

Review Article

Enhancement of biogas production by means of ultrasonic pre-treatment: State of art and review

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ABSTRACT

Currently, fossil fuels are being used in an aberrant manner and greenhouse gases are negatively impacting the environment, causing research into renewable energy sources to be pioneered. As highlighted by the International Treaty called the UN Framework Convention on Climate Change during the Conference of Parties (COP) meetings every year, the international community is striving to expedite worldwide climate action by reducing emissions, intensifying adaptation initiatives, and increasing the availability of suitable financial resources. Production of biogas is a way to help overcome these challenges by reducing wastage and preventing environmental damage. The anaerobic digestion technique faces some barriers like elongated reaction times, cost of heating or low biogas yields. Several techniques have been used to enhance the biogas yield. Among these techniques the use of ultrasound technology as a pretreatment in biogas production revolutionized the process with its widespread application, providing an environmentally friendly and low-cost alternative. This paper reviews research studies on ultrasonic pretreatment of several substrates categorized according to different sources, for biogas production and puts an effort to compare and advise various operation parameters. Low frequency ultrasound (10-50kHz) results in an increase between 15-800 % biogas production from several substrates, at varying operation periods of 2-300 minutes. The operators are advised to test different operation conditions to obtain the best yield in real case applications.

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INTRODUCTION

Biogas is a mixture of gases, primarily consisting of CH₄ (50-60%), CO₂, (40-50%), and the trace of other gases such as H₂S₂, O₂, and H₂ [1,2]. It is produced from materials such as livestock/agricultural waste, industrial, institutional and commercial waste, waste-activated sludge from wastewater treatment plants, landfill leachate and algae. It is a renewable energy source because the “production- and- use cycle is continuous”. From a carbon standpoint, the growth of the primary bio-resource absorbs an equal amount of carbon dioxide from the atmosphere as it emits when it is eventually transformed into energy.

Typically, biogas is generated by the anaerobic digestion of biodegradable organic waste either at landfill sites or in specially-designed reactors [2]. In an anaerobic digestion process, microbial fermentation of the organic residual occurs in the absence of oxygen, as a result the mixture of gases is produced [1]. This process is a complex biological process that includes many consecutive steps of conversion and biodegradation of the organic waste namely hydrolysis, acidogenesis, acetogenesis and methanogenesis [3]. If we analyze each stage, hydrolysis emerges as the initial step in which extracellular enzymes facilitate the conversion of complex organic polymers and lipids into essential building blocks such as fatty acids, monosaccharides, amino acids,

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and other related compounds. These transformed substances serve as a valuable energy source and contribute to the formation of cellular tissues [4]. During acidogenesis, fermentative bacteria break down soluble organic monomers of sugars and amino acids, resulting in the production of volatile fatty acids such as propionic, butyric, and valeric acids. Additionally, acetate (CH_3COO^-), H_2 , and CO_2 are generated. The degradation of amino acids in this process also leads to the production of ammonia (NH_3) [5]. In the acetogenesis step, both long-chain fatty acids and volatile fatty acids are destroyed, resulting in the production of acetate, carbon dioxide, and water [6]. Methanogenesis is the last step, and it involves bacteria breaking down the hydrogen and acetic acid to methane gas and carbon dioxide. The bacteria that cause this conversion are called methanogenic because they are strict anaerobes [7].

The hydrolysis step serves as the rate-limiting step, wherein hydrolytic enzymes secreted by specific groups of bacteria break down complex organic compounds (such as carbohydrates, proteins, and fats) into their respective constituent monomers. These monomers are molecules that are more soluble and can be easily transported through the cell membrane. There are several methods to enhance the rate of hydrolysis and ultrasonic pre-treatment has become one of the most popular methods among them.

The principle behind sonication of heterogeneous reactions lies in the disruption of the solids, caused by the jetting phenomenon resulting from the collapse of cavitation bubbles. It is worth mentioning that numerous similar effects can be observed when the heterogeneous medium is exposed to low frequency or power ultrasound (US) within the 20–100 kHz range. [8]. The impact of ultrasonic phenomenon on the fluid can be expressed as follows:

- Compounds are physically destroyed due to the presence of very high hydrodynamic shear stress [9].
- Chemical reactions are advanced as a result of high stress and extremely high local heating, reaching temperatures of nearly 5000 °C [10].

In particular, when low frequency ultrasound is applied to organic-rich samples, it enhances the ratio of Soluble COD/Total COD (SCOD/TCOD) by converting particulate matter into soluble COD. Another phenomenon that occurs during ultrasonic irradiation is the formation of acoustic flow. This acoustic flow is generated at the interface between a solid and a liquid when the solid interface vibrates with regular oscillations. The main advantage of acoustic flow is its ability to effectively mix substances in an aquatic environment, ensuring that ultrasonic energy is evenly distributed throughout the entire volume of a heterogeneous medium. Additionally, it facilitates the rapid distribution of heat. [11].

Ultrasound helps to break the large compounds into more biodegradable forms via the physical effects mentioned above. In this way, both the particle sizes and the biodegradability properties of the substrate are changed and the time for hydrolysis is reduced. Therefore, this review paper aims to recover a broad range of scientific enhanced work on

the production of biogas through ultrasound pretreatment. Many researchers working on this subject, have created a rich content for the scientific literature by using different measurement parameters and examining different environmental factors. A large number of studies were examined in order to see the similarities and differences by bringing these studies together.

