



Review Paper

Comparison of pretreatment methods that enhance biomethane production from crop residues - a systematic review

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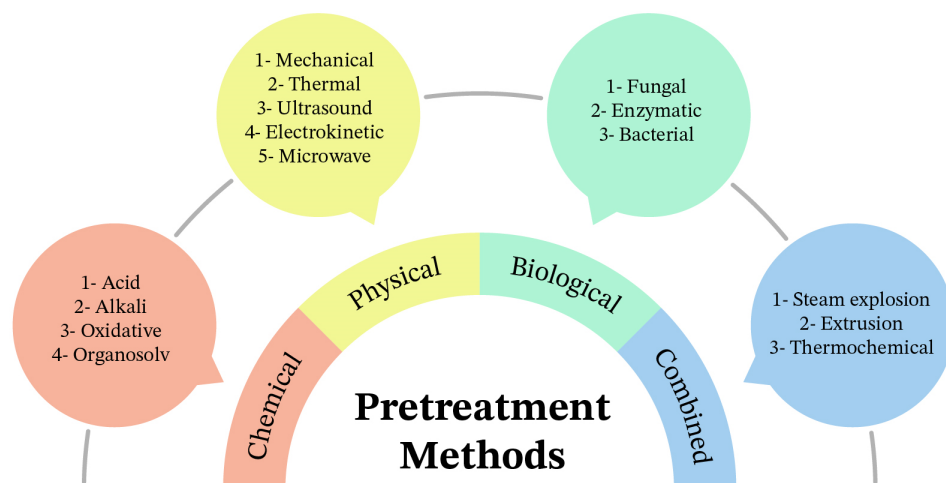
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HIGHLIGHTS

- Efficacy of biological, chemical, physical and combined pretreatments in enhancing biomethane production from crop residues compared.
- Physical and chemical pre-treatment methods are the most effective and fastest.
- Disadvantages of physical and chemical pretreatments are high cost of resources, operation and energy as well as formation of inhibitory by-products.
- Combined pretreatment processes are fast and cost-effective but have limited utility due to generation of toxic compounds.
- Biological pretreatment is inexpensive, eco-friendly and low energy-consuming process.

GRAPHICAL ABSTRACT



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ABSTRACT

A systematic literature review was conducted to compare the efficacy of biological, chemical, physical, and combined pretreatments in enhancing biomethane production from crop residues (CR). Three electronic databases *viz.*, Science Direct, EBSCOhost, and PubMed were used to identify the studies in literature. The pretreatment methods were compared in terms of their advantages and disadvantages with reference to techno-economic aspects. The techno-economic aspects considered included rate of hydrolysis, energy use, effectiveness, cost, and formation of toxic compounds. A total of 3167 studies, covering the period 2014 - 2018, were screened for relevance to the study. Forty-four records (n=44) consisting of 36 research papers (n=36) and eight narrative reviews (n=8) met the inclusion criteria. The results show that physical and chemical methods are the most effective and fastest. These methods have limited utility due to high cost of resources, operation, and energy as well as formation of inhibitory by-products. Despite generation of toxic compounds, combined methods are regarded as fast and cost-effective. Biological method is inexpensive, eco-friendly, and low energy-consuming. However, it is a nascent technology that is still developing. A combination of trends in research and development provide the best pretreatment alternative to improve the biomethane production from CR.

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1. Introduction

Biogas is a renewable fuel with wide applications the world over (Weiland, 2010; Achinas et al., 2017; Alhassan et al., 2019). Over 60.8 billion m³ of biogas are produced annually in the world (WBA, 2018). WBA data show that biogas production is increasing throughout the world. Global biogas production increased 3.7 times from 0.28 EJ to 1.31 EJ during 2000 to 2016 (WBA, 2018). Almost 54% of biogas is produced in the Europe. Africa accounts for only 0.03% of the annual global biogas production (WBA, 2018), yet it has vast resources for biogas production. Biogas is produced by anaerobic digestion (AD) of organic matter (OM). AD is a biochemical process whereby OM is degraded under anaerobic conditions by microbial consortia (Fitzgerald, 2013; Gould, 2015; Hagos et al., 2017; Mulat and Horn, 2018). It is an eco-friendly process and one of the most efficient methods for conversion of biomass to methane (CH₄) (Horváth et al., 2016).

OMs vary in their potential to produce biogas by AD (Gould, 2015; Strong et al., 2016). Parameters used to estimate the potential of OM to produce biogas include anaerobic biogasification potential (ABP) and biochemical methane potential (BMP). These parameters allow direct evaluation of biogas yield, which can be achieved by the AD process (Jingura and Kamusoko, 2017). BMP is the maximum volume of CH₄, which can be produced per gram of volatile solids (VS) in a substrate (Esposito et al., 2012). BMP provides an indication of the biodegradability of a substrate and its potential to produce CH₄ via AD (Sell et al., 2010; Páledal et al., 2013). BMP is an important indicator of the quality of feedstock for biogas production (Triolo et al., 2013). Methods that can be used to determine BMP of feedstock were reviewed by Jingura and Kamusoko (2017). In their review, Jingura and Kamusoko (2017) indicated that the BMP test is a simple, repeatable, and inexpensive method.

