly phenolic compounds that are usually inhibitory to anaerobic microbial populations [34].

Bougrier et al. suggested that thermal pretreatment at high temperatures (>170 °C) might lead to the creation of chemical bonds and result in the agglomeration of the particles [42]. One of the most known phenomena is the Mallaird reaction, which occurs between carbohydrates and amino acids, resulting in the formation of complex substrates that are difficult to be biodegraded. This reaction can occur at extreme thermal treatment at temperatures exceeding 150 °C, or longer treatment time at lower temperatures (<100 °C) [3,25,34,67,68].

In addition to these chemical reactions, thermal pretreatment can also result in loss of volatile organics and/or potential biogas production from easily biodegradable substrates. Therefore, the effects of thermal pretreatment depend on the substrate type and temperature range.

3.2. Thermal pretreatment at lower temperatures (<110 °C)

Protot et al. suggested that thermal pretreatment at temperatures below 100 °C did not result in degradation of complex molecules, but it simply induces the deflocculation of macromolecules [64]. Barjenbruch and Kopplow obtained a similar conclusion with pretreatment at 90 °C. Their results showed that the filaments are not disintegrated, but they were only attacked with thermal pretreatment [44]. Neyens and Bayens reported that thermal pretreatment resulted in the solubilization of proteins and increased the removal of particulate carbohydrates [60].

Thermal pretreatment of sludge even at lower temperatures (70 °C) has a decisive effect on pathogen removal [24]. Probably based on such results, the EU Regulation EC1772/2002 requires OSW to be pretreated at least an hour at 70 °C. In this regard, numerous studies on thermal pretreatment at 70 °C were conducted. For instance, pretreating household waste and algal biomass at 70 °C for 60 min and 8 h, respectively, did not result in enhancement of the biogas production [18,69]. Appels et al. obtained a negligible increase of biogas production from sludge pretreated at 70 °C for 60 min, whereas the biogas production was improved 20 times when applying a 60 min pretreatment at 90 °C [19]. Rafique et al. achieved a maximal enhancement of 78% higher biogas production with a 60% methane content by pretreatment at 70 °C [10]. Ferrer et al. obtained a 30% higher biogas production with a 69% methane content [17], whereas Climent et al. obtained a 50% biogas volume increase with pretreatment at 70 °C prior to thermophilic AD [70]. Gavala et al. reported that pretreatment of primary and secondary WWTP sludge at 70 °C has a different effect on the thermophilic and mesophilic methane potential. Thermal pretreatment at 70 °C was shown to have a positive effect on mesophilic AD of primary sludge, but not on its thermophilic AD; whereas it enhanced both the thermophilic and mesophilic methane production of secondary sludge. This can be explained by the chemical composition of the OSW substrates: primary sludge contains higher amounts of carbohydrates, whereas secondary sludge contains higher amounts of proteins and lipids [26].

3.3. Thermal pretreatment at higher temperature (>110 °C)

Liu et al. studied the thermal pre-treatment of FW and fruit and vegetable waste at 175 °C; they obtained a 7.9% and 11.7% decrease of the biomethane production, respectively, due to the formation of melanoids [62]. Ma et al. obtained a 24% increase of the biomethane production with FW pretreated at 120 °C [9]. Rafique et al. studied pretreatment of pig manure at temperatures higher than 110 °C [10]. They observed hardening and darkening of manure, which resulted in a low biogas yield. Hardening and the dark brownish color development of the substrate indicated the occurrence of Mallaird reactions.

4. Chemical pretreatment

4.1. Process description and mode of action

Chemical pretreatment is used to achieve the destruction of the organic compounds by means of strong acids, alkalis or oxidants. AD generally requires an adjustment of the pH by increasing alkalinity, thus alkali pretreatment is the preferred chemical method [71]. Acidic pretreatments and oxidative methods such as ozonation are also used to enhance the biogas production and improve the hydrolysis rate. The effect of chemical pretreatment depends on the type of method applied and the characteristics of the substrates. Chemical pretreatment is not suitable for easily biodegradable substrates containing high amounts of carbohydrates, due to their accelerated degradation and subsequent accumulation of VFA, which leads to failure of the methanogenesis step [72]. In contrast, it can have a clear positive effect on substrates rich in lignin [13].

4.1.1. Alkali pretreatment

During alkali pretreatment, the first reactions that occur are saponification and saponification, which induce the swelling of solids [31]. As a result, the specific surface area is increased and the substrates are easily accessible to anaerobic microbes [34,73,74]. Then, COD solubilization is increased through various simultaneous reactions such as saponification of uronic acids and acetyl esters, as well as neutralization of various acids formed by the degradation of the particulates [75]. When substrates are pretreated with alkali methods, an important aspect is that the biomass itself consumes some of the alkali [34], thus higher alkali reagents might be required for obtaining the desired AD enhancement.

