Review on pretreatment techniques to improve anaerobic digestion of sewage sludge

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ABSTRACT

Anaerobic digestion (AD) of sewage sludge is one of the most efficient, effective, and environmentally sus-tainable remediation techniques; however, the presence of complex floc structures, hard cell walls, and large amounts of molecular organic matter in the sludge hinder AD hydrolysis. Consequently, sewage sludge pre-treatment is a prerequisite to accelerate hydrolysis and improve AD efficiency. This review focuses on pre-treatment techniques for improving sewage sludge AD, which include mechanical, chemical, thermal, and biological processes. The various pretreatment process effects are discussed in terms of advantages and dis-advantages, including their effectiveness, and recent achievements are reviewed for improved biogas production.

1. Introduction

Anthropological activities generate vast amounts of sewage, which seriously endanger human health and the environment. It is estimated that approximately 85 km^3 of sewage per year is generated in North America, and in the Middle East and North Africa, approximately 22.3 km^3 of sewage per year is generated from various sources. In Asia, China alone produces a huge volume of sewage (58.9 km^3 per year) [1]. These large amounts of sewage produced have created challenges for effective management and efficient treatment [2,3]. Most sewage treatment methods are based on physical and biological processes. During sewage treatment, large amounts of semisolid residue, known as sludge, are generated, and sludge deposition in the environment is an alarming threat causing soil and water pollution [4]. Sewage sludge (SS) management and treatment is expensive and accounts for nearly one-half of the total sewage treatment cost [5]; therefore, progress in cost-effective SS treatment techniques represents an important research area for waste management companies.

Basic SS disposal practices include agricultural use, landfills, composting, anaerobic digestion (AD), recycling as a construction material, and incineration [2,6,7]. All these management practices have specific practical constraints related to human and environmental health [8]. Among the treatment methods, AD is considered an effective, economical, and eco-friendly technology. AD stabilizes sludge, aids in odor and pathogen removal, and noticeably generates methane gas. Methane gas can be used as a biofuel, which has commercial value as a renewable bioenergy source [9], and due to this beneficial feature, AD is a promising method that offsets a portion of the wastewater treatment capital costs.

However, the complex organic matter (OM) composition of SS creates obstacles for efficient AD. Several studies have identified that the presence of a complex floc structure (extracellular polymeric substances, EPSs), recalcitrant cell walls, and other high molecular weight OM in sludge hinders AD hydrolysis [6,10–12]. This hydrolysis problem leads to a longer retention time, requires a larger bioreactor, and results in a smaller biogas yield. To improve hydrolysis and accelerate methane production, several studies have suggested sludge pretreatment prior to AD (Fig. 1) [13–15].

Pretreatment processes, such as chemical, thermal, biological, or a combination of these techniques, disintegrate the complex sludge structure [5,16–18]. In a pilot-scale laboratory experiment, these pretreatment strategies have been proven to reduce solid mass, rupture complex EPSs, and increase methane production during AD. Pretreatment is required to manage substrates in order to maximize their use in AD and also improves substrate biodegradability, increases the soluble substrate amount, decreases SS viscosity, increases accessibility for microbial degradation, and lowers the overall sludge management cost [16,19]. Because of these beneficial aspects, SS pretreatment is considered to be essential for efficient AD and has consequently attracted global attention. This study reviews several pretreatment methods, which are prerequisites for SS management, and the main objective is to provide deeper insights into currently implemented pretreatment technologies for SS digestion.

Table 1
Properties of sewage sludge. Adapted from [23].

Parameters	Values
Total solids	2–12% (liquid SS) 12–40% (dewatered SS)
Volatile solids	75–85% dry weight (d.w.) basis
Pathogens:	
Virus	2.5×10^3–7×10^4 (no./100 mL)
Coliform	1×10^9 (No./100 mL)
Salmonella	8×10^3 (No./100 mL)
Helminth	2.5×10^2–1×10^3 (No./100 mL)
Nutrients:	
Total nitrogen	3.9% (d.w.)
Phosphorus	2.5% (d.w.)
Potassium	0.40% (d.w.)
Sodium	0.57% (d.w.)
Calcium	4.9% (d.w.)
Iron	1.3% (d.w.)
Metals:	
Arsenic	9.9 (mg/kg, d.w.)
Cadmium	6.94 (mg/kg, d.w.)
Chromium	119 (mg/kg, d.w.)
Copper	741 (mg/kg, d.w.)
Lead	134.4 (mg/kg, d.w.)
Mercury	5.2 (mg/kg, d.w.)
Molybdenum	9.2 (mg/kg, d.w.)
Nickel	42.7 (mg/kg, d.w.)
Selenium	5.2 (mg/kg, d.w.)
Zinc	1202 (mg/kg, d.w.)

2. Sewage sludge

Biological wastewater treatment releases effluent and biosolids, the latter of which are composed of complex biological and organic chemical structures. The generated SS usually represents approximately 2% of the treated sewage volume [6]. According to the United Nations-Habitat data, the sludge production in the USA accounts for more than 6.5 million tons of dry solids per year [20]. In China, sludge production is approximately 3.0 million tons per year [21]. In Japan and Germany, approximately 2.0 million tons of sludge are produced annually [6]. These representative data clearly indicate that a large SS volume is produced globally, which imposes a global challenge for its management.

SS is characterized by the presence of solid and organic compounds, pathogens, microbial aggregates, filamentous bacteria, EPSs, nutrients, and heavy metals [22]. The basic properties of unstable SS are listed in Table 1. Sewage pH varies depending on the wastewater source and may be acidic (< 6.5) or alkaline (greater than11). Various types of OM that are important for improving the physical properties of soil are present in sludge. Several recalcitrant solids that are difficult to break down and manage are present. The various types of toxic substances, microorganisms, and OM produce unpleasant odors, cause environmental pollution, and endanger human health. After treatment, SS can

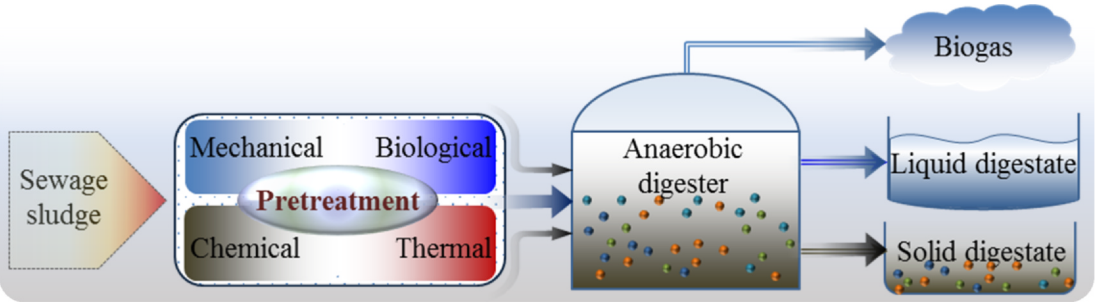

Fig. 1. Pretreatment implementation for improving sewage sludge anaerobic digestion.

be utilized for agricultural purposes because it contains various beneficial nutrients [6,23,24]. SS may possess hazardous organic chemicals such as those existing in pesticides, polychlorinated naphthalene, polycyclic-aromatic hydrocarbons, benzene, toluene, trichloroethylene, nitrobenzene, and heavy metals [25,26]. Because SS contains various nutrients, organic fractions, and other components, it is regarded as both beneficial and harmful; therefore, SS treatment before deposition in the environment is a crucial factor for proper wastewater and sludge management.

