

Review

Disintegration of Wastewater Activated Sludge (WAS) for Improved Biogas Production

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Abstract: Due to rapid urbanization, the number of wastewater treatment plants (WWTP) has increased, and so has the associated waste generated by them. Sustainable management of this waste can lead to the creation of energy-rich biogas via fermentation processes. This review presents recent advances in the anaerobic digestion processes that have led to greater biogas production. Disintegration techniques for enhancing the fermentation of waste activated sludge can be apportioned into biological, physical and chemical means, which are included in this review; they were mainly compared and contrasted in terms of the ensuing biogas yield. It was found that ultrasonic- and microwave-assisted disintegration provides the highest biogas yield (>500%) although they tend to be the most energy demanding processes (>10,000 kJ kg⁻¹ total solids).

Keywords: biogas; renewable energy; anaerobic digestion; waste activated sludge; disintegration

1. Introduction

The continuous increase in the use of fossil fuel in modern society and the harmful effects of greenhouse gases on the environment has prompted the search for alternative energy sources which are becoming increasingly important and even mandatory in the future. In this context, research focusing on the improvement of biogas production has become essential. Moreover, due to the increase in urbanization, the number of wastewater treatment plants (WWTP) have increased, and since the wastes (e.g., waste activated sludge (WAS)) generated from the WWTP are considered hazardous for the environment, it is important to develop efficient processes for their pre-treatment [1].

Untreated activated sludge is hydrated to a level of 97–99% and the rest is comprised of solid and dissolved matter, minerals and organic substances, coagulants, gels and trapped gas bubbles. The stabilized sediment, on the other hand, is often hydrated only to a level of 60–88% [1–3].

The basic process of sludge utilization predominantly involves spreading it over the surface of the soil to fertilize it or improve its properties. Final disposal of sewage sludge is used in the following areas:

- In agriculture, for growing crops,
- For the reclamation of land, including land for agricultural purposes,

- For the adaptation of land to specific needs resulting from waste management plans, spatial development plans or decisions on building and land development conditions,
- For the production of compost,
- For the cultivation of flora not intended for consumption [2].

Unless it contains excessive amounts of heavy metals, stabilized sludge can improve the agrotechnical state of fertilized soils, because it contains a high concentration of micro- and macronutrients [4].

Most waste generators, including WAS, are obligated to handle it in a manner consistent with the principles of waste management and the requirements of environmental protection and waste management plans [5]. Firstly, waste should be subject to a recovery process and if it is impossible for technological reasons or is not justified for ecological or economic reasons, it should be subjected to disposal [2]. The composition of WAS varies enormously and depends on many factors, for example, the type and origin of WAS (e.g., the impact of industry in a settlement unit) and the method of treatment applied in the WWTP [6]. There are many issues concerning WAS including, dissolved heavy metals and/or toxic organic substances [7]: dioxins and furans, polychlorinated biphenyls (PCBs), and pesticides. At present, there are 500 substances classified in 15 different categories. With the exception of heavy metals, European Union regulations still lack strict limits on these substances in WAS.

Anaerobic digestion (AD) is one of the most commonly applied processes for WAS treatment as it is considered to be sustainable. Although its main purpose is the stabilization of WAS, however, it does decrease the water content of the sludge and associated toxicity. Another advantage of this method is that it produces biofuel (biogas), rendering the whole operation not only an environmentally feasible but also cost-beneficial. In this context, it is not surprising that the number of WWTPs producing and storing biogas is increasing each year. For example, in 2009 there were 6227 of these plants in Europe, by 2015 there were 17,376, an almost three-fold increase in only six years [8]. In China alone there are currently more than one million of these plants [9].

Biogas can be made from a range of organic substances and can be used to produce heat, power, heat and power (combined) or as a fuel for vehicles. Biogas contains methane, carbon dioxide, and nitrogen in different proportions and in trace concentrations also hydrogen sulphide, hydrogen, ammonia, oxygen, and carbon monoxide [10], siloxanes and aromatic and halogenated compounds (also depending on the fermentation/pre-treatment type) [11]. Primary production of biogas in the European Union is shown in Figure 1 and the typical characteristic of biogas can be found in Table 1 [12].

