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Review

A Review of Pretreatment Methods to Enhance Solids Reduction during Anaerobic Digestion of Municipal Wastewater Sludges and the Resulting Digester Performance: Implications to Future Urban Biorefineries

Bimi Shrestha [1], Rafael Hernandez [1], Dhan Lord B. Fortela [1], Wayne Sharp [2], Andrei Chistoserdov [3], Daniel Gang [2], Emmanuel Revellame [4], William Holmes [5] and Mark E. Zappi [1,*]

1 Department of Chemical Engineering, University of Louisiana at Lafayette, Lafayette, LA 70504, USA; bimi.shrestha1@louisiana.edu (B.S.); rafael.hernandez@louisiana.edu (R.H.); dhan.fortela@louisiana.edu (D.L.B.F.)
2 Department of Civil Engineering, University of Louisiana at Lafayette, Lafayette, LA 70504, USA; wayne.sharp@louisiana.edu (W.S.); daniel.gang@louisiana.edu (D.G.)
3 Department of Biology, University of Louisiana at Lafayette, Lafayette, LA 70504, USA; andrei.chistoserdov@louisiana.edu
4 Department of Industrial Technology, University of Louisiana at Lafayette, Lafayette, LA 70504, USA; emmanuel.revellame@louisiana.edu
5 Energy Institute of Louisiana, University of Louisiana at Lafayette, Lafayette, LA 70503, USA; william.holmes@louisiana.edu
* Correspondence: mark.zappi@louisiana.edu

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Abstract: The rapid increase in the population is expected to result in the approaching of design capacity for many US wastewater treatment plants (WWTPs) over the next decade. WWTPs treat both municipal and industrial wastewater influents, resulting in the production of biosolids after digestion. Biogas, a potential recovered alternative energy source, is also produced as an output from successful anaerobic digestion. More than 7M of dry tons/year of biosolids produced in the US are most often disposed in either landfills or land-applied (~80%). These options are becoming more challenging to implement due to increases in transportation costs and tipping fees, decreases in the availability of landfill/landfarm space, and most importantly, increased regulations. This situation is strongly encouraging WWTPs to find alternatives for the disposal of biosolids. Developing alternative management/disposal options for biosolids are evolving. One of the most attractive alternative option from a sustainability perspective are biorefineries (converts waste to commercial products), which are a fast-growing option given the push toward circular urban source economies (little to no waste generation). Anaerobic digestion has been widely applied in WWTPs to reduce the volume of activated sludge due to its low energy requirements, effective handling of fluctuations due to organic loading rate, relative flexibility with temperature and pH changes, and since biogas is produced that can be transformed into energy. Various pretreatment methods for waste sludges prior to digestion that have been studied to reduce solids production and increase the energetic content of the biogas are presented and discussed. Solids handling and management, which comprises ~60% of the operational cost of a WWTP, is estimated to save more than $100 M annually by achieving at least 20% reduction in the annual production of biosolids within the US. This review incorporates an assessment of various pretreatment methods to optimize the anaerobic digestion of waste sludges with a focus on maximizing both biosolids reduction and biogas quality.

Keywords: anaerobic digestion; biosolids volume reduction; biogas production; sustainability of urban waste management

1. Introduction

Wastewater treatment plants (WWTP) are designed to treat municipal and industrial wastewater influents to lower the ecological risks associated with discharging the treated effluents into receiving water sources. Within these plants, the wastewater (WW) influent goes through a series of unit processes (mainly bio-based) to reduce the organic matter content, odor, and pathogen levels with the resulting end-materials exiting the plants being water effluent, biosolids, and biogas. Effluents are the treated water that are released after meeting applicable treatment standards. Biogas is the gaseous product produced during the anaerobic, methanogenic-based biodegradation of organic wastes and/or waste sludges. The gas is typically composed mainly of methane and carbon dioxide at levels in the 50–70% (*v*/*v*) and 30–50% (*v*/*v*) levels, respectively, with residual amounts of hydrogen, sulfur dioxide, and volatile organics. Biosolids are biologically digested sludges (end-process sludges), technically derived from either anaerobic or aerobic digestion processes that are typically dewatered to the 15 to 20% (*w*/*w*) range. In the context of this paper, only anaerobic digestion is considered. Over 7M dry tons of biosolids (the majority being anaerobic products) are produced annually in the United States [1]. With increasing urbanization due to population growth/densification, the amount of produced wastewaters is rising within a reduced area which makes the management of the biosolids one of the biggest concerns in WWTP industry. By the 2032, 56 million more people are expected to input into centralized urban-based WWTPs which is an overwhelming load for the WWTP industry to manage at its current capacity which is currently treating the generated wastewater from nearly 240 million Americans [2].

There are almost 14,700 WWTP facilities in the United States that produce approximately 33 billion gallons of WW effluents daily [3]. One key challenge at present is managing the solids that are being generated at the WWTPs. Biosolids contain low percent levels (*w*/*w*) of organic nitrogen and phosphorus and can be applied as fertilizers and soil amendments via land application. Other proven methods of handling biosolids are incineration, forestry application, and disposal in landfills [1]. The incineration of dried solids results in emission of greenhouse gases (GHG) which contributes to global warming. As seen in Figure 1 which shows the areas for biosolids use and disposal, agriculture is the mostly used area which requires the biosolids to be of high standard, free from pathogens, and rich in nitrogen and phosphorus. The second most popular choice for biosolids management is landfills which is not a sustainable option since available landfill volumes in the US are decreasing, thus becoming expensive as well as facing increasing regulatory and policy pressure to recycle or reduce wastes instead of landfilling. In addition to the unavailability of viable land, the cost to haul the dried solids is also of concern to WWTP managers. There is an average of $50 per dton tipping fee inclusive of hauling costs of the dried biosolids and landfill use fee which represents a significant expense to the WWTP. This also represents a waste of valuable carbon that potentially could have been used to make new, sustainable chemical products produced from urban biorefineries. Table 1 shows the increase in tipping fees from 2018 to 2019 in different regions of the US which is expected to increase in the future [4]. The carbon footprint associated with the hauling of solids to landfill makes this option carbon-intensive and unsustainable. The disposal costs for biosolids generated at US WWTPs generally represent about 60% of the annual operational cost of the WWTP [5] and is a major reason to look into disposal/post-production use alternative which includes processes that can potentially reduce the generation of solids, yet still support biorefineries. Any significant reduction of solids would lessen the burden of disposal and cost of transportation while being sustainable and environment friendly with the potential production of a value-added product(s) furthering the level of system sustainability.

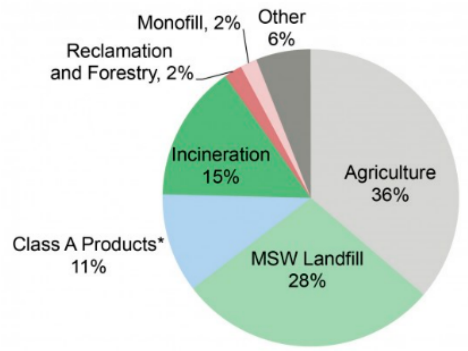

Figure 1. Biosolids use and disposal (note: * denotes Class A sludges; source: US Wastewater Treatment[3]).

Table 1. Rise in average US tipping fees: 2018 to 2019 (Source: wastetodaymagazine.com).

Region	Average Cost per Dry Ton (US $)	Percent Increase
Pacific North	70	0%
South Central	41	15%
Mountain	51	17%
Overall	56	6%

1.1. Anaerobic Digestion in WWTP

Anaerobic digestion (AD) is the popular treatment process within WWTPs due to its proven efficiency to further reduce pollutant levels, yield a fairly stabilized sludge, substantially reduce sludge tonnage needing disposal, use of minimum input energy, and the production of biogas. Most WWTP ADs are actually treating waste sludges in the form of primary sludge and secondary sludge (i.e., waste activated sludge or WAS) [6]. Within an AD, anaerobic microbial consortia degrade the organic content within the input wet sludges under strict anaerobic conditions to produce a fairly stable solid (i.e., biosolids) and biogas. The anaerobic digestion process is based on four key microbial-based steps; hydrolysis, acidogenesis, acetogenesis, and methanogenesis which are shown in Figure 2. Hydrolysis is the first step where complex organic sludge constituents such as carbohydrates, proteins, and lipids are broken into smaller compounds such as fatty acids, amino acids, and glucose. This step is the rate limiting step since it takes the most amount of time due to the lysing of the recalcitrant cell walls. Following hydrolysis is acidogenesis where the acidogenic bacteria feed on sugars, fats, and amino acids to produce complex organic acids, alcohols, carbon dioxide, and ammonia. Next, acetogenic bacteria feed on the products of acidogenesis to produce low molecular weight fatty acids (such as acetic, lactic, and propionic acids), hydrogen, and carbon dioxide. Methanogenesis is the final step where methanogenic microbes, archaea (also known as methanogens), convert the organic acids, hydrogen, and carbon dioxide into biogas. Biogas in non-optimized AD systems typically produce a biogas with ~55% methane (*v*/*v*) and ~45% carbon dioxide (*v*/*v*). Optimized systems can produce a biogas with ~80% methane and ~20% carbon dioxide. The calorific value of biogas is generally 500–800 BTU/ft^3 which is comparable to the calorific value of natural gas which typically ranges between 950–1150 BTU/ft^3.

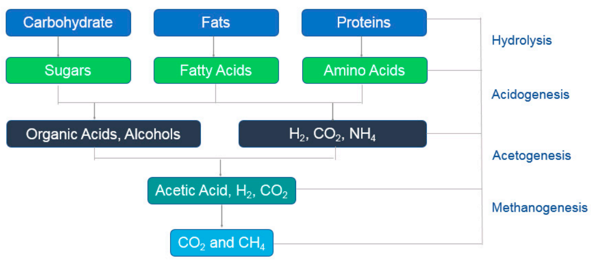

Figure 2. Anaerobic digestion reaction processes.