3. Anaerobic digestion of sewage sludge

3.1. Anaerobic digestion

AD is a chemo-biological process by which complex organic wastes are transformed into simple soluble compounds in an anaerobic environment [27–29]. The diverse microbial consortium during AD degrades organic waste material resulting in the production of biogas and other energy-rich organic compounds [30,31]. The main goal of SS AD is to stabilize OM and reduce pathogens [32], which is accomplished by the biological conversion of waste organic materials to methane and carbon dioxide under anaerobic conditions. The conversion of wastewater organics to methane involves several bacterial groups performing specific enzymatic reactions [33–35]. The microorganisms responsible for hydrolysis and acid fermentation include both facultative and obligate anaerobic bacteria [36]. Some genera found in anaerobic digesters include *Clostridium, Corynebacterium, Actinomyces, Staphylococcus*, and *Escherichia*. The microorganisms responsible for methane conversion include *Methanosarcina, Methanothrix, Methanococcus, Methanobacterium*, and *Methanobacillus*. *Methanosarcina* and *Methanothrix* utilize acetate to produce methane and carbon dioxide, while *Methanococcus, Methanobacterium*, and *Methanobacillus* oxidize hydrogen with carbon dioxide as an electron acceptor [34,37–39].

AD is an intricate process and consists of four stages: hydrolysis, acidogenesis, acetogenesis, and methanogenesis (Fig. 2). The hydrolysis stage reduces both insoluble OM and high molecular weight compounds such as polysaccharides, proteins, and lipids into amino and fatty acids; however, hydrolysis is generally considered a rate-limiting step [40]. In the second stage, components formed during hydrolysis are split during acidogenesis, where the acidogenic bacteria produce volatile fatty acids (VFAs), carbon dioxide, hydrogen sulfide, ammonia, and other by-products [41]. Acetogenesis is the third step in AD, where the higher organic acids and alcohols are digested to produce acetic acid, carbon dioxide, and hydrogen. This conversion is to some extent controlled by the partial pressure of hydrogen in the mixture. The final step in AD is methanogenesis. When methanogenesis reaches its end phase, methane gas is produced by two methanogenic bacterial groups: the first breaks down acetate into methane and carbon dioxide; and the second uses hydrogen (electron donor) and carbon dioxide (acceptor) to produce methane [40].

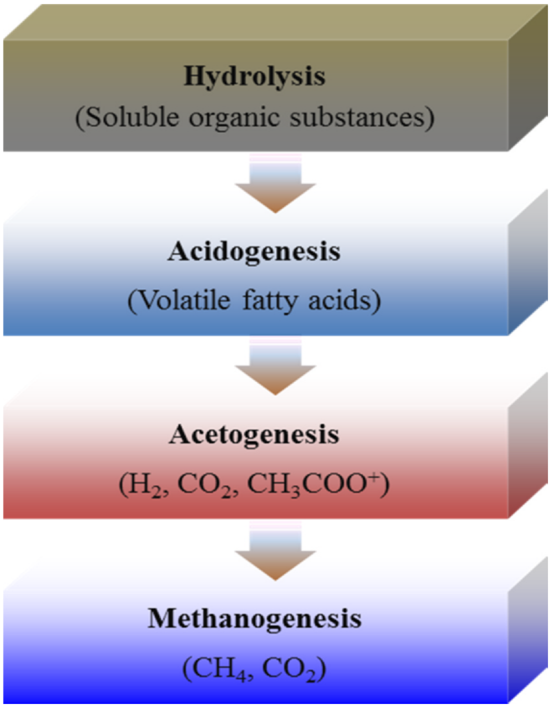

Fig. 2. Four basic stages of anaerobic digestion.

There are several AD types depending on the waste type. Based on the substrate source, the AD technique can be classified as solid-state or liquid-state AD. Solid-state AD typically occurs when the feedstock contains more than 15% solids and liquid-state AD occurs when the solid concentrations in the feedstock are between 0.5% and 15%. Liquid-state AD is used to treat SS, animal manure, and food waste. Organic elements of solid waste and lignocellulose such as crop residue can be treated through solid-state AD [42]. The AD of these organic wastes is an environmentally useful technology [43]; however, this process possess disadvantages including long retention times and low organic compound removal efficiencies [44]. In addition, various parameters such as pH, alkalinity, temperature, retention time, nitrogen, and carbon/nitrogen (C/N) ratio affect the stabilization rate of organic wastes during AD [45].

To improve AD efficiency, studies have implemented a co-digestion strategy. Co-digestion, also referred to as co-fermentation, is a waste treatment method in which different waste materials are mixed and treated together [46,47]. Co-digestion is mainly utilized to improve AD of solid organic waste because of its numerous benefits [48]. It can stabilize the feed in a bioreactor by improving the C/N ratio and decreasing the nitrogen concentration [49]. The use of a co-substrate having a low nitrogen and lipid content increases the biogas production amount. This reduces constraints associated with volatile organic compound accumulation and high ammonia concentrations during AD [46,50]. During co-digestion of plant material and animal manure, the plant material provides a large amount of carbon, resulting in a better C/N ratio balance and the animal manure provides buffering capacity and various nutrients [51,52]. Co-digestion with substrates that have a buffering capacity can be a good alternative for effective treatment of highly biodegradable materials. In addition, two-stage biogas production from SS AD could be a useful strategy to overcome the productivity limitation [53].

3.2. Biogas

Pretreatment processes increase biodegradability, dewaterability, and biogas production [54]. Biogas production during organic waste AD is a beneficial aspect that can offset a portion of the waste management costs. Waste sources treated by AD include solid waste (manure, SS, and organic fraction) and liquid waste (agricultural and industrial wastewater and sewage) [55]. The production of biogas, bioethanol, biodiesel, and other biomasses from these wastes during AD plays a crucial role in the renewable energy field [56]. The methane content in biogas can be directly used as a source of heat, energy, and electricity [57,58]. Currently, biogas is produced mostly via SS digestion and biogas produced by AD consists of approximately 50–75% methane and 25–50% carbon dioxide. For this reason, the SS AD utilized for waste management and bioenergy production has garnered worldwide attention [40]. SS conversion to biofuel (biogas) could be a sustainable approach to transform sewage waste into an alternative fuel [56]. Most importantly, biogas produced from AD could become an alternative source for natural gas (Table 2).

