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Pretreatment strategies for enhanced biogas production from lignocellulosic biomass

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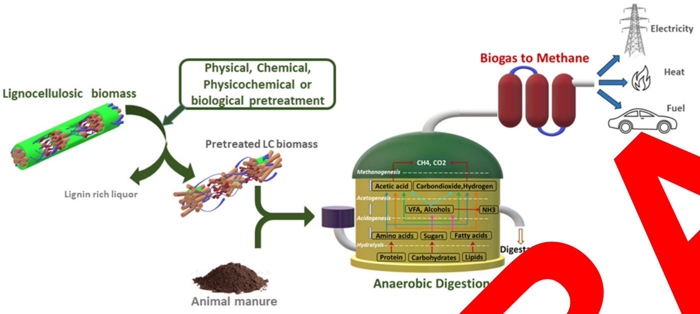
GRAPHICAL ABSTRACT



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ABSTRACT

The inclusion of a pretreatment step in anaerobic digestion processes increases the digestibility of lignocellulosic biomass and enhances biogas yields by promoting lignin removal and the destruction of complex biomass structures. The increase in surface area enables the efficient interaction of microbes or enzymes, and a reduction in cellulose crystallinity improves the digestion process under anaerobic conditions. The pretreatment methods may vary based on the type of the lignocellulosic biomass, the nature of the subsequent process and the overall economics of the process. An improved biogas production by 1200% had been reported when ionic liquid used as pretreatment strategy for anaerobic digestion. The different pretreatment techniques used for lignocellulosic biomasses are generally grouped into physical, chemical, physicochemical, and biological methods. These four modes of pretreatment on lignocellulosic biomass and their impact on biogas production process is the major focus of this review article.

1. Introduction

The population and economic growth in many countries will increase the demand for energy in the future. The current global energy consumption is 542 quadrillion BTU (QBTU) and is expected to increase 50% by 2050 (Energy Information Administration (EIA), 2013). Fossil-based fuels are used to generate 85% of current energy needs, an excessive use has a significant role in greenhouse gas (GHG) production

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and a deterioration in regional air quality (International Energy Agency (IEA), 2013). Energy security is another major issue for developing countries, with renewable energy technologies being promising alternatives to address environmental problems and energy scarcity in the future. The renewable energy production will increase at a fast rate, with 85% of energy demands estimated to be met with renewable energy by 2050 (IRENA, 2018). The major renewable resources used for power generation are wind, solar, hydro, and biomass.

Biomass sources like energy crops, agriculture and forest residues, sewage sludge, animal and food waste, municipal solid waste etc is generally used for energy production. The high carbon capture capacity of algal biomass makes it an emerging feed stock for bioenergy production (Sepehri et al., 2019). Lignocellulosic biomass resources are considered as an abundant and renewable feed stocks for bioenergy (biomethane, biohydrogen, bioethanol, etc.) production. The global production of lignocellulosic biomass is approximately120 × 10^9 ton per annum, which is equal to 2.2 × 10^{21} Joule, 300 times more than the existing global energy requirement (Guo et al., 2015). Crop residues generated after agricultural practices are considered as a larger source of lignocellulosic biomass and it does not have other uses. Residues from cereal crops (rice, wheat, etc.) are mainly used for fodder and manure applications and its surplus availability is less compared to other crop residues (Pandey et al., 2009). In most of the developing countries, unutilized crop residues left in the field become a potential source of greenhouse gases or openly burned residues cause serious air pollutions (Sukumaran et al., 2009).

Anaerobic digestion (AD) is a source of renewable energy from biomass resources (Martínez-Gutiérrez, 2018) and is a microbial process under anaerobic conditions to produce two major products: energy-rich biogas and nutrient-rich digested residues. Biogas is directly used as a source for heat or electricity is generated through a combined heat and power system. Digestate can be mechanically separate and use as a fertilizer or soil improver in the agriculture field. The AD process [illegible] performed using a microbial community that uses complex metabolic pathways under anaerobic conditions (Theuerl et al., [illegible]). AD occurs under many natural conditions, but controlled design and engineering approaches are used to produce biogas commercially in a digester. The major components of biogas are methane (50 ~ [illegible]) [illegible] carbon dioxide (35 ~ 50%), with few amounts of hydrogen, nitrogen, ammonia, hydrogen sulfide, and water vapour. Although AD processes have been operated for decades, AD remains an alternative approach for energy production as an eco-friendly substitute [illegible] fossil-based energy (Tabatabaei et al., 2015). The production of biogas from biodegradable waste materials improves waste management practices in different countries (Tun et al., [illegible]). Anaerobic digestion is considered as prime option for energy production due to reduced need for space and less complexity in processing. Compared to other renewable energy sources, biogas production [illegible] independent of seasonal fluctuations and can be stably produced. In its raw form, biogas has traditionally been used for cooking and other heating applications. Using several novel technologies, the quality of biogas [illegible] improved and it increase the range of applications (Kougias and Angelidaki, 2018). Various technologies can upgrade biogas to [illegible] grade bio-methane, which consists of 95–99% methane and 1–3% CO_2 (Khan et al., 2017). Bio-methane can substitute fossil fuel for transportation, gas fuel for electricity generation, and feedstock for various chemical industries. The global bio-methane market was 1.68 billion USD in 2018 and is expected to reach 2.61 billion USD by 2025 (Zion Market Research, 2019).

Biogas production from lignocellulosic biomass is considered as an eco-friendly second-generation technology for energy production (Florian et al., 2013). Bio-methane production is an efficient means of energy generation from biomass compared to other processes, exhibiting a high energy output/input ratio (Deublein and Steinhauser, 2011). Lignocellulosic biomass co-digested with nitrogen-rich feedstock can maintains the optimum C/N ratio and increasing the stability and biogas production in anaerobic digester (Zhang et al., 2016b). Many pretreatment strategies are available for cellulosic biomass (Julie et al., 2018) and in recent years many studies are evaluating the feasibility of these methods for accelerating the digestion process and improving biogas production from lignocellulosic biomass. Even though pretreatment can increase the biogas yield in anaerobic co-digestion, the development of an economically feasible pretreatment strategy is essential for lignocellulosic biomass. The present paper is reviewing the latest research on various pretreatment strategies of lignocellulosic feedstock for biogas production.

2. Lignocellulosic biomass for biogas production – Challenges

The production of biogas from lignocellulosic substrates creates a good opportunity to convert vast biomass resources into renewable energy. Even though lignocellulosic biomass has high methane potential, it has not been properly exploited for biogas production from AD processes (Sawatdeenarunat et al., 2015). Recently, the major difficulty with the lignocellulosic substrate is the poor digestion of biomass during the anaerobic process due to the complex and recalcitrant nature of feedstock (Patinvoh et al., 2017). The lignocellulosic biomass is made up of polymers such as cellulose, hemicellulose, and lignin. Cellulose forms the core portion [illegible] cellulose, which is bounded by a hemicellulose matrix and an outer lignin layer (Saini et al., 2015). Cellulose is the major constituent in biomass and forms liner homopolymer chains of 100 to [illegible] units. Each unit is made up of a glucose disaccharide (cellobiose), which are linked by a β-1,4-glycosidic bond (Somerville et al., 2004). The presence of higher numbers of hydroxyl group makes hydrogen bond between lateral fibres and makes the structure more stable. Even though cellulose is hydrophilic, but its large [illegible] makes it less soluble in water. In nature, crystalline and amorphous forms of cellulose are common and the high packing density of the crystalline structure is maintained by the unique hydrogen-[illegible] pattern that occurs in fibres. The crystalline nature of cellulose increases its resistance to biological degradation (Karimi and Taherzadeh, 2016a,b).