Anaerobic digestion can be conducted at 3 different temperature ranges: (1) psychrophilic (10–15 °C), (2) mesophilic (28–45 °C), and (3) thermophilic (>60 °C). Mesophilic AD is the most popular due to low energy requirements, the production of stable solids, and good biogas production. The metabolic activity of microorganisms is reduced with decreasing temperatures. Hence, temperature is an important parameter in the digestion process for maximum biological activity from microorganism leading to optimized pollutant removal and biogas production. For the same amount of organic pollutant removal, lower temperature kinetically slows down the hydrolysis process thus requiring longer hydraulic retention times (HRT) and solids retention times (SRT) compared to digestion at mesophilic temperatures (i.e., 35 °C) [7]. In addition, low temperature leads to less utilization of the substrate and changes the microbial community structure. Thermophilic temperature range produces higher biogas volumes; however, the high energy requirement (secondary heating often using the produced biogas) for the increased temperature increases the operation and management (O&M) costs. There also exists the potential for the formation of refractory compounds at very high temperatures.

1.2. Solids Composition

The treated non-gaseous products exiting an AD consist of liquid effluent and treated sludge streams [8]. Influent bulk solids concentrations into an AD at most WWTPs are in the 5% solids range which has the appearance and handling of a thick slurry that is pumpable. After digestion, the biosolids are typically dewatered to about 15 to 25% (*w*/*w*) solids with the liquid effluent either recycled back to the head of the WWTP or discharged depending on pollutant and pathogen levels. Dewatering of the slurries exiting an AD is often done using either a belt press or centrifuge [8]. Supernatant (liquid) pollutant levels exiting the AD are generally in the 500 mg/L 5-day biochemical oxygen demand or BOD5 range. The Wisconsin Department of Natural Resources (WDNR) quotes the US EPA as describing biosolids as "primarily organic solid product yielded by municipal wastewater treatment processes that can be beneficially recycled" as soil amendments (fertilizer and conditioners)" [8]. As this description infers, biosolids has or should have a use beyond simple disposal. Without a use beyond production, biosolids and hence WWTPs cannot become truly sustainable. The sludge that exits the AD, once dewatered, is considered at that point to be the "biosolid" product. The sludge is often measured as total solids (TS) comprised of, an organic fraction also called volatile solids (VS), and an inorganic fraction or fixed solids [9]. Total Suspended Solids quantifies suspended solids (particulates) but not soluble solids while Volatile Suspended Solids (VSS) quantifies suspended organic solids that does not include soluble solids or inorganic solids. Other measurements such as Total Chemical Oxygen Demand (tCOD) which measures the total amount of oxygen equivalent as potassium permanganate required to oxidize the organic pollutant; Soluble COD (sCOD) which is same as tCOD but without oxidizing particulates (only soluble chemicals); and Total Organic Carbon (TOC) which measures the organic carbon present in wastewater (excluding inorganic carbon), are used to define the characteristics of the sludge. However, VS and TS are the most common measurements used to quantify and qualify sludges.

Typical performance goals for AD systems at most US-based WTTPs are the >38% removal VS; 30% to 60% removal of TS; 85% to 99% pathogen removal; and biogas with a methane content >50% [8,10]. The solids profile entering and exiting the AD are generally 75% volatile and less than 40% volatile, respectively, which indicates more VS removal (in other words, additional organics) could be achieved with further system optimization of a typical AD unit (there still generally remains 40% volatile fractions which tend to be fairly biodegradable). As stated above, once dewatered, the sludges exiting a typical AD range from 15% to 30% (*w*/*w*) solids content. At 15% solids, the material feels like a wet soil with 30% solids having the feel of a damp soil. At these higher solid concentrations, mechanical bulk handling, such as conveyance or bucket loaders, are often used for sludge transport.

It is important to note that the primary composition of sludge inputs into an AD system at most WWTPs is wasted activated sludge which is mainly aerobic microbial cells. The aerobic microbial cells in the form of activated sludge are wasted to maintain a target cell density within the aerobic treatment phase of the plant (generally in the 1 to 5 g/L range). Sometimes, WWTPs will also have primary sludge inputs integrated within the waste activated sludge (aka. WAS or secondary sludge). Primary sludge is the sludge produced when the solids within the overall WWTP influent is settled thus it is composed mainly of feces, food wastes, bacterial cells, and light paper goods. Usually, WAS is the majority feed into an AD input sludge followed by primary sludge. These organic constituents of input sludge to an AD make up the bulk of the VS fraction. Thus, WAS and primary sludges are mainly composed of proteins, carbohydrates, and lipids with some fixed solids present in the form of grit (generally low levels). It must be noted that more often co-digestion (combining waste sludge with an organic feed, such as food wastes) is occurring at US municipal WWTPs thus increasing the complexity of AD sludge inputs but also potentially increasing the BTU output of the system [11]. Co-digestion is opined to be beneficial when attempting to enhance the biorefinery aspects of a municipal WWTP.

1.3. Potential Benefits of Further Optimization

Anaerobic digestion has been used in treating wastewater sludges since it has a lower energy usage compared to aerobic digestion (no aeration). The solids characteristics of a typical biosolid exiting an AD system has significant organic content left that can be further removed to achieve less sludge needing disposal and likely produce more biogas/methane within the exiting biogas. Based on the above information, assuming a $50/dton cost for disposing of the biosolids is a reasonable assumption. At 7,000,000 dtons of biosolids requiring disposal each year within the US, this comes to an estimated cost of $350 M US dollars spent each year within the US. If some form of AD system optimization could be performed that reduces this tonnage by 20%, then ~$70 M of savings could be realized within the US each year. Additionally, since most optimization efforts will result in higher VS conversion into methane, this adds more recovered energy to the utilities and thus potentially reduces net operating costs. Hence, this study of the literature was performed to identify promising pretreatment methods that could easily be applied at most US WWTPs to reduce the amount of biosolids tonnage requiring disposal while enhancing the potential for AD system to be further integrated into developing urban biorefinery design strategies. Pretreatment strategies have been studied over the years with increasing the energetic content and amount of biogas from AD systems. A secondary, but much less considered result, these same pretreatment strategies could open opportunities to reduce the total tonnage of biosolids needing disposal. With more and more environmental policies and laws encouraging better performance through reduced sludge production and increased secondary product development while decreasing landfill volume usage and/or reduce open landfarming of biosolids, the need for defining the most promising pretreatment methods that can be reasonably implemented and integrated into current WWTP designs. Biogas production from anaerobic digestion makes the process an energy generation process which produces clean energy such as methane and/or hydrogen. The benefits of AD to WWTPs has attracted applications of different methods to optimize the digestion process. This paper focuses on reduction of generated solids by anaerobic digestion using treatment methods prior to anaerobic digestion along with some other performance metrics (when reported), such as biogas quality and quantity.

2. Pretreatment Methods for Solids Reduction

With the increase in research on pretreatment technologies to improve AD treatment of wastewater sludges, several methods that can be categorized as mechanical, chemical, biological, or thermal have been studied including combinations of these processes. Physical methods can be categorized as mechanical and thermal methods since they use external energy sources for treatment. Chemical pretreatment methods utilize the addition of various chemicals for reaction prior to digestion while biological pretreatment methods use the metabolism of microorganisms and their environmental preference for treatment. Physical and chemical methods have been most widely studied while biological treatment methods appear to have been studied to a lesser extent [12]. Thermal processes have also seen some application, but often in conjunction with other mechanisms. Physical-chemical and thermo-chemical integrated processes are the most popular combination of different methods that have shown positive results [2]. In selecting an optimal pretreatment process, the costs associated with the additional treatment/handling, additional process equipment O&M, overall energy requirements, capital investment, and the influent sludge characteristics, such as the organic loading rate (OLR), TOC, COD, working temperature, and pH all need to be considered for the optimized selection and design of a candidate pretreatment system. The goal of the pretreatment methods is to make more organic sludge constituents available to the microorganisms for digestion thus striving toward a more complete conversion into biogas and microbial cells (albeit tending to be minimal) which reduces the amount of solids requiring disposal. By comparing the results obtained from the published studies, optimal methods can be established that use minimal energy and require low costs to implement.This paper focuses on solids reduction from studies on using various pretreatment methods prior to AD processing. It also reports on the resulting biogas production (where reported) in terms of quality (methane content) and quantity. The COD removal and biogas production from the various pretreatment methods discussed in this paper have been summarized in Table 2.

2.1. Mechanical Disintegration

Mechanical disintegration pretreatment reduces particle size by using external stresses and pressures to convert a higher percentage of the sludge constituents into soluble fractions thus making more of the sludge mass available for microbial digestion resulting in an increase in biogas production and a reduction of solids [13]. Decreasing the particle size increases the surface area for the microorganisms to feed on the organic sludge components thus improving the extent of bioavailability and biodegradability (such as cell lysis). This processing also ruptures microbial cells in the influent sludge, thus releasing more biodegradable substrate that would have been shielded within the cellular boundaries (walls and/or sheaths). The smaller the radius of the particle, the higher the extent of the substrate utilized by the microorganisms [14]. This pretreatment process category accelerates hydrolysis and acidogenesis reactions which leads to faster production of VFAs which can inhibit digestion or create AD system instability by overwhelming the kinetic capacity of the methanogenic step. Ultrasound, microwave, and high-pressure homogenization are the most used equipment with this pretreatment process category. The extent of sludge disintegration depends on the power supply and treatment time. The higher the power, the shorter the treatment time. However, very high-power applications and/or excessive treatment times can have an adverse effect on the AD microorganisms, and thus, a negative result can be observed. One drawback of this pretreatment method is high energy consumption, but this can be compensated by the production of quality biogas and reduction in solids [13,15].