For ease of assessment, the research has been categorized according to the raw material (substrate) content as below:

1. Livestock/agriculture waste
2. Activated sludge from wastewater treatment plants
3. Industrial, institutional and commercial waste (including food waste)
4. Algae
5. Landfill leachate

Each category has unique specifications and the concluded enhancements will be considered specifically for each of them, as well as the final conclusion of comparison among all raw materials. Some studies fall into mixed categories of substrate, however they are mentioned when evaluated in the text when considering them. Several other environmental factors effecting the biogas production such as temperature, reaction retention time, concentration of initial substrate etc. have been referred to, while perusing the ultrasonic contribution to the process. The energy balance of ultrasonic pretreatment is determined by the surplus energy generated after pretreatment, subtracted by the energy consumed during the pretreatment process. This balance is greatly influenced by the concentration of Total Solids (TS) in the substrate. Too high concentrations can produce adverse effects on sludge rheology and transportability as well as on possible inhibitor concentrations, while too dilute substrates can absorb the energy in the liquid portion of the liquor. It is obvious that for each substrate, the optimum conditions must be examined with great care, to produce highest possible energy potential.

This paper reviews research studies on ultrasonic pretreatment of several substrates categorized according to different sources, for biogas production and puts an effort to compare and advise various operation parameters for the enhancement of biogas production.

REVIEWED PAPERS ACCORDING TO SUBSTRATE CATEGORIES

Livestock/agriculture waste

This category defines the wastes produced in the farm/agricultural facilities (i.e., manure, crop residues, husks, agricultural production residues etc.). Improperly managed livestock wastes can lead to fecal contamination of waters receiving agricultural runoff. Livestock waste poses a significant threat as it emits noxious gases, harmful pathogens, and unpleasant odors. Consequently, it raises concerns for both public health and the environment. Therefore, it is crucial to

effectively manage livestock waste to reduce the production of these pollutants and safeguard the environment. One effective approach is to properly utilize livestock waste in the production of biogas and compost, which can greatly enhance crop yield and promote sustainability.

Castrillón et al. [12] studied the influences of fragmentation of substrate on biogas production in the presence of ultrasound waves as a pretreatment method under mesophilic and thermophilic conditions using the batch reactor. The cattle manure was either screened or ground. Several experimental parameters were investigated. The results showed limited effect of sonication time, however, the higher solubilization ratios were obtained using higher ultrasonic energy. Regarding the biogas production and methane content, the best result was obtained at 35°C for the sonicated mixture of screened manure with “365.4 L biogas/kg COD” with a methane content of around 60% (v/v). However, high temperature of 55°C produced a high percentage of gases of 581.6 L biogas/kg COD from a mixture of sonicated ground manure with the same methane content.

Braeutigam et al. [13] reported the impact of sonication time (1-2 min), amplitude (63, 94, and 125 μ m) and consumed energy on biogas yield from biological waste material (5% chicken manure), at room temperature of 25°C using frequency at 24kHz with 200W. The better outcomes were obtained at 2 min of reaction time with an amplitude of 94 μ m leading to an increase in biogas yield to 41% with respect to the non-sonicated sample. However, an intensive energy input could result in acidification, which causes less methane production. Therefore, low amplitudes and high reaction times, or vice versa, could yield the best outcomes. Moreover, the generated energy was higher than the consumed energy for disintegration by 36.6% per kg of chicken manure (2.72 kWh/kg substrate). The correlation of the biogas yield with soluble chemical oxygen demand (SCOD) and particle size was also investigated and the results showed that a linear relationship was observed for both SCOD increase and particle size reduction. The biogas yield was measured as 393 L/kg VS.

Zieliński et al. [14] carried out an experiment on ultrasonic pretreatment and hydrodynamic cavitation pretreatment to improve the production of biogas from dairy cattle manure mixed with wheat straw residues in a small-scale agricultural biogas plant. The energy balance with or without substrate pretreatment was analyzed. The small-scale plant was designed to include a special cavitation chamber and contained 60 transducers each with 10 kW power. Ultrasonic pretreatment and hydrodynamic cavitation pretreatment were found to significantly improve the production of biogas to 430 and 460L/kg VS, with a value greater than the other researchers working on agricultural substrates. Therefore, this study implies that cavitation operations were efficient for agriculture residues pretreatment.

You, Z. et al. [15] carried an experiment for the production of biogas through the enhancement of the fermentation of corn stover using pretreatment of sodium hydroxide in the company of additive calcium oxide together with 40 kHz US

(150 W). Sodium hydroxide is a costly technique applied at high temperature and long retention time (a few days) for lignin dissolution. Addition of calcium oxide along with US method decreased the cost of pretreatment of sodium hydroxide on corn stover at low temperature considering low time treatment (30 min). The result implies that the process of the pretreatment was able to intensify the transformation of lignin to about 60%, promoting production of biogas to 500 mL/g TS. Benefit to cost ratio was determined through a method related to the process and the pretreatment came about an increase in the ratio from 1.40 to 1.65, indicating a cost effective technique.