The BMP of feedstock is affected by several biochemical characteristics (Gould, 2015; Strong et al., 2016; Jingura and Kamusoko, 2017). These include nutrient content, VS content, chemical oxygen demand (COD), biological oxygen demand (BOD), carbon to nitrogen ratio (C/N), and presence of inhibitory substances (Babae and Shayegan, 2011; Kwietniewska and Tys, 2014). Amongst these characteristics, the C/N ratio plays a critical role in regulation of the microbial population of autotrophs and heterotrophs (Sepehri and Sarrafzadeh, 2018; Sepehri and Sarrafzadeh, 2019). Differences in biochemical characteristics make it possible to categorize feedstock on the basis of their BMP. This type of characterization places feedstock on different positions on a BMP spectrum. Feedstock at the lower end of the BMP spectrum are those, which present challenges in the AD process, whilst those at the upper end are highly biodegradable.

Different types of OM can be used as substrates for biogas production (Weiland, 2010; Páledal et al., 2013; Gould, 2015; Achinas et al., 2017). Feedstock for biogas production include animal manure and slurry, municipal solid waste, food waste, sewage sludge, and various types of crops and their residues (Demirbas and Balat, 2009; Achinas et al., 2017; Rabii et al., 2019). Over 200 billion tons of agricultural crop residues (CR) are produced annually in the world (Horváth et al., 2016; Patinoh et al., 2017), presenting a vast resource for biogas production. CR are largely at the lower end of the BMP spectrum because of high lignocellulose content. Lignocellulose limits degradation by anaerobic bacteria (Wang, 2014; Achinas, 2017). CR are heterogeneous in nature limiting their use as feedstock for AD (Horváth et al., 2016). Sahito et al. (2013) reported a BMP range of 142 to 322 mL CH₄ g VS⁻¹ for various CR. By comparison, feedstock such as residual fats which are at the higher end of the BMP spectrum have BMP of up to 800 mL CH₄ g VS⁻¹ (Muzenda, 2014).

There are several pretreatment options that can optimize the biomethane production from CR (Ariunbaatar et al., 2014; Ge et al., 2016; Achinas et al., 2017; Wagner et al., 2018; Liu et al., 2019). These can be classified as physical, chemical, biological, and combination of these processes (Amin et al., 2017; Patinoh et al., 2017; Kumar et al., 2018; Mustafa et al., 2018; Venturin et al., 2018; Dahunsi, 2019; Karrupiah and Azariah, 2019). Most of the pretreatment methods have limited applications due to high energy demands, need for special equipment, and emission of several adverse by-products (Liu et al., 2019). The efficacy of pretreatment technologies varies from study to study (Amin et al., 2017; Karrupiah and Azariah, 2019). This heterogeneity limits the utility of information available in literature for planning and designing the AD of CR.

Few attempts have been made to compare the efficacy of pretreatment methods used on CR for the purpose of biogas production by AD. There are also limited aggregated results that provide a comparative analysis on the

efficacy of the pretreatment methods with reference to CR. In view of these gaps, it is prudent to conduct a comparative analysis of the efficacy of pretreatment approaches by reviewing extant literature. This will provide empirical evidence on the comparability of existing pretreatment methods. Such information is useful for planning and enhancement of AD plants that utilize CR. To the best of our knowledge, this is the first systematic review study that has been undertaken to compare the efficacy of pretreatment methods in enhancing biomethane production from CR.

2. Methodology

2.1. Procedure

The standard procedure on performing a systematic literature review (Kitchenham and Charters, 2007; Okoli and Schabram, 2010; Mittal et al., 2018) was used. The search period was January 2014 to November 2018. Most recent work on enhancing biogas production was published during this period (Prasad et al., 2017; Kougias and Angelidaki, 2018). Three databases were chosen on the basis of their availability in the university library and are among the top ten online research databases. These are Science Direct, EBSCOhost and PubMed. The search was delineated to online full-text journal articles. Gray literature covering government reports, conference proceedings, graduate dissertations and unpublished papers related to biogas was excluded. The challenges of searching gray literature were pointed out by Mahood et al. (2014) and Paez (2017).

A summary of the search protocol for identification and selection of articles for inclusion in this study is shown in Figure 1. The process for inclusion of a study was in accordance with the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) statement (Moher et al., 2009; Forbes et al., 2018). The key search terms used for extracting the relevant articles are shown in Table 1. These were selected using the model of a concept map (Asiksoy, 2019). 'Biogas' and 'crop residues' were the major search terms. The major search terms were held as constant in order to eliminate articles not concerned with the use of CR for biogas production.

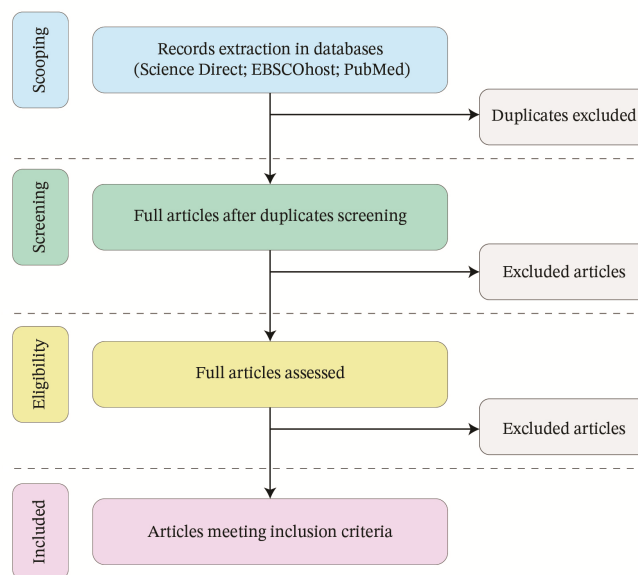


Fig. 1. Flow diagram of systematic literature review according to PRISMA (Source: Kougias and Angelidaki, 2018).