2.1.1. Ultrasound Pretreatment

The ue of ultrasound produces cavitation in liquid which creates high pressure and temperature pockets necessary to create destruction shear forces on the sludge components. Hydroxyl radicals are also created from water during ultrasonic treatment hence this treatment uses cavitation and chemical reaction mechanisms for cell lysis within the sludge which breaks down cell walls and thus releases cell components, such as DNA [15]. Sonic frequencies in the range of 20–40 kHz have been reported to be optimal for disintegrating the microbial cells for later solids reduction [13]. Tiehm et al. studied the use of ultrasound pretreatment of sewage sludge within an AD showing a 60% reduction in the required digestion retention time, from retention time of 22 days to 8 days and an increase sludge VS removal by 44%.The biogas produced in the treated WAS for use a 8 day retention time was twice that of the biogas produced without pretreatment with a 22 day retention time [14]. In terms of sonic power needs, a sonication density of 0.51 W/m and sonication intensity of 4.8 W/cm^2 for 15 min resulted in 24.6% increase in sludge VS reduction during AD treatment of WAS [16]. Kim et al. treated WAS prior to AD with 42 kHz of ultrasound for 120 min and achieved a 39% VS enhanced reduction during AD [17]. The effects of 20 kHz ultrasonic treatment of gravity thickened WAS showed an insignificant change in TS and VS removal; however, the applied energy had positive effect on particulate solubilization with an increase in sCOD, which means COD was transferred from particulates to soluble form by increasing ultrasonic specific energy to 8800 kJ/kg TS [18]. This means although there was no effect on mineralization and evaporation phenomena occurring, there was an increase in disintegration degree. Li et al. pretreated WAS using a 20 kHz ultrasound system with an energy density of 0.5 W/mL for 0 to 100 min. They reported an improvement in sludge COD removal between 30% to 50% as pretreatment time was increased, along with an improvement in VS removal ranging from 4.9% to 29.8% compared to no pretreatment [19]. This study concluded that an increase in ultrasonic treatment time increased the COD removal efficiency, likely due to an increase in sludge compositional soluble fractions. Studies have also indicated that solubilization is directly dependent on the energy applied with higher energy leading to higher disintegration. Pretreatment of WAS prior to AD at 53,000 kJ/kg TSS increased sCOD by 45% according to Foladori et al. [16]. An increase in energy density from 0.12 W/mL to 1.5 W/mL for 30 min applied to WAS increased sCOD from 10.78% to 15.11% according to a study by Xu et al. [20] These studies show that higher input energy leads to higher solids disintegration and thus better solids reduction during subsequent AD treatment. Sonication also was observed to increase soluble COD and sludge disintegration with an increase in methane generation during AD of WAS. The overall value of the process must break even in terms of the cost of energy consumed for this process versus biosolids volume reduction thus reduced tipping fees to be economically viable. Additionally, the economic benefit of biogas used as a recovered energetic product should be considered.

2.1.2. Microwave Pretreatment

Microwave pretreatment has been applied as an alternative to thermal pretreatment due to its lower consumption of energy and lesser reaction times. The production of heat through conventional heating is susceptible to excessive heat losses due to energy transmission which does not occur in heating using microwave treatment because it is an internal heat mechanism (activates water molecules causing heating). Faster heating, compact size of the instrument, and improved reduction in pathogens over traditional heating have made this technology applicable in decomposition of organic pollutants, sterilization of medical waste, water disinfection, and inactivation of pathogens [21]. Microwave pretreatment uses a set microwave frequency that ranges between 0.3 GHz to 300 GHz (home ovens use 2.45 GHz) which falls between infrared and radio waves [22]. Thus, the commonly used frequency is generally at 2450 MHz level with power ratings in the 700 W to 1000 W range to disrupt cells within the sludge. The power and treatment time dictate the level of energy applied on the sludge. Microwave can also have both thermal and non-thermal positive effects on sludges [15]. The thermal effect includes breaking of the cells by friction and molecular rotation caused by the increased interaction of electric field. The use of microwave-based pretreatment also is reported to increase the solubilization of organic matter within the sludge which aids in the reduction of solids after digestion. Organic matter such as intracellular matter can be exposed by the lysing of cell membranes and denaturalization of membrane proteins. High temperature can lead to boiling leading to the formation of bubbles which create high pressure and stress on the cell walls resulting in the rupturing of cell walls/membranes. The non-thermal effect occurs due to rapid change in dipole orientation of cell membranes which breaks the hydrogen bonds between molecules leading to cell membrane rupture [12]. Internal resistance to rotation converts microwave energy to heat through bond decomposition and repositioning [22]. Non-thermal effects in microwave irradiation are difficult to monitor since the heating mechanism is different than conventional heating. Coelho et al. pretreated primary and secondary sludge mixtures for testing within mesophilic, thermophilic, and temperature-phased AD systems using SRTs of 5, 10, 15, and 20 days. SRT did not show a significant increase in VS removal which was about 48% for the mesophilic two-stage system and about 51% for the thermophilic two stage-system [23]. Similarly, Appels et al. treated sludge at microwave energy of 336 KJ/kg, power of 800 W, and the feeding of thickened sludge weekly using an HRT of 20 days [24]. Their results indicate the achieving an increased solubilization of sludge as observed via the sludge sCOD increasing by 214% over the untreated sludge [25]. A study by Serrano et al. on the solubilization of sludge organic matter composed of primary and secondary sewage sludge, using microwave specific energies up to 20,000 J/g TS, and power outputs in the 400 to 700 W range, showed solubilization of the whole sludge organic matter over a digestion period of 90 days [26] A 30% increase in sCOD was observed at 400 W and specific energy of 30,000 J/kg TS. When using a higher energy, 700 W at a specific energy of 10,000 J/kg, the resulting TS increased the sCOD concentration by 67%; thus, indicating a more effective pretreatment at the higher power but at a lower applied specific energy [26] Reports indicate that microwave pretreatment does not require as much capital investment as ultrasonic treatment, consumes less energy compared to ultrasonic pretreatment, thus, also keeping O&M costs down. Unfortunately, microwave pretreatment has no field demonstrations performed to date as it has only been applied at the laboratory scale to date [15].

2.1.3. High Pressure Homogenization

High pressure homogenization (HPH) is a less energy intensive technique compared to ultrasonic treatment that uses pressures up to 10 MPa and homogenizes substrate under strong depressurization to break down the particles in the sludge influent. Pressure and homogenization cycles, which is the number of times the sludge fluid is passed through the homogenizer, are the main factors that determine the effectiveness of this pretreatment method [27] Pressure is applied externally with an air displacement pump that forces fluid to pass through a narrow orifice. The pressure which can be controlled by a homogenizing valve drops instantly causing strong shear forces within the fluid. The intercellular materials are released when the external pressure is greater than the internal resistance pressure which ruptures the cell membrane [28]. The process uses three mechanisms to break down the microbial cells within the sludge: eddies, impingement, and cavitation [27]. The energy applied by the valve is converted into kinetic energy which creates turbulence in the sewage sludge that results in formation of mixing eddies. The high-energy eddies are responsible for the shear forces that break the cells and make intercellular materials available. Impinge stress when applied on a plate creates an impact which results in a liquefied homogenate. The sudden drop in pressure causes forces within the bulk fluid to collapse onto the plate near the exit orifice creating cavitation and shearing. Cavitation is formed near the exit orifice and in spaces between valves in the homogenizer where the fluid pressure drops below the liquid vapor pressure (hence cavitation). The increase in surface area by sludge particle disintegration creates a finer emulsion thus accelerating the hydrolysis step with the AD. This pretreatment method requires an air displacement pump which is relatively easy to use, does not require the use of chemicals, effectively increases solubilization, requires low capital investment, and has lower O&M costs over the other two option discussed thus far which makes it a favorable candidate pretreatment option [29].

In a study by Zhang et al., anaerobic sludge was pretreated with HPH at pressures ranging from 0–50 MPa for one and two homogenization cycles. VS removal increased from 36% to 42% when HPH was applied at 50 MPa for one and two homogenization cycles, respectively. However, at 40 MPa pressure, VS removal was 33% at first homogenization cycle and then increased to 40% after the second homogenization cycle [29]. Sludge disintegration was analyzed with the researchers concluding that as the pressure was increased, the disintegration degree was increased; however, it was not as linearly as it performed during the lower pressure range of 0–30 MPa than it did when the pressure was changed from 40–50 MPa. The disintegration of sludge and increase in VS removal indicated that the pretreatment solubilized sludge matter. A subsequent study by the same team at 80 MPa and four homogenization cycles decreased TSS by 31% and the VSS by 37% during subsequent AD processing. A linear relationship of VSS and TSS indicated 86% of the TSS was contributed by the VSS which means the organic matter was transferred from the sludge solid matrix into the liquid matrix. The increase in homogenization cycles and high pressure resulted in an increase in both VS and TCOD removals [27].

2.2. Thermal Pretreatment

Thermal pretreatment has been reported to improve anaerobic digestion by breaking the gel structure in sludge which releases the water present in the cells [30]. Early studies on thermal pretreatment of sludge have reported to have a greater impact on carbohydrates and proteins than lipids. The effectiveness of this pretreatment method depends on the temperature and the length of treatment time; however, Carriere et al. concluded that treatment time had less effect on sludge solubilization than temperature. Increased solubility due to thermal pretreatment results in increased solids reduction when digested within the AD (thus reduces biosolids tonnage produced) along with pathogen removal, increased production of biogas, and improved the removal of odors [31]. Optimal temperature for thermal pretreatment ranges between 70–200 °C [30]. Heat can be applied through water immersion within a heated tank, autoclaving via pressure and heat, heating within heat exchangers, radiant heating via electric heat, or heating via the application of steam. Lower temperature pretreatment ranges from 70–95 °C while higher temperature pretreatment ranges from 100–200 °C [12]. High energy usage is necessary to maintain high sludge temperatures which can make this pretreatment process costly. However, quality biogas produced can be used to generate electricity on-site using a biogas-driven genset, which may breakeven the cost to use this process. In addition, thermal pretreatment increases efficiency of digestion, producing Class A solids that is pathogen free which makes this the most widely studied pretreatment method to be applied in full scale.

2.2.1. Low Temperature Heat Application

Low temperature pretreatment is applied at temperatures between 70–95 °C and is also dependent on the length of heat application time. Temperature and treatment time are often reported by past studies reviewed as the two key factors that determine the solubilization of sludge which in turn impacts biodegradation potential with a higher soluble substrate being more bioavailable. This technology can be applied over a few hours to as much as a few days. Lower temperatures generally are reported to require longer treatment times to gain a similar hydrolysis effect compared to pretreatment at higher temperatures. Appels et al. studied pretreatment at 70 °C, 80 °C, and 90 °C of waste activated sludge for up to 90 min which resulted in effective solubilization of organic and inorganic compounds. For a treatment time of 30 min at 70 °C, the sCOD increased by almost 18 times the sCOD of untreated sludge. However, a treatment time of 60 min at 70 °C did not produce higher sCOD than the 30 min of treatment. Similarly, a high release of heavy metals in the water phase was reported at higher temperature [32]. Heavy metals are present in sludge in various forms that are carbonate bound, including iron and manganese oxides. At high temperatures, the accelerated rate of sludge solids solubilization is likely due to the increase in ion diffusivity which is the mechanism for transportation of heavy metals from sludge flocs into aqueous forms as the sludge solid form degrades. Another study by Xue et al. on thermal hydrolyzation of dewatered high solid concentration sludge (raw sludge TS at 16.7%) pretreated at 60, 70, 80, and 90 °C for up to 72 h was reported to have reduced the sludge VS by 31% [33]. Thermal pretreatment also made intracellular materials more available in the liquid phase which was indicated by an increase in sCOD from 4.5% in the raw sludge to 29.6%, 30.3%, 34.8%, and 41.1% at 60, 70, 80, and 90 °C within 24 h, respectively. The increase in soluble proteins and soluble carbohydrates was also reported with more stable concentrations of soluble protein and carbohydrate at lower temperature than at higher temperatures.