Similar to You, Z et al. [16], Dong et al. [17], also carried out a research about improvement of the production of biogas from the corn stalk by dual US frequency (20 kHz and 57 kHz) in the company of alkaline pretreatment. However, the study documented the procedure and highlighted that the stimulation program demonstrated the superiority of energy over frequency. The corn stalk underwent anaerobic fermentation for a duration of 53 days after being pretreated with dual-frequency US and alkali. The findings revealed that the dual frequency process resulted in a cumulative biogas yield that was respectively 11.1%, 28.2%, and 56.6% higher than that of single-frequency US combined with alkali pretreatment, alkali pretreatment alone, and no pretreatment. The simulation program also attributed this outcome to the larger radius of the cavitation bubble in the case of dual frequency.

Zou, S. et al. [18] carried out research in anaerobic co-digestion of dairy manure and maize straw for biogas production at various ratios, through ultrasonic pretreatment. 50 kHz, 250 W US was applied on the dairy manure and maize straw separately and in combination at certain time intervals and power intensities. The authors have implied that ultrasonic pretreatment improved biogas production via changes in surface structures before and during anaerobic digestion process. Scanning electron microscope was used to detect the surface structure changes. The highest biogas produced of co digestion obtained was (240.32mL/gVS) when the dairy manure was pretreated for 30 min. However, the operation was affected by some other factors such as pH, reducing sugar, cellulosic activity at numerous stages digestion, as well.

Amirante, R. et al. [19] studied the biogas production from a 3-phase of olive pomace with and without ultrasonic pretreatment. The olive pomace produced in an olive oil mill in the southern regions of Italy was observed during anaerobic digestion for 43 days. During the pre-treatment, a frequency of 37 kHz and an installed power of 256 W were utilized to apply low-frequency and high-power US. The duration of this application was 1.88 minutes, aiming to achieve a specific energy range of 15-18 kJ/kg. It was crucial to prevent the destruction of bacterial flora. Consequently, the thermal conditions of the ultrasonic process were continuously monitored using an infrared thermometer throughout the experiments. The observations have proved an increase of 45% in biogas volume caused by the ultrasonic addition. In the same paper effect of nanoparticles, the addition was also investi-

gated and the authors found that nanoparticles showed positive effects for the first week of the process but after 23 days the effect was negligible.

The research conducted by Azman S. et al. [20] focused on exploring the potential of using US disintegration as a strategy for treating manure digestate. Digestate treatment involves the application of physical, chemical, or biological methods to treat and recycle the effluents in anaerobic digesters. This process aims to enhance the utilization potential of the digester by providing supplementary feed for energy conversion. The study found that applying 20 kHz US with a specific energy input of 1500 kJ/kg TS for a duration of 30 days significantly improved the efficiency of biogas production compared to the system without US assistance. On the other hand, the continuous process set-up had a negative energy balance due to the high energy requirements of the US digester, so the operation was not sustainable at all.

A recent study by Bianco F. et al. [21] showed that biogas production from Hazelnut Shells was improved by 2.3 folds after pretreatment by 20 kHz, 200 W ultrasound system for 10 minutes in combination with hydrothermal treatment (HT). The optimum solid to liquid ratio was 1:10 (w:w), resulting in the highest COD solubilization (9526 mg/L) at 100°C at the end of 16 days of anaerobic digestion (AD). Authors have declared production of 137 mL CH₄ per g VS under the optimized US+HT+AD conditions, which is a high value for a waste of high cellulose and lignin content.

Industrial, institutional and commercial waste

This category includes organic wastes that can be used for biogas production from households, institutions and industries. In this category food wastes, municipal solid wastes, and residues of institutional and production activities can be considered.

In the research conducted by Cesaro et al. [22], the effects of sonication were investigated on both municipal solid wastes and a combination of solid waste and sewage sludge. The experiments were conducted using different sonication durations (30 and 60 minutes) and various ultrasonic densities (0.1, 0.2, and 0.4 W/mL). To ensure comparability, the total solid percentage was maintained below 10% for all tests. Notably, a significant improvement of 60% in SCOD (Soluble Chemical Oxygen Demand) was observed when sonication was performed for 60 minutes with an ultrasonic density of 0.4 W/mL. Additionally, after a period of 32 days, the biogas production from the sonicated mixture was found to be 24% higher compared to the untreated mixture. Furthermore, comprehensive analysis of full-scale studies revealed that utilizing 1 kW of ultrasonic energy for sludge pretreatment resulted in the generation of approximately 7 kW of electrical energy after accounting for losses. The system was highly energy efficient.

Joshi & Gogate [23] examined the use of the 20 kHz US for the pretreatment of food waste from a university canteen. The results showed that biogas production increased twice as much in the US-supported system as without it. After 15 days 80% COD removal was recorded at the optimum con-

ditions of 200W and 10 min, while the anaerobic digestion process without the US obtained 10% COD removal at the same conditions. As the applied power increased, biogas production increased and reached its highest point at 14800 mg/L at 0.04 W/mL power density. The authors have suggested that, due to an increase in the cell wall porosity by the US irritation, more amount of nutrients can be transferred from fermentation media to microbial cells, promoting the production of biogas.