4.1.2. Acid pretreatment

Acid pretreatment is more desirable for lignocellulosic substrates, not only because it breaks down the lignin, but also
because the hydrolytic microbes are capable of acclimating to acidic conditions [76]. The main reaction that occurs during acid pretreatment is the hydrolysis of hemicellulose into perspective monosaccharides, while the lignin condensates and precipitates [34,77]. Strong acidic pretreatment may result in the production of inhibitory by-products, such as furfural and hydroxymethylfurfural (HMF) [73,76]. Hence, strong acidic pretreatment is avoided and pretreatment with dilute acids is coupled with thermal methods (see also Section 2.5). Other disadvantages associated with the acid pretreatment include the loss of fermentable sugar due to the increased degradation of complex substrates, a high cost of acids and the additional cost for neutralizing the acidic conditions prior to the AD process [73,78,79].

4.1.3. Effects of accompanying cations present in the acid/alkaline reagents

In addition to the effects of the alkali and acid themselves, the AD might be affected by the accompanying cations present in these reagents including sodium, potassium, magnesium, calcium, since the chemicals are added mostly as salts or hydroxides of these cations. Therefore, the inhibitory concentrations of these cations should be considered [3,80]. Kim et al. studied the inhibition of the sodium ion concentration on the thermophilic AD of FW, and reported that more than 5 g/L of sodium resulted in lower biogas production [38]. Sodium is more toxic to propionic acid utilizing bacteria as compared to other VFA degrading bacteria [81]. The inhibitory level of the potassium ion starts at 400 mg/L, though anaerobic microbes are able to tolerate up to 8 g/L potassium [82]. The potassium ion is more toxic to thermophilic anaerobes as compared to mesophilic or psychrophilic anaerobes [83].

The optimum concentrations of calcium and magnesium ions have been reported to be 200 mg/L and 720 mg/L, respectively [84,85]. Excessive amounts of calcium ions can cause precipitation of carbonates and phosphates, which results in scaling of the reactors, pipes, and biomass; thus it reduces the specific methanogenic activity and results in a loss of buffer capacity [113]. Also high concentrations (>100 mM) of the magnesium ion can cause disaggregation of methanogens, thus the conversion of acetate is inhibited [85].

Furthermore, AD could also be enhanced indirectly due to the supplementation of trace metals such as cobalt (Co), molybdenum (Mo), selenium (Se), iron (Fe), tungsten (W), copper (Cu) and nickel (Ni), which play a role in many biochemical reactions of the anaerobic food web. For instance Zhang and Jahng used supplements of trace metals such as Fe, Co, Mo and Ni to stabilize a single-stage reactor treating FW, and concluded that Fe was the most effective metal for stabilization of the AD process [86]. Facchin et al. achieved a 45–65% higher methane production yield from FW with supplementation of a trace metals (Co, Mo, Ni, Se, and W) cocktail [87]. Nevertheless, supplementing trace metals to solid waste AD plants should not be considered as a pretreatment method, though it could be an effective method for achieving higher biogas production rates with a higher methane content.

4.1.4. Ozonation

Another chemical pretreatment method is ozonation [3], which does not cause an increase of the salt concentration and no chemical residues remain as compared to other chemical pretreatment methods. Moreover, it also disinfects the pathogens [88,89]. Hence, ozonation has gained great interest for sludge pretreatment [3,50], and to a lesser extent of OFMSW. Ozone is a strong oxidant, which decomposes itself into radicals and reacts with organic substrates [90] in two ways: directly and indirectly. The direct reaction depends on the structure of the reactant, whereas the indirect reaction is based on the hydroxyl radicals. As a result, the recalcitrant compounds become more biodegradable and accessible to the anaerobic bacteria [91].

4.2. Chemical pretreatments of OSW

Chemical pretreatments are widely applied on wastewater sludge and lignocellulosic substrates [3,34,73], while very limited research has been conducted on OFMSW. Ozonation pretreatment has only been conducted on wastewater or sludge from WWTP. In general, the optimal ozone dose for enhancing AD of WWTP sludge ranges between 0.05 to 0.5 gO3/gTS [3,91–93]. Cesaro and Beligiorno reported that the optimum ozone dose for source-sorted OFMSW is 0.16 gO3/gTS, which resulted in a 37% higher cumulative methane production [56]. Lopez-Torres and Llorens obtained a 11.5% increased methane production with alkaline pretreatment of OFMSW [74]. Neves et al. achieved 100% of the potential production with alkaline (0.3 gNaOH/gTS) pretreated barley waste [28]. Patil et al. studied the effect of alkaline pretreatment of water hyacinth, which has a lower lignin content as compared to other plants [94]. They found that the alkaline pretreatment had a smaller effect than mechanical pretreatments. Therefore, acidic and alkaline pretreatment are not suitable for substrates with a low lignin content.

5. Biological pretreatment

Biological pretreatment includes both anaerobic and aerobic methods, as well as the addition of specific enzymes such as pepsidase, carbohydrolase and lipase to the AD system. Such conventional pretreatment methods are not very popular with OFMSW, but have been applied widely on other types of OSW such as WWTP sludge and pulp and paper industries.