2.2. Comparison criteria

The main variables of interest were advantages and disadvantages of each pretreatment method in terms of techno-economic aspects. The

Table 1.
Number of articles extracted from databases using different search terms.

Search terms	No. of articles scooped			
	Science Direct	EBSCOhost	PubMed	Total
“Pretreatment” and “crop residues” and “biogas”	384	214	6	604
“Chemical pretreatment” and “crop residues” and “biogas”	365	651	1	1 017
“Biological pretreatment” and “crop residues” and “biogas”	309	521	2	832
“Physical pretreatment” and “crop residues” and “biogas”	220	493	1	714

techno-economic aspects used were: rate of hydrolysis, energy use, effectiveness, cost input, and formation of toxic compounds. These were adapted from Wagner et al. (2018) and Venturin et al. (2018). Wagner et al. (2018) considered factors such as low capital and energy use, applicability to a range of feedstock, high product yields, and low waste management. Venturin et al. (2018) considered avoidance of inhibition of AD steps, high degree of simplicity, and effectiveness as some of the characteristics of an ideal pretreatment method.

3. Results and discussion

3.1. Identification and selection of articles for inclusion

A total of 3167 full text articles were screened for relevance to the study as shown in Figure 2. Disaggregation of the number of scooped articles by search terms is shown in Table 1. It is clearly shown that the majority of the publications (n = 1017) were extracted by using the search terms, “chemical pretreatment” and “crop residues” and “biogas”.

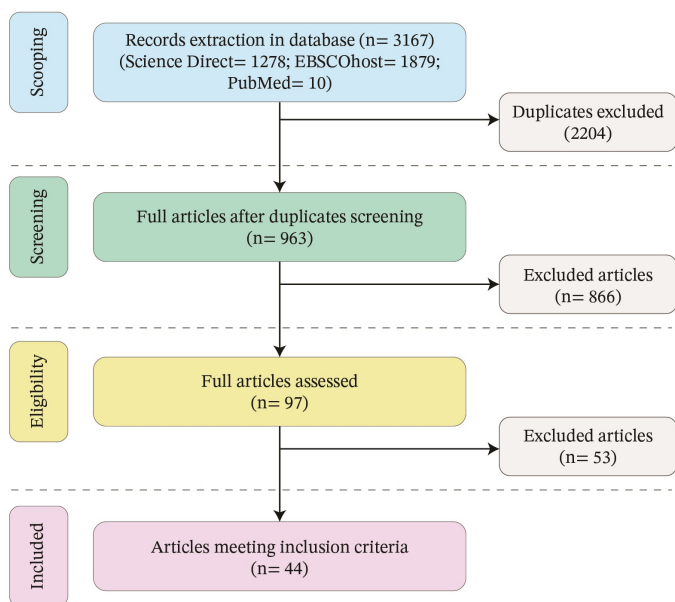


Fig. 2. Summary of identification and selection of articles for the study inclusion process.

Forty-four full text articles met the inclusion criteria. Thirty-six of the articles were research papers and eight were narrative reviews. The trend in the number of publications on pretreatment of CR for biogas production from 2014 to 2018 is shown in Figure 3. The number of publications regarding pretreatment of CR for biogas production rose between 2014 and 2017. This points to sustained research effort on the subject. This can be ascribed to the

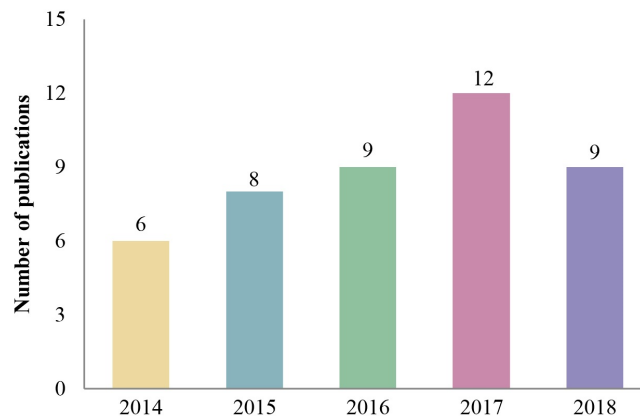


Fig. 3. Number of publications in which “pretreatment of crop residues for biogas production” appeared, based on search results by Science Direct, EBSCOhost, and PubMed.

fact that pretreatment of feedstock is a focal technology in biogas production (Kigozi et al. 2014; Horváth et al. 2016; Achinas et al. 2017). Comparably, Kougias and Angelidaki (2018) reported an increase in the number of articles retrieved from Scopus and Web of Science databases based on the keyword “biogas” between 1977 and 2017. Carrere et al. (2016) also reported a similar phenomenon from 2009 to 2014 based on search terms “anaerobic digestion” and “pretreatment” in the Web of Science database. Ge et al. (2016) reported an increase in the number of articles in which the phrase “solid state anaerobic digestion” appeared in the Google Scholar database from 2005 to 2015. They noted a predominant and steady increase in the period 2010 and 2015. This indicates the increasing focus on pretreatment technology.

3.2. Characteristics of selected articles

The number of selected articles and the authors are shown in Table 2. The articles are organized according to the four pretreatment methods. Various pretreatment techniques are used to depolymerize CR into simple components. These are shown in Figure 4. The key pretreatment methods are chemical, physical, biological, and a combination of these processes (Montgomery and Bochmann, 2014; Yuan et al., 2014; Kumar et al., 2018).