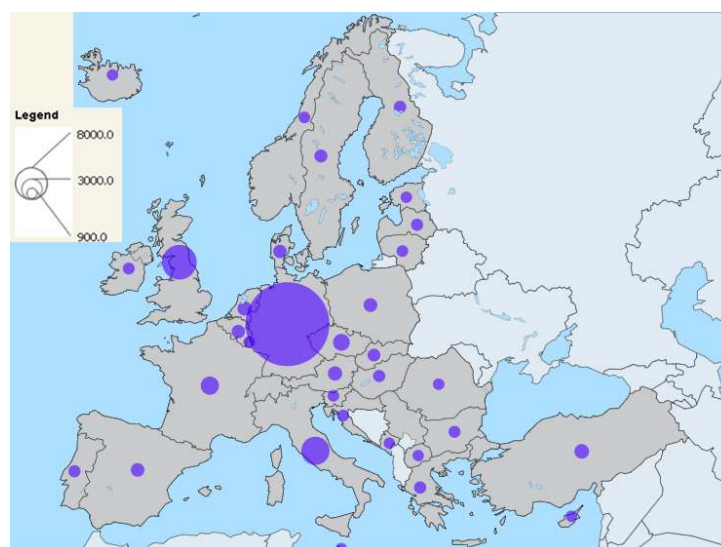
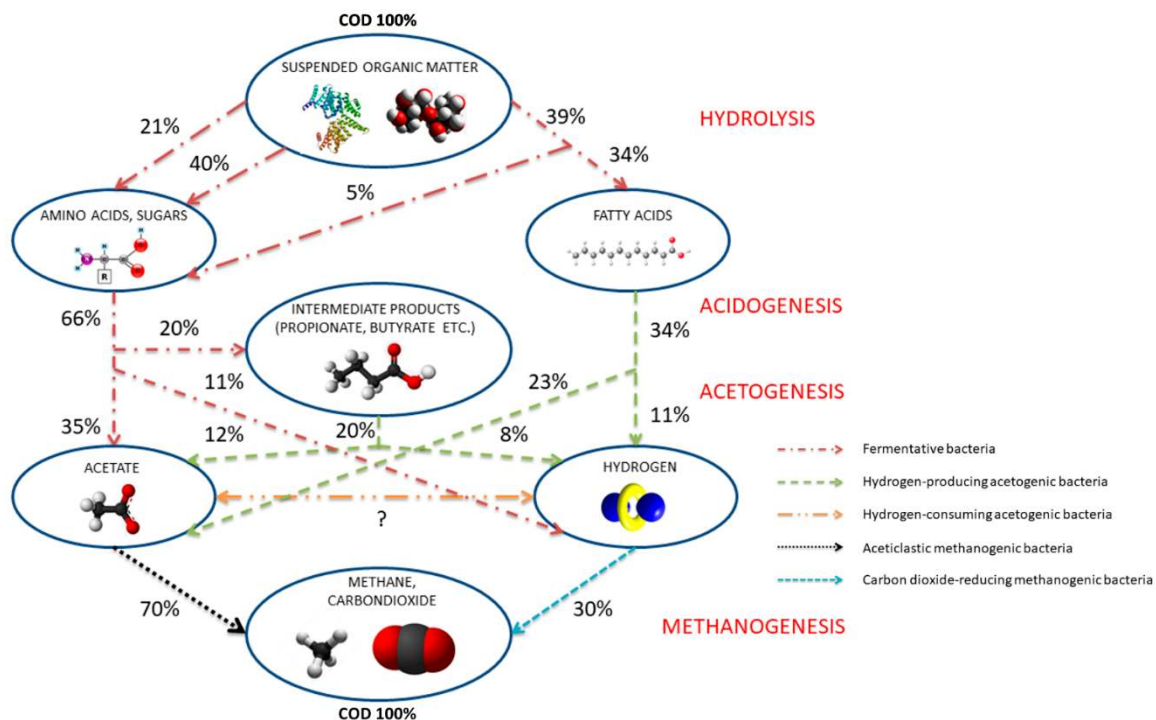


Figure 1. Primary production of biogas in the EU (tonnes of oil equivalent·1000; source: <https://ec.europa.eu/eurostat>; 2018).