The hydrolytic-acidogenic step (first step) of a two-phase AD process is considered as a biological pretreatment method by some researchers [3,95–97], while others consider it as a process configuration of AD, but not a pretreatment method [31]. Physically separating the acidogens from the methanogens can result in a higher methane production and COD removal efficiency at a shorter hydraulic retention time (HRT) as to conventional single-stage digesters [98]. Parawira et al. reported that optimizing the first hydrolysis stage could stimulate the acidogenic microbes to produce more specific enzymes, thus resulting in more extended degradation of substrates [99]. Therefore, in this review paper the first step of the two-phase AD systems are considered as a pretreatment method.

5.1. Conventional biological pretreatment

Aerobic pretreatment such as composting or micro-aeration prior to AD can be an effective method to obtain a higher hydrolysis of complex substrates due to the higher production of hydrolytic enzymes, which is induced by the increased specific microbial growth [100]. Fdez-Guelfo et al. reported that pretreatment by composting resulted in a higher specific microbial growth rate (160–205% as compared to untreated OFMSW) than by thermochemical pretreatment [14]. Lim and Wang also affirmed that the aerobic pretreatment yielded a greater VFA formation due to the enhanced activities of the hydrolytic and acidogenic bacteria [100]. However, according to the results obtained by Brummeler and Koster, a pre-composting treatment of OFMSW resulted in a 19.5% VS loss [101]. Mshandate et al. also observed a loss of potential methane production with a longer aerobic pretreatment of sial pulp waste [102].

Miah et al. investigated the biogas production of SS pretreated with aerobic thermophilic bacteria closely related to Geobacillus thermodenitrificans [15]. According to their results, the highest
amount of biogas (70 ml/gVS) with a 80–90% methane content was achieved at 65 °C. Melamane et al. studied the AD of wine distillery wastewater pretreated with the fungus Trametes pubescens. This fungal pretreatment obtained a 53.3% COD removal efficiency, which increased the total COD removal efficiency of the AD system up to 99.5% [103]. Muthangya et al. used pure cultures of the fungus Trichoderma reesei to aerobically pretreat sisal leaf decortication residues. Their results showed that aerobic incubation for 4 days resulted in a 30–40% cumulative biogas increase with a higher (50–66%) methane content [104]. Romano et al. studied two types of enzymes capable to hydrolyze plant cell walls to enhance the biomethanation of Jose Tall wheat grass [16]. They did not obtain a significant biogas enhancement or VS reduction, though the hydrolysis step was accelerated [15].

5.2. Two-stage AD

A two-stage AD system consists of a hydrolytic-acidogenic stage followed by the methanogenic stage. The advantages of such systems include: (i) increased stability with better pH control; (ii) higher loading rate; (iii) increased specific activity of methanogens resulting in a higher methane yield; (iv) increased VS reduction and (v) high potential for removing pathogens [6,105–109]. The disadvantages include: (i) hydrogen built-up resulting in inhibition of acid-forming bacteria; (ii) elimination of possible interdependent nutrient requirements for the methane forming bacteria; (iii) technical complexity and (iv) higher costs [110,111].

Verrier et al. compared two-stage methanization of vegetable wastes with mesophilic and thermophilic single stage continuously stirred tank reactors (CSTR). They found that for easily biodegradable wastes, a two-stage reactor converted 90% of the wastes to biogas, which outperformed both the mesophilic and thermophilic single stage CSTR and could withstand higher organic loads [112]. Zhang et al. investigated the effect of pH on two-phase AD of FW, and suggested that adjusting the pH to 7 in the hydrolysis stage can improve both the total solids (TS) loading rate and biogas production yield [113].

5.2.1. Temperature phased anaerobic digestion (TPAD)

Recently more research is being conducted on temperature phased anaerobic digestion (TPAD). This method usually consists of a primary digester at thermophilic (or hyper-thermophilic) temperature followed by a mesophilic secondary digester. The advantages of TPAD include not only higher methane production yields, but also a pathogen free high nutrient digestate [114]. Riau et al. suggested that TPAD is preferred if the purpose is to achieve pathogen free digestate, which can be directly used as soil conditioner [115].

Schmit and Ellis reported that TPAD outperformed conventional AD processes including dry digestion of source separated OFMSW [116]. Lee et al. investigated TPAD of FW and excess sludge at 70 °C in the primary reactor, followed by a secondary reactor with temperatures of 35 °C, 55 °C and 65 °C [117]. The best result was achieved at a solid retention time (SRT) of 4 days and 70 °C in the primary reactor, followed by a secondary reactor at 55 °C. Wang et al. compared the conventional thermophilic digestion with TPAD (hyper-thermophilic (80 °C) and thermophilic (55 °C) primary reactor followed by a mesophilic reactor), treating FW with polylactide. They obtained a COD solubilization of 82%, 85.2%, 63.5% with TPAD with a hyper-thermophilic first stage, TPAD with a thermophilic first stage, and a conventional thermophilic digester, respectively. Moreover, 82.9%, 80.8%, 70.1% of the organics were converted into methane with TPAD with a hyper-thermophilic first stage, TPAD with a thermophilic first stage and a conventional thermophilic digester, respectively [118]. Song et al. compared the biogas production and pathogen removal of WAS with TPAD compared to a single stage mesophilic and thermophilic digester [119]. TPAD yielded a 12–15% higher VS reduction and it was as stable as the single stage mesophilic reactor, whereas pathogen removal was as high as in the single stage thermophilic reactor.