Most of the selected articles (21) focused on chemical pretreatment methods, with the alkali technique being dominant. The dominance of the alkali technique arises from its efficacy (Kumar et al., 2018). Alkali pretreatment has been reported to have high efficiency and ability to improve the degradation of complex compounds (Amin et al., 2017). Physical and biological methods follow with 20 and 15 articles, respectively. However, fewer articles (11) focused on combined pretreatment methods. The main limitation of combined processes is their complexity (Montgomery and Bochmann, 2014).

Table 2. Characteristics of articles meeting inclusion criteria for comparison of pretreatment methods for crop residues

Pretreatment method	No. of articles	Reference
Chemical	21	Pei et al. (2014); Sahito and Mahar (2014); Song et al. (2014); Wikandari et al. (2014); Yuan et al. (2014); Li et al. (2015); Sträuber et al. (2015); Ge et al. (2016); Constant et al. (2016); Talha et al. (2016); Yao and Chen (2016); Amin et al. (2017); Chen et al. (2017); Gumisiriza et al. (2017); Ismail et al. (2017); Kumar and Sharma (2017); Den et al. (2018); Nair et al. (2018); Perendeci et al. (2018); Thomas et al. (2018); Zhang et al. (2018)
Biological	15	Kudanga and Le Roes-Hill (2014); Singh et al. (2014); Ge et al. (2016); Li et al. (2016); Mulakhudair et al. (2016); Thomsen et al. (2016); Amin et al. (2017); Gumisiriza et al. (2017); Kumar and Sharma (2017); Oszust et al. (2017); Rouches et al. (2017); Speda et al. (2017); Zieminski and Kowalska-Wentel (2017); Byrne et al. (2018); Wagner et al. (2018)
Physical	20	Sahito and Mahar (2014); Yuan et al. (2014); Chandra et al. (2015); Dumas et al. (2015); Luo et al. (2015); Xi et al. (2015); Wu et al. (2015); Baeta et al. (2016); Ge et al. (2016); Eskicioglu et al. (2017); Gaworski et al. (2017); Kostas et al. (2017); Li et al. (2016); Mulakhudair et al. (2016); Amin et al. (2017); Gumisiriza et al. (2017); Kumar and Sharma (2017); Zieminski and Kowalska-Wentel (2017); Paul et al. (2018); Sadhukhan et al. (2018)
Combined processes	11	Ge et al. (2016); Siddhu et al. (2016); Amin et al. (2017); Gaworski et al. (2017); Gumisiriza et al. (2017); Kumar and Sharma (2017); Zieminski and Kowalska-Wentel (2017); Byrne et al. (2018); Kim et al. (2018); Paul et al. (2018); Zhang et al. (2018)

3.3. Comparison of pretreatment methods

3.3.1. Rate of hydrolysis

Table 3 provides information on the pretreatment methods with reference to rate of hydrolysis. According to information in Table 3, physical methods appear to be the fastest amongst the pretreatment methods. This is more so for microwave (MW) pretreatment (Wu et al., 2015; Kumar and Sharma, 2017). This is in agreement with Yuan et al. (2014) who posited that short duration time is an advantage of mechanical pretreatment. Conversely, Gumisiriza et al. (2017) reported that irradiation processes are slow.

Two studies (Yuan et al., 2014; Li et al., 2015) reported that chemical pretreatment is a fast process, and three studies reported that it is a slow process (Amin et al., 2017; Gumisiriza et al., 2017; Kumar and Sharma, 2017). This can be explained in terms of variation in treatment conditions. For example, retention time of chemical pretreatment is affected by reaction temperature (Sambusiti, 2013; Theuretzbacher et al., 2015). Data on combined methods is rather limited. However, combined processes were reported to be fast methods (Kumar and Sharma, 2017; Kim et al., 2018).

As shown in Table 3, studies considered in this review cited the major drawback of biological pretreatment as being a slow process (Kudanga and Le Roes-Hill, 2014; Singh et al., 2014; Mulakhudair et al., 2016; Amin et al., 2017; Gumisiriza et al., 2017; Kumar and Sharma, 2017; Den et al., 2018; Wagner et al., 2018). Generally, the required residence time is 10 - 14 d (Amin et al., 2017). In order to enhance the rate of hydrolysis, it is recommended to optimize parameters such as nature and composition of biomass, type of microorganism involved, incubation temperature, pH, incubation time, inoculum concentration, moisture content, and aeration rate (Sindhu et al., 2016).

3.3.2. Energy use

Seven studies indicated the main advantage of biological pretreatment as low energy consumption method (Table 4). Biological pretreatment enables savings on chemicals and energy (Gumisiriza et al., 2017; Kumar and Sharma, 2017; Zieminski and Kowalska-Wentel, 2017; Den et al., 2018; Wagner et al., 2018). Singh et al. (2014) pointed out that fungal