Table 1. Typical characteristic of biogas.

Constituents	Energy (kW m ⁻³)	Fuel Equivalent (L oil m ⁻³ biogas)	Ignition Temperature (°C)	Critical Pressure (bar)	Critical Temperature (°C)	Normal Density (kg m ⁻³)
CH ₄ : 55–70%, CO ₂ : 30–45%, other gases	6.0–6.5	0.6–0.65	650–750	75–89	−82.5	1.2

Anaerobic digestion process entails four steps, i.e., hydrolysis, acidogenesis, acetogenesis and methanogenesis [13]; the complex process is attainable only under strict anaerobic conditions, with hydrolysis being a rate limiting step [14]. During hydrolysis, lipids, proteins, polysaccharides and soluble organic matter are all degraded, with the final products being further treated through acidogenesis to yield volatile fatty acids (acidogenesis = generation of acids; VFAs) and other by-products [15,16]. The acidogenesis step is followed by acetogenesis, during which the VFAs are digested by acetogenic microorganisms producing an even simpler molecule, acetate. The last step is methanogenesis, during which methane is generated; the process involves two methanogenic microorganisms, one group uses acetate to generate methane and CO₂ while the other uses hydrogen to produce methane [17]. This whole process, including the contribution of organic substances (chemical oxygen demand (COD)) and bacteria involved in it, is illustrated in Figure 2.

**Figure 2.** Proposed diagram of methane production during anaerobic digestion (based on [18]).

Biogas is a renewable fuel, which is considered to be more eco-friendly than conventional energy reserves; therefore, rapid development in this field is based on improving the biogas yield and especially by improving the pre-treatment process of WAS.

Pre-treatment of WAS is one of the most crucial steps before AD as it eliminates the disadvantages of the rate-limiting, hydrolysis step, and can often significantly enhance the yield of biogas with a simultaneous reduction of the sludge cake. In addition, several of these methods can efficiently reduce the toxicity of WAS by degrading the toxic and persistent microorganisms and molecules.

For various reasons, it is hard to compare the effectiveness of different pre-treatment methods used in wastewater technologies. The most important variables that make unambiguous comparison impossible are the type of sludge (inter alia: waste activated, primary, digested, sludge age) and

the operational conditions used (inter alia: temperature, continuous/batch, hydraulic retention time (HRT)).

However, this paper focuses only on the recent advances in WAS pre-treatment methods, which positively impacts subsequent biogas production.

2. Pre-Treatment Methods

Pre-treatment methods, be either separate or hybrid processes (not discussed here), are applied to provide the optimum results for various purposes [1]. Before selecting the technology, the pre-treatment goals must be clearly defined, because improvement of one of the processes can negatively affect another treatment stage.

As mentioned above, selection of the disintegration method depends heavily on the type of sludge. Müller [19] suggests that the most effectively method for primary sludges and sludges with high lignocellulose content is enzymatic pre-treatment. However, it could be less suitable for WAS or secondary sludges as they degrade themselves before enzymatic hydrolysis starts.

The vast majority of studies devoted to disintegration methods focus on the implementation of these methods for activated sludge or secondary sludge. This is due to the fact that primary sludges are most frequently composed of easily-degradable components (no treatment needed) or secondary sludges, and are mainly formed by microorganisms whose cell walls prevent rapid degradation.

One of the major components of WAS flocs are extracellular polymeric substances (EPS) comprised mainly of a proteins (e.g., enzymes), carbohydrates, humic matter and, to a smaller amount, uronic and deoxyribonucleic (DNA) acids and lipids. Multivalent cations and EPS interact and the associated hydrogen bonds results in the formation of a network of polymeric substances in the waste activated sludge [20,21]. All of this leads to longer retention times required for biological stabilisation. Many pre-treatment methods have received attention recently pertaining to EPS degradation and there are also many methods for their extraction [22,23].

Several different pre-treatment/disintegration methods are used specifically to improve anaerobic digestion e.g., biological, chemical and physical methods as well as combinations thereof. Essentially, sludge pre-treatment is used to break down the cell walls of microbes generally to reduce the molecular weight of substances in WAS, releasing the intracellular matter, which becomes more accessible to anaerobic microbes and consequently enhancement of the anaerobic digestion.