Table 2

Biogas properties and composition.

Properties/Composition	Values	References
Calorific value	22.6 MJ/m^3	[171–173]
Ignition temperature	650–750 °C	
Lower explosive limits	6–12%	
Density	1.2 kg/m^3	
Critical pressure	7.3–8.9 MPa	
Methane	50–75%	
Carbon dioxide	25–50%	
Nitrogen	0–5%	
Hydrogen	0–1%	
Hydrogen sulfide	0–1%	
Oxygen	0–2%	

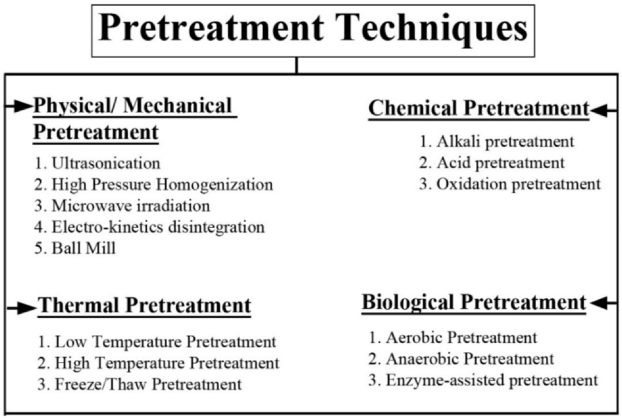

Fig. 3. Techniques of sewage sludge pretreatment for improved anerobic digestion.

4. Sewage sludge pretreatment – Anaerobic digestion enhancement

SS pretreatment prior to AD is essential to overcome the limitation encountered during AD hydrolysis [13,15,16]. Studies have reported effective pretreatment techniques that are physical, mechanical, chemical, thermal, and biological in character (Fig. 3) [5,6,9,17], which can accelerate SS solubilization. Enhancing the digestion process increases the substrate solubility and accelerates the biodegradation degree of organic solid waste during AD [48,59,60].

4.1. Physical and mechanical pretreatment

Physical and mechanical pretreatment disintegrates the solid particles, reducing their size and thus increasing the particle surface area to enhance the AD process [16]. Various studies have suggested that larger particles result in a lower chemical oxygen demand (COD) and less biogas production [61,62]. Physical and mechanical pretreatment techniques utilized for SS AD improvement are listed in Table 3. The most widely used physical and mechanical pretreatment processes for SS AD are explained in detail in Sections 4.1.1–4.1.5.

4.1.1. Ultrasonication

Ultrasonication is one of the most extensively studied and effective mechanical pretreatment methods for enhancing sludge biodegradability. Ultrasonication creates hydro-mechanical shear forces in cavitation that disrupt the sludge structure [16,63]. Different physical parameters such as ultrasound frequency, temperature, and density have been reported to influence the cavitation process during sludge pretreatment [64,65].

Ultrasonication exerts physical, chemical, and biological effects on the sludge, resulting in reduced particle size, higher organic compound

Table 3

Physical or mechanical pretreatment techniques for improvement of anaerobic digestion of sewage sludge.

Techniques	Mechanism involved	Effects	Drawbacks	References
Ultrasonication	-creates hydro-mechanical shear forces to disrupt sludge structure -specific energy, temperature, application time, ultrasonic frequency, and substrate nature are major factors influencing the ultrasonication mechanism -formation of cavities and their subsequent implosion	-exerts physical, chemical, and biological effects on sludge -reduces particle size, stimulates biological activity, solubilizes organic compounds, and releases enzymes -increases biogas production by 40–58%. -increases sludge dewaterability - VSS destruction	-high energy cost -not all studies confirm the enhancement of biogas production and VSS destruction	[5,64–67,70,73,174]
High pressure homogenization	-abrupt pressure gradient (up to 900 bar) -involves captivation, strong shearing forces, high turbulence, and subsequent depressurization -hydrolyzes macromolecules	-enzymatic hydrolysis of lignocellulosic biomass -increases biogas production by 43–90% -reduces odor-causing volatile substances	-less significant effect on pathogen removal -high energy consumption	[6,9,74,78,175,176]
Microwave irradiation	-operates within the wavelengths of 1 mm to 1 m -corresponding frequencies of operation are 300 MHz to 300 GHz	-increases organic compound solubilization -VS removal -increases biogas production by 20–53% -destruction of pathogenic bacteria such as *Clostridium perfringens*, *E. coli*, and *Salmonella* spp	-distribution of microwave power for materials is not uniform -may develop standing waves -no direct biomass degradation.	[79–83,177]
Electro-kinetic disintegration	-involves a high-voltage electric field of 20–30 kV -destroys ionic bonding of cell walls by changing the charge	-disrupts the rigid sludge and cellular membranes -changes microbial diversity during AD -increases biogas production by 30–31% -destroys COD -chemical neutralization is not needed	-no direct biomass degradation	[5,6,84–87]
Ball mill	-increases the sludge specific surface area	-increases biogas production by 20–41% -increases OM solubilization by 45%	-high energy requirement -fine particles may lead to acidification during AD	[74,91,94,95,178,179]

solubilization, biological activity stimulation, and enzyme release [5,64,66]. The impact of ultrasonication on the COD particle size could shift the peak at the particulate fraction (greater than1600 nm) to the lowest size range (< 2 nm) [67].

Sonification frequency and time play an important role in enhancing the AD process. At a density of 0.5 W/mL, frequency of 20 kHz, and sonification for 80 min, Li et al. achieved a 53.8% increase in methane production with a rapid decrease in Methanocorpusculum abundance and dewaterability deterioration in waste activated sludge (WAS) [65]. According to Appels et al., ultrasonication enhanced biogas production by more than 40% at a low specific energy input and approximately 15% at a moderate specific energy input [68]. Moreover, ultrasonic WAS pretreatment reduced WAS quantities, generated better dewaterability, and triggered the release of COD from the biosolids [69].

Based on previous studies, ultrasonication is the most widely used pretreatment process for enhanced biogas production during WAS AD and for enhanced sludge dewaterability [64,70,71]. However, the main drawback of ultrasonication pretreatment is the high energy cost [72]. Furthermore, not all studies confirmed the enhancement of biogas production and volatile solid (VS) destruction during ultrasonication pretreatment. Sandino et al. observed only a negligible increase in mesophilic methane production and VS destruction after WAS ultrasonication [73].