Hemicellulose is a heteropolysaccharide with a degree of polymerization of between 200 and 700 and is composed of different combinations of monomers, such as pentoses, hexoses, and sugar acids with xylan as the major structural unit (Somerville et al., 2004). Hemicellulose is non-covalently attached to cellulose fibres and acts as a matrix material in lignocellulosic biomass. The amorphous structure and lower degree of polymerization of hemicellulose causes it to be more susceptible to physical, chemical, and biological degradation than cellulose (Li et al., 2015a).

Lignin is a heteropolymer consisting of monomeric units of coniferyl, sinapyl, and coumaryl alcohols (Somerville et al., 2004). Lignin is hydrophobic and fills the space between cellulose and hemicellulose structures in lignocellulosic biomass, acting as a physical barrier against biological decomposition that restrict its utilization in the AD process (Thomsen et al., 2014). The AD process can digest both cellulose and hemicellulose portion of lignocellulosic substrate, whereas lignin remains undigested. Lignin rich residues either from pretreatment or anaerobic digester is a potential by-product in the lignocellulosic biomass based AD process. The valorization of lignin-rich fraction can improve the overall economics of the AD process. Lignin can be used as an organic matter in agriculture and a source of carbon fibre in the industry (Strassberger et al., 2014). Lignin can be used for energy production via combustion, gasification, and pyrolysis. Lignosulfonate is commercially used as a plasticizer in the cement industry, a binder in animal feed and as a substrate for the production of a flavoring agent, vanillin (Strassberger et al., 2014; Bjørsvik and Minisci, 1999).

Along with its structural features, the high C/N ratio of lignocellulosic biomass also limits its efficient use in AD processes for methane production. An optimum C/N ratio (20–30) of feed stock promote efficient methane production in AD, while most lignocellulosic feed stocks maintaining higher values. The high C/N ratio negatively affects methane production, and the mono-digestion of lignocellulosic substrate is not an efficient route for biogas production (Hagos et al., 2017). Currently, different processes, such as co-digestion (Zhang et al., 2016b), solid-state anaerobic digestion (Brown et al., 2012), bioaugmentation (Nzila, 2017), and nutrient supplementation (Zieliński et al., 2019) are used to promote the efficient degradation of lignocellulosic feedstock in AD process. Even though different techniques are practicing for the degradation and utilization of lignocellulosic feedstock via AD processes, the pretreatment of biomass remains the best option for improving its digestion rate and methane production (Paudel et al., 2017).

3. Pretreatment of lignocellulosic biomass for biogas production

To modify the recalcitrant structure and enhance the digestibility of biomass for anaerobic digestion, various pretreatment strategies have been introduced. Increasing the surface area of feedstock is a key approach, where more enzymes or microbes attach to the surface of biomass to enhance cellulose degradation (Yang et al., 2015). Size reduction is a common approach to enhance the surface area of biomass, in addition to which the wetting of biomass can increase its surface area due to expansion, while drying adversely affect biomass degradation (Selig et al., 2008). Even though particle size reduction can enhance the biomass digestibility, the energy consumption associated with size reduction needs to be thoroughly investigated to outweigh the cost of particle size reduction (Yang et al., 2015). Biomass decrystallization is another mechanism used to increase the digestibility of biomass. Different pretreatment techniques can reduce biomass crystallinity and improve its digestibility during the AD process. For instance, ionic liquid pretreatment is an example to breaks the hydrogen bonds between the cellulose microfibers (Feng and Chen, 2008), causing cellulose dissolution, reduced crystallinity and increased porosity, increasing the digestibility of biomass (Zhu, 2008).

The removal of hemicellulose and lignin is another approach used [illegible] increase the accessibility to cellulose present in feedstock, which enhances the digestibility of cellulose. Exclusion of hemicelluloses from lignocellulosic biomass is achieved using dilute acid or thermal pretreatments with temperatures ranging from 1[illegible]0 to [illegible] °C ([illegible] 2014). However, the formation of inhibitors is the main disadvantage of the high temperature pretreatment process (Yang et al., 2015), with waste liquors having to be further treated before their discharge into the environment. The removal or alteration [illegible] lignin is accomplished using alkaline, oxidation or biological methods. During alkaline treatment, the cleavage of lignin-carbohydrate linkage increases the porosity and surface area in pretreated biomass (Boladorodríguez et al., 2016). Other approaches used to remove the lignin present in biomass are the wet oxidation and [illegible]ical approaches. In the wet oxidation process, the ether bonds present in the alkyl aryl bond are cleaved by free radicals ([illegible]ng et al., 2015). During biological pretreatment, lignin fraction in the biomass [illegible] removed by microbial enzymes and enhance its digestion. An advantage [illegible] biological process is the application of mild process conditions compared to the chemical process, although much longer time [illegible] required than other modes of pretreatment (Xu et al., 2019). Numerous pretreatment studies have been performed with the goal of increasing biogas yield from biomass during the AD process. The efficiency of pretreatment varies in different biomasses and pretreatment methods are generally classified into physical, chemical, physiochemical, and biological methods (Fig. 1).

3.1. Physical pretreatment

The physical pretreatment methods can increase the surface area of biomass by reducing the particle size. This size reduction can improve the accessibility of the biomass and increase its susceptibility to microbial and enzyme attacks, promoting the digestion of biomass during AD. Importantly, physical pretreatments do not generate any toxic compounds, which inhibit the AD process (Zheng et al., 2014). The different physical pretreatment methods on biomass and its effect on biogas production are listed in Table 1.

3.1.1. Mechanical pretreatment

Grinding and milling are the most commonly used mechanical pretreatments for lignocellulosic substrates. The choice of grinding or milling techniques depends upon the moisture content of the biomass (Neshat et al., 2017). Mechanical pretreatment is highly reliable with respect to particle size reduction, but the improvement in biogas production is minimal compared to that obtained using other methods. For instance, an approximate 3% increase in [illegible] production was obtained when rice straw was pretreated via grinding prior to the AD process (Zhang and Zhang, 1999). The effect of reducing the size of hay biomass using a knife mill on the AD process, with a negative correlation observed with respect to particle size and lignin content (Menind and Normak, 2008). Tsape[illegible] et al. (2015) investigated the results of mechanical pretreatment [illegible] meadow grass using different commercial plates (sandpaper, me[illegible]h gra[illegible]e, and a [illegible]cker plate), resulting in a maximum rise [illegible] methane production of [illegible] than the untreated conditions. The mechanical pretreatment [illegible] six different lignocellulosic biomasses were performed by Dahun[illegible] (2019), who observed a methane production increase up [illegible] 22%. The energy consumed during the size reduction of miscanthus, [illegible] stalks, alfalfa, and willow using a 22 kW articulated hammer mill with [illegible] 10-mm mesh sieve was investigated, the results of which showed that miscanthus (50–65 kJ/kg) required significantly higher amounts of energy compared to other biomass (3[illegible] kJ/kg) ([illegible]ceanu et al., 2019). Mechanical pretreatment can result [illegible] positive response, but the requirement of a higher energy demand [illegible] it a costly addition to the AD process (Kratky and Jirout, [illegible]).