5.2.2. Biohythane production

Optimizing two-stage conditions may result in the production of bio-hydrogen from the primary reactor and biomethane from the second reactor, making it a very attractive biohythane producing system. Numerous studies have been conducted on the optimization of such systems with reactors both at mesophilic and/or thermophilic temperatures. Liu et al. obtained 43 mlH2/gVS and 500 mlCH4/gVS from household waste (HHW) [120], whereas Wang et al. obtained 65 mlH2/gVS and 546 mlCH4/gVS from FW [110]. Chu et al. reported that the optimum hydrogen production from FW is achieved at pH 5.5–6 in thermophilic AD. The biohydrogen content was 52% with no methane in the first stage, whereas the methane content was 70–80% in the secondary reactor. Based on a mass balance, 9.3% of the COD was converted to hydrogen and 76.5% converted to methane [121]. Escamilla-Alvareado et al. studied the optimization of two-stage AD of OFMSW, and obtained an overall biogas production of 661 ± 2.5 and 703 ± 2.9 ml/gVS for mesophilic and thermophilic operations, respectively. The biogas produced from the primary reactor contained 3–10% hydrogen, whereas the biogas from the secondary reactor contained 25–61% methane [122].

6. Combination of various pretreatments

6.1. Thermo-chemical pretreatment

Different pretreatment methods rely on various mechanisms to solubilize particulate organic matter [11,27]. Hence pretreatment methods in combination have also been studied to obtain a further enhancement of biogas production and faster AD process kinetics. Shahriari et al. investigated the AD of OFMSW pretreated with a combination of high temperature microwaves and hydrogen peroxide pretreatment [123]. The combination of microwaves with chemical pretreatments as well as the microwave irradiation at temperatures higher than 145 °C resulted in a larger component of refractory material per gCOD, causing a decrease of the biogas production. A similar trend was observed with pig manure pretreated with lime and heated at temperatures higher than 110 °C [10,124]. This could be explained by the increased hydrolyses of proteins and carbohydrates due to the chemical pretreatment, and in the presence of heat the produced amino acids and sugars reacted together forming complex polymers such as melanoidins. However, alkaline pretreatment coupled with thermal methods at a lower temperature (70 °C) could result in a higher (78%) biogas production with a higher (60%) methane content as compared to the best results (28% increase of biogas production with 50% methane content) obtained by thermal pretreatment at higher temperatures (>100 °C) [10]. This enhancement of the AD process is due to the reduction of the hemicellulosic fraction [10,124].

6.2. Thermo-mechanical pretreatment

Mechanical pretreatment combined with thermal treatment have also been studied to enhance the AD of OFMSW, though this combination is not popular for OFMSW. Zhang et al. obtained the highest enhancement of biogas production (17%) by grinding (up to 10 mm) rice straw and heating it to 110 °C [103]. Chiu et al. compared the hydrolysis yield of sludge pretreated with a combination of ultrasonic and alkaline pretreatment. Simultaneous
ultrasonic and alkaline pretreatment of sludge resulted in the highest hydrolysis rate of 211 mg/l min \([25]\). Wett et al. (2010) studied the disintegration of sludge pretreated at 19–21 bar pressure and achieved a 70% higher biogas recovery at 5 days shorter digestion as compared to AD of untreated SS \([126]\).

6.3. Various pretreatments combined with a two-stage AD

Considering the first stage of two-stage AD as biological pretreatment, three-stage processes can be classified as a combined pretreatment system. Kim et al. studied semi-anaerobic CSTRs followed by two-stage upflow anaerobic sludge blanket (UASB) reactors treating FW, and obtained a 95% COD removal and a biogas production of 500 mg/gVS at HRT of 16 days \([127]\). The same research group also reported that the same amount of biogas with a higher methane content (67.4%) could be obtained at a lower HRT (10–12 days) by increasing the temperature of the acidogenic stage from mesophilic to thermophilic \([128]\). Kvesitadze et al. studied the two-stage thermophilic co-digestion of OFMSW and pretreated corn stalk by freeze explosion. The best results of 104 mlH\(_2\)/gVS and 520 mlCH\(_4\)/gVS were obtained with alkaline (pH = 9) pre-hydrolysis, which could increase the heat and electricity production by 23% and 26%, respectively, as compared to the single stage process design \([129]\). Kim et al. investigated the hydrogen and methane production by a two-phase AD system fed with thermally pretreated FW; they found at least 3.4 days were necessary to produce hydrogen from FW \([130]\). Moreover, recycling the methanogenic effluent to the hydrogenesis step was applied to reduce water usage, which further increased the hydrogen production by 48% \([130,131]\).

