Bioresource Technology, Volume 357, August 2022, Article number 127345
DOI: 10.1016/j.biortech.2022.127345

Energy feasibility and life cycle assessment of sludge pretreatment methods for advanced anaerobic digestion

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Abstract

Energy sustainability is one of the critical parameters to be studied for the successful application of pretreatment processes. This study critically analyzes the energy efficiency of different energy-demanding sludge pretreatment techniques. Conventional thermal pretreatment of sludge (~5% total solids, TS) produced 244 mL CH_4/gTS, which could result in a positive energy balance of 2.6 kJ/kg TS. However, microwave pretreatment could generate only 178 mL CH_4/gTS with a negative energy balance of -15.62 kJ/kg TS. In CAMBI process, the heat requirements can be compensated using exhaust gases and hot water from combined heat and power, and electricity requirements are managed by the use of cogeneration. The study concluded that <100℃ pretreatment effectively enhances the efficiency of anaerobic digestion and shows positive energy balance over microwave and ultrasonication. Moreover, microwave pretreatment has the highest global warming potential than thermal and ultrasonic pretreatments.

Keywords : Thermal pretreatment, Microwave, Ultrasonication, Methane, Energy analysis

1. Introduction

Anaerobic digestion (AD) is one of the most promising technologies for stabilizing the sludge, removing odor, and generating energy-rich methane gas and nutrient-rich digestate. However, sludge hydrolysis is the rate-limiting step in the AD process and reduces the breakdown of organics to methane. Sludge pretreatment by physical, chemical, mechanical, and biological means can effectively enhance the sludge pre-hydrolysis and, eventually, the efficacy of AD for methane production (Tyagi and Lo, 2011; Atelge et al., 2020; Wahab et al., 2020). Sludge pretreatment results in disintegrating the sludge matrix and releasing the intracellular material into the liquid phase, where soluble organics are readily available for anaerobic degradation (Pilli et al., 2015). Various pretreatments methods such as thermal (Chen et al., 2020), ultrasonication (Celebi et al., 2021), microwave (Bicakci et al., 2019), ozonation (Sun et al., 2022), freeze-thaw (She et al., 2020), ball milling (Tyagi and Lo, 2011), lysate centrifuge (Jenicek et al., 2013), high-pressure homogenizer (Fang et al., 2015), thermal hydrolysis (Yan et al., 2022), microsludge (Stephenson et al., 2005), and pulse electric field (Ozlem et al., 2021) have been studied to improve the rate-limiting hydrolysis step.

The energy feasibility of a sludge pretreatment method depends on the degree of sludge disintegration and methane production and the energy and environmental benefits associated with anaerobic digestion (Zhen et al., 2017). For instance, a thermal pretreatment process is considered energy-efficient if thermal energy recovered using methane from AD is sufficient to fulfill the energy demand of the pretreatment process. Moreover, the process is self-sufficient if the heat recovered from the exhaust gases is used to satisfy the steam requirement (Diaz et al., 2021). Due to the low-energy requirement and overall positive energy balance, thermal hydrolysis has been referred to as one of the most promising pretreatment methods (Cano et al., 2015). Various sludge pretreatment techniques can be compared using specific energy, the amount of energy utilized to treat a specific volume of sludge (Muller, 1998). The energy input mainly depends on the pretreatment method, operating conditions, sludge composition, equipment used, etc. Earlier studies on sludge pretreatment focused primarily on the performance of AD, sludge dewatering, transportation, and disposal, and have not usually considered the energy feasibility analysis (Rittmann et al., 2008; Appels et al., 2013; Cano et al., 2015; Pilli et al., 2016). Therefore, there is a prerequisite to review and analyze the energy efficacy and environmental sustainability of variable pretreatment methods, which will impact the overall performance of AD.

This work extensively reviews the energy balance of different pretreatments, namely thermal, ultrasonication, microwave (MW), ozonation, pulse-electric field, freeze-thaw, ball milling, lysate centrifuge, microsludge, high-pressure homogenizer, and thermal hydrolysis process (THP). Also, the net energy balance of the pretreatment methods such as thermal, microwave, and ultrasonication has been computed based on the data collected from the literature. To broadly analyze the upscale feasibility of the pretreatment techniques, it is also necessary to include an environmental impact assessment. To the best of the authors' knowledge, limited studies have been carried out in life cycle assessment (LCA) of sludge pretreatment technologies. Thus, substantial efforts have been made to summarize the earlier work and carry out the impact assessment of various sludge pretreatment methods.

2. Energy analysis of different pretreatment methods

2.1. Thermal pretreatment

2.1.1. Thermal pretreatment (< 100 °C)

During low-temperature pretreatment, the cell wall of a part of bacterial biomass gets ruptured. Hence, a slight increase in biodegradability has been achieved. The pretreatment of primary and waste activated sludge (WAS) below 100°C has shown positive increment in hydrolysis rate, methane production, and pathogens removal (Prorot et al., 2011; Vrieze et al., 2016). Earlier studies show that reaction temperatures have a greater effect on biogas production over reaction time (Valo et al., 2004). The energy requirement during the thermal sludge pretreatment can be calculated by using the formula given by Zupancic and Ros (2003),

$$Qs = \rho . V. Cp (t_{final} - t_{initial}) \quad [1]$$

Where,

Qs is the heat required for sludge heating (kJ), ρ is sludge density (kg/m^3), V is the sludge volume treated (m^3), Cp is the specific heat of sludge in kJ/kg °C (4.18 kJ/kg/ °C), $t_{initial}$ and t_{final} are initial and final temperature of sludge (°C), respectively.

Biswal et al. (2020) studied the effect of different temperatures of 60, 80, 100, and 120°C on sludge pretreatment with respective energy inputs of 401, 750, 1098, and 1447 kWh. At 80, 100, and 120°C, the energy output from methane was approximately two times higher than control. The energy analysis revealed that thermal pretreatment at 100 and 120°C was more energy-intensive, while 80°C pretreatment was energetically feasible. Pilli et al. (2015) reviewed the impact of thermal pretreatment on sludge biodegradability, biogas production, and dewaterability. They reported that sludge pretreatment is energetically self-sustained and produces excess energy at total solids (TS) percentage of higher than 3%. The energy ratio and net energy increase with total solids concentration. An energy ratio higher than one infers that the net energy produced is greater than the input energy provided. An energy ratio of less than one indicates that the output energy produced is less than the input energy supplied (Kavitha et al., 2019). For a sludge sample pretreated at 170^{o}C for 30 min, the net energy balance was positive and greater than 1 at the TS concentration >1.5%. Authors suggested that optimizing of the TS concentration of sludge (to be pretreated) is necessary to obtain a positive energy balance and an energy ratio greater than 1. Leite et al. (2016) compared the performance of single and two-stage digesters under thermophilic conditions. The greater electrical energy generation of 0.4 MWh/day was achieved in a two-stage system with 15% higher energy over the single-stage process. However, the energy losses from the walls of the two-phase reactor were 14% higher than the single-stage system owing to the greater surface area of earlier. The equation EPT < (0.37 *c) kWh/m^3 sludge gives the energy consumed by pretreatment using heat. The equation can check if the energy balance is satisfied, making it energetically self-sufficient after pretreating the sludge (Cano et al., 2015).

2.1.2. Thermal Pretreatment (> 100 ^{o}C)

More extensive cell rupture is caused by temperatures above 100^{o}C, which increases biodegradability and intracellular content. High-temperature pretreatment was initially used to sterilize the sludge and generate Class A biosolids (Prorot et al., 2011). However, most energy is used in water vaporization; hence it is less desirable (Ariunbaatar et al., 2014). Heat exchangers or steam injection is used to increase the temperature of the sludge during high-temperature pretreatment. The high requirement for heat energy can be compensated by the increase in sludge biodegradability and subsequent methane production, which can be used to generate heat and electricity (Ariunbaatar et al., 2014). The use of a heat exchanger could efficiently offset the costs associated with the energy requirements of thermal pretreatment. The pretreatment using higher temperature (160-170^{o}C) and pressure (600-800 kPa) also results in higher energy gain due to higher biogas production (Appels et al., 2013). Increased energy requirements can be balanced by using the residual heat of sludge to maintain the digester temperature (Haug et al., 1983). Table 1 shows the energy balance for the temperature-based pretreatment studies.