3.1.2. Irradiation pretreatment

During microwave irradiation pretreatment, microwave energy is directly delivered to the biomass to enable its rapid heating with a minimal thermal gradient. Due to the rapid heating of biomass, the pretreatment time and energy investment are reduced in the irradiation pretreatment. During the process of microwave irradiation, deviations in the dipole orientation of polar compounds increase the solubility of lignocellulosic biomass (Pellera and Gidarakos, 2016; Feng et al., 2018). The application of a microwave pretreatment for methane production has been investigated by many research groups, including Eskicioglu et al. (2007), Kuglarz et al. (2013), and Sapci (2013). Biomethane yield from microwave-pretreated switchgrass and wheat straw was assessed at 150 °C, resulting in a 30% methane yield increase for wheat straw, whereas an increase was not observed for switchgrass. Li et al. (2012) studied methane yield from hybrid varieties of *Pennisetum* using microwave pretreatment, although the observed methane yield was lower than that of the control. In another study by Jackowiak et al. (2011), no significant variation in the methane volume was obtained when microwave-pretreated switchgrass was used for AD, but the digestion process was faster compared to the untreated samples. A possible reduction in methane content may be due to the formation of heat-induced inhibitor molecules, sugar degradation products or phenolic compounds (Qi and Xinyang, 2007; Ximenes et al., 2010). An assessment of biogas production from paddy straw using a combination of sodium hydroxide and microwave pretreatment resulted in 55% increase in biogas production (Kaur and Phutela, 2016). In addition, a combination of alkaline and microwave pretreatments on swine manure and rice straw increased the biogas yield by 25% in high solid AD than control (Qian et al., 2019). Sapci (2013) evaluated the result of microwave pretreatment on the AD process, with no major difference observed in biogas production from agricultural straw.

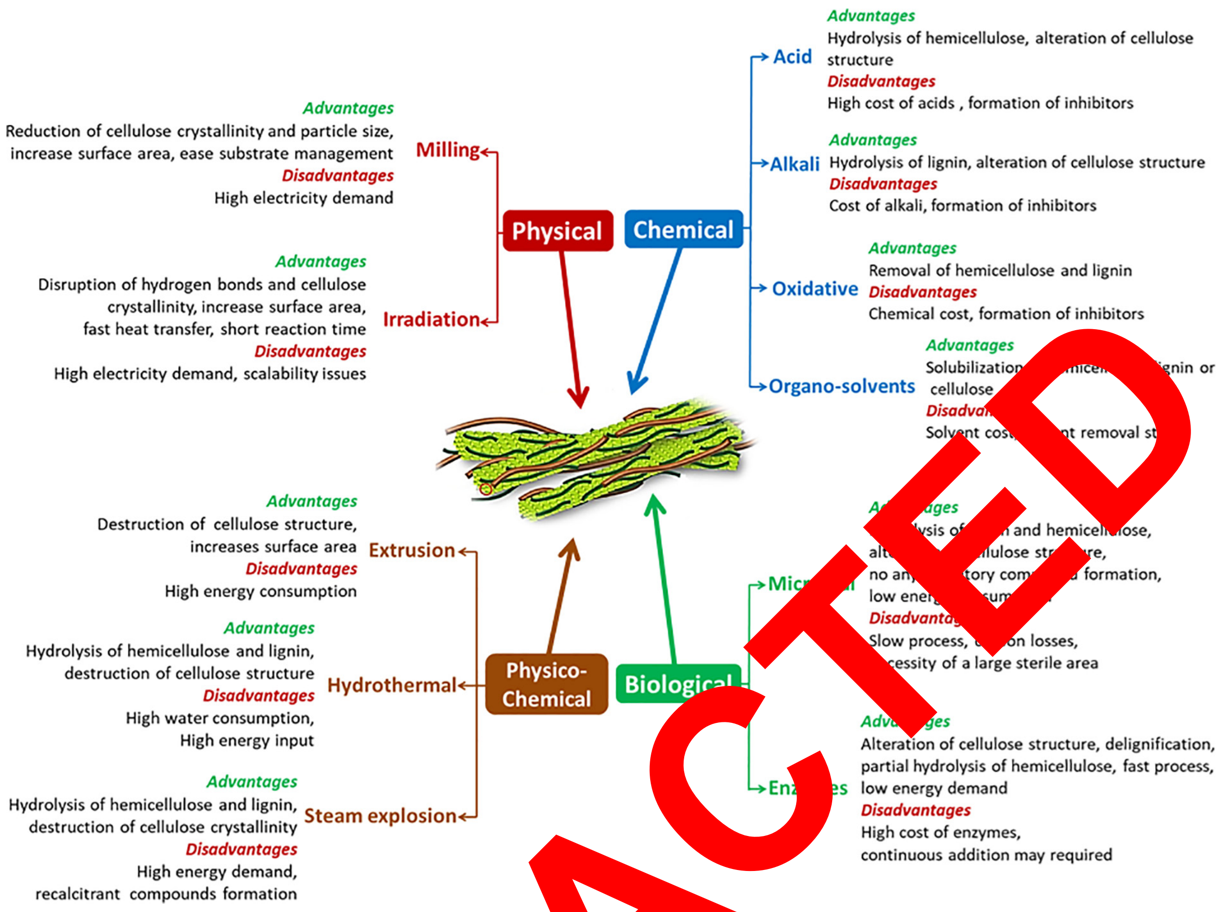

Fig. 1. Different pretreatment techniques [illegible] lignocellulosic biomass for biogas production.

3.2. Chemical pretreatment

Chemical pretreatments are primarily classified into [illegible], oxidative, and organo-solvent treatments based on the type of chemical being used (Table 2). Major acidic agents involved in biomass pretreatment include sulfuric, hydrochloric, [illegible], and nitric [illegible]. During alkaline pretreatment, agents such as sodium hydroxide and ammonia are commonly used. These treatments primarily [illegible] by removing the lignin or hemicellulose present in the biomass, where the mode of action depends upon the chemical being used and the operating conditions in the pretreatment process.

3.2.1. Acid pretreatment

Acid pretreatment [illegible] the [illegible] of Van der Walls, hydrogen and covalent bonds [illegible] are bonded together in the biomass. This treatment [illegible] [illegible] disintegration and cell lysis, resulting in the elimination of [illegible] hemicellulose portion from the lignocellulosic biomass. Wheat straw pretreated with sulfuric acid at high temperature, prior to mesophilic digestion, resulting in significant increase (16%) in methane production (Taherdanak et al., 2016). The effect of dilute sulfuric acid pretreatment on water hyacinth was studied by Sarto et al. (2019), with an increased biogas yield of 131% observed compared to the control. An acid pretreated *Salvinia molesta* and rice straw showed 5% increase in biogas production during AD process (Syaichurrozi et al., 2019). A comparison of chemical pretreatments involving four different acid reagents and three different alkaline reagents with respect to methane yield from corn straw were studied by Song et al. (2014), with 3% H_2O_2 and 8% $Ca(OH)_2$ providing the maximum yields of 115 and 105%, respectively. Ikeda et al. (2019) studied the effect of various pretreatments, including hot water, acid, and alkaline treatments on methane production from spent mushroom substrates, with the alkaline pretreatment observed to yield higher methane production compared to the other pretreatment techniques.

3.2.2. Alkaline pretreatment

Alkaline pretreatment enabling the delignification and causes more porosity, surface area and reduction in the degree of polymerization of lignocellulosic biomass. Chandra et al. (2012) improved the digestibility of wheat straw by NaOH pretreatment, which resulted in 88 and 112% increases in biogas and methane yield, respectively. In addition, methane yield was enhanced by 38% when 5% NaOH was used to pretreat grass silage (Xie, 2012). Khor et al. (2015) assessed the performance of $Ca(OH)_2$ pretreatment on extruded biomass, resulting in a maximum increase of 37% in methane yield at an alkaline loading of 7.5%. The ammonia pretreatment on wheat straw was investigated by Li et al. (2015b), with a 40% increase in biogas yield attained. In another study, the AD of pretreated wheat straw with a 4% ammonia solution enhanced biogas production by 52% compared to control (Yang et al., 2014). Pretreatments on rice straw using of NaOH and KOH resulted in a biogas yield enhancement of up to 50% compared to control (Dong et al., 2009). In general, NaOH pretreatment is effective for enhancing biogas production, but care must be taken to avoid inhibiting AD due to the formation of the Na^+ ions during the pretreatment process.