7. Comparison of pretreatment methods to enhance anaerobic digestion of OFMSW

A systematic comparison of pretreatment methods in terms of their efficiencies, economic feasibility and environmental impacts are necessary for choosing the desired pretreatment method. To the best of our knowledge, no comparison of pretreatment efficiencies to enhance the AD of OFMSW has been conducted so far. The efficiency of the AD process can be evaluated through the methane yield per amount of removed or initial feed of TS, VS, and COD. The substrate solubilization rate and anaerobic biodegradation is also used to evaluate the AD process performance. Table 2 compares the efficiency of pretreatment methods including mechanical, thermal, biological and a combination of them for enhancing the AD of OFMSW in terms of biogas production enhancement per amount of initial feed VS.

In general, OFMSW results in 280–557 ml/gVS biogas production, which is 70–95% of the organic matter in the feed. The pretreatment effects vary depending on the substrate characteristics and the type of AD system. The most commonly used mechanical pretreatment methods are size reduction by beads mills, electro-winning, pressurization, disc screen, screw press and shredder with magnetic separation. Mechanical pretreatments result in a 20–40% increased biogas yield as compared to the untreated substrates. Both chemical and thermochemical methods could yield up to 11.5–48% higher biogas yield depending on the pretreatment conditions and substrate characteristics.

In thermal pretreatment, temperature plays a major role in the enhancement of biogas production. Low temperature (70 °C) pretreatment can result in a 2.69% higher biogas production for FW, whereas it does not have any significant biogas production enhancement for HHW or commingled OFMSW. Pretreating FW at high temperature results in 24% and 11.7% increased biogas production at 120 °C and 150 °C, respectively. Higher temperatures (175 °C) result in a decreased biogas production, due to formation complex polymers such as melanoids.

Conventional biological pretreatments are not very popular for OFMSW, whereas two-stage AD systems with hydrogen recovery have become an interesting research field among the scientific community. Pretreatments such as composting could result in higher microbial activities \([14]\); but also result in a loss of volatile organics, and thus a potential methane production \([101]\). Moreover, for easily biodegradable wastes such as FW, hydrolysis is not necessarily the rate-limiting step, thus the increased hydrolysis due to pretreatment may lead to VFA accumulation, which subsequently inhibits the methanogens. Therefore, a two-stage AD is preferred for easily biodegradable OFMSW, as compared to conventional single-stage digesters coupled with other pretreatment methods \([132,122]\).

8. Feasibility of a full scale application

This review showed pretreatment methods can enhance the AD performance. Nevertheless, the high capital cost, high consumption of energy, required chemicals and sophisticated operating conditions (maintenance, odor control etc.) are the major factor hindering their full-scale application \([77,124,133,134]\). There are only a few examples of the thermal hydrolysis process (THP) that have been applied at a full-scale such as the Cambi, Porteous, and Zimpro process, and thermochemical pretreatment methods such as Synox, Protox, and Krepro. It should be noted that these methods are all applied for WWTP sludge. Concerning OFMSW, only a few mechanical pretreatment methods (Fig. 1), Cambi THP (Fig. 2), and an AD with a pre-hydrolysis stage (two-stage AD, Fig. 3) have been applied at a full scale.

8.1. Energy balance

The required energy depends on the desired pretreatment temperature. If it is above 100 °C, most of the energy is utilized in water vaporization, thus making it less desirable \([138]\). Some researchers report that microwave heating has advantages over conventional heating due to the direct internal heating with no heat loss \([22]\). However, according to Mottet et al., neither microwave nor ultrasound was energy incentive for pretreating mixed sludge, as the enhanced methane yields were not enough to compensate the required energy \([66]\). Yang et al. reported that thermal pretreatment significantly improves the total amount of biogas produced, and the extra biogas produced can be utilized to reduce the costs through an efficient heat exchanger \([135]\).