4.1.2. *High-pressure homogenization*

High-pressure homogenization (HPH) pretreatment involves an abrupt pressure gradient (up to 900 bar), cavitation, strong shearing forces, high turbulence, and subsequent depressurization resulting in a high soluble COD (sCOD) concentration and hydrolysis of the macromolecules [6,74,75]. Zhang et al. discovered that the most energy-efficient HPH treatment was at a homogenization pressure of 30 MPa with a single homogenization cycle for SS with a total solid (TS) content of 2.48% [76]. Moreover, a maximum sludge disintegration degree (COD) of 43.94% was obtained at 80 MPa with four homogenization cycles for a 9.58-g/L TS sludge [77]. HPH not only increased biogas production but also reduced the odor causing volatile sulfur compounds in the digester headspace from municipal waste sludge [78]. However, it has been reported that HPH wields a less significant effect on pathogen removal during the AD process [9].

4.1.3. *Microwave irradiation*

Microwave irradiation pretreatment is an alternative pretreatment process for WAS AD. Microwave irradiation pretreatment operates at wavelengths of 1 mm – 1 m with corresponding frequencies of 300 MHz and 300 GHz [79]. A 50% increase in biogas production has been reported with microwave pretreatment, leading to effective organic compound solubilization [80]. Additionally, the microwave pretreatment of SS AD increased the methane yield and biodegradability by 20% and 70%, respectively, in a semi-continuous mode [81].

Park and Ahn investigated the microwave pretreatment effect on the mixture of primary and secondary sludge during AD and observed a 3.2-fold increase in the sCOD to total COD (tCOD) ratio and VS removal of 41% [82]. This was accompanied by a 53% increase in daily biogas production at a reduced hydraulic retention time of 5 d. In addition to the enhanced biogas production, microwave irradiation helps destroy the pathogenic microorganisms during AD. Kuglarz et al. reported that microwave pretreatment reduced the *Clostridium perfringens* by 50%, total bacteria by 77%, and *Salmonella* spp. and *Escherichia coli* by 100% after irradiation at 70 °C (900 W; hydraulic retention time = 15–25 days) before AD. Additionally, 35% more methane was produced in comparison to untreated sludge [83].

4.1.4. *Electro-kinetic disintegration*

Electro-kinetic disintegration, also termed electric pulse, is a high-voltage pulsing electric field (20–30 kV) method, which disrupts WAS rigid sludge flocs and cellular membranes [5,6]. Lee and Rittmann studied the electro-kinetic disintegration effect at 34 kWh/m^3 and achieved a 110–460% increase in soluble compounds when applying the pulsed pretreatment to WAS AD [84]. Their study resulted in a 10–33% methane production improvement and an 18% increase in tCOD removal. Similarly, electro-kinetic disintegration of AD resulted in a 2.5-fold increase in biogas production and a 4.5-fold increase in the sCOD/tCOD ratio at 19 kV and 110 Hz for 1.5 s [85].

The electro-kinetic disintegration method causes a significant change in microbial diversity during AD. Zhang et al. studied the effects of focused-pulse sludge pretreatment at 16 kWh/m^3 on bacterial diversity [86] and discovered that the electro-kinetic pretreatment altered the methanogenic genera to acetate-cleaving Methanosaeta and away from the H_2-oxidizing Methanoculleus. Furthermore, a higher abundance of Ruminococcus was observed and methane production was increased by 30%.

Charles et al. investigated WAS AD enhancement using a 12 V two-chambered electrolysis process with an ion exchange membrane, in order to avoid chemical addition to change the WAS pH [87]. The pH dropped from 6.9 to 2.5 in the anode chamber and increased to 10.1 in the cathode chamber within a 15 h period. Chemical neutralization was not necessary because mixing the sludge from both chambers resulted in a pH of 6.5. In addition, they achieved a 31% higher methane production compared to untreated sludge.

4.1.5. *Ball mill*

Milling is a mechanical pretreatment process in which the large substrates can be reduced to fine particles leading to an increase in the AD surface area. Dry- and wet-milling processes consist of various milling pretreatment approaches such as ball, two-roll hammer, and colloid milling for WAS [88,89]. Ball mill disintegration has emerged as a beneficial pretreatment process for reducing microbial growth in SS AD treatment processes [90].

Ball mill pretreatment of SS increased sCOD from 2000 to 9000 mg/L and TSs increased from 1% to 4% at 75.8 KJ/g total suspended solids (TSSs) [91]. A study by Baier and Schmidheiny achieved an approximate 45% increase in sCOD for SS after wet-milling [92]. In addition, Kopp et al. achieved a 100% increase of SS biodegradation after 2 d and an improvement of approximately 20% after 4 d utilizing the stirred ball mill pretreatment [93]. Milling pretreatment of SS also increases biogas production. Milling pretreatment studies are limited and the major drawbacks are the high energy requirement during the pretreatment process and the formation of excessively fine particles, which may lead to a risk of acidification and VFA accumulation during AD [74,94,95].

4.2. *Chemical pretreatment*

Chemical pretreatment is the most promising method for complex organic waste destruction, which utilizes chemical reagents such as acids, alkalis, and oxidants to hydrolyze the sludge and also increases the biogas production by improving cellulose biodegradability [5,9,96]. For AD, different chemical processes have been studied, including alkali and acid pretreatment, and ozonation [9,97]; however, chemical pretreatment is not suitable for easily biodegraded substances [98]. The chemical pretreatment outcome mainly depends on the organic compound characteristics, applied method type, and chemical variety used. Table 4 summarizes the chemical pretreatment techniques for improving SS AD. Sections 4.2.1–4.2.3 describe the chemical pretreatment methods that have been widely used to improve AD.

4.2.1. *Alkali pretreatment*

Alkali pretreatment is a broadly used method for the disruption of sludge cells and EPSs leading to OM solubilization without producing toxic residues for the downstream processes. The reaction can be conducted at normal room temperature and pressure, and the energy requirements are modest [5,13,99]. Alkali pretreatment is effective for

Table 4
Chemical pretreatment techniques for improvement of anaerobic digestion of sewage sludge.

Techniques	Mechanism involved	Effects	Drawbacks	References
Alkali pretreatment	-disrupts sludge cells and EPSs. -can be carried out at a normal temperature and pressure. -low energy is sufficient for activities.	-solubilizes OM without producing toxins -solubilizes the COD and increases soluble macromolecules -improves sludge suitability for disposal -increases dewaterability of WAS -inhibition of pathogens such as *E. coli*, helminth eggs, and *Salmonella* spp. -increases biogas production by 38–80%.	-extremely high doses of alkali reduce AD activity -residual chemicals that can destroy the buffer system. -high cost of alkaline catalyst -alteration of lignin structure	[13,16,99,101,104,109–112,180]
Acid pretreatment	-involves the use of dilutes or concentrated acids -hydrolyzes the hemicellulose. -dissolves the cellulose -breaks down the lignin	-increases the solubilization of AD -supports accumulation of hydrolytic microbes -reduces tCOD and VSS -increases biogas production by 14–24%.	-reactor construction is expensive -acids are corrosive in nature -high cost of neutralization process	[5,113,114,118,181]
Oxidation	-involves the use of ozonation, peroxidation, and Fenton oxidation -based on hydroxyl radicals -	-disintegrates sludge and increases soluble COD. -does not increase salt concentration. -no chemical residue remains -results in more biodegradable recalcitrant compounds. -removes pathogenic microorganisms -increases biogas production by 20–200%.	-corrosiveness of peroxidation limits processes -requires high energy for ozone generation. -ozone is unstable	[5,9,74,119,121,127,131]

solubilizing COD [100]; however, the solubilization efficiency during alkali pretreatment depends on the type of chemicals used for pretreatment, which are listed in descending order of efficiency: NaOH > KOH > $Mg(OH)_2$ > $Ca(OH)_2$, and their concentrations [5,101–103].