In their research, Zeynali et al. [24] investigated the effectiveness of ultrasonic pre-treatment in improving the production of biogas from waste generated by the fruits and vegetable wholesale market. The substrate was subjected to three different sonication times (9, 18, 27 min) at a frequency of 20 kHz and an amplitude of 80 µm. The results showed that the highest methane yield was achieved with 18 minutes of sonication, while longer exposure to sonication resulted in a decrease in methane yield. This biogas production was achieved within a batch time of 12 days. Furthermore, the energy content of the biogas obtained from this reactor was twice the amount of energy input required for sonication.

Martinez-Jimenez et al. [25] conducted a study to assess the effectiveness of ultrasonic (US) treatment on the anaerobic co-digestion of sugarcane straw and vinasse substrates under thermophilic conditions (55°C) in a semi-continuous process. Vinasse is a residual liquid that remains after the fermentation and distillation of alcoholic liquors, while biohythane is a mixture of H₂ (5–20%) and CH₄ (80–95%). The researchers evaluated two different US pretreatments: one with 180 W of ultrasonic power irradiated at 37 kHz for 30 minutes, and the other with 800 W of ultrasonic power irradiated at 19 kHz for 15 minutes. They observed significant differences in the composition of biogas produced by the two pretreatments. Over a period of 42 days of hydraulic retention time, the methane concentration in the biohythane consistently increased in the first setup.

Rasapoor et al. [26] performed a study focusing on the on the solid concentration of organic fraction of municipal solid waste (4–10 %), different ultrasonic power intensities and time intervals. The outcome insinuated (*p* less than 0.01) strong effects sonication of time and power density on the final production of biogas after seventy-two hours' digestion. The applied US was 20 kHz and 400W. In conclusion the study implies that the specific input energy between 5000 and 10,000 kJ/kg TS, the optimum power intensity of 52 W/mL will effectively increase the production of biogas for 6% in 30 min. The final biogas production after US pretreatment reached as high as 440 mL/g VS after 25 days of digestion, which was 24% higher than plain operation.

In some studies, hydrogen gas production was observed instead of biogas or methane. Gadhe et al. [27] examined biohydrogen production from food waste. The substrate of organic complex was characterized by TCOD, SCOD, TS, and VS equivalent to 98600, 52900, 72500 and 51106 mg/L, respectively, while the US power and frequency for this study were equivalent to 1200W, 20 kHz. The ultrasonic pretreatment efficiency was evaluated in terms of improvement in

rate and hydrogen yield. The response surface methodology results demonstrated that when TS content and ultrasonic time were increased, so did the hydrogen yield. The ideal conditions were as follows: with TS 8% and sonication time 12 min., the best hydrogen yield was 149mL/g VS (corresponding to 1500 mL biogas/g VS approximately) and the rate was 5.23mL/h, with 104% increase compared to initial conditions.

The effectiveness of US (in combination with other pretreatment methods) on food residues, such as hazelnut skin, almond shell, and spent coffee grounds, was investigated by Oliva et al. [28] to determine their potential in terms of methane production and polyphenol and sugar solubilization using 35 kHz, 200W, at ambient temperature and 80 °C, in aqueous and methlyalcohol medium. The authors did not observe the COD or TS variations during the process, but the structural sugars and the lignins were recorded throughout the study. The results showed that the water-based liquors exhibited a higher methane potential than the MeOH-based liquors, achieving maximal 255.4 (± 7.4) and 366.2 (± 4.2) mL CH₄/g VS, respectively. Although, the highest polyphenols and sugar solubilization were obtained in the MeOH-based medium, due to the inhibition effect of MeOH in anaerobic digestion process, it resulted in low methane potential.

In India, Deepanraj et al. [29] examined the effect of three pretreatment methods (autoclave, microwave and ultrasonication) on the food waste and poultry manure. The results revealed that, better degradation efficiency (of VS and COD) and biogas yield were achieved by ultrasonication compared to others. So, the highest total biogas yield was obtained with the ultrasonication technique of 9926 mL using 20 kHz and 130 W power for 30 min. The initial VS was 41.96 mg/L. Biogas produced from ultrasonically-treated samples contained 62.47% methane and 35.07 % carbon dioxide. The ratio is an indication of high quality biogas production.

Buller et al. [30] investigated the ultrasonication of brewer's spent grains as a pretreatment prior to anaerobic digestion using 19 kHz, 800W US. The results revealed that the US pretreatment increased the production of biomethane from 29% to 56% compared to the untreated condition. Also, the pre-treated system showed higher potential in energy recovery pathways, including electrical energy and thermal energy, with 0.23 MWh/t and 1200 MJ/t, respectively, whereas system without pretreatment generating 0.15 MWh /t electrical energy and 790 MJ/t thermal energy. Further, pre-treatment with US reduces Green House Gas (GHG) emissions by up to 80% compared to anaerobic digestion alone.

Waste Sludge

Waste activated sludge (WAS) is a polluting waste that has significant negative impacts on the environment and must be managed in a manner that prevents pollution and health problems. To treat this waste, the sludge obtained from primary and secondary water treatment is collected and subjected to anaerobic digestion. Prior to being introduced into the digesters, the sludge is occasionally screened and then concentrated to achieve a dry solids content of up to 7%. This

is done to prevent excessive energy consumption for heating, which would occur if the sludge had a high water content. Additionally, this sludge has the potential to produce biogas and can also be utilized as a soil stabilizer through compost generation.