Escamilla-Alvarado et al. obtained a better energy balance with two-stage AD systems treating OFMSW \([122]\). However, the higher gross energetic potential was due to the higher performance in the methanogenic reactor rather than the hydrogen production from the first stage. Nasr et al. also estimated the energy balance of two-stage AD of thin stillage, and concluded that optimizing the two-stage AD process can increase the energy balance by 18.5% \([136]\). Lu et al. reported that a two-stage reactor showed a better energy balance with a surplus of 2.17 kJ/day, as compared to a single stage system for treating SS \([27]\).
<table>
<thead>
<tr>
<th>Substrate (source-sorted)</th>
<th>Pretreatment condition</th>
<th>Type of AD system</th>
<th>Results</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>OFMSW</td>
<td>Disc screen</td>
<td>Thermophilic batch</td>
<td>80.63% VS reduction with 338 mlCH₄/gVS</td>
<td>[51]</td>
</tr>
<tr>
<td>OFMSW</td>
<td>Screw press</td>
<td>Thermophilic batch</td>
<td>63.2% VS reduction with 354 mlCH₄/gVS</td>
<td></td>
</tr>
<tr>
<td>OFMSW</td>
<td>Screw press</td>
<td>Thermophilic batch</td>
<td>461 mlCH₄/gVS</td>
<td>[53]</td>
</tr>
<tr>
<td>OFMSW</td>
<td>Disc Screen</td>
<td>Thermophilic batch</td>
<td>428 mlCH₄/gVS</td>
<td></td>
</tr>
<tr>
<td>OFMSW</td>
<td>Shredder with magnetic separation</td>
<td>Thermophilic batch</td>
<td>487 mlCH₄/gVS</td>
<td>[49]</td>
</tr>
<tr>
<td>OFMSW</td>
<td>Screw press</td>
<td>Thermophilic batch</td>
<td>461 mlCH₄/gVS</td>
<td>[53]</td>
</tr>
<tr>
<td>OFMSW</td>
<td>Disc Screen</td>
<td>Thermophilic batch</td>
<td>428 mlCH₄/gVS</td>
<td></td>
</tr>
<tr>
<td>OFMSW</td>
<td>Shredder with magnetic separation</td>
<td>Thermophilic batch</td>
<td>487 mlCH₄/gVS</td>
<td>[49]</td>
</tr>
<tr>
<td>OFMSW</td>
<td>Semi-composting with Rotary drum</td>
<td>Mesophilic batch</td>
<td>Negligible effect on the enhancement of biogas production was achieved. However the kinetics of the process was faster at semi-continuous experiments</td>
<td>[52]</td>
</tr>
<tr>
<td>OFMSW</td>
<td>Semi-composting with Rotary drum</td>
<td>Mesophilic batch</td>
<td>5–11.5% VS higher reduction, and 18–36% higher biogas production</td>
<td>[50]</td>
</tr>
<tr>
<td>OFMSW</td>
<td>Composting</td>
<td>Thermophilic dry batch</td>
<td>160–205% Higher specific microbial growth rate</td>
<td>[14]</td>
</tr>
<tr>
<td>OFMSW</td>
<td>Composting</td>
<td>Mesophilic dry batch</td>
<td>19.5% VS loss, which is 40% loss of methane</td>
<td>[101]</td>
</tr>
<tr>
<td>Food waste</td>
<td>4 days microaeration with 37.5 mlO₂/Ld</td>
<td>Mesophilic batch</td>
<td>21% Higher methane yield for inoculated substrate, and 10% higher methane yield for non-inoculated substrate.</td>
<td>[100]</td>
</tr>
<tr>
<td>OFMSW (synthetic)</td>
<td>Thermophilic pre-hydrolysis</td>
<td>Thermophilic (continuous 2-stage)</td>
<td>81.5% COD removal with 95.7% VSS destruction and 2 times higher biogas production</td>
<td>[111]</td>
</tr>
<tr>
<td>OFMSW (synthetic)</td>
<td>Mesophilic and thermophilic pre-hydrolysis</td>
<td>Mesophilic and thermophilic (continuous two-stage)</td>
<td>Mesophilic pre-hydrolysis performed better in producing hydrogen, whereas thermophilic resulted in better solubilization. The highest methane production from second stage was 341 mlCH₄/gVS</td>
<td>[122]</td>
</tr>
<tr>
<td>OFMSW and corn stalk</td>
<td>Freeze explosion followed by thermophilic pre-hydrolysis</td>
<td>Thermophilic</td>
<td>104 ml-H₂/g-VS and 520 mlCH₄/gVS</td>
<td>[129]</td>
</tr>
<tr>
<td>OFMSW</td>
<td>Alkaline</td>
<td>NA</td>
<td>11.5% Higher COD solubilization, methane yield of 0.15 m³ CH₄/kgVS (172% higher than untreated)</td>
<td>[74]</td>
</tr>
<tr>
<td>OFMSW</td>
<td>Microwave pretreatment at 115–145°C for 40 min</td>
<td>Mesophilic batch</td>
<td>4–7% Higher biogas produced than untreated</td>
<td>[123]</td>
</tr>
<tr>
<td>OFMSW</td>
<td>Pre-hydrolysis at 55°C (TPAD)</td>
<td>Mesophilic continuous</td>
<td>47.5–71.6% VS destruction and methane yield of 299–418 ml/g-VS</td>
<td>[85]</td>
</tr>
<tr>
<td>OFMSW</td>
<td>Sonication at 20 kHz for 30–60 min</td>
<td>Mesophilic batch</td>
<td>60% Increased COD resulted in 24% higher methane yield</td>
<td>[56]</td>
</tr>
<tr>
<td>Household waste</td>
<td>70°C for 60 min KOH until pH = 10 at 70°C, 60 min</td>
<td>Thermophilic batch</td>
<td>Methane yield of 500 mlCH₄/gVS, no enhancement due to pretreatment</td>
<td>[69]</td>
</tr>
<tr>
<td>Household waste</td>
<td>160–200°C, 40 bar for 60 min</td>
<td>Mesophilic continuous</td>
<td>55–70% COD solubilization, and 3% higher biogas production</td>
<td>[126]</td>
</tr>
<tr>
<td>Household waste</td>
<td>Mesophilic pre-hydrolysis (hydrogenogenic)</td>
<td>Mesophilic continuous</td>
<td>43 mlH₂/gVS from first stage, 500 mlCH₄/gVS from second stage which is 21% higher than single stage system</td>
<td>[120]</td>
</tr>
<tr>
<td>Food waste</td>
<td>Microwave with intensity of 7.8°C/min</td>
<td>Mesophilic batch</td>
<td>24% Higher COD solubilization and 6% higher biogas production</td>
<td>[63]</td>
</tr>
<tr>
<td>Food waste</td>
<td>Size reduction by beads mill</td>
<td>Mesophilic batch</td>
<td>40% Higher COD solubilization and 28% higher biogas production</td>
<td>[8]</td>
</tr>
<tr>
<td>Food waste</td>
<td>Addition of HCl until pH = 2</td>
<td>Thermophilic batch</td>
<td>13 ± 7% Higher COD solubilization and 48% higher biogas production</td>
<td>[9]</td>
</tr>
<tr>
<td>Food waste</td>
<td>120°C, 1 bar for 30 min, HCl until pH = 2 at 120°C</td>
<td>Thermophilic batch</td>
<td>19 ± 3% Higher COD solubilization and 24% higher biogas production</td>
<td>[9]</td>
</tr>
<tr>
<td>Food waste</td>
<td>Pressurized until 10 bar and depressurized</td>
<td>Mesophilic batch</td>
<td>32 ± 8% Higher COD solubilization and 40% higher biogas production</td>
<td>[118]</td>
</tr>
<tr>
<td>Food waste</td>
<td>Frozen at ~80°C for 6 h, and thawed for 30 min</td>
<td>Mesophilic batch</td>
<td>12 ± 7% Higher COD solubilization and 48% higher biogas production</td>
<td>[118]</td>
</tr>
<tr>
<td>Food waste</td>
<td>Hyper-thermophilic/thermophilic pre-hydrolysis</td>
<td>Thermophilic (TPAD)</td>
<td>15–18% Higher methane conversion ratios than conventional thermophilic digesters</td>
<td>[118]</td>
</tr>
<tr>
<td>Food waste</td>
<td>Semi-aerobic and anaerobic pre-hydrolysis</td>
<td>Mesophilic continuous</td>
<td>95% COD destruction which resulted in methane yield of 500 ml/gVS</td>
<td>[127]</td>
</tr>
<tr>
<td>Food waste</td>
<td>Thermophilic pre-hydrolysis</td>
<td>Thermophilic</td>
<td>HRT can be reduced to 10 days</td>
<td>[128]</td>
</tr>
<tr>
<td>Food waste</td>
<td>Thermophilic pre-hydrolysis</td>
<td>Mesophilic</td>
<td>61.3% VS destruction, methane yield of 280 ml/gVS</td>
<td>[148]</td>
</tr>
<tr>
<td>Food waste</td>
<td>Mesophilic pre-hydrolysis (2 stage system)</td>
<td>Mesophilic continuous</td>
<td>93 and 133 Higher biogas production than mesophilic and thermophilic AD, respectively</td>
<td>[112]</td>
</tr>
<tr>
<td>Food waste</td>
<td>Mesophilic pre-hydrolysis</td>
<td>Mesophilic</td>
<td>Best results of 520 mlCH₄/gTS was achieved at pH = 7</td>
<td>[113]</td>
</tr>
</tbody>
</table>
8.2. Economic feasibility