The chemical dosage is associated with the amount of solubilization. Generally, large doses are associated with higher solubilization but extremely high doses reduce AD activity [101,104]. A study by Wei et al. examined the alkali pretreatment impact on hydrogen production from SS and achieved a maximum hydrogen production of 10.32 mL/g COD at an initial pH of 11.0 [105]. Another study indicated an sCOD increase from 200 to 8000 mg/L after alkali pretreatment of WAS with a pH of 12.0 [106]. Li et al. achieved SS organic degradation at a rate of 38.3% and a biogas yield of 0.65 L/g volatile suspended solids (VSSs), whereas these values for the control were 30.3% and 0.64 L/g, respectively [107].

Alkali pretreatment not only solubilizes the COD but also increases soluble macromolecules such as carbohydrates and proteins. Xu et al. achieved increases of 201.1% and 179.4% for soluble proteins and soluble carbohydrates, respectively, after alkaline pretreatment of sludge when adjusted to pH 10.0 by 5 N NaOH for 8 d [108]. Moreover, the biogas production increased by 41.41% compared to that of the control. Therefore, alkali pretreatment can improve sludge suitability for disposal and WAS dewaterability after alkali pretreatment [109]. Moreover, pathogens such as *Escherichia coli*, viable helminth eggs, and *Salmonella* spp. were killed after alkaline pretreatment, but *Azospira oryzae*, *Dechloromonas denitrificans, Geothrix* spp., and *Geobacter* spp. survived even at a high pH (greater than12.0) [110]. Alkali pretreatment, however, is associated with the presence of residual chemicals that can destroy the buffer system leading to anaerobic microbe inhibition, lignin structure alteration, and high alkaline catalyst costs [111,112].

4.2.2. *Acid pretreatment*

Acid pretreatment has received considerably less attention than alkali pretreatment for SS AD; however, this method is more effective at treating lignocellulosic substances present in SS because it supports hydrolytic microbe accumulation under acidic conditions and lignin breakdown [5,113]. The solubilization of COD and other macromolecules is related to the applied pH and 58% and 52% reductions were achieved for tCOD and VSS solubilization, respectively, during acid pretreatment of SS at pH 3.3 [114].

The increased solubilization also increases biogas production. Devlin et al. achieved a 14.3% increase in methane yield after acid pretreatment of WAS at pH 2.0 [115]. Acid pretreatment of SS not only increased the methane production but also elevated hydrogen-producing bacteria [116]. In contrast, strong acidic pretreatment may result in inhibitory by-product production, such as furfural and hydroxymethylfurfural [9,117]; however, concentrated acid is not preferred for acid pretreatment due to its corrosive nature and because it may result in increased costs in the neutralization process, which may undermine downstream processing [118].

4.2.3. *Ozonation*

Ozone (O_3) is a strong oxidant that has received significant interest with regard to WAS pretreatment. This method does not increase the salt concentration and no chemical residues remain compared to other chemical pretreatments [119]. Ozonation reacts with organic substances both directly and indirectly. The ozone indirect reaction is based on hydroxyl radicals, whereas the direct reaction involves rapid ozone decomposition into radicals. This process depends on the reactant structure, which facilitates recalcitrant compounds in becoming more biodegradable [9,120]. Ak et al. found that biogas production increased by 200% after sludge ozonation before AD [121] and when coupled with a mild ozone treatment (at 1.33 mg O_3/g VSS), biogas production doubled. Moreover, an ozone dose of 0.15 g O_3/g TSs resulted in an sCOD increase from 4% to 37% and biogas production increased 2.4-fold for WAS treatment [122]. Ozonation not only increases biogas production and sludge solubilization, it also removes pathogenic microorganisms from the WAS [123]. Although ozone has significant effects on SS AD, ozone instability and the high energy requirement for ozone generation are the major drawbacks of ozonation [5].

Peroxidation via H_2O_2 utilization has been reported as useful for disintegrating sludge and leads to increased sCOD [74,124]. The developed process was studied by Siciliano et al. and achieved phenol abatements of up to 80% and up to 90% VFA production using H_2O_2 (0.05 gH_2O_2/g COD) with lime [125]. Other alternative peroxidants utilized for chemical pretreatment are peroxymonosulfate and dimethyldioxirane, which increase the solubilization and biogas production of WAS. Dewil et al. achieved a 2- and 2.5-fold increase in biogas production in WAS when pretreated with peroxymonosulfate and dimethyldioxirane, respectively [126]. Another oxidizing agent, peracetic acid (PAA), forms a hydroxyl radical that reacts with organic compounds. Oxidation utilizing PAA could be an excellent pretreatment method because it does not produce any by-products [74]. Reductions of 24.5% and 39.0% in sludge solid and VS concentrations, respectively, were obtained with a 30 mg PAA/g SS dose after 120 min, as well as a 20% increase in biogas production compared to that of the control WAS AD [127].

A mixture of H_2O_2 and ferrous ions, generally known as the Fenton process, is frequently used for advanced oxidation [5,128,129]. The removal of 70% mixed liquor VSS was achieved by the addition of 725 g H_2O_2/kg TS and a H_2O_2/Fe^{2+} molar ratio of 80 and under ultraviolet light irradiation for 40 min for WAS [130]. Notably, Fenton oxidation pretreatment has been frequently applied to enhance WAS dewatering and sludge biodegradability [131–134].

All of the research studies noted in this section indicated that oxidation pretreatment (ozonation, peroxidation, PAA, and the Fenton process) results in an enhanced sludge solubilization effect and higher biogas production; however, peroxidation corrosiveness may limit these pretreatment processes.

4.3. *Thermal pretreatments*

Thermal pretreatment is a well-known commercially established technique used for improving AD hydrolysis [6,9,135]. In the thermal pretreatment technique, SS and other wastes are subjected to high temperatures, which induce hydrolysis and increase the digestibility of SS and other wastes [13,136,137]. This pretreatment strategy disintegrates cell membranes resulting in soluble organic substrates that are easily hydrolyzed during digestion [9,138]. Thermal pretreatments are beneficial in terms of pathogen sterilization, sludge volume reduction, odor removal, and enhanced sludge dewaterability [6,139,140]. Various temperatures (50–250 °C) have been implemented for the thermal pretreatment of SS [9]. Thermal treatment can be categorized as low-temperature (< 100 °C), high-temperature (greater than100 °C), and freeze/thaw pretreatment (Table 5) [5].