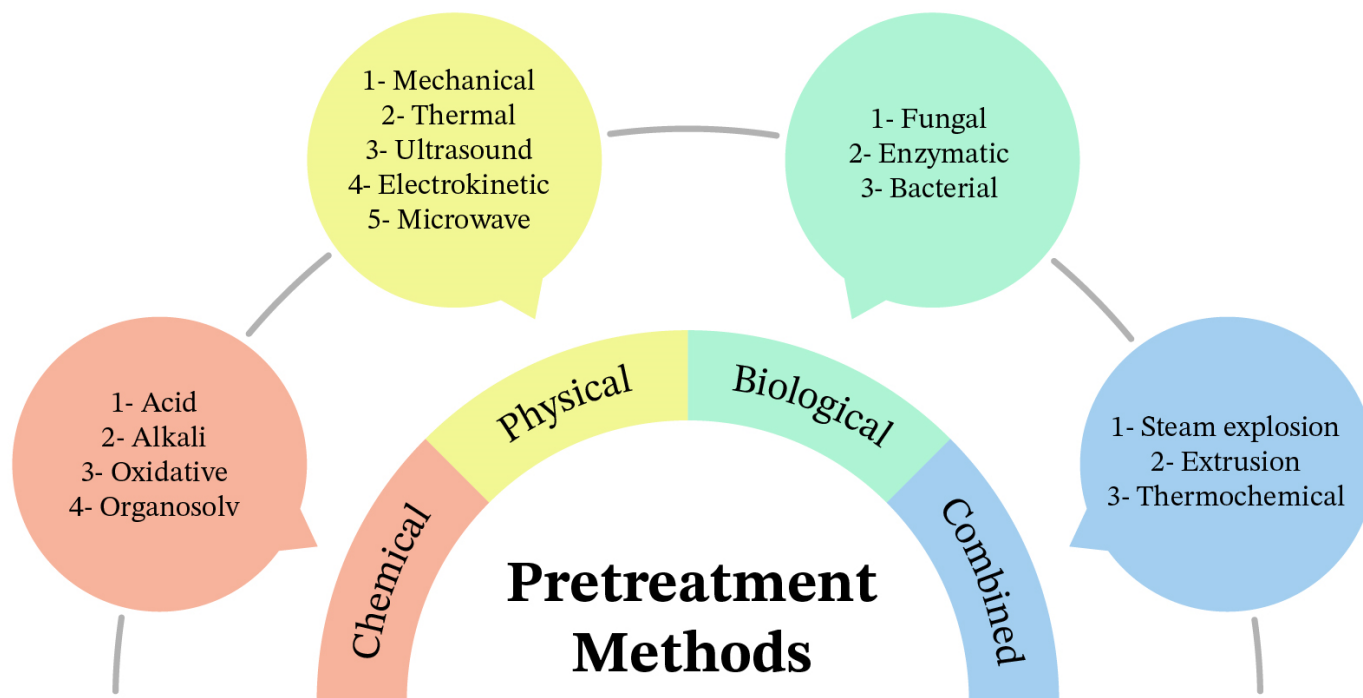


Fig. 4. Summary of pretreatment methods (Source: Montgomery and Bochmann, 2014).

Table 3.
Comparison of pretreatment methods in terms of rate of hydrolysis.

Pretreatment method	Observations	Reference
Chemical	Chemical pretreatments are fast	Li et al. (2014); Yuan et al. (2015)
	Chemical pretreatments are slow	Amin et al. (2017); Gumisiriza et al. (2017); Kumar and Sharma (2017)
Biological	Biological pretreatment is a slow process	Kudanga and Le Roes-Hill (2014); Singh et al. (2014); Mulakhudair et al. (2016); Amin et al. (2017); Gumisiriza et al. (2017); Kumar and Sharma (2017); Den et al. (2018); Wagner et al. (2018)
Physical	Physical pretreatments are fast	Yuan et al. (2014); Wu et al. (2015); Kumar and Sharma (2017)
	Physical processes are slow	Gumisiriza et al. (2017)
Combined processes	Combined processes are fast	Kumar and Sharma (2017), Kim et al. (2018)

pretreatment of biomass offers many advantages including low energy needs and mild reaction conditions, compared to abiotic pretreatments. Amin et al. (2017) reported that microaerobic pretreatment can be considered to be a pretreatment option for AD of corn straw due to low energy requirements and limited supply of oxygen. As an example, Hua et al. (2016) reported increased biogas yield from biomass pretreated by microbial consortium and ascribed this to minimal energy needs.

Physical pretreatment was reported as a high-energy consumption process (Table 4). Physical processes are very energy intensive given high temperatures and pressures involved (Mulakhudair et al., 2016; Gumisiriza et al., 2017; Speda et al., 2017; Wagner et al., 2018). For example, milling, extrusion, and crushing used for particle size reduction of straw to make it more accessible to microbial attack have high energy demands (Luo et al., 2015). Kostas et al. (2017) reported MW pretreatment to have a challenge of high energy demand. It was reported in three studies (Yuan et al., 2014; Chandra et al., 2015; Kumar and Sharma, 2017) that size reduction by mechanical comminution of lignocelluloses requires a considerable amount of energy. For instance, material such as corn stover and switch grass consumes energy worth of 11.0 and 27.6 kWh/metric ton, respectively (Baruah et al., 2018). This is equivalent to a third of the total electricity required for the whole biogas production process (Zheng et al., 2014). For this reason, physical pretreatment is considered to be economically unviable for large scale application (Zheng et al., 2014). On the other hand, Wu et al. (2015), Kostas et al. (2017), and Kumar and Sharma (2017) reported that physical pretreatment with respect to MW pretreatment are energy efficient.