As the pretreatment of OFMSW is relatively new, its cost estimation is still based on lab-scale level data. For instance, Ma et al. estimated the net profit of various pretreatments to enhance the biogas production of FW, and obtained the best result (10–15 euro/ton FW) with less energy intensive methods (acid and freeze–thaw) [9]. However, they have not considered thermal pretreatment at lower temperatures, which could have been more economic.

<table>
<thead>
<tr>
<th>Substrate</th>
<th>Pretreatment condition</th>
<th>Type of AD system</th>
<th>Results</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Food waste</td>
<td>Mesophilic pre-hydrolysis</td>
<td>Mesophilic continuous</td>
<td>65 mLH₂/gVS and 546 mLCH₄/gVS</td>
<td>[110]</td>
</tr>
<tr>
<td>Food waste</td>
<td>Thermophilic pre-hydrolysis</td>
<td>Mesophilic continuous</td>
<td>205 mLH₂/gVS and 464 mLCH₄/gVS</td>
<td>[121]</td>
</tr>
<tr>
<td>Food waste</td>
<td>400 pulses with electroporation</td>
<td>Mesophilic continuous</td>
<td>20–40% Higher biogas production due to substrate cell breakage</td>
<td>[46]</td>
</tr>
<tr>
<td>Food waste</td>
<td>70 ºC for 2 h</td>
<td>Mesophilic continuous</td>
<td>2.69% Higher methane production</td>
<td>[21]</td>
</tr>
<tr>
<td>Food waste</td>
<td>150 ºC for 1 h</td>
<td>Mesophilic continuous</td>
<td>11.3% Higher methane production</td>
<td>[149]</td>
</tr>
<tr>
<td>Food waste</td>
<td>Frozen/thawed and pre-hydrolysis for 7 days</td>
<td>Mesophilic continuous</td>
<td>10% Higher COD solubilization, 23.7% higher biogas production</td>
<td></td>
</tr>
<tr>
<td>Food waste</td>
<td>Frozen/thawed and pre-hydrolysis for 12 days</td>
<td>Mesophilic continuous</td>
<td>4% Higher COD solubilization, 8.5% higher biogas production</td>
<td></td>
</tr>
<tr>
<td>Food waste</td>
<td>70 ºC thermal and mesophilic pre-hydrolysis</td>
<td>Mesophilic continuous</td>
<td>91% of FW was converted to biolythane with 8% hydrogen and 83% methane</td>
<td>[127]</td>
</tr>
<tr>
<td>Food waste</td>
<td>175 ºC, 60 min</td>
<td>Mesophilic batch</td>
<td>7.9% Decrease in biogas production</td>
<td>[62]</td>
</tr>
<tr>
<td>Fruits and vegetables waste</td>
<td></td>
<td></td>
<td>11.7% Decrease in biogas production</td>
<td></td>
</tr>
</tbody>
</table>