The most important objectives of disintegration/pre-treatment methods include:

- Simple access to the organic substances that were trapped inside the biomass and their release into the supernatant/liquid phase, as well as to intracellular enzymes that cause direct decomposition of pollutants,
- Release of organic substrate (in the case of disintegration of surplus activated sludge; often represented as chemical oxygen demand (COD)) that can be an easily digestible organic carbon source for the denitrification process. The increase in COD solubilization can be often correlated with the increase in methane production [24],
- Removing activated sludge foam generated on the surface of bioreactors as well as elimination of foaming in digestion chambers and secondary settling tanks,
- Increase in the biogas production and biogas yield and hence energy production with faster digestion.

In recent years, several pre-treatment/disintegration methods have been applied both on a bench and technical scale, for example by using thermal energy [25,26], enzymes [27], ozonisation [28], acidification [29], alkalization [30,31], high pressure [32,33], mechanical grinding [34] and ultrasound energy [35,36]. In general, these techniques can be classified into three categories: biological, physical and chemical (Figure 3).

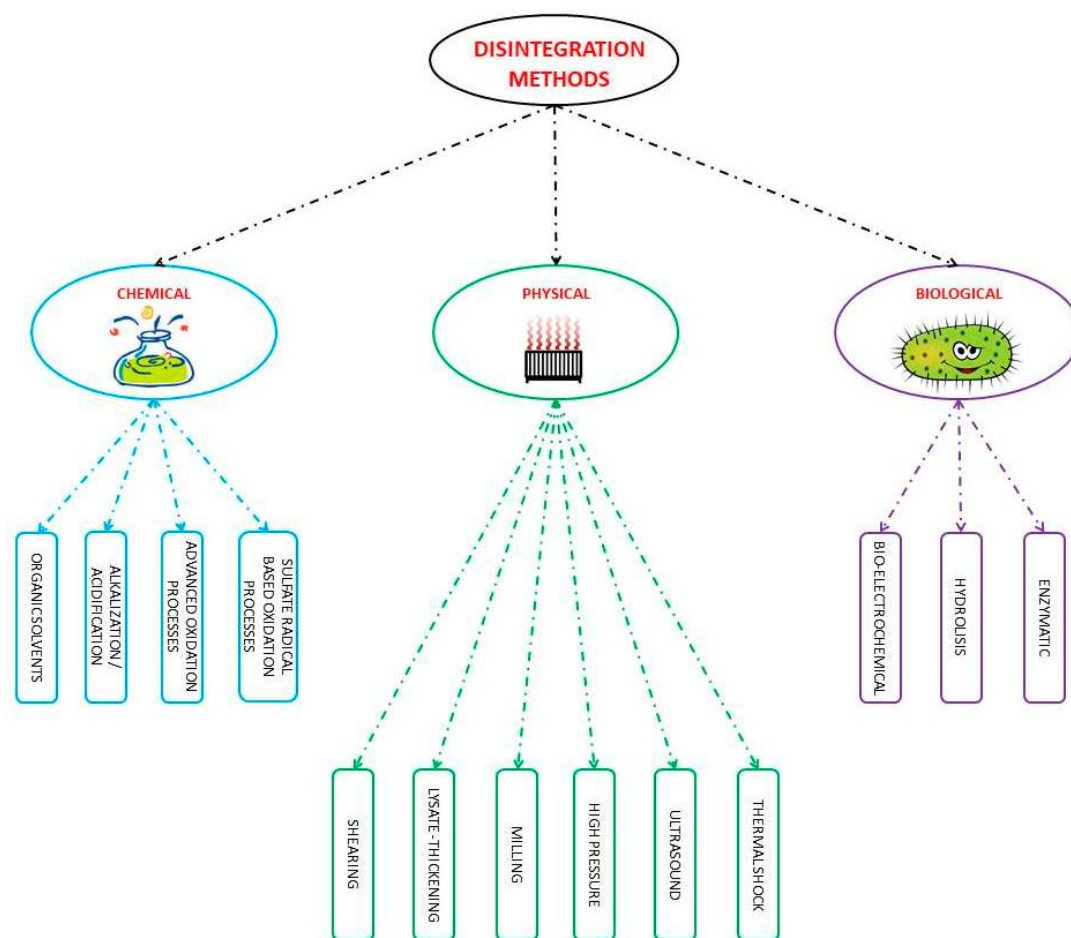


Figure 3. Diagram showing the available disintegration methods for waste activated sludge.