2.2.2. High Temperature Heat Application

High temperature thermal pretreatment has proven to be more effective within the temperature range of 160 °C to 180 °C with treatment times of less than an hour. At a temperature of 175 °C, sludge production post-AD treatment was reduced by 50–70% depending on the composition of the feed [30]. However, at high temperatures, defined as over 200 °C, formation of refractory products due to caramelization can cause inhibition instead enhancement. According to Braguglia et al., thermal pretreatment of WAS at 135 °C using an autoclave as pretreatment to thermophilic digestion at 55 °C via a 20 day HRT increased the post-AD VS removal rate by 46% [18]. They also defined the final sludge product to be a "stable" sludge since VS removal rates >40% at thermophilic temperatures is defined by the USEPA as a stabilized sludge [18]. Results from the AD treatment at the mesophilic temperature of 37 °C obtained 37% more VS removal at an HRT of 20 days over no pretreatment. Decreasing the HRT to 10 days adversely impacted digestion in terms of VS removal likely due to the increased organic loading rate (ORL) which over-loaded the digester. Another study by Bougrie et al. pretreated sludge containing 14.5 g/L TS initial levels at 135 °C and 190 °C for 30 and 15 min, respectively, at an HRT of 20 days and an OLR of 1 g COD/L/day. The total COD removal increased from 52% to 64% at 190 °C while also obtaining better filterability results while also observing a decrease in post-AD sludge production by 30%. An 84% increase in soluble carbohydrate and lipid levels, and a 46% increase in proteins supported the observed higher sludge AD-based biodegradation yields at 190 °C. These improved rates were higher for the lipid and carbohydrate sludge fractions than the protein fraction. The TS removal for the 190 °C treatment was 49% which is higher compared to the TS removal of 35% observed at the treatment temperature of 135 °C. However, formation of recalcitrant compounds at elevated temperatures was observed as well as an increase in stable proteins [30]. Research on high temperature pretreatment on WAS has shown that 160–180 °C appears to be the optimal temperature requiring less time for higher COD solubilization, an increase in dewaterability, and significant VS reduction [34].

2.2.3. Full Scale Heat Application Case Studies

The Cambi^{THP} process is a full-scale commercial process that has been available worldwide since 1995 which uses thermal pretreatment for enhancing digestion. The process operates at 150 to 165 °C temperature range using residence times ranging from 20–30 min at an operating pressure in the 8.5 bar range. The results reported indicate that the process increases biodegradability, reduces sludge volume, increases digester throughput by almost three times, reduces foaming problems, improves sludge dewaterability to the 40% solid level, and yields a final sludge product that is eligible for Class A designation [25]. The US has three WWTP locations using this process with three more under construction. Biothelys is another commercial process available since the late 1990s that operates at a temperature of 165 °C and pressure of 9 bars. This process is reported to improve digestion performance by producing 25–35% less dry sludge needing disposal, and enhances sludge dewatering solids content from 22% to 30% corresponding to a 46% reduction in sludge volume compared to a typical digestion process [35]. Exelys is a second-generation version of the Biothelys process in an early stage of development. Both processes solubilize solids prior to digestion. The former process uses a series of batch units and the latter uses a continuous plug flow reactor. Cambi process has more application history because it has been commercially available for a longer period; however, the Exelys process shows potential as it is reported to have a lower capital investment compared to the Cambi process [36].

2.3. Chemical Pretreatment

Chemical pretreatment uses chemical additives such as acids, alkali, oxidizers (such as hydrogen peroxide, ozone, and Fenton's reagent), trace metals, and nutrients to improve the disintegration of the complex molecules in wastewater sludges prior to AD processing. The use of chemicals is mainly done to accelerate the rate limiting hydrolysis reaction. The increase in the extent and rate of hydrolysis results in the higher production of biogas, shortens the HRT, and reduces dry sludge production due to the availability of more soluble compounds and the later conversion into biogas. Chemicals break down the cell membrane which makes the organic matter present in the cell walls more bioavailable hence increasing the solubility of some sludge components. This method is preferred for sludges containing high lignin levels because lignin tends to be difficult to biodegrade [14]. In easily biodegradable sludges that contain mainly carbohydrates, the accumulation of VFAs within the AD can adversely affect methanogenesis due to the resulting reduced pH levels which tend to inhibit methanogens. Chemical pretreatment increases the O&M cost related to the purchase of chemicals; however, the additional costs can break-even or become cost-advantageous with the production of a high-quality biogas and reduction of sludges requiring disposal (lower overall disposal costs).

2.3.1. Acidic and Alkaline Addition

The use of acids to pretreat the wastewater sludges prior to the AD step has been studied. Acids are reported to break down recalcitrant compounds, such as lignin and cellulose, into smaller, easier to degrade by-products. Acidic environments are also favorable to organic acid degrading organisms (such as acetogens and acidogens) by allowing them to acclimate to AD conditions faster and flourish within a slightly acidic microenvironment. However, low pH levels also make the digester "sour" which leads to methanogenic inhibition as well as the formation of refractory compounds such as furans requiring this treatment to be paired with other pretreatment methods [14,31]. Devlin et al. studied the effects pH levels within the 1–6 range using hydrochloric acid prior to the AD treatment of WAS at 37 °C for 21 days for batch digestion and HRT of 12 days at 35 °C for semi-continuous digestion [37]. Addition of an alkali after acid pretreatment is required to bring the influent sludge pH back to a neutral level prior to introduction into the AD to prevent inhibition of the microorganisms, mainly the methanogens. The results on semi-continuous digestion reported a slight to no increase in VS destruction when pretreated atpH of 2 (33% for untreated and 34% at pH 2); but they observed an increase in soluble carbohydrates and proteins by up to six-fold compared to the control. The amount of biogas produced by the semi-continuous digester digesting WAS that was pretreated at pH 2 after 36 days yielded a 14% increase in biogas methane content which suggests an increase in the bioavailability of the sludge organic fractions. Another study by Sun et al. used peracetic acid (PAA) at doses ranging between 10–50 mg PAA/g SS on WAS digested at 35 °C for 28 days (batch) to investigate its effect on biogas production and resulting solids removal post-AD [38]. This pretreatment resulted in the biogas volume increasing by 20%, the sludge solids concentration and volatile solids concentrations decreasing by 25% and 39%, respectively, after 120 min of contact at a 30 mg PAA/g SS dose. These results suggest an enhanced biodegradation and disintegration of the WAS by the PAA treatment. The acids which are produced in-situ during anaerobic digestion can be reused which can lower the investment cost on purchasing acid for treatment. Free nitrous acid (FNA) can be produced during AD by nitration of the liquor which has proved to cause cell lysis and EPS destruction [39]. A study by Wei et al. on full scale continuous WAS pretreatment by FNA for 24 h at FNA concentrations of 1.8 mg NN/l resulted in 17% of VS reduction and dewaterability increment from 12% to 14% compared to the untreated WAS. The VS reduction was supported by the increase in methane production by 16% [39].

Alkaline treatment is a well-studied chemical pretreatment method that uses alkali such as NaOH, $Ca(OH)_2$, and KOH, which aids in the breakdown of the complex pollutants and accelerates hydrolysis reactions [13,38,40,41]. Solvation and saponification are the mechanisms that occur within sludge matrices via the introduction of alkalis which increases the surface area of solids making the substrate more accessible to the anaerobes [14]. The saponification of acids and esters and the neutralization of VFAs formed by the degradation of particulates during sludge transport/storage results in the increase of sCOD. A high dosage of alkali is necessary since the biomass itself consumes some amount (scavenging reactions). The high pH also aids in the lysis of the microbial cells within the sludges and increase solubility of proteins [42]. The performance of the digestion is determined by the strength of the alkali with NaOH being the strongest base and the most effective alkali for pretreatment for subsequent AD treatment from a sludge reduction perspective. However, high concentrations of alkali can overly increase the pH leading to the point of inhibition of the microorganisms present in the sludges. Tulun et al. investigated chemical pretreatment at the pH levels of 2, 5, and 10 using a wide range of temperatures and contact times: 25, 40, 50, and 60 °C temperatures and 5, 10, 15, and 45 min of mixing time, respectively [40]. Pretreatment using NaOH at pH 10 for 15 min and a temperature of 40 °C resulted in an increase in sCOD by almost seven-times compared to untreated WAS with an sCOD of 2000 mg/L. This increase in solubility suggests that more organic material within the sludge is made available for the AD microorganisms. This observation is supported by the reported increase in methane composition within the produced biogas which was enhanced by 44% compared to the AD control. Temperatures between 25 and 45 °C at pH of 10 was determined to be the optimal temperature based on maximum biogas production from the alkaline pretreatment similar to previous studies done by Navia and Kim [40]. The operating time and temperature showed better solubility and biogas production as they were increased; however, in the presence of NaOH, the pH level played an important role. At the lower pH level of 2, the particles congregated and decreased dissolution which was the reason for the reported lower biogas production and lower sCOD/tCOD ratio, even when the temperature was increased.

Li et al. studied the extent of organic matter degradation and biogas increase at different concentrations of NaOH on a mixture of sludge consisting mainly of primary sludge [43]. The optimal concentration of NaOH was 0.1 mol/L yielding a pH of 8. The organic degradation increased to 38% and biogas yield to 0.65 L/g VSS compared to the control which had 30% organic degradation and 0.64 L/g VSS biogas yield. In another study by the same research group, the impacts of NaOH and $Ca(OH)_2$ on the sludge disintegration of WAS at 0–40 °C was examined. A NaOH dose of 0.05 mol/L achieved 60% to 71% solubilization of organic matter during the first 30 min of the treatment [44]. At $Ca(OH)_2$ doses of 0.02 mol/L, 0.05 mol/L, 0.3 mol/L, and 0.5 mol/L, the sCOD increased from 275 mg/L to 1375 mg/L, 1365 mg/L, 984 mg/L, and 821 mg/L respectively. This proves that $Ca(OH)_2$ treatment decreases sludge dewaterability at doses over 0.02 mol/L (which was overall lower compared to NaOH). Free ammonia (FA) has been used as a pretreatment for secondary sludge digestion which obtained increased methane production according to a study by Wei et al. [45]. They concluded that FA pretreatment at 250 to 680 mg N/L increased methane production by increasing the hydrolysis rate. The solubilization at 680 mg N/L for one day was 10 times more than untreated sludge (0.03 mg sCOD /mg VS). The FA can be obtained from the AD liquor without any additional processing unlike FNA extraction.