2.1.3. Microwave pretreatment

Microwave (MW) refers to the part of an electromagnetic spectrum occurring in the frequency range of 300 MHz to 300 GHz (Remya and Lin, 2011). The hydraulic retention time (HRT) of the digester can be decreased from 20 days to even five days due to significant improvement in the rate of solubilization and biodegradation with the use of microwave heating. Thus, sludge can be stabilized in smaller digesters, which would reduce the capital and operational cost significantly (Toreci et al., 2009). Although microwave pretreatment of thickened activated sludge increases biogas production, the treatment is energy-intensive (Ara et al., 2014). Banu et al. (2017) performed an energy balance analysis for microwave pretreatment of sludge. A negative net energy production of −466.02 kWh per ton of sludge was reported. Tang et al. (2010) stated that minimum microwave specific energy of 1000 kJ/kg sewage sludge (SS) is needed to rupture the cell membrane. Chang et al. (2011) noted that a longer pretreatment duration and higher microwave irradiation power could enhance the release of intracellular material. However, higher microwave irradiation power for longer duration results in higher energy consumption. Climent et al. (2007) observed that 13000 kJ/kg SS energy was needed to obtain a 311% increase in fixed volatile solids (FVS) to total volatile solids (TVS) ratio. However, only a 211% increase in FVS/TVS ratio was recorded under 7800 kJ/kg SS energy input. Specific energy of 13000 kJ/kg SS was considered the maximum energy applied in the studied conditions without sludge boiling.

Appels et al. (2013) carried out an energy balance and stated that the energy consumed by 1m^3 of sludge for microwave pretreatment would be 336000 kJ. However, it increased biogas production by 2760 L, resulting in an energy output of 57141 kJ, significantly less than the energy supplied. Hence, the net energy production is negative (-

278,859 kJ), which states that the system is not self-efficient. Kavitha et al. (2018) carried out energy analysis of microwave only and integrated ultrasonic- microwave process for sludge pretreatment. The microwave process demands 362.7 kWh energy input to achieve 20% sludge lysis. However, ultrasonic-assisted microwave pretreatment required 189 kWh of energy input to achieve the same lysis. Total energy consumed by ultrasonic- microwave process and microwave only were 425kWh and 598kWh, respectively. However, the total energy recovered was 461 kWh for both ultrasonic- microwave and microwave-only processes. The study reported an energy ratio of 0.77 and 1.08 for microwave and ultrasonic-microwave, respectively. They concluded that the ultrasonic-microwave process resulted in higher methane generation (0.3 L/g chemical oxygen demand, COD) over microwave only (0.2 L/g COD). However, microwave (2620 kJ/kgTS) results in four to fivefold greater cell disintegration over ultrasound (2370 kJ/kgTS) pretreatment (Cella et al., 2016). Increased energy consumption is one of the main disadvantages of microwave based pretreatment (Eswari et al., 2017). Table 1 revealed that low-temperature pretreatment shows a significantly positive energy balance than microwave pretreatment. For instance, when the sludge with ~5% TS was pretreated at 65^{o}C using the conventional thermal pretreatment technique, methane generation of 244 mL/gTS can be achieved. It could result in a net positive electricity balance of 2.6 kJ/kg TS. For sludge with similar TS content, microwave pretreatment at 96^{o}C has generated only 178 mL CH_4/gTS with a net energy balance of -15.62 kJ/kgTS owing to its high electricity requirement (17.51 kJ/kg TS). Thus, the low-temperature pretreatment (<100°C) positively affects the energy balance of the entire process.

2.1.4. Freezing and thawing

Freezing and thawing is a promising method of sludge pretreatment. In this method, sludge freezing occurs at around -20°C followed by thawing. Physical damage to the cells is caused by ice crystals (Vaclavik and Christian, 2008). When sludge is frozen, the tiny unfrozen regimes in the intracellular solution have been continuously dehydrated because of the extracellular ice fronts. The freeze-thaw pretreatment causes effective cell disruption and release of intracellular material into the medium (Ormeci and Vecilind, 2001). Sludge dewaterability increases and organic matter gets solubilized in the sludge matrix. The formation of recalcitrant or other by-products can also be avoided using freezing and thawing pretreatment (Hu et al., 2011).

The rate of freezing, temperature, and pretreatment time are the factors that affect the process (Hu et al., 2011; Wang et al., 2001). Freeze and thaw cycles can increase ice crystals' size, thereby promoting more sludge solubilization (Vaclavik and Christian, 2008). Wang et al. (2001) pretreated the sludge at -10°C for 24h and reported a fourfold increase in soluble carbohydrates and 25 fold increase in soluble proteins. The higher sludge solubilization was observed at -10°C compared to -80°C. Sludge volume can be reduced to one-tenth using the flotation thickened method, followed by freezing and thawing. It will result in reduced energy consumption. Freezing and thawing using natural conditions involves no energy input and enhances methane production; hence it has a positive energy balance. In another study, sludge was frozen at -25°C for 24h in a laboratory freezer, followed by thawing for 12h at 20°C. The biogas yield of the pretreated sludge was 1.3 m^3/kg VS removed. The increase in biogas production can be attributed to the increase in solubility caused by freeze-thaw pretreatment (Montusiewicz et al., 2010). This technique is restricted to cold weather conditions only when natural conditions are used. Warmer or tropical conditions need the use of artificial freezing. However, artificial freezing is not feasible because of the high energy requirements and space constraints required for sludge storage. Most studies reported the effectiveness of freeze-thaw pretreatment in sludge dewatering ability (Ormeci and Vesilind, 2001; Wang et al., 2001). Montusiewicz et al. (2010) stated that freeze-thawing pretreatment in anaerobic digestion seems to have counter effects because of its energy intensiveness.

2.1.5. Thermal hydrolysis process: Pilot and full scale experiences

Compared to the other pretreatment methods, the thermal hydrolysis process (THP) has gained interest among the scientific community and industry for the past twenty years, with a significant rise in the number of THP-based systems in wastewater treatment plants, WWTP (Abelleira-Pereira et al., 2015). During thermal hydrolysis, the energy applied in the form of heat increases the reactivity significantly. It results in the breakdown of complex molecules to produce simpler compounds (Ngo et al., 2021). Partial solubilization of the sludge and improved dewaterability take place at 150ºC to 180ºC. The disintegration of the sludge gel composition and release of trapped water happens during the process (Tyagi and Lo., 2011; Carrere et al., 2016). CAMBI and BIOTHELYS processes apply in the temperature range of 150-180ºC for 30-60 min through steam injection. Figure 1 shows the schematic representation of a WWTP equipped with thermal hydrolysis pretreatment.

The formation of inhibitory compounds and high energy demand are some of the drawbacks of the THP process. Oosterhuis et al. (2014) stated that increased energy consumption in the THP process owing to steam generation causes a negative impact on energy balance. They reported that when a THP pretreated mixture containing 60% WAS and 40% primary sludge (PS) is digested, the heat generated by the combined heat and power (CHP) process will be sufficient to generate steam for pretreatment of waste activated sludge (WAS) only. In the advanced thermal hydrolysis process (ATHP), the formation of recalcitrant compounds and energy consumption could be minimized under reduced operating temperatures (Yan et al., 2022). Barber et al. (2016) stated that increasing the percentage of dry solids (DS) will reduce the energy requirements. Sludge is usually thickened to a dry solids (DS) content of 15-18%, and additional thickening could result in heat transfer constraints. The steam consumption is affected by the feed sludge temperature, thermal difference, sludge viscosity, and thermodynamic and physical properties of the fluid.

The feed sludge temperature presents a linear response against steam requirement with a negative slope, as given below:

$$Q= -10.476\ T+ 1729 \qquad [2]$$

Where Q= Steam demand / tonnes dry sludge at %DS; T= Inlet sludge temperature (°C)

Using heat balance calculations, the effect of heat recovery within thermal hydrolysis on steam requirement is linear,

$$S= 10.85\ \Delta T \qquad [3]$$

Where S is the kg steam/tonne of dry solids processed, ΔT = internal temperature. The factor of 10.85 is relevant for a loss free system processing 60:40 primary sludge: waste activated sludge mix. Taking efficiency losses into account, the steam demand can be expressed as:

$$S = \frac{10.85\Delta T}{911\ \eta} (134*DS^{-1:05}) \qquad [4]$$

S= Steam requirement (kg/metric tonne), ΔT internal temperature difference (°C), system efficiency, dry solids (DS) of sludge entering thermal hydrolysis expressed as a decimal.