4.3.1. *Low-temperature pretreatment*

Low-temperature pretreatment employs temperatures below 100 °C for AD improvement. This technique can stimulate thermophilic bacteria, solubilize organic particles, and improve biodegradability [14,139]. Pathogens can also be removed from the sludge by applying low-temperature pretreatment at 70 °C [141]. De los Cobos-Vasconcelos et al. conducted a study on class A biosolid production without pathogens [141], in which SS was pretreated at various temperatures and a treatment at 70 °C for 1 h was found to be effective; however, pathogen reactivation was observed in the anaerobic mesophilic digester. Furthermore, their study applied a rapid cooling step utilizing an ice bath with NaCl followed by thermal pretreatment at 70 °C for 1 h and found no pathogen reactivation during anaerobic mesophilic digestion [141].

Nazari et al. established the ideal conditions for low-temperature thermal pretreatment of municipal wastewater sludge [139]. Their study reported that the optimal temperature, time, and pH for pretreatment were 80 °C, 5 h, and pH 10, respectively. Under these conditions, sCOD increased to 18.3 ± 7.5% and VS decreased to 27.7 ± 12.3% [139]. This indicates that organic fraction solubilization favors a higher temperature, longer reaction time, and alkaline pH conditions. Liao et al. treated SS for 30 min at 60, 70, and 80 °C and reported disintegration rates of 9.1, 13.0, and 16.6%, respectively [14] and biogas production increased by 7.3, 15.6, and 24.4%, respectively. In another study, sequential ultrasound and low-temperature (55 °C) thermal pretreatments were applied as options for sludge AD and improved outcomes for sludge solubilization, enzymatic activities, and AD were found. The methane yield after ultrasound and low-temperature pretreatment following AD resulted in an increase of up to 50% compared to raw SS digestion [142]. These studies clearly highlight the efficacy of low-temperature pretreatment for accelerating AD and

Table 5
Thermal pretreatment techniques for improvement of anaerobic digestion of sewage sludge.

Techniques	Mechanism and pretreatment conditions	Effects	Drawbacks	References
Low-temperature pretreatment	-sludge is treated below 100 °C -stimulates thermophiles for organic particle degradation	-removes pathogens from sludge -methane production increases by 10–100% -VS is reduced by 20–150%	-possibility of pathogen reactivation -occasionally results in low degradation of complex organic molecules	[5,139]
High-temperature pretreatment	-sludge is treated above 100 °C -disrupts the cell walls and membranes forming proteins	-complete pathogen removal without the possibility of reactivation -large amount of proteins are degraded -methane production increases by 10–150% -VS is reduced by 10–160% -Protein solubilization increases by 30–40%	-despite higher protein solubilization, ammonia is not released at a desirable level -requires time to maintain higher temperature	[5,6,145,146]
Freeze/thaw pretreatment	-sludge is subjected to freezing and thawing process -increases dewaterability and separates solid and liquid fraction -Freezing at a slow rate under −20 °C produces good results	-can solubilize OM -reduces VS by 16.9% -best for sludge in cold environments	-no adequate improvement in biogas yield -not considered effective for SS pretreatment in normal environments	[149–151]

increasing biogas production.

4.3.2. High-temperature pretreatment

This pretreatment technique applies a temperature greater than 100 °C to the sludge. High-temperature pretreatment typically promotes physical disintegration and solubilization of organic particles [5,143,144]. Thermal pretreatment within a temperature range of 125–175 °C transformed WAS to become readily and slowly biodegradable [140]. Thermal treatment disrupts the bonding of the cell wall and the membrane, thereby making proteins accessible for biodegradation [145]. Furthermore, the pretreatment temperature and sludge characteristics determine the ammonia solubilization. At a higher temperature, an adequate number of proteins are degraded and solubilized but very few degrade to ammonia. Graja et al. reported a 32% increase in protein solubilization at 175 °C; however, only 20% of this was transformed into ammonia [146].

Sludge solubilization relies on treatment time and applied temperature [6]. The higher the temperature, the higher the COD and VFA ratios will be. Aboulfoth et al. reported the optimal temperature range to be 175–200 °C for OM solubilization in a mixture of primary sludge and WAS [145]. Their study showed that at 175 °C the COD solubilization ratio increased from 11.25% to 15.1% and 25.1% within the 60–120 and 60–240 min treatment periods, respectively. Analyses have documented some variation in the biogas production results due to the effects of high-temperature pretreatment. Climent et al. reported that high-temperature treatment had no effect on methane production, whereas Carrère et al. found that high-temperature pretreatment increased biogas production by up to 150% [147,148].

4.3.3. Freeze/thaw pretreatment

In cold regions, freeze/thaw techniques can function as alternative approaches for sludge pretreatment. This method improves dewaterability and separates solid and liquid fractions by forming ice crystals [149,150]. Freezing improves floc conversion into a highly compact form and reduces the sludge water content. Slowly freezing sludge from −10 to −20 °C produces better results than rapidly freezing sludge at −80 °C [151]. Montusiewicz et al. evaluated the freeze/thaw treatment effect on mixed SS and found reductions of 12% and 16.9% in tCOD and VS, respectively [149]; however, the biogas yield only increased by 1.5%. This suggests that freeze/thaw pretreatment could contribute to a reduction in biomass but cannot adequately increase biogas production. Hu et al. showed that freeze/thaw treatment not only enhances sludge dewaterability but also improves OM solubilization from the sludge matrix [150]. Freezing and thawing are common natural phenomena in a cold environment; therefore, SS deposited in an external cold region environment could benefit from the freeze/thaw mechanisms prior to AD [5].

4.4. Biological pretreatments

Biological pretreatments are eco-friendly techniques that utilize aerobic, anaerobic, and enzymatic methods to predigest and enhance the AD hydrolysis stages [9]. These steps can be improved by implementing a complex matrix of microbes that play a synergistic role during the floc structure disintegration of sludge and other organic compounds [74]. This pretreatment technique, although eco-friendly and cost-effective, is time-consuming and requires optimal parameters for microbial proliferation [13]. Broadly, biological pretreatments are categorized into aerobic, anaerobic, and enzyme-assisted pretreatments (Table 6) and are described in Sections 4.4.1–4.4.3.

4.4.1. Aerobic pretreatments

Aerobic pretreatments can be performed by treating sludge with air and aerobic or facultative anaerobic microbes prior to AD [15]. The micro-aeration technique involves injection of oxygen into the treatment system, which aids in accelerating complex organic compound

Table 6
Biological pretreatment techniques for improvement of anaerobic digestion of sewage sludge.