2.1. Biological

Biological pre-treatment (Table 2) can utilise either anaerobic or aerobic processes [37,38]. Anaerobic pre-treatment is one of the most commonly used in sludge pre-treatment [39], this process can effectively destroy pathogens, reduce volatile solids and enhance biogas production [40–44]. So-called bioaugmentation can be used as a pre-treatment of WAS for reducing the molecular weight of substances therein [45,46]. Indeed, many authors have reported on the effectiveness of this technique e.g., addition of cellulolytic bacteria, that are capable of generating an enzymatic complex, cellulosome, can enhance the digestion rates of cellulose (and subsequent improvement of biogas production) [47].

Ai et al. [48] concluded that adding *Bacillus* coagulants can promote the hydrolysis and acidogenesis process with no negative effect on the methanogenesis process. In order to obtain the sludge disintegration, it is possible to use enzymes, which are biologically derived molecules that work as a catalyst (also termed biocatalyst). It is possible to classify these enzymes into six basic classes: oxidoreductases, ligases, transferases, lyases, hydrolysases, and isomerases. Enzymatic lysis by the enzyme catalysing the reaction leads to the breaking down of bonds and compounds constituting the cell wall of the microorganisms. The enzymes can help decompose the organic matter, i.e., turn it into smaller molecules [27,38,39,49–62]. Recently, Prajapati et al. [63] successfully used a bio-electrochemical process to enhance the methane production from sewage sludge by augmenting it with food waste.

2.2. Chemical

Chemical pre-treatment is another efficient and cost-effective process for hydrolysing the membranes and the cell walls and thus increasing the solubility of the organic matter that is

located inside the cells; the most common used before anaerobic digestion are alkalization [64] or acidification [58,65,66].

The often used alkaline reagents are NaOH [67], Ca(OH)₂ [68] and KOH [68]. Alkali pre-treatment can solubilize the cell membranes, releasing the intracellular matter from the cells in solution, which becomes available for the fermentation process. The intracellular matter essentially formed by lipid proteins and hydrocarbons is decomposed in the soluble substances that are available for the microorganisms [69]. The advantages of alkali methods are their high efficiency and the ease of performance during the process [70].

On the other hand, oxidation techniques are used because of their enormous efficiency. Traditional oxidation techniques include Fenton reactions, and photocatalytic and ozonation processes. All the aforementioned techniques have one thing in common, namely the formation of a hydroxyl radical; these processes are also known as advanced oxidation processes (AOP). Ozonation pre-treatment includes two different types of oxidation processes i.e., ozonolysis and hydroxyl radical reactions, which depend mainly on the pH values. It has been reported that at low pH ozone reacts selectively with the organic molecules comprising C=C, -OH, CH₃, -OCH₃ and other functionalities [71], and at pH > 8 the ozone generates 'O₂⁻ and HO₂' radicals [72]; more details are available [71,73–86]. On the other hand, Fenton reactions involve the reaction of hydrogen peroxide with divalent iron producing hydroxyl radicals, and have been used in environmental matrices (including WAS) for a long period of time [87,88]. Although, these methods have been tested for many years for WAS pre-treatment, there are still some innovative solutions being uncovered. For example, in a recent study Hallaji et al. [89] reported the possibility to increase the methane production by 72% with a combined free nitrous acid (FNA)/Fenton reaction pre-treatment process. The possibility to increase methane production by ~200% with a combination of micro aerobic hydrolysis and the addition of trace metals has also been reported [90]. Anjum et al. [91] recently reported the possibility to enhance the biogas production using photocatalytic disintegration of WAS, whereby this procedure increased the biogas production by 1.6 times.

It should be noted that whereas the hydroxyl radical pre-treatment of WAS have been known for some time, sulphate radical pre-treatment (activated peroxydisulfate or peroxymonosulphate (persulphates)) has only been introduced in the last few years; however, several limitations disable their in situ application. Peroxydisulphate (PDS) is a strong oxidant used with success for the disintegration of WAS by several authors [22,92–95]. In order to form sulphate and hydroxyl radicals, persulphates need to be activated, usually by heat [96], metal [97], UV [98] and alkaline conditions [99], among others [100].