The energy benefit of the THP process is relatively neutral because the surplus biogas produced after the pretreatment is partly used in generating reaction temperature for sludge pretreatment. A significant energy benefit from the technology is the improved dewaterability of the digested sludge. Pérez-Elvira et al. (2008) conducted an energy balance analysis using a different configuration of thermal hydrolysis and anaerobic digestion. It was suggested that the feed sludge TS concentration must be 7% to produce enough biogas to make the system self-energy-sufficient. Moreover, the energy recovery from the flash vapor outlet of the reactor, exhaust gases, and hydrolyzed sludge can reduce the energy demand of pre-heating the feed sludge. Thermal hydrolysis of waste activated sludge is energetically beneficial over a mixture of WAS and primary sludge. A 30% higher biogas production can be obtained from WAS, which generates 30% more electrical energy. Polanco et al. (2008) designed a thermal hydrolysis pilot plant and operated it in batch mode to study the effects of sludge type, temperature, solids concentration, and residence time. The optimal pretreatment conditions were observed as 170°C, and 30 min, which resulted in a 50% enhancement in methane production. During continuous operation, the biogas production increased by 40-50%. The increase in biogas production led to 40% more electrical energy and an energy self-sufficient system. Heat requirements of the thermal hydrolysis pretreatment can be offset by using exhaust gases and hot water from combined heat and power, CHP (cogeneration), to pretreat the sludge. Without a heat integration arrangement, the process could not achieve a positive energy balance (Cano et al., 2015). According to Taboada-Santos et al. (2019), volatile solids (VS) load and bio-methanation are the two main factors determining the total energy produced in an anaerobic process. The total energy produced is given by:

$$Et = V.S.L * BMP * \Delta Hc \quad [5]$$

Where VSL is the Volatile solids loading kg VS/m^3 sludge, BMP is the biomethane production m^3(N) CH_4/kg VS, E_T is the total energy produced kWh/m^3 sludge.

Considering a heat of combustion of 11 kWh/m^3 CH_4 and an electrical efficiency of 0.35, the net electrical energy produced is given by the difference between the energy produced by the pretreated and fresh sludge.

$$\Delta E_{elec} = VSL * (SMP_{pret} - SMP_{fresh}) \Delta Hc. H \quad [6]$$

Rather than the increase in energy production (maximum savings of 35,000–60,000 €/year), the main impact of thermal hydrolysis is mainly due to sludge disposal savings (270,000–430,000 €/year for 500,000 inhabitants WWTP). The payback period could be 2 to 4 years for a WWTP (1,000,000 inhabitants) and 15-30 years for a 1,00,000 inhabitants WWTP. It indicated higher profitability in large WWTPs installed with thermal hydrolysis unit and it was also concluded that the minimum total solids concentration of 1-2% is needed to reduce the operational costs (Taboada-Santos et al., 2019).

2.2. Mechanical pretreatment

Usage of mechanical pretreatment methods can degrade the complicated structure of the sludge by using shear stress, high pressure, and centrifugal forces. Compared to thermal pretreatments, mechanical methods have multiple advantages of no byproduct formation and minimal energy requirements (Muller, 2004). Disintegration by mechanical methods destroys the floc structure and increases the number of colloidal particles. Increased release of organic material and a high degree of disintegration with optimal energy consumption is necessary for the practical implementation of the method in AD (Lehne et al., 2001).

2.2.1. Ultrasonication

Ultrasound pretreatment using low energy input has been an effective tool for enhancing sludge solubilization and subsequent biogas production (Dhar et al., 2012). Mechanical disruption of cell structure and floc matrix takes place under ultrasonic pretreatment. Cavitation under low frequencies, hydrodynamic pressure and chemical reactions due to the formation of hydroxyl radicals (OH•, HO2•, H•) are the fundamental mechanisms behind ultrasonic treatment (Tyagi et al., 2014). The energy input, ultrasonic frequency, and substrate type are the main factors that influence the performance of ultrasonic pretreatment (Bougrier et al., 2005). The specific energy input depends on the ultrasonic power, sonication time, TS concentration, and sludge volume. According to Bougrier et al. (2005), the specific energy input can be calculated by using the following equation,

$$SE = \frac{P*t}{V*TSS} \quad [7]$$

According to Bougrier et al. (2005), the biogas production was improved upon increasing the applied specific energy from 0-7000 kJ/kg TS. However, further increment to 15000 kJ/kg TS resulted in no notable improvement in biogas production. The methane production of 325 mL/g COD_{added} was observed over control (221 CH_4/g CODadded) at 6250 kJ/kg TS. No significant change in methane production (334 CH_4/g COD_{added}) was observed at the increased specific energy input of 9350 kJ/kg TS. On the other hand, Bougrier et al. (2005) observed no improvement in biogas production over control at a specific energy of 1000 kJ/kgTS. However, a 40% improvement in biogas production was observed at a higher specific energy of 14000 kJ/kg TS. Celebi et al. (2021) stated that the sCOD concentration of WAS has been significantly improved with ultrasonic pretreatment. For a specific energy input of 12930 kJ/kg TS, a 32% increase in methane production was observed over control. However, a negative energy balance was reported despite the enhancement in methane production. The authors suggested that the implication of partial stream sonication at a full-scale system could improve the energy balance.

Pilli et al. (2016) reported that the energy input of the sonicated WAS (31 gTS/L) was 1907 kWh/mg of dry solids, while the energy output was 1915 kWh/mg of dry solids, resulting in the energy ratio (output/input) of 1.0. A comparison of sonicated mixed, primary, and WAS revealed that maximum net energy of 7.9 kWh/mg TDS was achieved for sonicated WAS. The primary sludge contains fiber, inert and inorganic materials, while WAS mainly has organic matter. It leads to higher biogas production over primary sludge, and hence the positive energy balance was achieved. The authors concluded that net energy was positive for sonicated WAS with an energy ratio of 1.0. However, the net greenhouse gas (GHG) emissions were higher than the control. Braguglia et al. (2015) stated that the energy applied had a greater influence on organics solubilization. The protein solubilization was also observed to increase significantly by increasing the sonication energy. Similarly, COD solubilization was increased from 10% to 90% by increasing the specific energy from 1000 kJ/kg TS to 100000 kJ/ kgTS (Bougrier et al.,2005, Lehne et al., 2001, Muller, 1998). During ultrasonic pretreatment, a minimum specific energy of 1000 kJ/kg TS is necessary to break the sludge flocs (Bougrier et al., 2005).

Dhar et al. (2012) stated that combined thermal-ultrasonic pretreatment (119000 kJ/kg TSS) increased the volatile fatty acids (VFA) concentration by 230% over control. Moreover, the soluble proteins and carbohydrate concentrations were increased by 2.8 and 4.5 folds on increasing the specific energy input from 1000 to 10000 kJ/kg TSS. The integrated thermal-sonication pretreatment resulted in better organics solubilization, VFA production, and biogas generation over thermal and sonication pretreatment alone. A 30% increase in methane production was obtained using the combined pretreatment at 90 ^{0}C- 30 min with a sonication energy input of 10000 kJ/kg TSS. Salsabil et al. (2010) compared the energy requirements of ozone and ultrasonic pretreatment and stated that the ultrasonic treatment is energetically costly. However, the digestion time can be reduced. As per Table 2, ultrasonic pretreatment shows a negative energy balance for sludge with 2-4%TS content owing to its high electricity requirement. During the ultrasonic pretreatment, two energy conversions are carried out, electrical to mechanical energy vibration, and further mechanical energy into cavitation. It leads to significant energy losses while electricity is required for pretreatment instead of heat generation (Pérez-Elvira et al., 2010).