2.3.2. Ozone Oxidation

Ozonation is an oxidation pretreatment method that utilizes ozone which is a strong gaseous oxidant and widely applied within drinking water treatment plants to disinfect water. This process has also been used in wastewater treatment processes to oxidize the organic pollutants. The application of ozone disrupts the cell membranes, allowing the increased exposure of released inner cellular material to the AD microorganisms [12]. The reaction mechanism of ozone for organic decomposition is a two-step process: solubilization via the disintegration of suspended solids and the partial degradation of the parent substrate likely to more biodegradable by-products due to the oxidation of the soluble organic matter [12]. Ozone is unstable and thus generated using an ozone generator on-site prior to use which requires energy. Microbubble generation can be applied that provides better mass transfer due to a increased surface area to volume ratio (i.e., bubble to water contact area). Additionally, venture in-line injectors can also be used which simplifies the process footprint and eliminates the need for contact chambers.

Geol et al. applied 0.015 g O_3/g TS on waste activated sludge which solubilized 19% and 37% of solids, respectively, and increased the TVS up to 30%. Only partial mineralization occurred which was indicated by 3% decrease in TVS concentrations compared to the original TVS concentration of the feed sludge. When the digestion was continued after another phase of oxidation via the addition of 0.05 g O_3/g TS, TVS destruction increased to 59% using a 28-day SRT during AD. The overall TVS destruction efficiency improved by 85% to 90% compared to AD alone [46]. Significant energy is required to generate ozone (12.5 kWh/kg O_3) which could make this pretreatment process costly. This pretreatment method has been applied at the full scale in WWTPs plant with significant biogas production and solids reduction observed [25]. Commercially available ozone pretreatment systems for solids reduction includes Aspal Sludge Inc. who reports improved dewaterability and low energy consumption. Also, Praxair Lyso Inc. reports 80% sludge reduction and improved settling and dewatering characteristics of the treated sludge [25].

2.3.3. Fenton's Reagent Pretreatment

The Fenton's Reagent process is an oxidation process that uses hydrogen peroxide (H_2O_2) in the presence of iron ions (Fe^{2+}) to produce hydroxyl radicals which are a very strong oxidizers that can disintegrate recalcitrant organic pollutants to more soluble species. The addition of iron (Fe, via scrap iron) also enhances the biodegradation of complex substrates and diversifies acetoclastic bacteria useful in acetogenesis reaction with AD systems. Both H_2O_2 and iron ionic solutions are unstable chemicals that can easily lose activity which can lead to wastage of reagents which is a drawback of this method. Also, waste sludge contains non-targeted chemical species (peroxidases and catalases) that can serve as parent oxidizers and radical scavengers which can increase costs and delay treatment time. The extent of sludge disintegration depends on the applied concentrations of H_2O_2 and Fe^{2+} (both in amount and respective dose-ratio), treatment time, and system pH. Due to iron catalyst deactivation and radical (OH) scavenging, the organic reactants present in the sludges cannot be treated effectively at both low and high pH; therefore, studies report that the optimal pH range to be 2–4 for best results [47]. Fenton's reagent oxidation using 50 g H_2O_2/kg DS and 0.07 g Fe^{2+}/g H_2O_2 at pH 3 showed a 75% increase in biogas production compared to the AD system using the untreated input sludge. A five times increase in sCOD with oxidation over the untreated control was reported by Dewil et al. [48]. Increasing the H_2O_2 dosage slightly increased biogas production which suggests that Fenton's reagent treatment breaks the sludge into smaller molecules that are easier to degrade. Additionally, the oxidation step appears to degrade the refractory COD into a more biodegradable by-products making up the COD. Negative aspects of the process include the low pH values required to minimize precipitation of Fe^{2+} (pH < 4), when using higher pH values, the production of iron (precipitation) sludge product [47], hydroxyl radical scavenging by the parent oxidizer (H_2O_2) or radical initiator (Fe^{2+}), higher chemicals costs (due to the needed quantities), and the potential need for waste iron sludge storage and transportation [25]. To summarize, Fenton's reagent pretreatment methods require a narrow pH range for an effective outcome, results in relatively higher chemical costs, and has the potential for producing an iron sludge which needs to be dewatered and disposed [47].

2.4. Biological Pretreatment

Some biological pretreatment methods use an anaerobic pretreatment step that focuses on hydrolysis reactions prior to input into regular AD treatment while other biological pretreatment methods involve enzyme amending that can aid in the reduction of ending sludge volumes. The anaerobic biological pretreatment process focuses on the separation and enhancement of the hydrolysis step from the remaining stages for more efficient and faster digestion [35]. Enzyme pretreatment, temperature phased anaerobic digestion, aerobic pre-digestion, auto-hydrolytic process, and dual digestion are some of the biological treatments that have been applied in pilot scale to achieve solids reduction [12]. The advantages of biological pretreatment are the reduction of inhibitory substances formed due to milder AD operating conditions compared to treatment with chemicals and lower capital costs since no additional chemicals or processing equipment are required [3]. The use of biological pretreatment in practice is limited due to low solubilization yield and difficulty in modeling thus causes challenges with predicting outcomes [16]. Longer treatment times are required with biological pretreatment compared to other pretreatment methods due to much slower kinetics than physical/chemical methods. This is likely the reason why this pretreatment method is sparingly studied [49].

2.4.1. Enzyme Addition

The qddition of enzymes such as cellulase, peptidase, carbohydrase, proteases, and lipase to AD influents have been studied to break down the organic components of the sludges thus making more soluble matter available for digestion along with increasing the rate of solubilization [50]. The dose of enzyme, pH, temperature, and exposure time are the factors that determine the efficiency of this pretreatment method, which has only been studied to date at the laboratory scale and has not been applied at full scale. Few studies have shown that a combination of enzymes tend to improve performance over single enzyme dosing. The primary functioning mechanisms are that the enzymes appear to break-down recalcitrant compounds and cell walls within the sludge making soluble particles readily available for AD. A study by Yin et al. on the effects of fungal mash pretreatment on waste activated sludge co-digested with food waste yielded a 19.9% reduction in the VS present in the sludge. The biogas increased from 0.24 L/g VS for the control to 0.37 L/g VS for the pretreated [51]. The fungal mash was produced in situ with the waste including the cake waste and was inoculated with *Aspergillus awamori*. This enzymatic solution effectively hydrolyzed the sludge mixture without later separation of the enzymes [52]. The fungal mash pretreatment of the mixed sludge projected up to 54.3% total sludge VS concentration reduction. The use of cellulase and Pronase-E on the degradation of recalcitrant sludge components for solids reduction was studied by Roman et al. The study used both the enzymes independently and in combination resulted in enhanced TSS and COD removal in an AD by accelerating the hydrolysis step. Total solids reduction was 80% when the enzymes were used, while the TSS reduction was 36% and 29% for Pronase-E and cellulase, respectively [53]. When applied in an upstream anaerobic sludge reactor, the available carbohydrate was consumed by endogenous microorganism which did not contribute to biogas production, but significantly aided the resulting solids reduction. Similarly, pretreatment using amylase and proteases on WAS at 37 °C and 28 h increased biogas by 23% [51]. Enzyme mixtures have been proposed for sludge reduction with some success noted, but the actual cost and the optimal dosages need to be investigated much more prior to full-scale implementation/commercialization.

2.4.2. Temperature Phased Anaerobic Digestion (TPAD)

Typically, AD is implemented in a single, semi-mixed vessel. All biological phases of digestion occur within this single-tanked system without any physical separation. With TPAD, the methanogenesis step is separated from the previous three primary reaction steps (hydrolysis, acidogenesis, and acetogenesis) to increase the efficiency of the digestion and enhance sludge reduction [15]. Most applications involve using a thermophilic digester which operates at temperatures higher than 50 °C which is placed in series prior to a mesophilic AD reactor that operates at 37 °C. The higher temperature of thermophilic digester promotes hydrolysis of the feedstock and the integration of the two different temperature ranges increases the syntropy of the acetogens and methanogens [25]. This configuration is reported to utilize low quality thermal energy, increase biogas production, increase solids reduction, and deactivate more pathogens as compared to a typical, single stage digester. TPAD systems are different from two phased anaerobic digestion systems. Thermophilic digestion maintains the methanogenic activity in TPAD while the methanogenic activity is suppressed in the first stage of a two phase AD system [54]. Song et al. compared TPAD applied toward WAS digestion with single stage mesophilic and thermophilic digesters operated using various HRTs. The input sCOD was higher in the TPAD digester compared to the single staged digesters as well as the achieved solids reduction, which was almost 12 to 15% higher compared to single stage digestion [55]. Healy el al. conducted temperature phased AD of primary sludge (PS) with the organic fraction of municipal solid waste (OFMSW) at various ratios. They obtained a 48% reduction of VS on 100% primary sludge and a 71% reduction of VS on 20% primary sludge [54]. The first phase was a completely mixed thermophilic digestion operated at 55 °C and was followed by a completely mixed mesophilic digestion operated at 35 °C. It was concluded that the hydrolysis step was accelerated due to the high temperature which resulted in an increase in solids reduction. A specific hydrolysis rate of 0.3 g particulate COD/g VS was achieved in this system with a maximum methane production of 0.4 L/g VS fed obtained. Mehari et al. compared TPAD treated sludge with thermally treated sludge. The first stage of the TPAD was thermophilic digestion 55 °C followed by 42 °C which obtained a 23% reduction in VSS. A thermophilic single digestion at 165 °C resulted in a 22% reduction in VSS [56]. The sCOD increased by 287% for the TPAD while the sCOD for the thermophilic digester increased by 378% compared to AD of untreated sludge. The methane yield of TPAD system and single thermophilic digester was 23% and 20% higher than AD fed the untreated sludge. This study presents that the methane production from a TPAD system operating at temperature range between 42 to 55 °C for 3 days is comparable to a thermal pretreatment at 165 °C for 30 min.