Generally, chemical methods are considered as high energy-consuming processes. This was reported in five studies (Wikandari et al., 2015; Gumisiriza et al., 2017; Rouches et al., 2017; Speda et al., 2017; Wagner et al., 2018) as shown in Table 4. Three studies (Table 4) show that combined processes are high energy-consuming methods (Gumisiriza et al., 2017; Speda et al., 2017; Perendeci et al., 2018). High temperatures and limited heat recovery during steam explosion (SE) pretreatment may lead to high energy use and reduced methane yield (Gumisiriza et al., 2017). Extrusion pretreatment also consumes high energy of about 10 to 15 kW of power to pretreat a tone of substrate (Montgomery and Boehmann, 2014). In contrast, three studies reported combined processes to be low energy-consuming. This observation was sustained by Kumar and Sharma (2017), Zhang et al. (2018), and Kim et al. (2019). It can be postulated that this variation in results is due to the complexity of combined methods and further research is needed.

Table 4.
Comparison of pretreatment methods in terms of energy use.

Pretreatment method	Observations	Reference
Chemical	- Chemical pretreatment has low energy needs	Sahito and Mahar (2014); Ismail et al. (2017)
	- Chemical methods are energy intensive	Wikandari et al. (2015); Gumisiriza et al. (2017); Rouches et al. (2017); Speda et al. (2017); Wagner et al. (2018)
Biological	Biological pretreatment is a low energy demand process	Singh et al. (2014); Amin et al. (2017); Gumisiriza et al. (2017); Kumar and Sharma (2017); Zieminski and Kowalska-Wentel (2017); Den et al. (2018); Wagner et al. (2018)
Physical	- Physical pretreatment saves energy	Wu et al. (2015); Kostas et al. (2017); Kumar and Sharma (2017)
	- Physical methods are very energy intensive	Yuan et al. (2014); Chandra et al. (2015); Luo et al. (2015); Mulakhudair et al. (2016); Gumisiriza et al. (2017); Kostas et al. (2017); Kumar and Sharma (2017); Speda et al. (2017); Wagner et al. (2018)
Combined processes	- Combined methods require low energy	Kumar and Sharma (2017); Kim et al. (2018); Zhang et al. (2018)
	- Combined processes consume high energy	Gumisiriza et al. (2017); Speda et al. (2017); Perendeci et al. (2018)

3.3.3. Effectiveness

Information in Table 5 suggests that chemical pretreatment is the most effective method. Song et al. (2014), Sträuber et al. (2015), Jiang et al. (2016), Amin et al. (2017), Ismail et al. (2017), Kumar and Sharma (2017), and Rouches et al. (2017) reported that chemical pretreatments are effective for enhancing biodegradation of complex compounds such as agricultural residues and herbaceous crops. For example, Perendeci et al. (2018) observed 78% enhanced BMP from alkaline H₂O₂ pretreatment of greenhouse crop waste. Ammonia pretreatment of wheat straw effectively increased total methane yield by 17.5% (Li et al., 2015). Thomas et al. (2018) obtained 32% more BMP from lime pre-treated *Mischanthus x giganteus* than non-treated. Den et al. (2018) reported that acid- and alkali-pretreated oil palm empty fruit bunches increased methane yield by 40% and 100%, respectively.

Data in Table 5 also show that combined processes are effective methods (Amin et al., 2017; Kumar and Sharma, 2017; Kim et al., 2018; Perendeci et al., 2018; Zhang et al., 2018). Steam explosion was found effective in pretreatment of agricultural residues (Amin et al., 2017; Kumar and Sharma, 2017). As an example, Zhang et al. (2018) reported improved cumulative methane yield of 226.6% and 216.4% at 1.2 MPa for 15 min and 1.5 MPa for 5 min, respectively, from steam-exploded crop straw.

As shown in Table 5, biological pretreatment is not as effective as the other methods. None of biological methods is efficient as standalone pretreatment method (Thomsen et al., 2016). For instance, no significant difference in methane yields between enzymatically-pretreated banana

stems and non-treated stems was observed by Li et al. (2016). Paul et al. (2018) reported that fungal pretreatment of agricultural biomass did not improve methane production. However, pretreatment of rice straw with fungal strains such as *Pleurotus ostreatus* and *Trichoderma reesei* increased methane yield by 120% (Wagner et al., 2018). This variation in results is expected as biological pretreatment is still under development.

Physical pretreatment appears to be generally effective as shown in Table 5. For example, hot water pretreatment increased methane yield from rice straw by 222% (Ge et al., 2016) and MW method increased methane yield by 28% (Wu et al., 2015). Baeta et al. (2016) reported that autohydrolysis is a highly effective process. As reviewed by Den et al. (2018), MW pretreatment at 200 or 300°C cannot increase biogas production. Furthermore, an inverse relationship between temperature increase and biogas production was noted during MW pretreatment (Den et al., 2018). This is because high temperatures can lead to production of heat-induced inhibitors such as phenolics and furfural. As a result, MW pretreatment has been used in combination with chemical pretreatment at fairly low temperatures (Den et al., 2018).

Table 5.
Comparison of pretreatment methods in terms of effectiveness.