2.2.2. High pressure homogenizer

High-pressure homogenization (HPH) is one of the well-investigated mechanical methods of sludge disintegration (Zhang et al., 2012a). Because of its easy operation, high energy efficiency, and low investment, HPH has been used in large-scale implementations over recent years. Under a high-pressure homogenizer, sludge pressure is increased to 900 bar, after which the sludge goes through a homogenization valve (Muller, 1998). The kinetic energy is produced because of the energy applied to the homogenizer valve, which further disperses into the liquid. This energy creates turbulence in the liquid phase resulting in the formation of eddies. These eddies result in the disruption of sludge flocs and microbial cells (Doulah et al., 1975). Homogenization pressure, cell concentration, and the number of homogenization cycles are the crucial factors that influence cell disruption during the HPH process (Middelberg et al., 1991). The energy input for a high-pressure homogenizer depends on the homogenization cycle number (N) and pressure (P, Pa). According to Anand et al., (2007), the energy consumption per unit sludge volume (Ev, J/m^3) can be formulated as,

$$Ev = P * N \quad [8]$$

Further, the specific energy consumption (Es, kJ/kg TS) shall be given as,

$$Es = \frac{Ev}{Tso * 1000} \quad [9]$$

Where, Ev is the energy consumption per unit sludge volume and Ts_o is the total solid concentration of the raw sludge.

Zhang et al. (2012b) investigated the energy efficiency of the high-pressure homogenizer (HPH) process. They found that more effective sludge disintegration can be obtained using higher energy input with higher total solids (TS) concentration. For the sludges of 10, 15, and 25 g/L TS concertation, the energy consumptions of 8450, 5351, and 3252 kJ/kg TS were needed to achieve the highest sludge disintegration degree of 25, 23, and 17%, respectively. The energy consumptions by ultrasonication and microwave processes were 18000 and 16000 kJ/kg TS, which were 236 and 395% higher than the HPH process, respectively (Ahn et al., 2009). Under similar operating conditions, HPH pretreatment of sludge with higher TS is more energy-efficient than ultrasonication and microwave. Nabi et al. (2020) pretreated the sludges of different TS content (1.0%, 1.5%, and 2.5%) with the energy efficiencies of 46.92, 55.31, and

77.18 g/MJ to achieve maximum COD solubilization at 30 MPa. An increment in homogenization pressure from 20 to 80 MPa resulted in significant sludge disintegration. However, the process was energy-intensive. An increase in cycles increases sludge disintegration; however, the cycle number needs to be optimized since the process is energy-consuming (Zhang et al., 2012a). Onyeche et al. (2003) observed that the energy consumed during the HPH pretreatment was lower than the energy produced, making the net energy balance positive. The HPH pretreated sludge showed positive energy of 790 and 510 kJ/kg TS compared to the control (290, 180 kJ/kg TS) at homogenization pressures of 10 and 20 MPa, respectively. Digester Volume and sludge digestion time can be significantly reduced with HPH pretreatment. Nevertheless, the high energy input can be compensated with the increased biogas production (Nah et al., 2000). Zhang et al. (2012a) investigated the effect of two homogenization cycles at 40 MPa pressure. They reported that the energy consumption of 3380 kJ/kg TS is lesser than the energy consumed by ultrasonication and microwave pretreatment. In other studies, energy consumption by the HPH process was higher than hydrothermal pretreatment; however, lower than the ultrasonic pretreatment (Cano et al., 2015; Zhang et al., 2012a). According to Cano et al. (2015) the energy consumed by a pretreatment using electricity is given by:

$$EPT < (0.20 * c)\ kWh/m^3\ \text{sludge} \quad [10]$$

The equation can be used to check if the energy balance is satisfied, and making the pretreatment energetically self-sufficient.

2.2.3. Ball mills

In Ball mills pretreatment, high critical tension is used to rupture the bacterial cells in the sludge. The diameter of the bead is one of the influential parameters in operating a stirred ball mill. Decreasing the bead size would result in less energy requirement (Lehne et al., 2001). Another critical parameter determining the energy consumption is the stress intensity obtained by multiplying the specific energy consumed by a single stress event and the number of stress events. Because the stress intensity is too low, very low cell disruption occurs despite high energy consumption. However, if the stress intensity is too high, a large amount of energy is consumed for a single stress event which may be higher than the energy needed for disruption (Lehne et al., 2001). Compared to ultrasonic and thermal pretreatments, the energy consumption in ball milling pretreatment is lower. Lee et al. (2010) reported that the ball milling process utilizes specific energy of 75800 kJ/ kg TSS to increase the soluble COD from 2000 mg/L to 9000 mg/L and TS from 1% to 4%. According to Lee et al. (2010), the energy input of a ball mill pretreatment can be determined by the following formula:

$$E\ (kJ/\ g\text{-}TSS) = P * T/\ TSS * V \qquad [11]$$

Where P is the power, V is the volume of sludge treated, T is the operation time. For similar COD solubilization, ultrasonic and thermal pretreatment utilizes higher energy over ball mills pretreatment (Muller et al., 1998). However, the full-scale application of ball mills is not considered energy efficient since the surplus energy recovered from enhanced methane production is negated by the energy utilized in the pretreatment. It leads to a negative net energy balance.

2.2.4. Lysate centrifuge

In the lysis centrifuge method, a centrifuge with a unique disintegrating device is installed to achieve partial disintegration (10-15%) of excess sludge and enhanced biogas yield of 15-26% (Zabranska et al., 2006). Muller et al. (2004) reported that the lysate centrifuge contributed to a slight increase in sludge degradation and resulted in lower energy demand. Sludge disintegration is often proportional to the energy applied. However, integrating the disintegrating device in a typical thickening centrifuge increases specific energy consumption by 216 kJ/kg TSS (Fabregat et al., 2011). Jenick et al. (2013) estimated the energy consumption and production from a full-scale WWTP in Prague, Czech republic. The thickening centrifuge was upgraded to a lysate thickening centrifuge. The total suspended solids (TSS) concentrations were increased using the centrifuge. Also, the operating temperature of the digester was increased to 55ºC. It was observed that 42% of the COD was converted to biogas, which was exceptionally high. The major fraction of the total energy demand of the WWTP can be covered by enhanced biogas production. The efficiency of electricity production from the plant was 31%. Zabranska et al. (2006) studied the use of a lysate thickening centrifuge in three full-scale WWTPs. Firstly, the Czech republic WWTP, with a capacity of 100000 PE, was installed with two anaerobic digesters of 4400 m^3 capacity, each having a disintegrating device mounted into the centrifuge (3140 rpm). The increase in the annual biogas production was around 217585 Nm^3. The process can be used to achieve a TS concentration of 9-11%, which further reduces the volume of sludge to be fed into the digester. Secondly, WWTP treated wastewater from 70000 PE, fitted with a centrifuge with a rotating speed of 2250 rpm, was investigated. Sludge disintegration degree of 8.5-10.7 % was reported. Thirdly, WWTP (Germany) treating wastewater from 650000 PE was investigated. The facility is installed with four digesters having a total volume of 20000 m3. Also, a sludge disintegrating device has been installed with two thickening centrifuges. The process resulted in enhanced biogas production of 1128 m^3/day, which led to an increased power generation of 2410 kWh/day. The authors concluded that installing a sludge disintegration device with a thickening centrifuge significantly enhances the biogas production, reduces the sludge volume to be disposed of, and results in a positive net energy balance of the whole system. An investigation was carried out to compare ball milling, ozonation, sonication, and lysate centrifuge for energy demand and sludge degradation efficiency (Muller, 2000). It was reported that the energy demand was in the order of lysate centrifuge < stirred ball mill < sonication < ozonation. The increase in sludge degradation was in the order of ozonation > stirred ball mill > sonication > lysate centrifuge. When comparing all the mechanical pretreatment methods, the lowest energy consumption was shown by lysate centrifuge and stirred ball mills, while an ultrasonic homogenizer showed the highest energy consumption.

2.2.5. Pulse electric field

A pulsed electric field (PEF) directly affects the basic building blocks of the cell membrane and cell walls. It also attacks phospholipids and peptidoglycans. These molecules exert a net negative charge on the cell’s outer surface (Bruce et al., 2008). The polar and charged nature of the cell membranes makes them susceptible to strong electric fields. Focused pulse (FP) technology is a modification of PEF, which ruptures cell membranes, cell walls, and macromolecules by using a high voltage electric field (20-30 kV). PEF provides reduced energy consumption than other pretreatment methods (Rittmann et al., 2008). PEF also reduces the pretreatment time compared to chemical-based pretreatment methods (24 h to 30 min). Cano et al. (2015) stated that the electrical energy consumption in ultrasound, ozone, microwave, and high-pressure homogenizer pretreatment processes is higher than in the PEF processes. Salerno et al. (2009) investigated the effects of treatment time and applied voltage of the PEF method on sludge solubilization. The pretreatment increased the COD solubilization by three times compared to the control, which resulted in an 80% increase in methane generation. The authors further concluded that optimizing pretreatment conditions would lead to greater solubilization with the least energy input. Bruce et al. (2008) conducted a full-scale study using focused pulse technology. They stated that the energy consumption in pretreatment could be compensated in two ways: (1)

additional methane production that can be used to generate heat and power, (2) as the pretreatment increases the input temperature of the sludge, the need for an external heat source can be avoided. They also stated that the energy recovered for a full-scale system treating 380 m^3/day of primary + WAS could be used for treating 95% of the feed sludge. The energy benefit and heat recovery were approximately eighteen times higher than the energy needed for pretreatment. The study concluded that no heat was recovered. The gross energy recovery ratio could be 2.7 times because of the 60% enhancement in biogas generation caused by pretreatment.