Elalami et al. [31] carried out a review on a number of studies about the production of biogas through pretreatment and co-digestion of wastewater sludge. They indicated that while co-digestion aims at enhancing methane production by adjusting several factors such as pH, moisture, the C/N ratio and nutrient availability, the efficiency is not guaranteed. The feasibility should be managed in detail about the techno-economic, out turn of the pretreatment on the sludge properties in the physical, chemical together with the digestant standard. Hence, it is very vital to make sure that the process is sustainable in fields of the legislatives as well socio environmental.

In one of the earliest studies, Tiehm et al. [32] used ultrasonic disintegration as a pretreatment for waste activated sludge with SCOD of 240 -622mg/L. The range of the ultrasonic frequency was 41-3217 kHz, with 30 – 150min application of 45 W power. Large cavitation bubbles are produced by low frequency US and when they collapse, they start strong jet streams that impose substantial shear stresses in the liquid while the smaller cavitation which prevents the beginning of strong shear forces, is thought to be the cause of the declining sludge disintegration efficiency seen at higher frequencies. Sludge floc deagglomeration was achieved with short sonication times without destruction of bacteria cells. The waste activated sludge's anaerobic digestion was substantially improved. Along with a rise in the breakdown of volatile solids (VS degraded 27.3 – 33.7%), a rise in biogas generation was observed, but the authors did not focus on this theme.

Lizama et al. [33] studied the biochemical methane potential of WAS through co-operation of US and anaerobic digestion with specific energy input, frequency and supplied US power, 5000- 35000 kJ/kg TS, 20kHz and 750 W, respectively. The authors found that the solubilization of SCOD increased from 888 to 12970 mg/L, when the energy applied changed from 15000 to 35000 kJ/kg TS. Consequently, the accumulative biogas production raised up from 501mL/gVS to 658 mL/gVS at the same level of applied energy. This corresponded to 31.43% increment in biogas yield.

Gallipoli et al. [34] carried out an experiment on the 200 kHz sonochemical breakdown of organic pollutants in secondary sludge sampled from the recycle stream before secondary clarifier in a domestic wastewater treatment plant. The sludge was gravity thickened for 24 h at 4 °C up to a total solids concentration of 21–24 g/L. The corresponding VS was about 12-14 g/L. The 90-100 W US application provided more readily biodegradable COD to the system while extra inoculum was added to the system to enhance the anaerobic digestion for biogas production. Authors have concluded that the sonication indicated better performance for VS removal at higher Substrate/Inoculum ratios, with an optimum of 0.9.

Riau et al. [35] carried out an experiment on biochemical methane potential in order to determine how ultrasonic pretreatment would affect the Temperature Phased Anaerobic Digestion of waste activated sludge. The initial characteristics of sludge were as follows: COD= 19.0 g/L, TS= 47.8 g/kg, VS= 41.0 g/kg. The ultrasonic parameters were as follows: Frequency= 24kHz, Power=400W, time= 5–30 min. The WAS was subjected to US before or after the thermophilic stage (specific energy 3380kJ/ kg TS). The results revealed that US application before the thermophilic stage was more effective where total production of methane improved by more than 50% and the elimination VS increased by 13%.

Martin et al. [36] also studied waste sludge with an initial COD of 160 g/L, TS of 132 g/L with 88 g/L of VS. The researchers sonicated the raw sludge in an ultrasonic bath at 150W for 0-15-30-45 and 60 min. The sludge digestion system had its optimum organic loading rate values of up to 4.1 kg VS/m³·d providing an increase in methane yield coefficient from 88 to 172 mL /g VS (STP, 0 °C, 1 atm) after the ultrasonic pre-treatment, in 110 h hydraulic retention time.

Algae

Biogas can be produced from sustainable biomass, especially materials that are locally available. Several substrates are considered in this category including plants and algae, which is a quickly reproducing specie in natural environments. Plant cells are completely enclosed in cell membranes and one or two cell walls which depend on the plant type. The cell wall should be broken down to obtain a source for biogas production potential. There are various types of algae species present in the nature but their common property is that they are in vast amounts especially in polluted surface waters, due to eutrophication.

In a reviewed study carried out by Rodriguez et al. [37] algae were investigated. Different pretreatment methods were taken into consideration to disrupt the structure of algae, categorizing them into mechanical, microwave and US, biological, chemical, thermal, and combined groups. Pretreatment with low energy input had a low impact on the production of biogas and methane compared to pretreatments with high energy demand. This study implied that the main technique applicable to macroalgae was mechanical pretreatment because it causes the size of the particles to be reduced from the feedstock which enhances the availability of the substrate from the lignocellulose materials, while thermal pretreatment was for microalgae which had the highest production for methane increasing with lower energy required. Almost all the pretreatment methods had the disadvantage of high energy requirements (except for biological and chemical). In another study by Rodriguez et al. [1] grassland has been investigated because it received considerable attention in recent years as a source of bioenergy due to its less need for irrigation. For instance, in the United Kingdom, there are 55 biogas plants working with grass as feedstock from a total of 913. The grass contains different amounts of sugar and fiber depending on when it is harvested. As a result, biogas production from grass can be improved by 50% or more with most pretreatment techniques. Mechanical methods increase

the improvement to about 60%. In regards to biological and chemical treatments, there is a need for new compounds and enzymes to be studied in order to increase the efficiency as well as toxicity reduction and pretreatment time shortening. Hence it is proposed that parameters for pretreatment should be taken into consideration, in order to obtain high production of biogas alongside a positive energy balance.