3.2.3. Oxidative pretreatment

The oxidative pretreatment destruct the lignin and hemicellulose structures in lignocellulosic biomass. The oxidative pretreatment promote the breakage of aromatic nuclei, electrophilic substitutions, the dislocation of side chains, and the cleavage of alkyl aryl ether bonds (Paudel et al., 2017). During oxidative pretreatment, hydrogen peroxide (H_2O_2) is converted into corresponding hydroxyl radicals (OH^-)

Table 1
Different physical pretreatment methods for enhanced biogas production.

Pretreatment methods	Pretreatment conditions	Biomass	AD conditions	Effect on methane or biogas production	References
Grinding/Milling	Mechanical	Elephant grass, Mexican sunflower, Siam weed	Batch, 37 °C, 30 days	22% increase in methane yield	Dahunsi (2019)
	Brush and a steel mesh [illegible]tioner	Meadow grass	Batch, 54 °C, 27 days	27% increase in biogas production	Tsapekos et al. (2018)
	knife mill 300 μm a[illegible]m partic[illegible]ze	Wheat straw	Batch, 37 °C, 28 days	49.3% increase in methane yield	Dell'Omo and La Froscia (2018)
	Plate method	Ensiled meadow grass	Batch, 54 °C, 20 days	25% increase in methane yield	Tsapekos et al. (2015)
	Grinding: 10 and [illegible] mm, 0-[illegible]	Rice straw	Batch, 35 °C, 25 days	17.5% increase in biogas production	Zhang and Zhang (1999)
Extrusion	Single screw extruder	Maize straw silage	Batch, 38 °C, 27 days	35% increase in methane yield	Pilarski et al. (2016)
	Co-rotating twin screw ext[illegible]	[illegible]heat straw, Deep litter	Batch, 35 °C, 90 days	1–16% increase in methane after 90 days	Wahid et al. (2015)
	Variable hydro-module, 0.[illegible]2.7 MP[illegible] min	[illegible] biomass	Batch, 38 °C, 20 days	33% higher biogas production	Maroušek (2012)
	Twin-screw extruder	Str[illegible] Grass, manure and deep litter	Batch, 35 °C, 90 – 110 days	9–28% increase in methane after 90 days	Hjorth et al. (2011)

and act upon lignocellulosic biomass. The oxidative mechanism of action is less selective towards biomass, and portions of hemicellulose and cellulose will lose after pretreatment process. Almomani et al. (2019) evaluated the results of advanced oxidative pretreatment on agricultural wastes using fenton, ozone, and ozone combined with H_2O_2 and Fe(II). A maximum increase of 30% in methane yield and a 25% enhance in digestion process were attained when O_3/H_2O_2 was used for oxidative pretreatment. An oxidative pretreatment using ozone on coffee husks improved the resulting AD performance, especially with respect to the hemicellullosic hydrolysate, resulting in a maximum methane production of 85 mLg^{-1} coffee husk being obtained (Santos et al., 2018). In another pretreatment study, H_2O_2 on rice straw was investigated, with an 88% increase in m[illegible]duction was observed during AD process (Song et al[illegible]013). Mo[illegible] et al. (2011) achieved a higher methane recovery [illegible]3% when [illegible]lower stalks pretreated with H_2O_2wereused f[illegible] AD. I[illegible]dition, a 7[illegible] increase in methane generation potentia[illegible]s obtain[illegible]hen [illegible]enhouse crop waste was subjected for H[illegible] pretr[illegible]tment [illegible]o AD (Perendeci et al., 2018).

3.2.4. Organic sol[illegible] pretreatm[illegible]

Organic sol[illegible] are used to [illegible]e high-purity cellulose from lignocellulosi[illegible]o[illegible] without cau[illegible]g significant cellulose degradation. Pinewood, elmw[illegible] and rice straw were pretreated with 75% ethano[illegible]1% H_2SO_4 at [illegible]-180 °C for 0.5 to 1 h, resulting in 85, 73, and [illegible]% increases in [illegible]gas yields being obtained after AD (M[illegible]ohamadsadeghi et al., 2014). Pretreatment on sweet sorghum st[illegible] performed [illegible]g ethanol (50%) at 160 °C for 30 min caused in a 27[illegible]ise in bio[illegible] yield than control (Ostovareh et al., 2015). Ionic liqu[illegible]s) h[illegible]he ability to dissolve cellulose and it can be completely re[illegible]ed after the pretreatment process, with no negative ef[illegible] observed on the subsequent AD process. Although various ILs are [illegible] N-methylmorpholine-N-oxide monohydrate (NMMO) is commonly used for the pretreatment of lignocellulosic biomass during AD process (Kabir et al., 2014). Wheat straw pretreated with NMMO showed a 47% enhancement in methane production during AD process than control conditions (Akhand et al., 2012). Teghammar et al. (2012) studied the result of IL pretreatment time on different types of biomass, observing that a longer pretreatment time improved the methane yield, with the exception of rice straw. The study observed that a maximal 400–1200% higher methane yield was achieved from the IL-pretreated biomass compared to the control. An ionic liquid pretreatment of waste textile materials with NMMO significantly improved the methane production by 16% compared to control material (Jeihanipour et al., 2010). Pretreatment of oil palm empty fruit bunch with NMMO achieved a maximum improvement of 167% in biogas production after the AD process (Purwandari et al., 2013). Gao et al. (2013) pretreated water hyacinth with the IL, 1-*N*-butyl-3-methyimidazolium chloride, for biogas production and 98% increase in methane yield was observed in AD process than untreated water hyacinth.

3.3. Physicochemical pretreatment

Physicochemical pretreatment is a combined approach used to breakdown the hemicellulose or lignin polymers within lignocellulosic biomass prior to the AD process (Table 3). During physicochemical pretreatment, the hydrogen bonds between the complex polymers are broken down by heat, increasing the surface area accessible for efficient enzyme or microbial action towards biomass (Rodriguez et al., 2017). Physicochemical pretreatment is performed over a wide temperature range (from 50 to 250 °C), with a large number of studies having reported from 150 to 180 °C temperature (Dahadha et al., 2017). The utilization of waste heat is crucial for efficient energy management in physicochemical pretreatment, i.e., heat from the pretreatment reactor can be successfully recovered and utilized for maintaining the digester temperature or heating the slurry during the pre-digester phase. The

Table 2
Different chemical pretreatment methods for enhanced [illegible]gas produ[illegible]n.