2.3. Chemical pretreatment

2.3.1. Ozonation

The ozone disintegrates the microbial cell wall due to its strong oxidant nature with high disruption capability (Yan et al., 2009). Exposure of sludge to highly oxidative conditions ruptures the cell wall, thereby releasing soluble COD. Smaller molecular weight compounds are produced because of the reaction of ozone with proteins, polysaccharides, and lipids (Goel et al., 2003). Mineralization of the released cellular compounds can also occur under high ozone dosages. Bougrier et al. (2006) stated that 300 mL of biogas/g COD added can be produced in 15-18 days with the ozone dosage of 0.10–16 g O_3/g TS. However, the process took around 24 days without ozonation. Boehler and Siegrist (2006) conducted an energy balance study for a Swiss wastewater treatment plant (35000 PE). The energy required for ozonation and to generate liquid oxygen was 12.5 kWh/kg O_3, and 0.5 kWh/Nm^3 O_2, respectively. The 30% sludge reduction increased the plant's energy consumption by 20%. Hodaei et al. (2021) investigated the effect of ozonation on sludge (WAS) solubilization and methane production. During the study, the energy analysis of the ozonation was also investigated to realize the process's sustainability. The energy consumed in ozonation was calculated as:

$$E_{ozonation} = W \times t \quad [12]$$

Where W, is the ozone generator power in watt and t is the pretreatment time in seconds.

The energy produced during anaerobic digestion was calculated by using the ideal gas law, Firstly methane density was calculated by using the following formula:

$$\rho act\ CH4 = \rho std\ CH4 * \frac{Pact}{Pstd} * \frac{Tstd}{Tact} \quad [13]$$

Where $\rho^{act}\ CH_4$ is the density of methane at standard temperature and pressure 0.72 kg m^3, Tstd is the standard temperature (273 K), Pstd is the standard pressure (101.325 kPa), the Tact is the gas temperature (303 K), and Pact is the total pressure equal to the gauge pressure plus air pressure. The total biogas energy produced was calculated by the equations:

$$Hu.act = \frac{VCH4}{Vtotal} * \rho act\ CH_4 * Hu,n \quad [14]$$

$$E_{biogas} = Hu.act \times V_{biogas} \quad [15]$$

where Hu,n is the normal calorific value of biogas equal to 50,000 kJ/kg, Hu,act is the actual calorific value of given biogas (kJ/kg), and Vbiogas is the biogas volume (L).Finally, the energy balance was calculated by using the equation [16]:

$$\text{Energy balance} = E_{ozonation} - E_{biogas} \quad [16]$$

The authors concluded that the sludges ozonated with 0.05 gO_3/gTS and 0.1 gO_3/gTS dosage could provide only 38 % and 29 % of the input energy, respectively. The net energy balance of the overall process was negative, stating that the ozone pretreatment method has higher energy demand. However, it results in increased release of soluble organics and improved sludge dewaterability. It was concluded that the biogas production reduced and the energy demand increased at high ozone dosage. The higher ozone dose promoted the hydrolysis, which resulted in the VFAs accumulation and inhibited the performance of methanogens under acidic pH. A study by Kannah et al., (2017b) stated that dispersion induced ozonation resulted in a positive net energy 152.65kWh/ton when compared to ozonation alone (−12.42kWh/ton). Thermochemical pretreatment demands an energy input of 1450 kWh to achieve a COD solubilisation of 30%. However, a combination of thermochemical and ozonation pretreatment led to the energy use of only 607 kWh. Hence, combination of ozonation along with other pretreatment methods will result in reduced energy input (Kannah et al., 2017a)

2.3.2. Microsludge

Microsludge is a combination of chemical and pressure pretreatment that causes a significant change in the extent and rate at which sludge is degraded in an anaerobic digester. The technique can result in rapid VS destruction with a higher degree of completion. During the microsludge process, alkaline pretreatment is used to weaken cell membranes, and a sudden change in pressure is exerted to burst the cells. The process requires significant energy for sludge solubilization under extremely high pressure of 12,000 psi (Saha et al., 2011). Stephenson et al. (2005) investigated the microsludge pretreatment process at a full-scale WWTP treating municipal wastewater from 70000 PE. The WWTP has two anaerobic digesters with working volumes of 1325 and 715 m^3. During the full-scale demonstration, sludge was transferred to a chemical conditioning tank, which was processed later in a homogenizer (12,000 psi), followed by mesophilic digestion. The energy analysis shows that 185% (1420 kWh) of the electrical energy required by the process can be recovered using the electricity generated from methane produced using the microsludge processed sludge. Also, 1650 kWh can be recovered as heat from the methane generated. Overall, the study concluded that 2075 kWh/dry tonne sludge of heat and 915 kWh/dry tonne sludge of electricity could be recovered using a full-scale microsludge-based anaerobic digestion system.

3. Life cycle assessment

Life cycle assessment (LCA) is a technique for assessing the environmental impact of activities with or without human interference. LCA has many advantages over other environmental assessment tools like material and substance flow analysis. It provides a systematic assessment of the product based on new information and scientific advancements and quantification of emission effects (Torabi and Ahmadi, 2020).

3.1. Life cycle assessment methodology

LCA is governed by ISO 14040:2006 standards, which define the necessary principles, framework, and guidelines (International Organization for Standardization, 2004). The framework includes four stages: goal and scope, inventory analysis, impact assessment, and interpretation. The functional unit (FU) is an essential element of the LCA, which helps define the scope of the study. It is a qualitative measure of the output function of the studied system, and helps in creating a benchmark or reference point for comparing different product inputs and output (Ding et al., 2021). Earlier works related to sludge pretreatment generally used mass/volume-based FUs. Volume-based FU is most common in the case of LCA of wastewater treatment (Corominas et al., 2013). In contrast, it is mass-based in the case of sludge pretreatment. After defining the FU, a rigorous definition of system boundaries is needed, significantly impacting the LCA results (Finnveden et al., 2009). Most LCA studies of sludge pretreatment include all sludge management processes like sludge thickening, sludge digestion, dewatering, and disposal except for treatment plants' construction and demolition stages.

However, some studies suggest that construction and transportation contribute significantly to environmental damage in the case of sludge management (Ding et al., 2021). Commonly used life cycle inventory (LCI) databases for wastewater treatment include the ecoinvent database and database provided by software like SimaPro and GaBi. Further, the LCI results are processed and generalized as environmental impacts (Nakakubo et al., 2012). The typical impact categories, which are considered for sludge pretreatment are global warming potential (GWP)/climate change (CC), stratospheric ozone depletion (SOD), ionizing radiation (IR), fine particulate matter formation (FPM), terrestrial acidification (TA), freshwater eutrophication (FE), and fossil resource scarcity (FSC)/ natural resources consumption (NRC), and human toxicity potential (HT)/ terrestrial ecotoxicity (TE) (Ding et al., 2021). Some of the standard impact characterization models are Eco-indicator 99, EDIP2003, CML 2001, IMPACT2002+, TRACI, and ReCiPe (Dong et al., 2021).