Microbial biomass, *Scenedesmus*, was subjected to US and thermal pretreatment methods using a batch system to improve its ability for biogas production as reported by González-Fernández et al. [38]. The US pretreatment experiment was operated at 20kHz with different energy levels at 35°C for more than 30d. While thermal pretreatment experiments were carried out by pumping hot water into the reactor. Higher energy level values had a high impact on cell wall disruption. The thermally pretreated samples at 80°C exhibited higher particle diameter (9.8 μ m) compared to the untreated sample (7.4 μ m). Additionally, SCOD increased 2.2- to 3.1-fold with increasing energy level. The methane production was much higher in sonicated samples (153 mL CH₄/g COD) than in the untreated sample of (81.8 mL CH₄/g COD) at 129 MJ/kg of specific energy level, but thermally treated biomass at 80 °C produced a lower amount.

Park et al. [39] carried out an investigation of the disruption and digestion of the single-cell microalgae (*Chlorella vulgaris*) that grows in swine wastewater. The unicellular microalgae were cultivated following the disintegration of the cell wall of the *Chlorella vulgaris* through the use of an ultrasonic homogenizer ranging from 5-200 J/mL. In a series of biochemical methane potential assays, it was found that the production of methane was limited from the batch anaerobic process due to the fact that the microalgae cell wall is very hard thereby causing high resistance to chemicals and enzymes. 20 kHz, 750 W US application showed higher soluble COD with higher energy applied. The cell disruption due to the application of US during batch digestion increased when compared to the untreated sample by 90% at 200J/ml of the energy. Addition of sludge increased the biogas production.

Lee et al. [40] did an experiment on the biomass of the filamentous algae through US pretreatment ranging from 10 – 5000J/mL (around 150W) for the enhancement production of biogas. When higher energy was applied, disruption of the cells increased, while the SCOD rose from 250 to 1000mg/L at 2500J/ml. The methane formation efficiency was improved by up to 2.3 times when the US changed the chemical composition and enhanced the substrate solubility.

El Nemr et al. [41] conducted a study in which they added sonication-modified sawdust-derived biochar into the biogas production system from the green algae *Cheatomorpha Linum* (*C. Linum*), either alone or in combination with another potential co-substrate. They discovered that the addition of biochar significantly increased biogas production. According to the results of the experiments, the best amount of biochar addition was 50.0 mg/L and increasing the amount resulted in a decrease in biogas production. With biochar, the biogas yield increased compared to the control sample which did not include the biochar, yielding 1059 and

629 mg/L VS, respectively.

Passos et al. [42] investigated the effect of US irradiation on algal solubilization, biodegradability, and methane output. US irradiation was used before the microalgae anaerobic digestion process to enhance the methane output at the specific energy range of 16-67 MJ/kg TS. A positive linear association was discovered between the applied specific energy and the methane output of microalgal biomass. The more the applied specific energy, the greater the methane output. Application of 20kHz, 70 W US increased the methane by 101%.

Cesaro et al. [43] carried out a research to evaluate how well solid organic substrates treated with US can improve anaerobic digestion yields for energy production. The substrate had TS 3.1% with VS 62.1%TS. US was applied at 20KHz, 1KW for 5-15 min, and it gave rise to the increment of production of biogas by 71% with ultrasonic energy inputs in the range of 31-93W h/L. For protein rich substrate, a smaller biogas increase in the range of 3-23% was discovered. The findings showed that the link between solubilization and anaerobic biodegradability varied greatly depending on the combination of US / anaerobic digestion process.

Alzate et al. [44] conducted research on the methane produced by microalgae, regarding the effects of biomass concentration, a substrate to inoculum ratio, and pretreatment (one being US). The authors did not give any detailed information about the frequency of the ultrasound but regardless of the microalgae studied, Substrate/Inoculum ratios between 0.5 and the microalgae concentration of 10 g TS/kg led to the highest ultimate methane productions ranging from 188 – 395mLCH₄/gVS. The ultrasound pretreatment increased CH₄ productivity by 6% - 24%, but no additional increases were seen at higher energy inputs. The outcomes obtained proved that thermal pretreatment is a better promising method than ultrasonic pretreatment for the generation of renewable energy.

A recent study by Roshni et al. [45] provided another point of view for anaerobic digestion of 2 species of algae. As SCOD efficiency it was observed that the two species had higher soluble COD at two different applied energies, where *T. suecica* had a SCOD efficiency of 5% at a specific energy of 10 MJ/kgTS, while *N. oceanica* had a SCOD efficiency of 18% at a specific energy of 27 MJ/kgTS. This could mean that with the increased SCOD efficiency, the cell wall was ruptured, and cellular material was released. However, from an AD perspective, for our study results, it could also mean that ultrasound energy was insufficient to achieve adequate cell disruption for increased specific methane production or that it resulted in the release of material that are inhibitive for AD. Therefore, additional pre-treatment could be needed to allow better digestion of the cell wall residues.