Pretreatment methods	Pretreatment Conditions	Biom[illegible]	AD conditions	Effect on methane or biogas production	References
Acid pretreatment	Dilute H_2SO_4 (5%), 121 °C, 60 min	[illegible] hyacin[illegible]	Batch, 28–30 °C, 90 days	131% increase in biogas production	Sarto et al. (2019)
	Dilute H_2SO_4 (2,4,6%) 30 °C, 2 days	[illegible]vinia molesta	Batch, 30 °C, 30 days	5% increase in biogas production	Syaichurrozi et al. (2019)
	Dilute H_2SO_4 (1%), 121 °C, 10–120 min	Wheat straw	Batch, 37 °C, 30 days	16% increase in methane yield	Taherdanak et al. (2016)
	Dilute H_2SO_4, HCl, CH_3COOH and H_2O_2 (1,2,3,4%), 25 °C, 7 days	Corn stover	Batch, 37 °C, 35 days	115% increase with 3% H_2O_2	Song et al. (2014)
Alkali pretreatment	NaOH (1.6%), 30 °C, 24 h	Whea[illegible]w	Batch, 37 °C, 40 days	15% increase in methane yield	Mancini et al. (2018)
	$Ca(OH)_2$ (7.5%), 10 °C, 20 h	Ma[illegible]raw, gr[illegible] stem	Batch, 37 °C, 30 days	37% increase in methane yield	Khor et al. (2015)
	Ammonia (0–30%), 20–80 °C, 6–48 h	Wheat straw	Batch, 30 °C, 28 days	40% increase in biogas production	Li et al. (2015a,b)
	Ammonia (2,4,6%), 35 °C, 7 days	Wheat straw	Batch, 35 °C, 60 days	52% increase in methane yield	Yang et al. (2014)
	NaOH (4%), 37 °C, 5 days	Wheat straw	[illegible]ch, 37 °C, 60 days	87.5% increase in biogas production	Chandra et al. (2012)
	NaOH (5%), 100 °C, 4–48 h	Grass silage	[illegible]tch, 35 °C, 90 days	38% increase in methane yield	Xie (2012)
Oxidative pretreatment	Fenton, ozone, and ozone combined with Fe(II) and H_2O_2	Agricultural waste[illegible]	[illegible]atch, Semi-batch, 35 °C, 20 days	23–30% increase in methane yield	Almomani et al. (2019)
	Ozone (6–81 mg O_3/g CH), pH 3–11	Hemicellullosic hydrolysate of [illegible] husk	Batch, 35 [illegible] 40 days	27% methane increase in two-stage anaerobic digestion	Santos et al. (2018)
	H_2O_2 (1%) , 50 °C, 6 h	Greenhouse crop waste	Ba[illegible] °C, 62 [illegible]	77% increase in methane yield	Perendeci et al. (2018)
	H_2O_2 (2.68%), 6–18 days	Rice straw	[illegible], 37 °C, 41 da[illegible]	88% increase in methane yield	Song et al. (2013)
	H_2O_2 (4%), 55 °C, 24 h	Sunflower stalks	[illegible]h, 35 °C, 35 d[illegible]	33% increase in methane yield	Monlau et al. (2011)
Organic Solvent pretreatment	50% ethanol, 180 °C, 1 h	Wheat straw	[illegible]h, 37 °C, 40 days	15% increase in methane yield	Mancini et al. (2018)
	NMMO, 120 °C, 3 h	Wheat straw	[illegible] 37 °C, 40 days	[illegible]% increase in methane yield	Mancini et al. (2018)
	50% or 70% ethanol	Sweet Sorghum Stalks	Ba[illegible]50 days	[illegible] increase in biogas production	Ostovareh et al. (2015)
	75% ethanol and 1% H_2SO_4	Pine & elm wood, rice straw	Batch, [illegible] °C, 55 days	[illegible]% increase in biogas production	Mirmohamadsadeghi et al. (2014)
	NMMO, 90 and 120 °C, 1,3,5h	Oil palm empty fruit bunch	Batch, 55 °C, 50 days	Up to [illegible]% increase in methane yield	Purwandari et al. (2013)
	BMIM.Cl/DMSO, 120 °C, 120 min	Water hyacinth	Batch, 35 °C, 30 days	98% increa[illegible] biogas production	Gao et al. (2013)
	NMMO, 90 °C, 7 h	Wheat straw	Batch, 55 °C, 45 d[illegible]	47% inc[illegible]ethane yield	Akhand and Blancas (2012)
	NMMO, 130 °C, 1–15 h	Softwood spruce, rice and triticale straw	Batch, 55 °C, 20 da[illegible]	400–[illegible] inc[illegible] in methane yield	Teghammar et al. (2012)
	NMMO, 120 °C, 2 h	Waste textiles	Batch, 55 °C, 6 days	[illegible]ncr[illegible] in meth[illegible] yield	Jeihanipour et al. (2010)

Table 3

Different physico-chemical pretreatment methods for enhanced biogas production.

Pretreatment Methods	Pretreatment Conditions	Biomass	AD conditions	Effect on methane or biogas production	References
Steam pretreatment	Steam explosion, 1[illegible]nd 5–15 bar, up to 15 min	Rice straw	Batch, 37.5 °C, 60 days	Up to 147% increase in biogas production	Aski et al. (2019)
	Steam explosio[illegible] °C, 10[illegible]	Birch wood	Batch, 62 °C, 50 days	118% increase in biogas production	Mulat et al. (2018)
	Steam explo[illegible] 220 °C, [illegible], 15, 20 min	Reed biomass	Batch, 37.5 °C, 63 days	89% increase in methane yield	Lizasoain et al. (2016)
	Steam ex[illegible]n, 1–[illegible]	Rice straw	Batch, 38 °C, 21 days	51% increase in biogas production	Zhou et al. (2016)
	Steam ex[illegible]sion, 16[illegible]10, 15 [illegible]n	Late harvested hay	Batch, 37.5 °C.	16% increase in methane yield	Bauer et al. (2014)
Irradiation pretreatment	Alkaline & Microwa[illegible]	Rice straw	Batch, 35 °C, 66 days	25% increase in biogas production	Qian et al. (2019)
	NaOH & Microwave [illegible]0 W	Paddy straw	Batch, 37 °C, 45 days	55% increase in biogas production	Kaur and Phutela (2016)
	Microwave	Wheat, Oat and Barley straw	Batch, 37 °C, 60 days	No net increase in biogas production	Sapci (2013)
	Microwave, 400–160[illegible] W	Switch grass	Batch, 35 °C, 42 days	No change in volume of biogas production	Jackowiak et al. (2011)
Hydrothermal Pretreatment	Hydrothermal, 125–200 °[illegible] min	Napier grass	Batch, 37 °C, 42 days	35% increase in methane yield	Phuttaro et al. (2019)
	Hydrothermal, 120–180 °C, [illegible]0 min	Safflower straw	Batch, 37 °C, 45 days	70% increase in biogas production	Hashemi et al. (2019)
	Hydrothermal, 90 – 130 °C, 1[illegible]in	[illegible]ice straw	Batch, 35 °C, 30 days	22% increase in methane yield	Luo et al. (2019)
	Hydrothermal, 120–180 °C, 60 m[illegible]	[illegible]at straw	Batch, 35 °C, 45 days	53% increase in biogas production	Rajput and Visvanathan (2018)

high temperature during pretreatment can remove the pathogens present in the biomass. The pretreatment time is a critical factor in the physicochemical process, as prolonged heat exposure can lead to unexpected reactions, such as Millard reactions, which leads to the formation of inhibitors harmful to the AD process (Fernández-Cegrí et al., 2012).

3.3.1. *Steam explosion pretreatment*

Steam explosion pretreatment changes the structure of lignocellulosic biomass with high pressure (5–50 bar) steam at 160–250 °C for different intervals (Paudel et al., 2017). The rapid release of pressure after a period of incubation with high temperature and pressure causes the destruction of lignocellulosic [illegible]. Steam explosion pretreatment on hay biomass was inves[illegible]ed for bi[illegible]production by Bauer et al. (2014), who displayed 16[illegible] in methane [illegible]ld compared to untreated biomass. The effect o[illegible] steam [illegible]eatment o[illegible]eed biomass at 200 °C for 15 min was i[illegible]tigated, w[illegible]n o[illegible]ved methane production increase of 89% [illegible] the u[illegible]treated [illegible]s (Lizasoain et al, 2016). Zhou et al. (201[illegible] [illegible]rform[illegible] steam explo[illegible]ion of rice straw at different temperature[illegible] (20[illegible] and ti[illegible]ntervals (1–4 min), observing a maximu[illegible]crease o[illegible]% in bi[illegible] yield at 200 °C with a 2-min incubation [illegible]. A combine[illegible] explosion and bioaugmentation pretreat[illegible]t [illegible]ch wood bi[illegible]ss enhanced the methane production by 140% than [illegible]trol experiment with native birch wood. The steam-[illegible]ed birch w[illegible] alone enhanced the methane yield by 118[illegible] [illegible]ating that stea[illegible] explosion contributed the majority of m[illegible]ane yield in the combined pretreatment process (Mulat et al., 20[illegible]). A maxim[illegible] methane yield recovery of up to 147% was obse[illegible] by Aski et[illegible] (2019) when steam-exploded rice straw was used for b[illegible]prod[illegible]on.