3.2. Life cycle assessment of sludge pretreatment

The LCA of energy and nutrient recovery in sludge management is gaining considerable attention, leading to a need to study closed-cycle sludge management, including sludge pretreatment methods, nutrient and other value-added products recovery strategies, and sustainable sludge disposal. Table 3 summarizes the LCA of previously studied pretreatment techniques, including ultrasonic, thermal hydrolysis, freezing and thawing, ozonation, and pressurize-depressurizing process. Carballa et al. (2011) conducted the LCA analysis of various pretreatment using operational performance data of lab-scale works with system boundaries. Based on their environmental feasibility, they recommended chemical (alkali, acid) and pressurize-depressurize processes. Moreover, ozonation, freeze-thaw, and thermal methods are not recommended owing to their adverse environmental impacts. Moreover, ozonation, freeze-thaw, and thermal methods are not recommended owing to their adverse environmental impacts. Mills et al. (2014) conducted LCA on sludge pretreatment. They concluded a need for a detailed LCA of pretreatment of full-scale operations to understand the impacts on operational economics, energy balance, and environmental health. They suggested that integration of THP with AD improves the environmental and economic benefits over conventional anaerobic digestion only. The electricity generation from biomethane and feeding to the grid is financially lucrative but causes substantial environmental damage. Li et al. (2017) performed LCA on sludge with or without pretreatment and compared their normalized impact factors. Among the processes studied, thermal hydrolysis pretreatment (THP) increases biogas production significantly and provides better environmental performance.

Moreover, energy productivity related to the organic fraction of sludge and biogas yield is considered the most sensitive factor, which defines the assessment outcomes. Mainardis et al. (2021) performed a detailed LCA on pretreatment techniques, which showed that ultrasonication has variable impacts on lab-scale and full-scale applications. The authors revealed that sludge composition played a crucial role in choosing the best pretreatment technology. Low-temperature thermal pretreatment was the best technology among others considering energy recovery. At the lab scale, ultrasonication shows the high environmental impacts due to energy-intensive operation. However, ultrasonication offers low environmental impacts at full-scale operation over thermal pretreatment due to the latter's heat and chemical requirements.

An LCA is performed using the data published earlier to understand the potential impact of the pretreatment process on the environment. The LCA had a system boundary limited to the pretreatment unit (such as thermal, microwave, and ultrasonic pretreatment), with the functional unit of 1 kg of total solids. The ReCiPe 2016 Midpoint (H) approach is applied to understand the impact of sludge pretreatment. The inventory data collected for net heat is supplied from coal or natural gas, while the Indian electricity grids are assumed for net electricity. The LCA results are presented in Figure 3, which shows the global warming potential (GWP) of different pretreatment technologies. The carbon footprints of various Indian grids vary between 0.7-1.7 CO_2eq/kWh (Hossain et al., 2019). In Figure 2, the error bars show sensitivity to carbon footprint by the Indian electricity grid. The global warming potential data for different pretreatment techniques have been tabulated and given in supplementary data (see supplementary material).

In contrast, different bars show the GWP of pretreatment utilizing coal and natural gas (for heating) with the Indian grid. The LCA findings of this study reveal that thermal pretreatment with total solids ranging from 4.8-5.8 % and temperatures around 35°C-65°C has less global warming impact than microwave pretreatment. The difference in effects is because microwave pretreatment uses a large amount of electricity compared to thermal pretreatment.

Further, thermal (190°C, 1.45% TS), microwave (190°C, 4.1% TS), and ultrasonic (1.9 kW/L, 3% TS) processes have high global warming impacts in their respective categories. The effects of the above three pretreatments on different impact categories of stratospheric ozone depletion, freshwater eutrophication, fossil resource scarcity, and terrestrial ecotoxicity are shown in Figure 3. The impact categories of different pretreatment techniques have been given in supplementary data (see supplementary material). The natural gas (for net heat) with an average of various Indian grids (net electricity) were considered to calculate the impacts of the three pretreatment methods. Results stated that microwave pretreatment causes more global warming impact than thermal pretreatment, requiring a relatively high amount of electricity to produce microwaves. In some cases, the effect of thermal pretreatment shows negative emissions as the net electricity generated is exported.

4. Research needs and perspectives

Even though thermal pretreatment at low temperature (<100oC) shows an increase in substrate biodegradability and methane generation, optimizing pretreatment temperature and time could be a critical factor due to the formation of Maillard reaction byproducts above 150^{o}C. It needs to be investigated thoroughly together with the mechanism of the recalcitrant formation. In CAMBI based thermal hydrolysis process, the heat requirements shall be compensated by using exhaust gases and hot water from combined heat and power (CHP) system, and electricity requirements are managed by the use of cogeneration. However, the refractory organics formed during thermal hydrolysis may have adverse effects when the concentrated sludge is returned to the wastewater headworks. Even though higher solubilization rates are achieved, pretreatments that use electricity (microwave, ultrasonication, etc.) may not be able to meet their energy demand from the increased biogas production in the same process. Hence, there is a need for systematically assessing the pretreatment options to decide the best one from an industrial point of view. Previous studies have used the combination of different pretreatment techniques. However, it does not always result in a direct additive effect on biodegradation; rather, it could increase energy consumption (Sahinkaya et al.,2015). The excessive use of energy input during pretreatment may lead to the production of inhibitory byproducts that may result in reduced AD process performance. In energy calculations, it is to be made sure that the actual energy (heat/electricity) supplied by the pretreatment equipment and, in the case of chemical-based techniques, the energy spent in manufacturing the chemicals all are needed to be taken into account. It is necessary to perform a life cycle assessment study to choose alternatives and minimize the adverse impacts of a pretreatment technique. However, studies on LCA of sludge pretreatment techniques are mostly missing in the literature, which could be a lucrative topic for future research.

5. Conclusions

Low-temperature pretreatment reduces electrical energy consumption and would result in a positive energy balance. Microwave increases the electricity demand and is not feasible for full-scale implementation. In freezing and thawing, a positive energy balance is only possible if natural freezing is performed, which is not possible practically throughout the year. THP would result in a positive energy balance with implementation of a CHP. LCA study revealed that microwave pretreatment results in higher global warming potential, thereby causing negative impacts on the environment.

E-supplementary data

E-supplementary data for this work can be found in e-version of this paper online.

Acknowledgment

Authors are thankful to Department of Biotechnology-GoI (Grant No. BT/RLF/Re-entry/12/2016) and Department of Science and Technology, GoI (Grant No. DST/IMRCD/India-EU/Water Call 2/ SARASWATI 2.0/2018/C) for financial support to this research.

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Table 1. Energy analysis of various thermal pretreatment methods

Pretreatment type	Conditions	TS (%)	VS/TS	Methane generation (L/kgTS)	Heat Consumed (kJ/kgTS)	Electricity Consumed (kJ/kgTS)	Heat generated (kJ/kgTS)	Electricity generated (kJ/kgTS)	Net heat Balance (kJ/kgTS)	Net electricity Balance (kJ/kgTS)	References
Thermal	Control	1.23	0.8	101	0	0	1430	1070	1430	1070	Kim et al., 2013b
	60ºC	1.23	0.8	136	16990	0	1940	1460	-15050	1460	
	75ºC	1.23	0.8	166	22090	0	2360	1770	-19730	1770	
	90ºC	1.23	0.8	124	27190	0	1760	1320	-25430	1320	
Thermal	Control	4.74	0.73	101	0	0	1430	1070	1430	1070	Ruffino et al., 2015
	80ºC	4.74	0.73	131	6170	0	1860	1390	-4310	1390	
	90ºC	4.74	0.73	133	7050	0	1890	1410	-5160	1410	
	Control	3.82	0.7	117	0	0	1660	1240	1660	1240	
	70ºC	3.82	0.7	142	6570	0	2010	1510	-4560	1510	
	80 ºC	3.82	0.7	139	6570	0	1980	1480	-4590	1480	
Thermal	Control	5.8	0.78	225	0	0	3190	2390	3190	2390	Bolzonella et al., 2012
	65 ºC	5.8	0.78	244	3960	0	3470	2600	490	2600	
Thermal	Control	4.8	0.81	164	0	0	2330	1750	2330	1750	Wang et al., 2014

	35 ºC	4.8	0.81	164	2180	0	2330	1750	-150	1750	
	55 ºC	4.8	0.81	167	3920	0	2370	1780	-1550	1780	
	70 ºC	4.8	0.81	175	5230	0	2480	1860	-2750	1860	
Thermal	Control	1.45	0.81	211	0	0	3000	2250	3000	2250	Bougrier et al., 2007
	135ºC	1.45	0.81	237	36030	0	3360	2520	-32670	2520	
	190ºC	1.45	0.81	264	51890	0	3760	2820	-48130	2820	
Thermal	Control	2.54	0.69	62	0	0	880	1670	880	1670	Ge et al., 2011b
	70ºC	2.54	0.69	117	9870	0	660	1250	-9210	1250	
Microwave	Control	5.14	0.7	134	0	0	1910	1430	1910	1430	Coehlo et al., 2011
	96ºC	5.14	0.7	178	0	17510	2520	1890	2520	-15620	
Microwave	Control	4.09	0.77	189	0	0	2680	2010	2680	2010	Chi et al., 2011
	190ºC	4.09	0.77	234	0	38290	3320	2490	3320	-35800	

*****Only the energy required for pretreatment was considered

*(-) sign indicates that the energy balance is negative.