Many oxidation techniques, including Fenton reactions [101]) and new reactions concerning peroxomonosulphate (PMS; [102]), nano-layered TiO₂ [103] and dimethyldioxirane (DMDO), can cause the alteration of refractory organic matter into easily accessible and soluble biochemical oxygen demand (BOD), and subsequently improve the biogas yield [88] (Table 2). Recently, a newly greener oxidant for this purpose, peracetic acid (PAA), has been proposed to improve anaerobic digestion [104].

2.3. Physical Methods

Physical and mechanical treatments of WAS work in basically the same way, the cell walls are broken and flocs break up by the application of force/external energy. Disintegration of the WAS with the use of mechanical forces causes the fragmentation of flocks and effective lysis of bacterial cells, leading to the release of organic substances and therefore an increase in biogas production.

The effect of mechanical disintegration of WAS on the efficiency of AD has been investigated for a long period. Mechanical shearing, lysate-thickening centrifuge [105], milling technology [106], and high pressure technology [61,107–112] are some of the main techniques.

Hydrodynamic cavitation triggered by the Venturi effect is a promising process for pretreatment of waste activated sludge prior to mesophilic fermentation according to many authors, including Machnicka et al. [113]. Furthermore, mesophilic digestion trials have reported a significant increase in

the biogas production of approximately 36.1% and 62.16% for 10% and 30% of the volume of foam added to the digestion chamber, respectively.

Ultrasonic methods have been included as physical treatment in this review but they can also be included as AOPs. These methods involve two different processes: cavitation, which is promoted at low frequencies, and the formation of radicals ($\cdot\text{OH}$, $\text{HO}_2\cdot$, $\text{H}\cdot$) due to the chemical reactions at high frequencies [38]. To induce cavitation, the process has to operate at a certain frequency (<100 kHz), the ultrasound creates gas bubbles that up on collapsing produce hydromechanical forces, which disintegrate the macromolecules [33,114,115]. The extreme conditions that occur during the cavitation process can cause generation of hydroxyl radicals, which can degrade volatile and non-volatile pollutants. Ultrasonication can enhance the WAS digestibility by damaging the physical, chemical and biological properties of the sludge; lysis accelerates the hydrolysis reactions by disrupting the cells. Within the explosion (cavitation) of transient bubbles, a certain amount of soluble particulate organic matter can be rendered completely soluble. Ultrasound is considered as one of the most efficient sludge pre-treatments for sludge floc disintegration [85,114,116–121]; Lizama et al. [122] reported an increase in biogas production after ultrasonic treatment of 560% (Table 2).

Table 2. Recent advances in the pretreatment of WAS for biogas production enhancement.

Disintegration Type	Treatment Type/Condition	Anaerobic Digestion Condition	Results	Reference
Biological	Amylase + protease	37 °C	+23% biogas yield	[27]
	Subtilisin	38 °C	+37% biogas yield	[123]
	Biological hydrolysis	35 °C	“significantly higher methane generation”	[124]
Chemical	Micro-aerobic hydrolysis	35 °C	38% methane yield	[90]
	Acidification: 0.52–1.42 mg $\text{HNO}_2\text{-N L}^{-1}$	37 °C	+12–16% methane yield	[66]
	Acidification: 2.5 mg L^{-1} HNO_2	37 °C	+25% methane yield	[89]
	Alkalization: 20 mg NaOH g^{-1} TS	37 °C	+35% methane yield	[125]
	Alkalization: 157 mg NaOH g^{-1} TS	37 °C	+34% methane yield	[126]
	Oxidation: H_2O_2 : 5 mg L^{-1}	37 °C	+27% methane yield	[89]
	Oxidation: 0.1 g $\text{K}_2\text{S}_2\text{O}_8 \text{g}^{-1}$ SS	35 °C	180% methane yield	[93]
	Oxidation: ZnO-ZnS@polyaniline	35 °C	62% methane yield	[91]
	Hybrid: $\text{HNO}_2/\text{H}_2\text{O}_2$	37 °C	+72% methane yield	[89]
	Thermal: 70 °C	55 °C	+148% methane yield	[127]
	Thermal: 90 °C	55 °C	+161% methane yield	[128]
	Physical and hybrid	Thermal: 100 °C	33 °C	+343% biogas production
Thermal: 120 °C		33 °C	+345% biogas production	[129]
Thermal: 134 °C		55 °C	+47% biogas yield	[130]
Microwaves: 14,000 kJ kg^{-1} TS		35 °C	+570% biogas yield	[131]
Ultrasounds: 96 kJ kg^{-1} Sludge		37 °C	+27% biogas yield	[132]
Ultrasounds: 750 kJ		37 °C	+52% methane yield	[133]
Ultrasounds: 1000 kJ kg^{-1} TS		35 °C	+95% methane yield	[134]
Ultrasounds: 25,000 kJ kg^{-1} TS		36 °C	+560% biogas yield	[122]
Hybrid: Alkalization + Ultrasounds		35 °C	+33% biogas yield	[135]
Hybrid: Free ammonia (135 mg L^{-1}) + 70 °C		35 °C	+25% biogas yield	[136]