Landfill Leachate

When the organic portion of domestic wastes are stored in the landfill, leachate occurs over time. It is a high organic matter and ammonium nitrogen strength wastewater formed as a result of percolation of rain water through landfills. It is possible to obtain biogas from leachate with high organic

content. Landfills are generally designed with biogas facilities. If the landfill contains the organic part of the separated household waste, only, the organic load of the resulting leachate will be quite high and will have favorable conditions for biogas production.

A set of experiments were conducted in Burkina Faso by Nikiema et al. [46] to improve the production of biogas by combining bovine dung and leachate from municipal organic waste. The contents of VS in cow dung and leachate were 72.41 and 70.48%, respectively. The volatile Fatty Acids (VFA) to alkalinity ratio in the leachate from organic waste was high (>3600). Increase in the organic matter level, improved microbial development, and increased stability of anaerobic biomass. The biogas yield of 152mL/g VS was obtained in the combined system. Since leachate contains high organic load in liquid form, it provides great benefit in increasing the efficiency of the existing systems by enriching the content of the substrates used for biogas production.

In cases when the waste separation is not effective and the leachate has high particulate matter and non-biodegradable organics, further improvements may necessitate. Oz & Yarimtepe [47] reported that the ratio of SCOD/TCOD increased from 47% to 63% when the landfill leachate was sonicated at 600 W/L for 45 min., whereas the biogas production in the ultrasonically pretreated reactor increased by 40% (the contribution of methane production was about 60% of the total increase). The particulate COD was successfully transferred into soluble COD and the optimum conditions were as follows: reactor temperature 35 °C, the pH 7. Authors have concluded that integration of ultrasound pretreatment with anaerobic processes can be suitable to enhance anaerobic treatability of other complex industrial waste waters, as well as leachate.

Scientists investigate various analytical parameters such as COD or BOD₅ to express the organic strength of the feedstocks. Effects of 20 kHz and 12 μm ultrasonic waves were also investigated by Grosser, et al. [48], to find the optimum conditions for the improvement of biological landfill leachate treatment. Reaction time varied between 0.5 to 15 min., with an optimum of 3 min. The BOD₅/COD ratio increased 2.7 times compared to that without ultrasound application. The practice of releasing landfill leachate into wastewater treatment plants (WWTP) is widely used in numerous countries as a means of managing leachate. According to the research conducted, subjecting landfill leachates to sonication resulted in a significant increase in leachate biodegradability (up to 270%—from 0.1 to 0.3) and a decrease in toxicity towards microorganisms present in the activated sludge. As a result, the initial conditioning of leachate not only improved the quality of sewage sludge but also enhanced the overall effectiveness of its treatment process.

Nazimudheen et al. [49] have found the following findings about ultrasonic pretreatment of leachate: Leachate properties are influenced by both physical and chemical effects of sonication. Pretreatment with low power US was more suitable for anaerobic digestion. Oxidizing radicals generated in high power US treatment degrade organic matter.

Kwarciak-Kozłowska et al. [50] conducted a study on the ultrasonic disintegration of leachate using a low-frequency ultrasonic field ranging from 10 to 50 kHz and an intensity exceeding 1 W/cm². Following a 300-second ultrasonic pre-treatment at an amplitude of 14 μ m, an anaerobic granular sludge bed reactor was operated. The application of this pre-treatment resulted in a 7% increase in COD removal efficiency compared to the fermentation of nonconditioned wastewater. Consequently, the COD removal efficiency reached 88.0%. Additionally, on the 8th day of the process, there was a 2.6% increase in biogas production, with the specific methane yield in sonicated leachate being 22% higher than that in non-conditioned leachate. Therefore, this study provided that ultrasonic irradiation has a positive effect on the hydrolysis phase of refractory compounds in leachate. Hence, in 8 days of digestion time, the efficiency of SCOD removal in the sonicated leachate increased to 88% from 81.1% in the non-sonicated leachate. This was accomplished at 14 μ m with 22 kHz frequency and in 300 seconds.

RESULTS

It had been concluded that the anaerobic digestion of the substrates with high organic content is a complex process which may be challenging to obtain high biogas yields. The researchers have added auxiliary techniques to their set-ups in order to find optimal solutions to overcome the challenges. Ultrasonic irradiation technique seems to be the most promising among the others. When the operational parameters in various papers were investigated, the organic contents of the substrates were expressed with various assay parameters such as COD, SCOD, TOC, TS, or VS. The summarized table below reveals the selected common operational parameters for biogas production set-ups. The papers reviewed here have investigated the topics from different aspects. In order to increase the comprehensibility of the table, the operating parameters given with different units have been converted into common units using the data in the papers. The most important operating parameters were categorized as those related to US and those to biogas production. The substrate with the lowest initial organic content was the algae with 55-250 mg/L COD. The authors have preferred to use this bio-source together with WAS because the two substrates supported the enrichment of the nutrient content for microorganisms with high Carbon from the sludge and high Nitrogen and Phosphorus from the algae. The biogas formation was limited to 326 L/kg VS (with 230 % increase by US irradiation). On the other hand, the complex food waste had reached 1500 L of biogas production per kg of VS, with 75% increase by US irradiation. The initial COD was around 100000 mg/L. Regarding the research papers, the indicated process parameters and the resultant finding were in diverse units and it was quite difficult to compare the various techniques with each other. Table 1 shows the gathered data together and the unit conversions to bring them at a base of common understanding.