[illegible]2. *Extrusion pretreatment*

[illegible]g extrusion pretreatment, the lignocellulosic biomass is exposed to a series of treatments, such as heating, mixing, and sudden pressure drop while releasing the material from the extruder. The pressure drop causes the release of intracellular water from the feedstock and results in structural breakdown, which can improve biomass digestion in the AD process (Zheng et al., 2014). The primary operating parameters for extrusion pretreatment are reaction time, pressure, and the moisture content of the biomass. The extrusion pretreatment of pelletized hay caused a rise of 33% in biogas production after the AD process (Marousek et al., 2012). The efficiency of the AD process using five different biomasses was investigated after extrusion pretreatment, resulting in approximately 18–70% increases in biogas yields being obtained after 28 days of digestion at the lab scale. When extrusion pretreatment was attempted at an industrial scale using the same biomass, 8–27% increase in methane yield was obtained after 30 days of AD process (Hjorth, 2011). The extrusion pretreatment of a wheat straw and deep litter mixture enhanced the methane yield by 1–16% after 90 days of AD process (Wahid et al., 2015). Anaerobic digestion of maize silage after extrusion pretreatment showed an approximately 35% enhancement in methane yield (Pilarski et al., 2016). The high pressure and the additional heat supply can improve the extrusion efficiency and methane yield from biomass. Even though methane yields increase with heat supply, prolonged heat causes the formation of toxic compounds such as furfural (Zheng et al., 2014).

3.3.3. *Hydrothermal pretreatment*

During the hydrothermal process, water is heated to temperatures of up to 200 °C is used to treat biomass under high-pressure conditions (Mansini et al., 2018). High temperatures cause the formation of undesirable phenolic compounds and furan derivatives during the pretreatment process. A 35% growth in methane yield was obtained from Napier grass pretreated using the hydrothermal method at 175 °C temperature for 15 min incubation (Phuttaro et al., 2019). The optimum temperature of hydrothermal pretreatment for biogas production from wheat straw was observed to be 180 °C, which resulted in a 53% rise in methane yield than untreated samples (Rajput et al., 2018). The pretreatment of safflower straw using hydrothermal method at 120 °C for 1 h increased the biogas production by 98% in AD process (Hashemi et al., 2019). Luo et al. (2019) compared the hydrothermal pretreatment of wheat straw at two different temperatures (100 and 130 °C) and observed an approximately 20% enhancement in methane production using both conditions compared to the control. In another study, an enhancement in methane production of up to 148% was obtained from paddy straw when pretreated at 200 °C for 15 min prior to the AD process (Bauer et al., 2014).

3.4. Biological pretreatments

Biological pretreatments involve the direct action of microbial metabolism or by-products on lignocellulosic substrates (Zheng et al., 2014). Microbial pretreatment of lignocellulosic biomass is a low-cost process used to increase biogas production during the AD (Mutschlechner et al., 2015). In particular, biological pretreatment is an eco-friendly process compared to physical or chemical processes due to the need for less energy consumption and the use of mild process conditions (Shrestha et al., 2017). Biological pretreatment strategies are more compatible with AD, as no toxic by-products are formed during the process that may affect AD. Compared to non-biological pretreatments, the resulting hydrolysate from biological pretreatment does not need a processing step before being used for anaerobic digestion (Wagner et al., 2018). In biological pretreatment, delignification is the primary objective and the removal of lignin can expose the cellulose and hemicellulose fractions for efficient digestion (Shrestha et al., 2017). The reduction in crystalline nature of cellulose and the decomposition of hemicellulose can improve the digestibility of the substrate during the AD process (Karimi and Taherzadeh, 2016a,b). In addition to the hydrolysis action, the biological pretreatment process can also improve the quality of the substrate by eliminating compounds that affect the efficiency of an AD process (Wan and Li, 201[illegible]. Biological pretreatment process in lignocellulosic biomass [illegible] improved biogas production by up to 141% compared to untreated conditions (Yuan et al., 2012). The major biological pretreatment strategies [illegible] microorganisms or enzymes before the AD process (Z[illegible] et al., 2014) (Table 4).

3.4.1. Microbial pretreatment

The microbial pretreatment approach primarily involves the use of single or a consortium of microbes to treat feed stock prior to the AD process (Fig. 2). The main advantages of microbial pretreatment are low inputs of energy and [illegible] of hazardous chemicals. However, a major drawback of this method is need for a longer incubation time to generate sufficient microbial growth [illegible] as well as a longer pretreatment process ([illegible]aurya [illegible] al., 2015). Another major concern is the loss of carbohydrates during microbial pretreatment, as microbes use nutrients from the same [illegible] biomass for their growth and metabolism. Fungal treatment [illegible] widely using as microbial pretreatment strategy for lignocellulosic biomass (Rouches et al., 2016). Fungi can be classified as cellulose- or lignin-degrading species, with major cellulolytic species being brown-rot fungi (Wagner et al., 2018). White and soft rot fungi act upon lignin and to some extent on cellulose polymers. Fungal pretreatment for delignification is a major goal rather than cellulose and hemicellulose removal due to the low digestion of lignin in the AD process (Kumar and Sharma, 2017). The white rot fungi are highly potential for delignification, and their efficiency may vary depend on the species used for pretreatment (Rouches et al., 2016). Fungal growth and metabolism are the crucial factors for the effective use of fungi in microbial pretreatment process. Moisture content is a crucial factor affecting the efficiency of fungal pretreatment, with other major factors including biomass characteristics, competition from indigenous microorganisms, availability of nutrients, pH, and oxygen concentration (Wan and Li, 2012). The pH is an important factor in microbial pretreatment which influence the microbial growth and metabolism. Maintaining the optimum pH during the pretreatment stage and the AD process is crucial for maximum efficiency. The biomethanation process is operating within a pH range of 6.5–8.5. The optimum pH range for the growth and ligninolytic activity of white rot fungus is between 4 and 5. The deviation from optimum pH affects the production of laccase enzyme and efficiency of microbial pretreatment (Patel et al. 2009).