Table 2. Energy balance of ultrasonic pretreatment of sludge

Pretreatment conditions	TS %	VS/ TS	Methane generation (L/kg TS)	Heat Consumed (kJ/kgTS)	Electricity Consumed (kJ/kg TS)	Heat generated (kJ/kg TS)	Electricity generated (kJ/kg TS)	Net heat Balance (kJ/kg TS)	Net electricity Balance (kJ/kg TS)	Reference
Control	2.8	0.68	116	0	0	1640	1230	1640	1230	Braguglia et al.,
0.5 kW/L	2.3	0.68	163	0	2500	2320	1740	2320	-760	2015
Control	4.2	0.83	183	0	0	2600	1950	2600	1950	Cella et al., 2016
1 kW/L	4.2	0.83	192	0	2370	2720	2040	2720	-330	
Control	3.0	0.87	99	0	0	1410	1060	1410	1060	Seng et al., 2010
1.9 kW/L	3.0	0.87	112	0	3800	1590	1200	1590	-2600	
Control	3.3	0.83	206	0	0	2920	2190	2920	2190	Perez Elvira et al.,
13.3 kw/L	3.3	0.83	290	0	2700	4110	3080	4110	380	2010

Notes: Only the energy required for pretreatment was considered. (-) sign indicates that the energy balance is negative.

Table 3. Summary of previously studied pretreatment techniques and their LCA

Pretreatment Techniques	System boundary	Function unit	Impact categories**	LCA methodology	Software	Inventory	Key finding	References
Ultrasonication, Conventional thermal pretreatment (65°C, 75°C, 85°C)	Anaerobic digestion with pretreatment and successive sludge handling	1 kg of fresh matter (FM) of sewage sludge	GWP, SOD NRC, FPM HT, FE, TE IR, TA	ReCiPe 2016 midpoint	GaBi	Sphera/ GaBi, Ecoinvent 3.6	LCA underlined the high environmental impact of ultrasonication	Mainardis et al., 2021
Thermal hydrolysis	pre-dewatering, pretreatment and dewatering	1 tonne total solid of the sludge	TA, CC, NRC FE, HT, TE	CML 2 baseline 2000 v2.05	OpenLCA	---	THP increase biogas yield with environmental performance	Li et al., 2017
Thermal hydrolysis	Pretreatment, anaerobic digestion, digestion of sludge and sludge transportation	1 Tonne dry solids (TDS) of the dry mass of sludge	GWP, SOD FE, TA, NRC	---	GaBi	GaBi	Producing methane for grid injection has worst environmental impact	Mills et al., 2014

Thermal pretreatment (120°C) Freezing and Thawing Ozonation pressurize-depressurize	anaerobic digestion process with energy recovery and the disposal of the digestate	10 L of solid waste	NRC, FE GWP, HT, TE	CML 2 baseline 2000 v2.05	SimaPro 7.3	---	Thermal pretreatment is most suitable for improvement of waste stabilisation	Carballa et al., 2011
Ultrasonication/ Ultrasound Conventional thermal pretreatment (65°C)	Anaerobic Digestion, dewatering, transport, and storage of dewatered sludge, spreading on agricultural land	1 kg of fresh matter (FM) of sewage sludge	GWP, SOD NRC, FPM HT, FE, TE IR, TA	ReCiPe 2016 midpoint	GaBi	Sphera/ GaBi, Ecoinvent 3.6	Ultrasonication had lower impact than thermal pretreatment (owing to heat requirements of latter)	Mainardis et al., 2021

973 ** GWP, Global warming potential; CC, Climate change; SOD, Stratospheric ozone depletion; IR, Ionising radiation; FPM, Fine particulate
974 matter; TA, terrestrial acidification; FE, Freshwater eutrophication; FSC, Fossil resource scarcity; NRC, Natural resources consumption; HT,
975 Human toxicity potential; TE, Terrestrial ecotoxicity

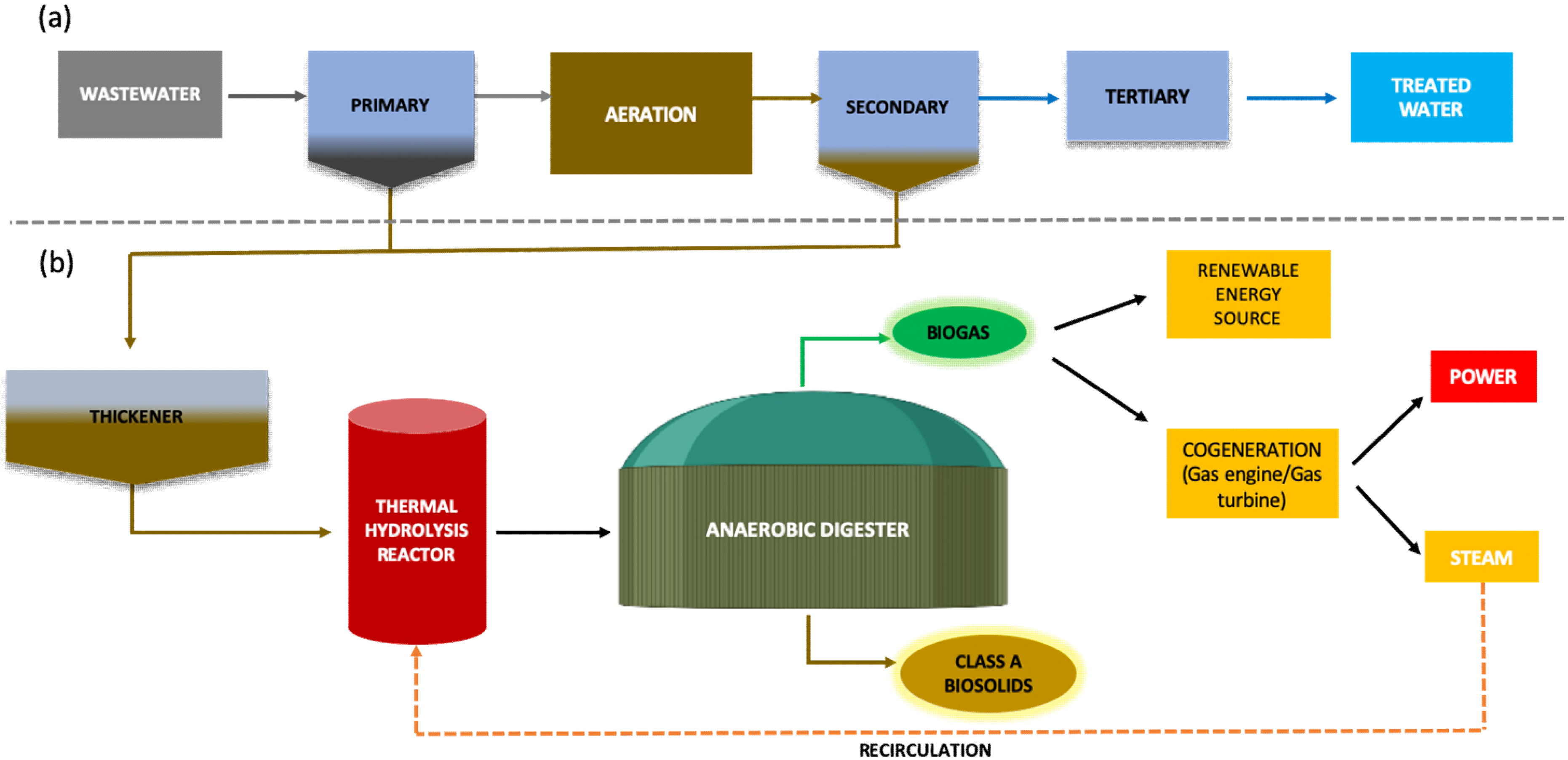

Fig. 1. Schematic representation of a wastewater treatment plant (WWTP) (a) without and (b) with thermal hydrolysis process (Ariunbaatar et al., 2014)