Techniques	Mechanism involved	Effects	Drawbacks	References
Aerobic pretreatment	-sludge is treated with air and microbes prior to AD -oxygen is injected in the treatment system -microbes produce hydrolytic enzymes and degrades substrates	-improves sludge solubilization -accelerates hydrolytic activities -increases methane yield by 20–50% -decreases VS by 21–64%	-requires instrumentation to ensure the amount of air supplied -extra installation of aeration equipment may increase costs -slow process -requires appropriate conditions for microbial activities	[15,153,154,156,157]
Anaerobic pretreatment	-employs dual temperature strategies -most common method is TPAD -treated under thermophilic conditions followed by mesophilic conditions	-improves floc and solid structure disintegration -increases methane yield by 20–50% -decreases VS by 10–70% -kills pathogenic microbes -uses low-quality thermal energy	-requires parameter optimization -installation of additional equipment for energy balance may increase costs -biokinetic parameters for microbial communities should be evaluated	[9,159,160,162,178]
Enzyme-assisted pretreatment	-hydrolytic enzymes are added to the pre-treatment system -carbohydrases, proteases, and lipases are key enzymes that degrade biosludge and improve biogas yield	-degrades polymeric substances and improves sludge solubilization -increases methane yield by 12–40% -decreases VS by 16–55%	-several parameters should be optimized before adding enzymes to the pretreatment system -enzyme specificity with the substrate should be assessed	[5,166,168]

hydrolysis by improving the hydrolytic activities of the endogenous microbial population [152]. The oxygenic environment improves the hydrolytic activities of both aerobic and facultative anaerobic microbes. These microorganisms are important biological resources that can be used for SS pretreatment prior to AD [19].

Micro-aeration pretreatment stimulates the excretion of exoenzymes that slowly biodegrade substrates that otherwise remain recalcitrant under anaerobic environments [152]. High temperatures (< 70 °C) in combination with oxygen stimulate the hydrolytic microbial population to produce hydrolytic enzymes (e.g., proteases). These hydrolytic enzymes improve sludge solubilization and enhance organic compound degradation during AD; therefore, this pretreatment is also referred to as the autohydrolytic process [5,15]. Several studies have shown that micro-aeration treatment prior to AD not only enhances the AD hydrolysis step but also increases the overall methane yield [152–154]. Lim and Wang found that micro-aeration pretreatment increased the methane yield by 21% and 10% with inoculated and non-inoculated substrates, respectively [152]. Another study also showed the positive effects of micro-aeration pretreatment and found a 20% increase in the methane yield, suggesting that short-term oxygen pretreatment does not decrease the methanogenic activity of anaerobic methanogens [153]. Montalvo et al. optimized the airflow rate, pretreatment time, and temperature for microaerobic pretreatment of SS [154] and determined the optimum conditions for higher hydrolytic activity to be 0.3 vvm, 48 h, and 35 °C, respectively. In this scenario, micro-aeration pretreatment of SS increased methane production by 211% compared to the process without pretreatment [154].

Studies have shown that a pretreatment using hydrolytic thermophilic microorganism improves AD and increases methane production (Table 7). Aerobic pretreatment of organic waste with *Trichoderma viride* enhances AD hydrolysis and subsequently increases methane production [155]. Another study reported that *Bacillus licheniformis*, a thermophilic proteolytic bacterium, was very useful for sludge pretreatment and resulted in better OM stabilization and gas production [19]. Bioaugmentation using *Geobacillus thermodenitrificans*, an aerobic thermophilic bacterium, for sludge pretreatment showed a 21% reduction in VS and a 2.2-fold increase in methane production [156]. The effect of aerobic thermophilic bacteria was reported in another study using a temperature of 55 °C to ameliorate the AD of mixed sludge, which produced a 12% improvement in biogas yield and 27–64% reduction in VS [157]. Overall, these studies imply that aerobic pretreatment with oxygen or a thermophilic hydrolytic microbial population overcomes the hydrolysis process problems during AD and increases biogas production.

4.4.2. *Anaerobic pretreatments*

Anaerobic pretreatments can be conducted by predigesting the substrates in mesophilic or thermophilic environments [5]. For anaerobic pretreatment of SS, the commonly used method is temperature-phased anaerobic digestion (TPAD). TPAD is an effective option for sludge pretreatment that is utilizes a primary or hyper thermophilic digester followed by a secondary mesophilic digester [9]. Employing dual temperatures enhances SS hydrolysis and acidogenesis under thermophilic conditions and ensures acetogenesis and methanogenesis improvement under mesophilic conditions. This pretreatment strategy is also known as two-stage AD [6,15]. There are several advantages of TPAD, which include higher biogas production, improved floc and solid structure disintegration, low-quality thermal energy requirements, and killing of pathogens during thermophilic digestion [6,158].

Several research studies have been conducted on utilizing TPAD to predigest SS. A recent study performed TPAD on wastewater sludge digestion and found a 77% reduction in VS along with a methane release rate of 3.55 ± 0.47 L CH_4 /L day at 45 °C [159]. In another study, TPAD enhanced the biogas yield by improving sludge AD, which resulted in a 37–43% higher methane production [160]. Bolznella et al. studied the effect of extreme thermophilic prefermentation and reported a 30–50% increase in methane yield compared to mesophilic and thermophilic single-stage tests [161]. Ge et al. reported optimal conditions to be a retention time of 1–2 d, pH range of 6–7, and temperature of 65 °C for enhanced degradability and higher methane production for TPAD [162]. All these studies determined that thermophilic-mesophilic TPAD helps improve hydrolysis, remove VS, and increase biogas production.

4.4.3. *Enzyme-assisted pretreatments*

Enzyme-assisted SS pretreatments have amassed a significant amount of interest for AD hydrolysis improvement [163]. The addition of hydrolytic enzymes in a pre-treatment system improves sludge solubilization, degrades EPSs, and increases biogas production [5,164]. Brémond et al. have documented the following four enzymatic addition methods: 1) addition in a dedicated pretreatment vessel, 2) direct addition in a single-stage process digester, 3) direct addition in a hydrolysis and acidification vessel of a two-stage process, and 4) addition in recirculated AD leachate [15].

For successful enzymatic pretreatment, several parameters such as activity, specificity, quantity, enzyme stability, temperature, and pH should be assessed and optimized [163,165]. Sludge in wastewater treatment plants is mainly composed of carbohydrates, proteins, and minimal lipids. WAS is comprised of EPS rich flocs that are less biodegradable [15]; therefore, carbohydrases, proteases, and lipases are the main enzymes used for enzymatic sludge pretreatment [13,165].