Compared to mechanical techniques, thermal disintegration processes consume more energy, but they can be deployed e.g., by using heat exchangers, or by the use of steam to the WAS. Thermal pre-treatment processes can occur at a wide range of temperatures from 60 to 270 °C; processes at a temperature of <100 °C are considered as low temperature processes and those taking place at a temperature of >100 °C are high temperature processes [60,61,85,137–141].

The optimal temperature treatment is frequently stated to be around 170 °C [142]; Ennouri et al. [143] reported that at a thermal pre-treatment temperature of 120 °C, the biogas yield increased by 37%.

However, Dwyer et al. [144] reported that COD solubilisation increased at temperatures above 150 °C but there was no increase in the methane production. According to Batstone et al. [145], the main disadvantages of thermal pre-treatment are associated with the costs and increased ammonia inhibition.

Another pre-treatment process that has already shown to improve the anaerobic digestion is microwave pre-treatment [146]; which is a good substitute for thermal pre-treatment, increasing the concentration of soluble proteins in solution [147] and improving biogas [148,149] production and disinfection [150]. Microwave irradiation has two main effects, thermal and non-thermal; they involve

the interaction of rapidly alternating electric field with polar proteins, fats and H₂O and the breakdown of hydrogen bonds and the consequent death of microorganisms [151]. Other recent methods involve the application of low temperatures for WAS treatment known as freezing/thawing [25,26,152].

3. Conclusions

In conclusion, we have evaluated various disintegration protocols pertaining to the waste activated sludge for the enhanced production of eco-fuel-biogas. The pre-treatments are focused mainly on augmenting the disintegration method and improving the hydrolysis and gaining more biogas in the anaerobic digestion (AD) process. Three types of pre-treatment processes (biological, chemical and physical) are presented with their associated strengths, weaknesses and recent advances. Some pre-treatment processes are more efficient in reducing the biomass, while the others work better for the solubilization of organic matter or for the cell disintegration. Additionally, the diverse pre-treatment processes generate fluctuating costs depending on e.g., the volume of WAS used, varying reaction times and effects on biogas generation. Biological pre-treatment is usually slower than other types of treatment and can last several days. Moreover, despite its advantages e.g., often lower costs, it is less effective in terms of solubilization of the organic matter and corresponding increase of the biogas yield (<40%). Regarding the chemical treatments, one could distinctly identify peroxydisulfate one, which produced 180% increase of biogas yield (the highest in comparison to all other chemical treatments reported herein), however, further studies are needed to verify the quality of thus produced biogas, since the sulfate ion (end-product of the treatment) can negatively influence anaerobic digestion process. Chemical and physical methods are faster and are rather easier to implement; however, they can be more energy demanding as described in recent findings, where the application of microwaves or ultrasonic energy increased biogas production by >500% but tend to be very energy-demanding (>10,000 kJ kg⁻¹ TS). In keeping with the aim of attaining sustainability, the newer pathways need to be explored which may use photochemical approach exploiting the visible light or solar energy, an abundant energy source, to facilitate the processability of the sludge and reduction of water contents.

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