Pretreatment method	Observations	Reference
Chemical	Chemical pretreatment is an effective method	Song et al. (2014); Li et al. (2015); Sträuber et al. (2015); Jiang et al. (2016); Talha et al. (2016); Amin et al. (2017); Ismail et al. (2017); Kumar and Sharma (2017); Rouches et al. (2017); Den et al. (2018); Perendeci et al. (2018); Thomas et al. (2018)
Biological	- Biological pretreatment is an effective process	Wagner et al. (2018)
	- Biological pretreatment is not an effective process	Li et al. (2016); Thomsen et al. (2016); Paul et al. (2018)
Physical	- Physical pretreatment is an effective method	Wu et al. (2015); Baeta et al. (2016); Ge et al. (2016)
	- Physical pretreatment is not an effective process	Den et al. (2018)
Combined processes	Combined pretreatment is an effective method	Amin et al. (2017); Kumar and Sharma (2017); Kim et al. (2018); Perendeci et al. (2018); Zhang et al. (2018)

3.3.4. Cost

Table 6 compares the different pretreatments in terms of cost of operation. Despite reported to be less effective, biological pretreatment is considered to be variable in cost-effectiveness. Biological pretreatments were reported to be inexpensive (Mulakhudair et al., 2016; Gumisiriza et al., 2017; Den et al., 2018; Wagner et al., 2018). For instance, Rouches et al. (2017) stated that fungal pretreatments are cost-effective. This is because use of fungi helps to reduce the number of pretreatment steps and costs by avoiding enzyme recovery steps (Carrerre et al., 2016). Kudanga and Le Roes-Hill (2014) reported that enzyme pretreatments have low utility costs of enzymes due to the use of mild conditions. As reported by Amin et al. (2017), microaerobic pretreatment (MP) is more economically and environmentally friendly than other pretreatment methods. During MP, microorganisms are partially exposed to O₂ or air under moderate operating conditions such as temperature and pressure. MP has also minimal enzyme and energy requirements (Mustafa et al., 2018). The process is designed to promote the hydrolysis stage through stimulation of cell growth and activity (Wagner et al., 2018). MP is likely to be one of the most promising

Table 6.
Comparison of pretreatment methods in terms of cost.

Pretreatment method	Observations	Reference
Chemical	- Chemical pretreatments are relatively inexpensive	Song et al. (2014); Amin et al. (2017); Gumisiriza et al. (2017); Ismail et al. (2017); Kumar and Sharma (2017)
	- Chemical methods are expensive	Sträuber et al. (2015); Wikaandari et al. (2015); Amin et al. (2017); Kumar and Sharma (2017); Rouches et al. (2017); Speda et al. (2017); Den et al. (2018); Sadhukhan et al. (2018); Wagner et al. (2018)
Biological	- Biological pretreatments are cost-effective	Kudanga and Le Roes-Hill (2014); Mulakhudair et al. (2016); Amin et al. (2017); Gumisiriza et al. (2017); Rouches et al. (2017); Den et al. (2018); Wagner et al. (2018)
	- Biological pretreatments are costly	Kudanga and Roes-Hill (2014); Sahito and Mahar (2014); Li et al. (2016); Mulakhudair et al. (2016)
Physical	- Physical pretreatments are inexpensive	Kostas et al. (2017)
	- Physical pretreatment methods are expensive	Sahito and Mahar (2014); Song et al. (2014); Yuan et al. (2014); Xi et al. (2015); Amin et al. (2017); Gumisiriza et al. (2017); Kumar and Sharma (2017); Speda et al. (2017); Perendeci et al. (2018); Sadhukhan et al. (2018)
Combined processes	- Combined processes are cost-effective	Gumisiriza et al. (2017); Kumar and Sharma (2017); Zhang et al. (2018)
	- Combined processes are expensive	Speda et al. (2017)

technologies in the foreseeable future. Contrarily, biological pretreatment was reported to be costly with respect to production costs due to the use of commercial enzymes (Kudanga and Le Roes-Hill, 2014; Sahito and Mahar, 2014; Li et al., 2016; Mulakhudair et al., 2016).

It is noted that, despite variations shown in Table 6, chemical and physical pretreatments appear to be predominantly expensive. Selected results show that chemical or physical pretreatments are expensive and not economically viable for biogas production from CR. Chemical pretreatments were reported to be expensive due to high costs of disposal of digestion residues (Wagner et al., 2018), use of extraneous agent which incurs higher cost of chemicals and downstream processing (Sträuber et al., 2015; Amin et al., 2017; Kumar and Sharma, 2017; Den et al., 2018; Sadhukhan et al., 2018), the need for expensive auxiliary equipment (Wikaandari et al., 2015; Speda et al., 2017), and high operational and maintenance costs (Rouches et al., 2017). Other studies reported chemical methods with respect to alkali pretreatment to be inexpensive (Song et al., 2014; Amin et al., 2017; Gumisiriza et al., 2017; Ismail et al., 2017; Kumar and Sharma, 2017). The benefit of Ca(OH)₂ pretreatment is the low disposal costs as Ca can be easily recovered from the hydrolyzate (Amin et al., 2017). Based on pretreatment of corn straw, Ca(OH)₂ could be a better option than NaOH although their pretreatment costs were not highly variable (Song et al., 2014). Alkali pretreatment was favored over other pretreatments due to low operational costs (Ismail et al., 2017). The low cost of lime and ease of recovery from the waste make it a better pretreatment technology than other alkalis (Kumar et al., 2017).

Table 7. Comparison of pretreatment methods in terms of formation of toxic compounds.