The researchers had different set-ups for anaerobic digestion

and the hydraulic retention time changed between 4 to 50 days. US practice mostly induces positive improvements in biogas production. Regarding diverse applications, the main findings were summarized below:

- 1-Increase in the applied US energy causes high COD solubilization.
- 2-Sonication time is effective in a limited way.
- 3-Probe type of sonicators are much more efficient in shorter time (30 sec. – 5min.) than ultrasonic baths (1 h.) and the provided energy is comparatively lower for the first one.
- 4-US creates changes in the rheology of the particulate portion of the substrates while solubilizing most of the substrates and leading to further biodegradation by anaerobic bacteria
- 5-The particle size of substrate is an important parameter; such that the screened substrate produces less biogas compared to ground substrate.
- 6-When the digestion temperature increases the biogas production efficiency is positively affected.
- 7-The pretreatment is preferable if the applied energy is not more than the potential energy of the biogas produced in the system.

Table 1. Summary of operational parameters from selected articles

US Related Parameters					Substrate Related Parameters			Biogas Related Parameters			
Reference Paper	Reaction Time (min)	Amplitude (μm)	Power (W)	Frequency (kHz)	TS mg/l	VS mg/L	COD mg/L	Retention Time (d)	Biogas Yield (l/kg VS)	Energy Content (kWh/kg VS)	% Increase before and after US
Industrial, institutional and commercial waste (Food waste in this categories)											
Buller et al. [30]	15	—	800	19	41000	30170	—	50	48,9	0,31	27
Joshi & Go-gate [23]	5	—	200	20	43000	27500	11500	15	—	—	100
Rasapoor et al. [26]	30	—	400	20	6000	5340	—	25	441	6000kJ/kg TS	24
Zeynali et al.[24]	18	80	100	20	18700*	14025*	—	12	396	—	80
Gadhe et al.[27]	30	—	1200	20	72500	51106	98600	—	1500	—	75
Deepanraj et al.[29]	30	—	130	20	75000	71340	69920	30	140*	—	15*
Landfill Leachate											
Oz & Yarimtepe[47]	45	—	600	20	2300	—	28500	4	477	—	40
Kwarciak-Kozłowska, et al (2005) [50]	300	14	1W/cm ²	10-50	—	—	—	8	—	—	22
Algae											
Lee et al.[40]	30	—	150	20	10000	87000	250	10	326,2	50J/mL	230
Park et al.[39]	—	—	150	20	—	49,1	55	25	366	—	65,1
Activated Sludge from wastewater treatment plant											
Lizama et al.[33]	33	—	750	20	—	—	46433	50	658	35000 kJ/kg TS	31.4
Gallipoli [34]	40	—	100	200	21300	12500	—	20	140	—	40
Livestock/Agricultural waste											
Braeutigam [13]	2	125	200	24	—	—	14550	—	393	—	41
Dong [17]	30	—	150	40	49500*	46150*	—	53	1000	—	56,6
(Castrillón et al.[12] for Screened manure	4	—	100	20	—	35000	45000	—	298.7	—	121
(Castrillón et al.[12] for ground manure	4	—	100	20	—	35000	45000	—	291.6	—	800 (w/ glyc)

*The numbers are re-calculated using the data given in the text in order to have comparable values with the rest of the table

CONCLUSION

The increasing concern for the environment has led to the widespread adoption of biogas as an alternative energy source. Biogas, being renewable and clean, offers an effective solution to combat global warming. By reducing reliance on fossil fuels like oil and coal, it helps in mitigating the negative impact on the environment. The production process of biogas involves converting waste into valuable resources, namely biogas and organic fertilizers. These fertilizers are the solid byproduct obtained after the anaerobic digestion process.

The technology used to produce biogas is categorized as part of the circular economic system, which emphasizes the reuse and regeneration of materials or products. This approach ensures sustainable and environmentally friendly production methods.

Biogas plants can be utilized even in small-scale settings, such as farms, where the waste produced by livestock can be converted into biogas. Remarkably, the waste from a single cow can generate enough energy to power a lightbulb for an entire day. In larger plants, biogas can be compressed to match the quality of natural gas and used as a fuel for automobiles.

The improvement of biogas systems is archived by various pretreatment techniques and Power ultrasound (20-100kHz) is the best alternative because it is easy to use and energy efficient. Operators can produce up to 1500 L of biogas per kg of VS in the substrate if the digesters are operated efficiently with help of US pretreatment.

DATA AVAILABILITY STATEMENT

No data was used for the research described in the article.

CONFLICT OF INTEREST

The author declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

USE OF AI FOR WRITING ASSISTANCE

Not declared.

ETHICS

There are no ethical issues with the publication of this manuscript.

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