2.5. Integrated Pretreatment

The integration of pretreatment processes has been popular due to the potential combined efficiencies of treatments that can complement the limitation of a single pretreatment method. Several studies have been conducted that use integrated pretreatment methods to improve solubility of sludge components and reduce biosolids production from AD systems [20,41,57–59]. These integrated systems often require less energy, lower capital costs, and easy operation. The most common integrated pretreatment technology is physical-thermal; however, other integrated pretreatment technologies tested include thermo-chemical, mechanical-chemical, and bio-chemical. The integrated pretreatment methods can be a combination of two or more single-mechanism pretreatment processes. Key to integrated processes are decisional factors such as energy requirements, capital investment, O&M costs, and the resulting complexity of the integrated process to ensure maximum benefit from the treatment combinations, and most importantly, the achieved extent of solid removal and potential increase in biogas production from the AD.

2.5.1. Mechanical-Thermal Pretreatment

This combined pretreatment system uses thermal (temperature) and mechanical (shear forces) mechanisms that are optimally integrated to break down complex molecules within the input sludges into smaller, easier to biodegrade by-products (both solid and liquid forms). With most example systems studied, the sludge was pressurized using mechanical methods, such as HPH which creates shear forces, and then heated to soften the sludge components and break-down the cell walls which ultimately solubilizes the sludge components prior to AD processing. Nagle et al. studied the performance of mechanical thermal pretreatment methods via shear force applications on the test sludge by both pressurizing and applying thermal heat using steam. They reported that pretreatment of a sludge having a solids content of 1 to 2% (*w*/*w*) at a temperature of 90 °C for 10 min increased the observed COD solubilization up to 90% [16]. The study concluded sludge disintegration was dependent on the input percentage solids of the sludge, treatment time, and the reaction temperature [16]. Similarly, another study by Wett et al. pretreated WAS at 19–21 bars of pressure and 160 to 180 °C temperature for one hour and reported an increase in biogas production by 75% compared to AD processing of the untreated sludge (control). An improvement in the dewatering characteristics of the input sludge was also obtained which was estimated by the authors to reduce in AD-processed sludge disposal costs by over 25% [60].

2.5.2. Thermal-Chemical Pretreatment

High temperature helps in abiotic hydrolysis of complex organic sludge constituents resulting in the sludge components becoming more soluble. This mechanism is often enhanced via the addition of a chemical(s) which breaks down cell membranes making the intracellular material more available for digestion. This integrated pretreatment can be applied using either low or high temperature additions such as 70 to 90 °C for low temperature treatment and 115 to 170 °C for high temperature treatment. The chemical pretreatment component can be the addition of an alkali, ozone, or other oxidation agent, such as hydrogen peroxide. The integration of thermal methods with chemical addition reduces the consumption of chemicals by up to six times over chemical addition alone, while achieving improved solubilization at the higher temperatures compared to chemical pretreatment alone. The use of thermal methods in combination with an alkali has been extensively studied as an integrated pretreatment system [34,40,61]. Acid has also been used in conjunction with thermal treatment; however, integrated alkaline/thermal pretreatment systems have shown the maximum VSS reductions. Low temperature addition coupled with chemical pretreatment requires a higher amount of chemical reagent to be added but is less energy intensive to achieve an improved sludge solubilization over chemical addition alone.

A study by Nagler et al. on the pretreatment of WAS over a temperature range between 39 and 200 °C and alkaline treatment dosing NaOH over different concentrations reported an optimal treatment condition for WAS pretreatment of 70 °C and 0.04 M NaOH mixed for one hour. This pretreatment strategy reduced sludge production post-AD by increasing the sCOD by 34% when compared to the untreated WAS but only increased the sludge degradation by 2% when compared to thermally treated sludge at 70 °C. This reduction resulted in the lowering of annual biosolids disposal costs by 21% [61]. Yi et al. using WAS achieved 26% reduction for VSS and 200% for sCOD by using alkali dosing of 0.05 g NaOH/g TS that was mixed/reacted for 24 h followed by thermal treatment step using a temperature of 70 °C that was then reacted for either 2 h and 9 h under mixed conditions [34]. A suspended solids (SS) reduction of 21% was achieved for alkaline addition with 2 h of thermal treatment, while a 68% SS removal was achieved for alkaline with 9 h of thermal treatment. The control which was alkalized for 24 h achieved a 2% SS reduction. The increase in SS reduction suggests more availability of substance for methane production during AD [34]. Biogas production, measured at the end of 30 days of digestion, was 45 mL for the control and 329 mL for the alkalized and thermally treated sludge for 9 h. The pretreated AD system had an average methane content of 64%. The increase in biogas production in the alkalized and thermally pretreated sludge suggests that the integrated use of alkali and thermal treatment makes more substrate available for anaerobic digestion via the increased sCOD.

Research by Rivero et al. used a thermal-chemical process via hydrogen peroxide dosing at an applied dose of 2 g H_2O_2/g influent VSS and the heating to a temperature of 90 °C as the thermal pretreatment step followed by AD using a SRT of 30 days [62,63]. The sludge mixture was composed of primary sludge and WAS at a 1:1 mass ratio. The achieved VSS removal for the control, which is untreated, and for the integrated pretreatments prior to AD were 48% and 75%, respectively. The single thermal pretreatment of 90 °C resulted in removing 49% of the sludge VSS with the control showing no removal. The volume of methane production per gram of VSS removed for the control and the integrated pretreated sludge mixtures were 650 and 293 mL, respectively. The lower value obtained using alkaline integrated with thermal treatment compared to the control indicates greater sludge component solubilization using the pretreatments than the control. Another study by the same research team applied thermo-oxidative pretreatment at a lower temperature, reduced alkaline dosage, and reduced SRT. A maximum of 67% VSS removal was achieved using 1.0 g H_2O_2/g sludge VSS at 90 °C which was then digested using a 15-day SRT. Decreasing the dosage to 0.5 g H_2O_2/g sludge VSS resulted in essentially the same performance in terms of VSS removal (64%) which is not significant compared to the VSS removal achieved at twice the dose. The investment and operating cost at a half the dosage and half the SRT might reduce processing costs but also reduced the overall solids destruction achieved by 12%. Therefore, a balance between the cost and performance must be evaluated [63]. Valo et al. studied the use of thermo-oxidative pretreatment on WAS for solubilization and later AD solids reduction [64]. An alkali pretreatment using potassium hydroxide at range of 30 to 65 meq/dm^3 was applied. Temperatures ranging from 135 °C to 170 °C were applied using an autoclave on WAS for one hour. Results from the integrated thermal-chemical pretreatment obtained increased COD solubilization by 63% at 130 °C and pH 12 and 83% at 170 °C and pH 12 compared to control raw sludge.

Table 2. Summary table of the biogas production and Chemical Oxygen Demand (COD) removal from different pretreatment methods.

Study Reference	Pretreatment Method	Biogas Production	COD Removal
Kim et al.	42 kHz ultrasound for 120 min	3657 L/m^3 control vs. 4413 L/m^3 for ultrasound	sCOD control-1136 mg/L, pretreated sCOD 4000 mg/L
Li et al.	20 kHz for 0 to 100 min	67 mg/g VS for 20 days HRT and sonication for 80 min	sCOD of 338 mg/L for control, sCOD of 8133 mg/L for 80 min sonication
Coelho et al.	MW pretreatment using domestic microwave at 100% intensity, 2450 MHz frequency on WAS at 20 days HRT Thermophilic temperature −55 °C Mesophilic temp −35 °C	0.5 L/d thermophilic treatment, 0.35 L/d thermophilic control 0.4 L/d mesophilic, 0.28 L/d mesophilic control	35% tCOD removal for thermophilic treatment with 29% tCOD removal for untreated 55.6% tCOD removal for mesophilic treated vs 55.3% tCOD removal for mesophilic control
Appels et al.	Microwave (336 kJ/kg sludge) 800 W for 3.5 min	Increase in biogas production by 50%	Increase in sCOD from 1353 mg O_2/L untreated to 4247 mg O_2/L MW pretreated, increase by 214%
Serrano et al.	Microwave pretreatment of WAS at 30,000 J/kg, 400 W	111 mL/$CH_{4\ STP}$/gVS of methane yield for control, 118 mL $CH_{4\ STP}$/g VS methane yield for pretreated	Increase in sCOD was 0.113 mg O_2/g TS
Zhang et al.	50 MPa homogenization pressure for 1 cycle, 7 days digestion	64% increase in biogas, 1546 mL biogas production for untreated sludge	45% tCOD removal for 1 cycle 17% tCOD removal for untreated
Zhang et al.	50 Mpa homogenization pressure for 2 cycle	3330 mL biogas produced in treated, 115% increase in biogas compared to untreated	62% tCOD removal for 2 cycles 17% tCOD removal for untreated
Appels et al.	Thermal treatment at 70 °C, 80 °C, 90 °C for 1 h	34 mL/g Organic Dry Solids (ODS) biogas for untreated 35 mL/g ODS for treatment at 70 °C 75 mL/g ODS when treated at 80 °C 377 mL/g ODS when treated at 90 °C	55,300 mg O_2/L COD for untreated mg O_2/L COD when treated at 70 °C mg O_2/L COD when treated at 80 °C mg O_2/L COD when treated at 90 °C
Xue et al.	Thermal pretreatment at varying temperature for up to 24 h	Biogas production increased from 0.96 L/g VS removed to 101, 0.99, 1.04, & 0.94 L/gVS removed at 60 °C, 70 °C, 80 °C, & 90 °C respectively	Solubilization of COD increased from 4.5% (control) to 29%, 30%, 35%, & 51% at 60 °C, 70 °C, 80 °C, & 90 °C respectively
Braguglia et al.	Thermal hydrolysis prior to thermophilic treatment	0.18 Nm^3/m^3 d biogas production for untreated sludge 0.36 Nm^3/kg VS fed	68% sCOD removal at HRT of 15–20 days
Devlin et al.	pH 2 pretreatment using 37% HCl	14% increase in methane yield compared to untreated WAS	30 g/L tCOD of the untreated digestate 29 g/L tCOD of treated digestate 48 g/L tCOD of feed WAS
Sun et al.	Pretreatment with 10–50 mg Peracetic acid (PAA)/g SS and reaction time of 30 min	20% increase in biogas with pretreatment by 30 mg PAA/g SS	Supernatant sCOD increased by 530%
Tulun et al.	Chemical pretreatment at pH 2, pH 5, and pH 10 using H_2SO_4 for acid treatment and NaOH for alkaline pretreatment	43% increase in BMP in pH 10, 60 °C and 15 min treatment time.	pH 2–194% increase in sCOD pH 5–567% increase in sCOD pH 10–708% increase in sCOD for 15 min pretreatment at 60 °C compared to untreated sludge
Li et al.	Alkaline treatment using 0.005–0.5 mol/L NaOH on WAS	Increase in biogas production was insignificant for days greater than 8 at all pH and untreated sludge (<10 mL)	Increase in sCOD from 3000 mg/L for untreated to 6000 mg/L for pretreatment using 0.4 moles/L NaOH of 80% primary sludge and 20% biofilm sludge.
Wei et al.	FA treatment maintaining 10 pH using NaOH at 25 °C	160 L CH_4/kg VS added for the untreated sludge in 50 days digestion time increased to ~185 L CH_4/kg VS added for FA treatment at 250 mg NH_3-N/L	10 times increase in solubilization of sludge compared with control and ammonium treatment 0.025 mg/mg VS sCOD of control 0.4 mg/mg VS increase at pH 10, and FA concentration of 250 mg NH_3-N/L