In wheat straw, microbial pretreatment with *Polyporus brumalis* BRFM985 improved the production of biogas during anaerobic digestion by 52%, and metal addition and incubation time positively influenced the AD process (Rouches et al., 2018). The fungus *Trametes versicolor* was evaluated for use in the pretreatment of corn silage and showed an increase on biogas production. While the methane production rate from untreated corn silage [illegible].167 m^{3}CH[illegible]$^{-1}$VS,that of the pretreated corn silage was significantly improved, with an observed methane production rate of up to 0.236 m[illegible] kg^{-1} [illegible] (Tisma et al., 2018). Fungi such as *Pleurotus eryngii*, *Pleurotus ostreatus*, and *Trametes versicolor* were screened for their pretreatment efficiency towards lignocellulosic biomass, with [illegible] *eryngii* showing a high rate of ligninolytic enzyme production [illegible] 19% [illegible] in biogas yield (Wyman et al., 2018). The impact of moisture content on the pretreatment of *Agropyron elongatum* with *Flammulina velutipes* was evaluated, with the maximum lignin and hemicellulose removal observed at 65% moisture content. When *Agropyron elongatum* was pretreated at the optimum moisture content, a 120% higher biogas production was obtained (Lalak et [illegible], 2016). Microbial pretreatment of Albizia biomass with the white-rot fungus *Ceriporiopsis subvermispora* exhibited enhanced biogas production via SS-AD. A maximum lignin removal of 24% was observed after [illegible] days of fungal pretreatment and lead to a 3.7-fold higher methane production than raw feedstock via SS-AD (Ge et al., 2015). The inoculation of yard trimmings with *Ceriporiopsis subvermispora* at different [illegible]tios showed a significant degradation of the lignin (14.8–20.2%), cellulose (8.1–15.4%), and hemicellulose (20.7–27.8%) components after 30 days of incubation, generating a methane yield of 34.9–44.6 LkgVS^{-1}viaSS-AD (Zhao et al., 2014). The microbial pretreatment on rice straw for biogas production was investigated using the fungal species *Pleurotus ostreatus* and *Trichoderma reesei* via SS-AD. Pretreatment with *Pleurotus ostreatus* removed 33.4% lignin and resulted in a methane yield of 263 LkgVS^{-1}, 120% higher than that observed in the untreated control. In contrast, the use of the fungus *Trichoderma reesei* resulted a 23.6% removal of lignin and a 78.3% increase in methane production (214 LkgVS^{-1}) under the pretreatment conditions (Mustafa et al., 2016).

Several aerobic bacterial species have been reported to have high degradation potential towards lignocellulosic biomass and have the major advantage of having a faster growth rate than fungi (Xu et al., 2018). The lignin degradation mechanisms of different bacterial species have been exploited and been shown to exhibit some unique features compared to those of fungal species, including the ability to promote C_{α}-oxidation and C_{β}-C_{β} bond cleavage in the lignin polymer. The efficiency of ligninolytic enzyme production is lower in bacterial species compared to fungi, and more research is needed to identify bacterial strains for the better delignification of lignocellulosic biomass. Shah et al. (2019) showed that the co-culture of *Bacillus* sp. in rice straw significantly reduced the lignin content and caused a 76% rise in methane production obtained via AD. A bacterial strain, *Citrobacter werkmanii* VKVVG4, isolated from the gut of silverfish improved the solubilization of water hyacinth during AD. The microbial-pretreated water hyacinth showed a three-fold rise in biogas yield than control conditions (Barua et al., 2018).

Pretreatment of lignocellulosic with microbial consortia is an effective method to improve biomass degradation performance (Zhang et al., 2011). A microbial consortium contains various species with different biomass degradation efficiencies and functions in different environmental conditions. The microbial consortia can use different

Table 4
Different biological pretreatment methods for enhanced bi[illegible] production.

Pretreatment methods	Microorganism used for pretreatment	Bi[illegible]s	AD conditions	Effect on methane or biogas production	References
Fungal pretreatment	*Polyporusbrumalis*	Wheat straw	Batch, 36 °C, 57 days	52% higher methane yield	Rouches et al. (2018)
	Trametes versicolor	Corn silage	Semi-continuous, 37 °C, [illegible]ys	Methane generation rate 0.236 m^3 CH4 $kgVS^{-1}$ (Control 0.167 m^3 CH4 $kgVS^{-1}$)	Tišma et al. (2018)
	Pleurotus eryngii	Corn sto[illegible]	Ba[illegible] Mesophilic, 40 days	19% higher biogas production	Wyman et al. (2018)
	Flammulina velutipes	*Agro*[illegible]*onga*	B[illegible] 37 °C, 24 days	120% higher biogas production	Lalak et al. (2016)
	Pleurotus ostreatus	Rice [illegible]w	[illegible]h (SS), 37 °C, 45 days	120% higher methane yield	Mustafa et al. (2016)
	Trichoderma reesei	Rice straw	Batch (SS), 37 °C, 45 days	78.3% higher methane yield	Mustafa et al. (2016)
	Ceriporiopsis subvermispora	Albizia chips	Batch (SS), 3[illegible]8 days	3.7-fold higher methane yield	Ge et al. (2015)
	Ceriporiopsis subvermispora	Yard trimming[illegible]	Batch [illegible] days	106% higher methane yield	Zhao et al. (2014)
Bacterial pretreatment	*Bacillus* sp.	Rice straw	[illegible]S), 37 [illegible]0 days	76% higher biogas production	Shah et al. (2019)
	Bacillus subtilis	Corn straw	[illegible]S), 3[illegible] 50 days	17.35% higher methane yield	Xu et al. (2018)
	Citrobacter werkmanii VKVVG4	Water hyacinth	Batch [illegible]philic, 80 days	3.07 times higher biogas production	Barua et al. (2018)
Microbial consortium pretreatment	Microbial consortium TC-5	Wheat straw	Batch, 45 [illegible]35 days	[illegible] higher methane yield	Kong et al. (2018)
	Microbial consortium	Saw dust	Batch, M[illegible]philic, [illegible]ys	25.[illegible]igher biogas production	Alia et al. (2017)
	Rumen fluid	Rice straw	Batch, 35 [illegible]C, 30 [illegible]	82.6[illegible]igher methane yield	Zhang et al. (2016b)
	Microbial consortium	Wheat straw	Batch, 37 °C, 20 [illegible]	80.34[illegible] high[illegible] methane yield	Zhong et al. (2016)
Enzyme pretreatment	Cellulase	Corn stover	Batch, 37 °C, 18 [illegible]	36.9% highe[illegible]as production	Wang et al. (2018)
	Endoglucanase + Xylanase + Pectinase	Spent hops	Semi-batch, 37 °C	13% higher bio[illegible]roduction	Ziemiński et al. (2012)
	Cellulase + Cellobiase	Switch grass	Batch, 50 °C, 30 days	Methane yield [illegible]L g^{-1} (VS), (Control 197.39 mL g^{-1} (VS))	El-Mashad (2015)
	Endoglucanase + Xylanase + Pectinase	Sugar beet pulp silage	Batch, 37 °C, 30 days	27.9% high[illegible]gas p[illegible]ction	Ziemiński and Kowalska (2015)
	Endoglucanase + Exoglucanase + Xylanase	Sorghum forage	Batch, 35 °C, 30 days	15% [illegible] methane yield	Rollini et al. (2014)
	Laccase	Corn stover	Batch, 37 °C, 30 days	25[illegible]her methane yi[illegible]	Schroyen et al. (2014)
	Mn Peroxidase + Versetile Peroxidase	Corn stover	Batch, 37 °C, 30 days	17% higher methane[illegible]	Schroyen et al. (2014)

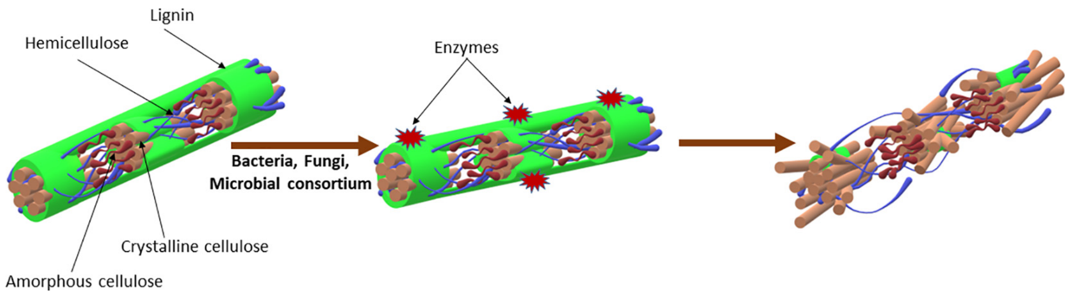

Fig. 2. Biologoical pretreatment process on lignocellulosic biomass.

lignocellulosic biomass degradation mechanisms and better ability to colonize the feedstock over the endogenous species. Zhong et al. (2016) reported improved biogas yield from wheat straw pretreated with a microbial consortium, obtaining a maximum methane yield of up to 41%. The increase of pretreatment time reduced the methane yield due to the consumption of nutrients released from wheat straw by microorganisms used for the pretreatment. The use of rumen fluid for the pretreatment of wheat straw improved biogas production by 66.5% and a decrease process time by 40%in AD (Zhang et al., 2016a). In another study, a microbial consortium isolated from rotten sawdust significantly increased the methane yield by 72.6% compared to that observed in the control treatment (Alia et al., 2017).