Enzymes can accelerate the anaerobic degradability of biosludge and improve biogas production, and enzymes such as protease, amylase, glycosidase, and glucosidase were reported to improve anaerobic digestibility and increase biogas production. A 26% increase in biogas production was achieved with a protease pretreatment obtained utilizing *Bacillus licheniformis* [163]. Chen et al. evaluated the effects of lysozyme, protease, and α-amylase pretreatments for enhancing WAS hydrolysis and degradability and found that lysozyme was the most effective compared to the other studied enzymes [166]. Lysozymes increase the sCOD concentration in the sludge by 2.23 and 2.15 times compared with protease and α-amylase, respectively, and improved the sludge flocculation disintegration [166].

Fungal mash (*Aspergillus awamori*) has been used for enzymatic pretreatment of activated sludge, food waste, and their mixture. These hydrolytic enzyme rich substrates, pretreated with fungal mash,

Table 7
Microorganisms for potential application in the pretreatment of sewage sludge.

Microorganisms	Functions	Outcomes	References
Bacillus stearothermophilus SPT2-1	-enhances sludge solubilization -secretes hydrolytic enzymes (proteases and amylases)	-VSS removed at 20–30% -increases biogas production by 1.5-fold	[182]
Geobacillus thermodenitrificans AT1	-improves sludge degradation rate	-VS reduced by 21% -increases biogas production by 2.2-fold	[156]
Clostridium straminisolvens CSK1	-degrades lignocellulosic substrates	-increases biogas production by 136%	[183]
Bacillus sp.	-degrades oil substrates	-increases methane yield by 280%	[184]
Clostridia spp., *Bacillus* spp., and *Methanosaeta concilli*	-improves sludge digestion	-increases biogas production -promotes VS reduction	[142]

achieved a VS reduction of 54.3% and an increased methane yield of 1.6–2.5 times [167]. Odnell et al. suggested that specific enzymes that are better adjusted to sludge environments are required for large-scale pretreatment [168]. Several enzymes (cellulose, α-amylase, protease, lysozyme, subtilisin, and trypsin) were assessed for enzymatic activity, lifetime, and biogas production. The study concluded that the activity lifetime of all the assessed enzymes was limited (< 24 h) in WAS and anaerobic digester sludge. Among the tested enzymes, only subtilisin reported a marked increase in biogas production (37%) [168]. The effects of endogenous enzymes such as amylase, protease, and the mixture of amylase/protease were evaluated for sludge pretreatment. It was determined that a combined enzymatic treatment proved to be better for biogas production; however, for sludge solubilization and acidification, amylase was better than protease or mixed enzymes [169]. All these studies suggest that enzymatic pretreatment can enhance sludge AD and increase biogas production; however, further research is needed to determine specific enzymes for specific substrates to develop more efficient SS AD.

5. Future perspectives

An increasing SS amount has resulted in global concern regarding its treatment and management. As reviewed in several studies, the major limitation in SS disposal is the slow hydrolysis process during AD. To improve the hydrolysis process and enhance biogas production, various disintegration processes have been explored to pretreat the SS prior to AD. The physical, chemical, thermal, and biological pretreatment processes are widely used and they improve AD efficiency [2,13]. These pretreatments, however, do not always enhance biogas yield during AD [170]. There are several constraints on establishing sustainable and promising pretreatment methods. Most of the physical treatment methods require large amounts of energy, which limits their flexibility on an industrial scale. A significant issue with chemical pretreatment is the failure to balance pH in the reactor, which can inhibit AD performance [104]. The corrosive effects of acidic treatment may limit the processes and cause increased costs for designing a corrosive tolerant reactor. Thermal pretreatments are time-consuming and do not always produce positive results [5].

The most critical aspect of biological pretreatment is to maintain optimal conditions for microbial activities, which requires a high installation cost that restricts its application [159]. Therefore, it is vital to develop cost-effective pretreatment strategies that require less energy and minimal instrumentation. Proper assessment of the various pretreatment methods is lacking, which creates difficulties and economic burden for deciding the best SS pretreatment; therefore, a systematic assessment of the various pretreatment methods is an important step to combat the economic burden. A rigorous study on up scaling the research and system from the laboratory to the industrial plant is also required to estimate the energy and economic requirements accurately. Successful SS management also depends on environmental issues [6].

Future studies should be conducted to evaluate the impact of anaerobic digestate deposition in the environment and additional investigations should be performed on sludge properties, pretreatment conditions, and process parameters to produce cost-effective and practical pretreatment strategies. Research must be focused on reducing energy requirements and addressing environmental issues. Additional studies on microbial communities involved in AD to understand the microbial processes existing in the reactors would also be beneficial and might be useful for optimizing the appropriate conditions that are essential for microbial activity. Pathogen revival issues associated with various pretreatment methods are additional health and environmental concerns. This issue may encourage researchers to work on pretreatment techniques that can kill pathogens without the potential for reactivation. Combining different phenomena to ameliorate biogas production could also be a beneficial research topic for SS pretreatment.

6. Conclusions

In conclusion, to accelerate the hydrolysis process and enhance biogas production, various disintegration processes have been explored to pretreat SS prior to AD. The physical, chemical, thermal, and biological pretreatment techniques accelerate SS solubilization, which enhances solid organic waste biodegradation and increases the substrate solubility. Thermal pretreatment has been extensively implemented on an industrial scale due to its ability to increase OM solubilization and pathogen inhibition. Acid or base supplementation to thermal pretreatments improves biogas production. Ultrasonication is a widely used pretreatment technique that enhances biogas production and dewaterability. HPH results in high sCOD and reduces odor-causing sulfur compounds. Electro-kinetic disintegration significantly increases microbial diversity during AD. Chemical pretreatment is another promising method for biogas production and sludge biodegradability. Anaerobic pretreatment notably improves AD, increases methane yield, and has a low energy requirement. All these pretreatments have the potential to increase biogas yield; however, depending on the implemented substrates and facilities, these techniques have shown variations in sludge solubilization and biogas production. These existing pretreatment methods require further research to address the economic and energy concerns. Finally, there is an utmost necessity to establish standardized techniques for each pretreatment technique in terms of energy balance and environmental sustainability perspectives.

CRediT authorship contribution statement

V. Khanh Nguyen: Conceptualization, Writing - original draft. **Dhiraj Kumar Chaudhary:** Conceptualization, Writing - original draft. **Ram Hari Dahal:** Conceptualization, Writing - original draft. **N. Hoang Trinh:** Writing - original draft. **Jaisoo Kim:** Conceptualization, Investigation, Formal analysis. **S. Woong Chang:** Conceptualization, Investigation, Formal analysis. **Yongseok Hong:** Investigation, Formal analysis. **Duong Duc La:** Validation, Writing - review & editing. **X. Cuong Nguyen:** Writing - review & editing. **H. Hao Ngo:** Conceptualization, Writing - review & editing. **W. Jin Chung:** Writing - review & editing. **D. Duc Nguyen:** Supervision, Project administration, Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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