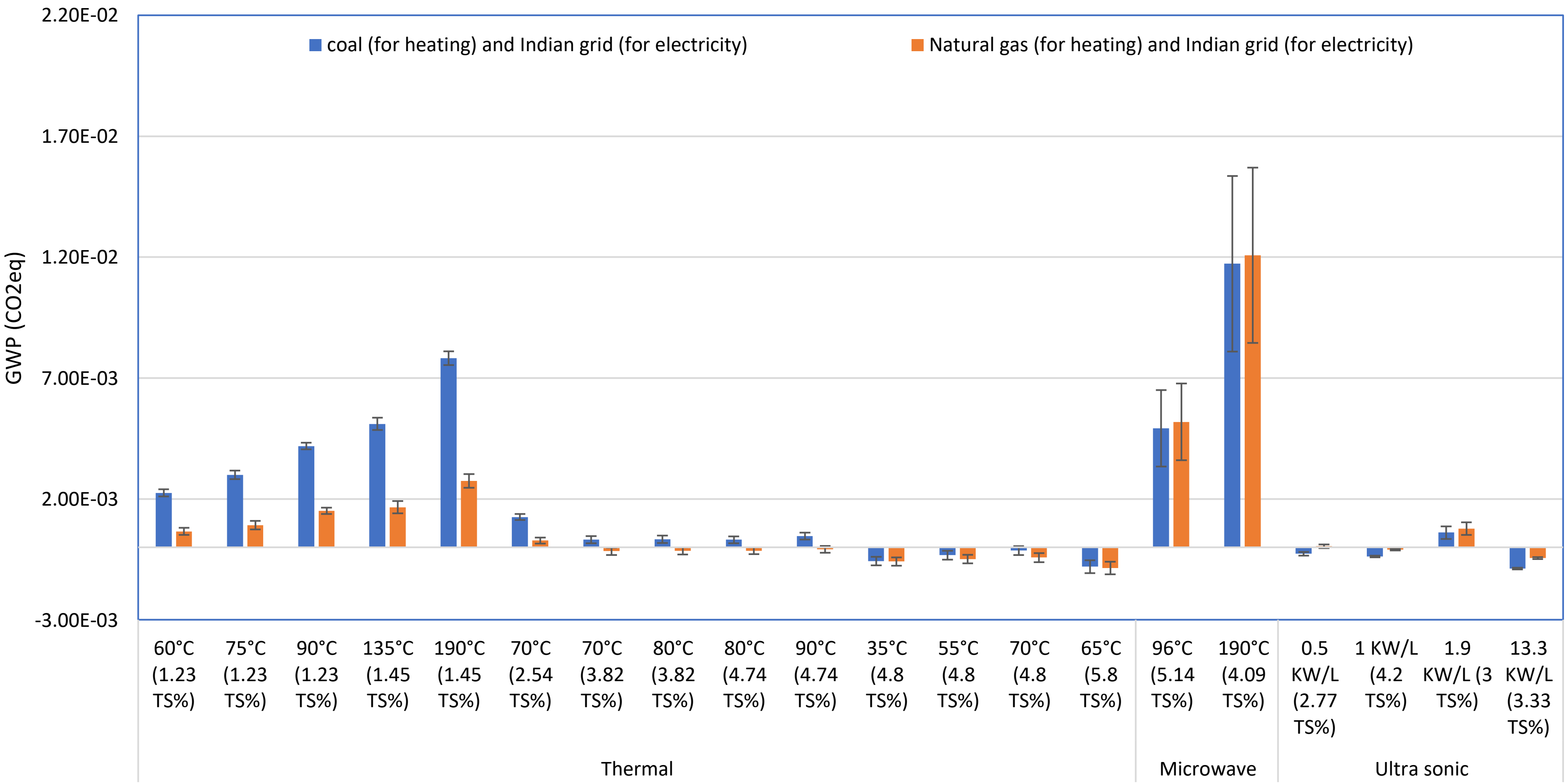

Fig. 2. Global Warming Potential of different types of pretreatment techniques

(Data source: Kim et al 2013b; Bougrier et al., 2007; Ge et al., 2011b; Ruffino et al., 2015; Wang et al., 2014; Bolzonella et al., 2012; Coehlo et al., 2011; Chi et al., 2011; Braguglia et al., 2015; Cella et al., 2016; Seng et al., 2010; Perez Elvira et al., 2010)

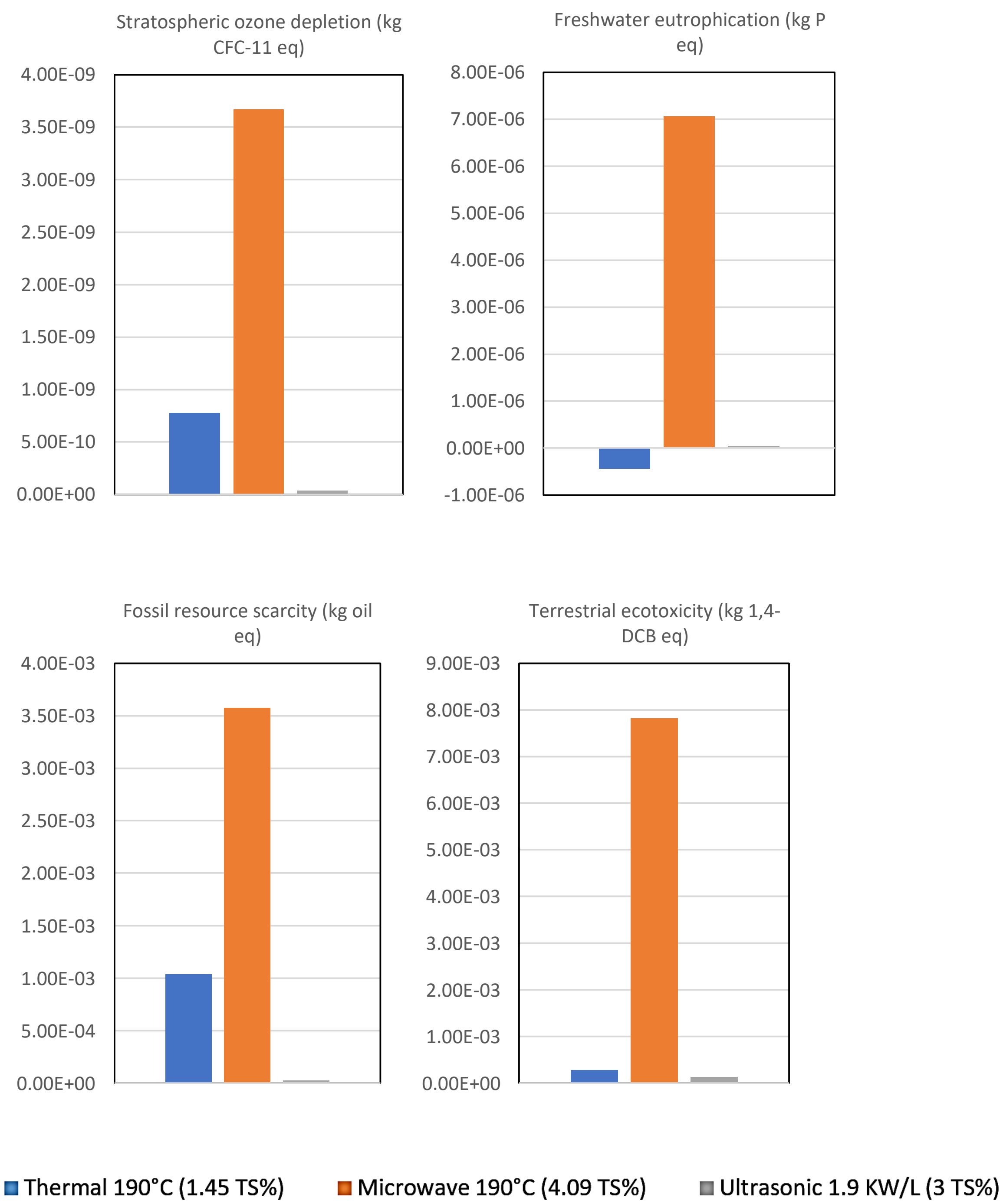

Fig 3. Impact categories of different pretreatment techniques (Data Source: Bougrier et al., 2007; Chi et al., 2011; Seng et al., 2016)