Ensiling is a traditional storage process that is now considered to be a low-cost pretreatment strategy to increase the digestion of lignocellulosic substrates (Kreuger et al., 2011). Ensiling involves maintaining conditions that favours lactic or acetic acid fermentation of the substrate, resulting in an acidic feed stock environment. The acidic conditions inhibit the microbial growth on biomass and increas[illegible] storage life of feedstock. In addition, the acidic conditions can [illegible]o improve the cellulose and hemicellulose digestion and favour AD p[illegible]cess. The ensiling method has not shown any effect on the structure [illegible] lignin in feedstock, and a few studies have shown a[illegible]ment i[illegible] biogas production during AD. Another major con[illegible] with [illegible]siling is the organic matter loss that occurs due to b[illegible]ial gr[illegible] in the feedstock (Kreuger et al., 2011). In one stud[illegible] en[illegible] h[illegible] uated for methane production and was sh[illegible] to ha[illegible]0% yield increase than that observed under the [illegible]trol conditi[illegible] (Pakarinen et al., 2011). In addition, the pretr[illegible]f giant reeds [illegible] ensiling resulted in a 4–14% rise in methane yield f[illegible] AD process (Liu et al., 2016).

3.4.2. Enzyme pretreatm[illegible]

The exogenous appl[illegible] enzyme[illegible] hydrolytic or oxidative classes, before the AD pro[illegible] an acce[illegible]te the degradation of lignocellulosic bi[illegible]der ana[illegible]ic [illegible]ditions (Koupaie et al., 2019). Enzyme pre[illegible]ment [illegible]ecoming [illegible]e attractive due to short reaction times an[illegible]inimu[illegible] of sugars during the digestion process. Enzymes have [illegible]ty to the substrate and higher mass transfer rate d[illegible] the reaction process (Romero-Güiza et al., 2016). The efficacy of en[illegible] pretreatment determined by the activity of the enzyme, its specific[illegible] towards the substrate, the amount of enzyme used for the treatment, the tolerance of the enzyme to various inhibitors, incubation time, the anaerobic digestion system, enzymatic stability at different temperatures, and pH (Kiran et al., 2015; Thomas et al.,2016). The efficacy of enzyme pretreatment on lignocellulosic biomass can be improved by multiple enzymes. Although enzymes have a number of advantages, the high cost is a key factor with respect to the development of an economically feasible strategy for biomass pretreatment (Romano et al., 2011). The use of enzymes with high specific activity and cross specificity can reduce the amount of enzyme needed for pretreatment, decreasing the pretreatment cost in the AD process. The endogenous microorganisms present in feedstock consume released nutrients and cause sugar loss during the enzyme pretreatment process (Binner et al., 2011). The enzymes a[illegible] categoriz[illegible]nto cellulose/hemicellulose-degrading or lignin-re[illegible]g enzymes [illegible]sed on their activity towards lignocellulosic [illegible]iomas[illegible] he enzym[illegible] pretreatment strategy can improve biog[illegible] productio[illegible] om [illegible]alcitrant lignocellulosic biomass (Koup[illegible]t al., [illegible]19).

Cellulase or hemicell[illegible] enzy[illegible] can break [illegible]own complex polymers in lignocellulose [illegible]iom[illegible]crease t[illegible] digestibility of complex substrates in an A[illegible]ocess. [illegible] pretrea[illegible]nt study on corn stover, cellulase enhan[illegible] he hydrolys[illegible] the substrate and improved the biogas p[illegible]uc[illegible] by up to 3[illegible]. The optimized pretreatment conditions fo[illegible] corn s[illegible] were an enzyme loading of $30FPUg^{-1}$substrate, [illegible] content of [illegible]L^{-1} and 24 h of incubation time (Wang et a[illegible]). The use of sp[illegible] hops pretreated with a mixture of endo[illegible]canase, xylanase, and pectinase enzymes increased the biogas p[illegible]uction in a[illegible] process (Ziemiński et al., 2012). In a combined pr[illegible]tment of [illegible]tchgrass with enzymes and NaOH, the reported biog[illegible]ducti[illegible] was 586 mL g^{-1}VS under the pretreatment conditions a[illegible]mL g^{-1} VS under the control conditions (El-Mashad, [illegible]). Pretreatment of sugar beet pulp silage and vinasse with enzyme [illegible] biogas production up to 27.9% than untreated conditions ([illegible]ieminski and Kowalska, 2015). The pretreatment of ensiled sorghum forage with different commercial enzymatic preparations resulted in maximum 15% rise in biogas yield compared to control (Rollini et al., 2014).

Lignin-removing enzymes can hydrolyse lignin to monomers, allowing other components of lignocellulosic biomass to become more accessible to microbes during anaerobic digestion (Parawira, 2012). Lignin shows less digestibility, and the enzymes targeting lignin are now being explored to increase biogas production during anaerobic digestion. Laccases and peroxidases are the primary enzymes used for lignin removal in lignocellulose biomass (Kudanga and Roes-Hill, 2014). Laccases (Lac) are copper-containing enzymes that catalyse the oxidation of phenolic components in lignin polymers. Peroxidases are mainly classified in to lignin peroxidase (LiP), manganese peroxidase (MnP), and versatile peroxidase (VP) (Zamocky et al., 2014). LiP is a haem-containing enzyme that catalyses the H_2O_2-dependent oxidative degradation of lignin. The Mn peroxidase enzyme aids in lignin bond breakage by the oxidation of Mn^{2+} to Mn^{3+}. During the lignin degradation process, phenolic rings are oxidised by MnP and non-phenolic regions by LiP. (Houtman et al., 2018) while VP oxidizes both compounds in lignin polymers (Zavarzina et al., 2018). The laccase pretreatment of manure fibre in combination with other pretreatments was shown to enhance methane yields by up to 34% in AD process (Bruni et al., 2010). The pretreatment of corn stover with laccase and peroxidase resulted in 25 and 17% improvements in methane production compared to the control conditions (Schroyen et al., 2014).

4. Conclusion

Lignocellulosic substrates show great potential for biomethane production, but their inherent recalcitrant nature limits their use in AD processes. Thus, pretreatment is a necessary stage for the efficient utilization of biomass in the AD process, and various treatments can modify the biomass structure by removing lignin, increasing surface area, decreasing the crystalline nature, and length of polymer chain. Each method has its pros and cons or not suitable when considering the overall economics of the AD process. In the future, more research is essential to make a cost-effective and sustainable pretreatment methods for the efficient biogas production from different LC substrates.

CRediT authorship contribution statement

Amith Abraham: Writing - original draft. **Anil K. Mathew:** Writing - original draft. **Hyojung Park:** Writing - original draft. **Okkyoung Choi:** Conceptualization. **Raveendran Sindhu:** Conceptualization. **Binod Parameswaran:** Conceptualization, Validation. **Ashok Pandey:** Writing - review & editing. **Jung Han Park:** Writing - review & editing. **Byoung-In Sang:** Conceptualization, Writing - review & editing, Supervision.

Declaration of Competing Interest

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

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