* NA – Not available.

![Mechanical pretreatment methods to enhance AD of OFMSW](image)
The estimation of the economic feasibility of pretreatment methods based on a full-scale application has only been reported for WWTP sludge. Rittman et al. estimated the operational and maintenance (OM) cost of a full-scale AD (3300 m$^3$) treating 380 m$^3$ sludge per day based on the application of focused-pulsed pretreatment technology, which could generate a benefit of 540,000 USD per year [137]. Muller reported that a rough cost estimate of pretreatment methods is between 70 and 150 US$/tonTS for capital and OM cost [33]. Bordeleau and Droste estimated the cost of pretreatment methods to enhance the AD of sludge, based on the existing literature. They concluded that the microwave (0.0162 US$/m$^3$) and conventional thermal (0.0187 US$/m$^3$) pretreatments were cheaper than ultrasound (0.0264 US$/m$^3$) and chemical (0.0358 US$/m$^3$) methods [22]. However, the amount of sludge is an important factor to consider when estimating the pretreatment cost. Ultrasound pretreatment could be energetically feasible if a typical value of 6 kWh/m$^3$ sludge for a full-scale application is considered [138]. If a higher energy is required, biological pretreatment such as adding hydrolytic bacteria could be a cheaper option [57,139].

The cost estimation of conventional biological pretreatment has not been reported to date. The economical feasibility of a two-stage AD were estimated by Bolzonella et al., who reported that the payback time for a full-scale two-stage AD system with hyper-thermo-philic pre-stage followed by mesophilic reactor is 2–6 years depending on the method of sludge disposal [140].

In addition to the calculation of net benefits, local circumstances such as labor, treatment capacity, transport, collection cost,
energy prices, tax, purchase tariffs, land price, market, price of digested material, disposal of residue, additional mixing and pumping should be considered as well [4,5,6,132,141–146].

8.3. Environmental aspects and sustainability of pretreatment methods

In addition to the energy balance and economic analysis, environmental consideration such as pathogen removal, use of chemicals, and the possibility for a sustainable use of the residues, impacts on human health and the environment should be considered as well when choosing a pretreatment method [4,22,133,141–149]. Moreover, the anaerobic residues have the possibility to be used as soil fertilizers. Thus, the soil type as well as the potential gaseous emissions such as N$_2$O should be considered [147]. Carballa et al. evaluated the environmental aspects of different pretreatment methods including chemical (acidic and alkaline), pressurize-depressurize, ozonation and thermal treatment in terms of Abiotic Resources Depletion Potential, Eutrophication Potential, Global Warming Potential, Human and Terrestrial Toxicity Potential through a life cycle assessment. They concluded that the pressurize–depressurize and chemical pretreatment methods outperformed ozonation, freeze–thaw and thermal methods [36].

Table 3 gives a simple sustainability assessment of pretreatment methods to enhance OFMSW was carried out based on existing literature. Pretreatment methods with higher efficiencies, and that are more economically as well as environmental friendly methods obtained more plus points. The pretreatment methods with the most number of plus points were evaluated as the most sustainable. Table 3 shows that the thermal pretreatment at low temperature and the two-stage AD system were assessed as the most sustainable methods to enhance the AD of OFMSW, followed by conventional biological methods and mechanical pretreatment. Chemical, thermochemical or thermal pretreatment methods at high temperatures could result in a higher enhancement of the AD process as compared to untreated substrates. However, the costs of the methods as well as the environmental considerations make it less desirable.

9. Conclusion

The growing global concerns on the increasing amount of waste, energy demand, and global warming have stimulated research on the acceleration and enhancement of the AD process. Pretreatment methods can be categorized as mechanical, thermal, chemical, biological or a combination of them. Among the widely reported pretreatment methods tested at lab scale, only few mechanical, thermal and thermochemical methods were successfully applied at full scale. Based on a simple sustainability assessment, thermal pretreatment (at low temperatures) and two-stage AD systems offer more advantages as compared to the other pretreatment methods. These include: (i) higher biogas yield; (ii) decisive effect on pathogen removal; (iii) reduction of digestate amount; (iv) reduction of the retention time; (v) better energy balance and (vi) better economical feasibility.

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References