Pretreatment method	Observations	Reference
Chemical	- Chemical pretreatment avoids formation of toxic inhibitory compounds	Sahito and Mahar (2014); Gumisiriza et al. (2017); Den et al. (2018); Paul et al. (2018)
	- Chemical pretreatment produces toxic inhibitory compounds	Kudanga and Roes-Hill (2014); Pei et al. (2014); Amin et al. (2017); Eskicioglu et al. (2017); Gumisiriza et al. (2017); Kumar and Sharma (2017); Rouches et al. (2017); Speda et al. (2017); Den et al. (2018); Nair et al. (2018); Paul et al. (2018)
Biological	Biological pretreatment reduces formation of inhibitory substances	Singh et al. (2014); Amin et al. (2017); Wagner et al. (2018)
Physical	- Physical pretreatment avoids formation of toxic inhibitory compounds	Baeta et al. (2016); Kumar and Sharma (2017)
	- Physical pretreatment produces toxic inhibitory compounds	Wu et al. (2015); Zieminski and Kowalska-Wentel (2017); Speda et al. (2017); Den et al. (2018); Perendeci et al. (2018)
Combined processes	Combined processes generates toxic inhibitory compounds	Amin et al. (2017); Eskicioglu et al. (2017); Gumisiriza et al. (2017); Kumar and Sharma (2017); Speda et al. (2017); Zhang et al. (2018)

Kostas et al. (2017) reported that MW heating proffers opportunities to reduce the capital cost of processing. However, most studies reported that physical pretreatment is an expensive process. This is mainly due to higher energy and capital cost (Sadhukhan et al., 2018). Milling, grinding, ultrasonic, and irradiation processes have high energy and equipment maintenance costs (Song et al., 2014; Amin et al., 2017; Gumisiriza et al., 2017; Ismail et al., 2017; Kumar and Sharma, 2017). Combined methods are affected by costs associated with the other three methods. SE pretreatment requires expensive auxiliary equipment (Speda et al., 2017), although it was regarded as inexpensive (Gumisiriza et al., 2017; Kumar and Sharma, 2017; Zhang et al., 2018).

3.3.5. Formation of toxic compounds

Table 7 provides information regarding formation of toxic inhibitory compounds. Biological pretreatment avoids formation of inhibitors of biogas production (Singh et al., 2014; Amin et al., 2017; Wagner et al., 2018). This is

Table 8. Aggregated comparison of the pretreatment methods.

Pretreatment method	Rate of hydrolysis	Energy use	Effectiveness	Cost	Generation of toxic compounds
Chemical	Fast	High	Very effective	Very expensive	Yes
Biological	Slow	Very low	Less effective	Cost-effective	No
Physical	Very fast	Very high	Moderately effective	Very expensive	Yes
Combined processes	Fast	Moderate	Effective	Cost-effective	Yes

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a positive attribute of biological pretreatment. While some chemical pretreatment methods do not produce inhibitory compounds (Sahito and Mahar, 2014; Den et al., 2018; Gumisiriza et al., 2017; Paul et al., 2018), 11 studies in Table 7 reported that chemical pretreatment causes formation of toxic compounds. As such, formation of inhibitory compounds is one of the disadvantages of chemical pretreatment. Production of inhibitors of methanogenic metabolism and growth can be ascribed to elevated levels of alkalinity and pH. The ultimate result is earlier process failure during biomethane production. The optimum pH range for methanogenic activity is 6.5 to 8.2. Alkaline addition provides buffering capacity and prevents inhibition during AD (Chen et al., 2015).

As shown in Table 7, physical processes generally lead to formation of inhibitory compounds. For instance, thermal pretreatment at temperatures above 160°C may lead to partial degradation of polysaccharides and lignin to phenolic and heterocyclic compounds (Zieminski and Kowalska-Wentel, 2017). However, some of the studies (Baeta et al., 2016; Kumar and Sharma, 2017) show that autohydrolysis and mill pretreatment do not generate toxic compounds. Seven studies in Table 7 show that combined processes generate inhibitory compounds, especially SE pretreatment. Zheng et al. (2014) reported that the efficacy of SE and extrusion may be affected by production of fermentation inhibitory substances such as furfural and hydroxymethylfurfural (HMF) due to sugar and lignin degradation. This can explain the CH₄ yield loss during high temperature SE pretreatment of late harvested hay observed by Bauer et al. (2014).

3.3.6. Aggregated results

Table 8 provides aggregated results of the five parameters for all the pretreatment methods. Putting together, biological methods have more techno-economic advantages across the five parameters compared to other methods. The advantages of biological pretreatments are tagged with low energy use, low cost, and ability to avoid formation of by-products that are toxic to methanogens. However, there is need to improve on the efficacy of biological pretreatment. Focus area should be enhancement of the rate of hydrolysis. Despite high effectiveness, the main limitations of chemical and physical methods are high energy use and cost, as shown in Table 8.

4. Conclusions

It is evident from this study that pretreatment methods used for CR are variable in their effects. As such, the multi-factor evaluation conducted in this study provides information that can assist selection of methods to use. Rate of hydrolysis, energy use, effectiveness, cost, and formation of toxic compounds are critical parameters that inform selection of a pretreatment method. Physical and chemical pretreatment methods have been utilized to some extent at industrial scale for delignification of CR to enhance biomethane production. However, these methods are energy intensive, expensive, not environmentally safe, and have the ability to generate toxic compounds including carboxylic acids, furans, and phenolic compounds which may be inhibit methanogenic activity. In comparison with other methods, biological pretreatment offers more techno-economic advantages. Biological pretreatment is regarded as inexpensive and low energy need process that can minimize formation of inhibitory compounds. Thus,

biological pretreatment is one of the most promising technologies for enhancing biomethane production of CR. Moreover, rigorous research is still needed for the development of novel microorganisms and more efficient pretreatment options for CR yielding potential results.

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