Table 2. *Cont.*

Study Reference	Pretreatment Method	Biogas Production	COD Removal
Dewil et al.	Fenton reagent pretreatment at 5, 25, 50 g H_2O_2/kg Dry Solids POMS and DMDO pretreatment at 30, 60 g/g DS and 330, 660 mL/kg DS respectively	Increase in specific biogas production from 644 for control to 668 mL/g ΔODS for 50 mg/kg DS H_2O_2, 716 mL/g ΔODS for 60 g/kgDS POMS, and 829 mL/g Δ ODS for 660 mL/kg DS of DMDO	187%, 405%, 595% increase in COD using 5, 25, 50 g H_2O_2/kg dry solids, respectively 385%, 506% increase in COD using 30, 60/g DS POMS respectively and 456%, 690% increase in COD using 330, 660 mL/kg DS DMDO compared to untreated sludge with 421 mg O_2/L COD
Yin et al.	Use of fungal mash as enzyme pretreatment on activated sludge and mixture of activated sludge and food waste	Net methane production increased from 240 to 367 mL/g VS after pretreatment fungal mash on activated sludge	220% more sCOD in pretreated co-digestion than untreated activated sludge
Song et al.	Temperature phased AD at single stages mesophilic temperature (35 °C) and thermophilic temperature (55 °C), and a co-phase AD system of mesophilic and thermophilic digester	Specific methane yield of 450 mL/g VS removed for single stage mesophilic temperature and mesophilic-thermophilic co-phase digestion while 416 mL/g VS removed for single staged thermophilic temperature.	sCOD of 2555 mg/L for single phased mesophilic, 5240 mg/L for single phased thermophilic treatment, 2100–2200 and 1700–2900 mg/L for co-phase mesophilic-thermophilic digesters
Healy et al.	Comparison of two phased and temperature phased AD of MSW and PS Thermophilic temperature of 55 °C followed by mesophilic temperature of 35 °C, pH 5.6 for first system of two phased system while pH 7 of the first stage of temperature phased reactor on mixture of OFMSW and PS	Maximum methane production rate of 0.4 L/gVS for temperature phase system and 0.3 L/g VS for two-phase system	64% particulate COD removal from temperature phased system and 65% particulate COD removal from two phase system on sludge mixture of 60% OFMSW and 40% PS
Mehari et al.	Temperature phased biological hydrolysis at 4255 °C, combination of 42 °C followed by 55 °C, and 55 °C followed by 42 °C and thermal hydrolysis at 165 °C for 30 min	Biological hydrolysis at 55 °C followed by 42 °C 23% higher methane production Untreated sludge and thermal treatment enhanced methane production by 20%.	sCOD increased by a maximum of 377% by thermal hydrolysis followed by biological hydrolysis at 55 °C which increased sCOD by 324%
Wett et al.	Ball milling (55 kW with specific demand of 0.49 kWh/kg TSS and thermal hydrolysis as pretreatment methods on Low loaded WAS. Thermo-pressure Hydrolysis (TDH) at a pressure of 19–21 bar, 170–180 °C for 60 min	TDH produced 75% more biogas and ball milling produced 41% more biogas compared to untreated sludge	COD was enhanced from 33% for untreated to 44% using ball milling and to 51% using TDH treatment.
Nagler et al.	Thermo-chemical pretreatment on WAS using 1M NaOH, aluminate, and ash as alkaline reagents and temperatures ranging from 39–200 °C	122% increase in biogas compared to the untreated sludge when treated at 70 °C and 0.04 M NaOH	COD disintegration of 34% compared to the control was achieved at 70 °C which was increased by 50% when 0.08 M NaOH was used.
Yi et al.	Comparison of alkaline, thermal + alkaline treatment on WAS at 0.05–0.25 g NaOH/g TS for 2 and 9 h	630% increase in biogas at 0.05 g NaOH pretreatment for 24 h and thermal treatment at 70 °C for 9 h compared to control	sCOD increased by 2 times at 0.05 g NaOH, by 17 times at 0.05 g NaOH and 70 °C for 2 h, and by 226 times at 0.05 g NaOH, 70 °C and 9 h.
Valo et al.	Thermal + oxidative pretreatment at 130 °C and pH10 using 1.65 g/dm^3 KOH	74% increase in biogas compared to untreated sludge	tCOD removal increased by 37% compared to raw WAS

2.5.3. Mechanical-Chemical Pretreatment

Mechanical pretreatment on sludge creates shear forces due to the applied pressurization and depressurization which results in the degradation of the cells. The integration of chemical(s) enhances the decomposition process by breaking the cell walls and increasing the solubility of the organic matter within the microbial cells [16]. The use of chemical(s) has shown an additional benefit along with mechanical forces on the hydrolysis step during AD [57,59,65,66]. Ultrasound and ozonation are an example of a mechanical-chemical pretreatment that has the simultaneous advantage of increased solubility of sludge components through the production of radical oxidizer species, enhanced ozone mass transfer, and increased reaction sludge solid surface area via particle disintegration. Xu et al. studied an ultrasound-ozonation pretreatment system applied on WAS and its effect on the particle size of the sludge solids. Their study indicated that reaction temperature, ultrasonic energy density, pH, and O_3 dose directly affected the outcome. An increase in sludge total COD of up to 20% was observed as the treatment time increased from 0 to 60 min and ultrasonic energy density was increased from 0.12 W/mL to 1.50 W/mL at a steady O_3 dosage of 0.6 g/h [20]. The doses of ozone used ranged from 0.05–0.5 g O_3/g TS and the tested specific energy output of the sonication unit ranged from 40 W to 600 W. However, at higher O_3 doses, an increase in treatment time did not increase biogas production suggesting the potential formation of refractory by-product compounds perhaps due to oxidizer overdosing. The use of ultrasound was reported to produce mechanical shear forces on the extra-cellular polymeric substances while the reaction of the O_3 molecules resulted in increased cell disintegration, and thus, higher solids removal post-AD. Chiu et al. pretreated WAS with NaOH and 20 kHz ultrasound, which resulted in sludge hydrolysis of 211 mg/L/minute [16]. They reported that the NaOH breaks the cell walls which releases the intracellular material available for lysis during sonication resulting in a net high solubility of sludge components for later AD treatment. Microwave pretreatment can also be applied with the dosing of a base as an example of the integration of a mechanical/chemical mechanisms as a sludge pretreatment that has shown promising results by increasing the solubilization of sludge components [59,67]. Alkaline pretreatment solubilizes the cells while MW irradiation increases the temperature through the friction caused by the rotation of polar molecules within the microwave electric field. The increase in temperature is faster and requires less energy compared to conventional heating and results in increased sludge component hydrolysis [12]. A study by Dognan and Sanin reported that the proteins available by integrated pretreatment using microwave and alkalis is greater than the proteins available by separate pretreatment which suggests the synergistic effect of the two treatments on increased sludge component solubilization [59]. Dognan and Sanin found that the sCOD to tCOD ratio was increased from 0.005 for the control to 0.37 using a pretreatment run at a pH of 12.5. Their results also showed a VSS reduction of 42% for the control and a 48% VSS reduction for the WAS pretreated with microwaves at a pH of 12 followed by 49 days of AD processing. The biogas produced after digestion (50 days) was also reported with the untreated WAS fed AD producing 65 mL of cumulative methane while the integrated pretreatment fed the same influent that was operated at a pH of 12 produced 80 mL of cumulative methane [12].

High-pressure homogenization (HPH) coupled with base dosing is another integrated treatment that has been shown to improve the solubilization of sludge components through solids degradation and increased bioavailability of the organic materials within sludges. organic material [41,57]. HPH is a simple yet efficient method that does not require any chemical reaction and uses high pressure, forces, and eddies to disrupt cells which when applied together with a base further breaks down the whole cells and their compositional materials. Stephenson et al. reported 80% solubilization of sludge SS along with the reduction in required AD retention time from 18 to 13 days (mesophilic digestion). They used a pressure of 12,000 psi and a pH level of 10 on a sludge that was reacted under this condition for one hour [16]. Fang et al. reported alkaline dosages at a higher dose range up to 0.05 mol/L. These dosing tests were reported to enhance the digestion of sludges via the conversion of the sludge components to lower molecule weight compounds (by-products) [41]. HPH was applied after alkaline pretreatment (0.04 mol/L of NaOH) and using a pressure of 60 MPa. After 14-days of subsequent

AD post-pretreatment, sludge tCOD removal was 43% and a sludge removal of 61% was achieved. The control which was only HPH treated did obtain the same 43% tCOD removal but only 31% sludge emoval. The methane content within biogas produced using the integrated pretreatment was 68% while the methane content in the biogas produced by HPH treatment only system was 58%. Clearly, the benefits of integrated these two mechanisms were highlighted by the enhanced quality of the biogas. Also, another improvement in AD performance observed between the integrated dual-mechanism pretreatment versus HPH pretreated conditions was with biogas production. At the end of digestion, the dual-mechanism systems achieved 50% more biogas volume production: 735 mL biogas/day with the HPH only pretreatment and 1105 mL biogas/day using dual-mechanism pretreatment. Cho et al. studied the combined effect of NaOH dosing integrated with a novel crushing device that was used as a mechanical secondary pretreatment mechanism. This dual-mechanism system was applied toward a thickened WAS via the following conditions: 40 g TS/L sludge, pH of 13, and 90 min of reaction time which achieved 64% of improvement of the amount of solids solubilized [65].The crushing device, which is suggested to be effective and economically feasible, consisted of four cutting blades rotating at 2500 rpm and tangential velocity of 5430 m/second. Using a reaction time of 90 min and a pH of 13 for pretreatment of WAS, the methane yield within the subsequent AD step was increased eightfold (233 mL/g sludge TS) compared to the AD control without any pretreatment at 50 g TS/L (25 mL/g sludge TS).

3. Comparison of the Pretreatment Methods for Solids Reductions

Pretreatment methods that have been reviewed in this paper were compared based on the extent of resulting degradation of the sludge components via pretreatment, the solubilization of complex sludge compositional molecules via the pretreatment step, the reduction of solids after subsequent AD treatment, and the quality of the biogas generated from the digester. Table 3 summarizes the assessment of the various candidate pretreatments by comparing the advantages and disadvantages of the various pretreatment methods and their status in terms of use in WWTPs [15,16,35,50]. Some pretreatment methods were found in the literature to have shown positive effects on improving AD performance; however, it was clear that recalcitrant by-product formation needs to be avoided which appears to occur at very high temperature during thermal treatment and/or using high base concentrations. These recalcitrant compounds inhibit the digestion process resulting in higher overall sludge processing costs thus placing an increased financial burden on the WWTPs. There are a few pretreatment methods were found to have been scaled up and applied at full-scale WWTPs (see Table 3). Among the single mechanism pretreatment methods applied, ultrasound and ozonation have been proven by research to show solid results with regard to proven benefits toward improving subsequent AD performance. These pretreatment technologies have the largest sector of commercial vendors, among the various systems assessed, that have a history of use at WWTPs. Thermal chemical pretreatment when used as one of the steps within an integrated pretreatment system was shown to be the most effective in terms of terms of post-AD reduction of solids. However, in general, it can be stated that numerous pretreatment processes indicated high potential, albeit most needing more development, to serve as effective pretreatment processes for AD systems–both in terms of sludge volume reduction and biogas generation. As biorefineries mature, these pretreatment options may be excellent tools for conditioning AD sludge outputs with targeting compositions that are more conducive toward biorefinery conversion into multiple beneficial products.

Table 3. Advantages and disadvantages of the pretreatment methods assessed via this review and their evaluated relative state of development with regard toward applicability at actual wastewater treatment plants (WWTPs).

Pretreatment	Advantages	Disadvantages	Applicability in WWTP
MECHANICAL			
Ultrasonication	High degree of solubilization Biogas production High solids removal	High energy requirement High investment cost Degradation of electrodes	Commercialized, Vendors: Biosonator Sonix Heizcher
Microwave	Improved sludge disintegration Low reaction time	High investment And operation cost	Lab scale application only
High pressure homogenization	Short reaction time Increased sludge disintegration	High investment and operation cost Degradation of equipment	Commercialized, Vendors: Crown Process Cellruptor
THERMAL			
Low temperature	Solids reduction achieved with low operation	Long treatment time	Lab and pilot scale
High temperature	Short contact time Higher solids reduction achieved compared to low temperature treatment	High operation cost High energy requirement Recalcitrant compounds formation	Commercialized, Vendors: CambiTHP Biothelys Exelys
CHEMICAL			
Acid/alkaline	Excellent sludge disintegration	Additional cost for chemicals Digester instability due to pH	Lab scale
Ozone	Improve sludge hydrolysis High solids reduction	High investment and operation cost	Commercialized, Vendors: Aspal Sludge Praxair Lyso Biolysis O-Process
Fenton's reagent	Low investment and operating cost compared to ozonation Easy to use Sludge disintegration	Chemical cost, Low pH requirement, Hydroxyl scavenging	Lab scale
Trace metal nutrients	Some sludge disintegration VS solids reduction Increase in methanogenesis	Cost for chemicals Not widely studied	Lab scale

Table 3. *Cont.*

Pretreatment	Advantages	Disadvantages	Applicability in WWTP
BIOLOGICAL			
Enzyme addition	Lysis of recalcitrant compounds TS, COD reduction	Specificity on enzyme High cost on enzyme purchase	Lab scale
Temperature phased AD	Low operating cost	Long reaction time Limited research for analysis	Lab scale
Integrated Pretreatment			
Mechanical-thermal	Low energy required compared to single pretreatment High solids reduction	High energy demand	Full scale
Mechanical chemical	Higher solids reduction achieved medium cost for chemicals and lower energy requirement compared to individual pretreatment	Additional chemicals required	Commercialized, Vendor: Microsludge process
Thermal chemical	Increased solids removal Low thermal energy required compared to thermal only	Cost for chemicals High energy demand Recalcitrant compound formation	Commercialized, Vendors: Krepro Process

4. Cost and Energy Demand

The goal of achieving maximum solids reduction and biogas production from AD by applying pretreatment methods that have been reviewed in this paper with the previous section summarizing technical performance. For these various pretreatment processes to be economically viable, their application must provide enough economic benefit to offset installation costs, operation and maintenance costs, and the energy requirements. Technical performance provides the entrance toward considering the use of a pretreatment system, but it is the additional cost of installing and operating the system that must provide economic benefits if their maturation to full commercialization is to be realized. Table 4 provides a brief comparison of capital cost, reduction in disposal cost, and overall saving using various pretreatment methods. The operation and maintenance cost had a higher impact on the net savings (which is dependent on the characteristics of the WAS). Therefore, it is challenging to present the exact savings when scaling up from the laboratory scale to actual commercial application. These relative assessments reflect the opinion of the author team toward the relative reported costs of each technology. However, a technically high performer sonication, ozonation, and the direct thermal systems will be expensive to implement—again, costs needed versus savings with the installation must be assessed via a solid techno-economic analysis (TEA). Phased heating, microwave, enzymatic, and mechanical-chemical all showed the best economic performance. However, it must be realized that these economics are often based on laboratory performances with minimal full-scale performance data to support these cost assessments. Clearly, more R&D is needed along with some level of demonstration histories at real WWTPs. Caution is given in that it is best to not be the first field application of a developing/new process unless a partnership between the vendor and user are developed along with shared costs during the demonstration between the WWTP, vendor, and potentially, government entities.

Table 4. Assessment of the cost of applying the various pretreatment methods (the vertical arrows represent our assessment of relative cost, ↑ means increase, ↓ means decrease).

Treatment	Method	Capital Cost	Electricity Generation	Disposal Cost	Net Saving	Reference
Thermochemical	70 °C and 0.04 M NaOH	22% ↑	22% ↑	27%↓	21% ↑	[61]
Alkaline-mechanical	pH 13, 90 min reaction time + crushing device for	$556,000	5 times↑	40%↓	40% ↑	[65]
Ultrasound †	1000 kJ/kg TSS	$20 ↑	N/A	55$ ↓	$54 ↑	[68]
Thermal †	70 °C	$43 ↑	N/A	$55 ↓	$67 ↑	[68]
Ultrasound-thermal †	1000 kJ/kg TSS + 70 °C	$63 ↑	N/A	$63 ↓	$51 ↑	[68]
Thermochemical ozone $^{\delta}$	0.0004 mg O_3/mg SS to 0.0016 mg O_3/mg SS, 50–100 °C, 0.1 N NaOH	$7 ↑	N/A	$139 ↓	$36 ↑	[69]

†: cost calculated in $/ton dry solid; $^{\delta}$: cost calculated in $/ton of biosolid.

5. Summary

The increasing human populations is slowly overwhelming existing WWTP capacities with the resulting increasing generation of wastewater volumes requiring treatment. Over 7,000,000 dtons of biosolids are generated each year within the US alone. Biosolids handling is a major concern for the WWTP operators which can be alleviated by more efficient removal of total solids within the AD processes of a WWTP. Reduction of solids not only lessens the O&M costs for the WWTP but also advocates for a sustainability and potentially coupling with developing biorefinery concepts. Moreover, in almost all cases, when the technologies assessed achieved higher post-AD solids reduction, they also yielded more and improved biogas production. Numerous studies have been conducted and more studies are being initiated to help facilitate improved wastewater sludge degradation (defined as solids removal) using anaerobic digesters. Pretreatment methods which have been presented in this paper showed a proven increase in the solubility of the sludge components thus increasing the biodegradation of complex matter within AD systems resulting in an increase in biogas production which can be utilized as energy. Reduction in biosolids production not only lessens the burden of disposal (with disposal options and volumes decreasing within the US) but lowers the carbon footprint when solids converted to methane used as an energy resource (particularly in the production biogas with higher BTU values). Although the addition of pretreatment processes for improving AD performance does require additional investment and operating costs, the benefits from an optimized digestion can potentially outweigh the operating and handling costs of biosolids at present. Additionally, increased biogas production after pretreatment can be also be valorized as an energy/biorefinery feedstock resource, which lowers the overall operating cost. Finally, none of the pretreatment technologies assessed were free of negative aspects. Their potential use must be supported by solid TEA analyses before seriously being considered for application at a WWTP. Note that regional biosolids disposal costs, along with projected long-term disposal costs, must be used within the TEAs because of the variability of these costs from region to region.

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Abbreviations

WWTP	Wastewater treatment plant
WW	Wastewater
AD	Anaerobic Digestion
TS	Total Solid
TSS	Total Suspended Solid
VS	Volatile Solid
SS	Suspended Solid
VSS	Volatile Suspended solids
COD	Chemical Oxygen demand
tCOD	Total Chemical Oxygen Demand
sCOD	Soluble chemical oxygen demand
WAS	Waste Activated Sludge
PS	Primary Sludge
OFMSW	Organic Fraction of Municipal Solid Waste
VFA	Volatile Fatty Acids
MW	Microwave
HPH	High Pressure Homogenization
OLR	Organic Loading Rate
HRT	Hydraulic Retention Time
SRT	Solids Retention Time
TPAD	Temperature Phased Anaerobic Digestion
2PAD	Two phased Anaerobic Digestion
TEA	Techno-Economic Analysis

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