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Insight into the recent advances of microwave pretreatment technologies for the conversion of lignocellulosic biomass into sustainable biofuel

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ABSTRACT

The utilization of renewable lignocellulosic biomasses for bioenergy synthesis is believed to facilitate competitive commercialization and realize affordable clean energy sources in the future. Among the pathways for biomass pretreatment methods that enhance the efficiency of the whole biofuel production process, the combined microwave irradiation and physicochemical approach is found to provide many economic and environmental benefits. Several studies on microwave-based pretreatment technologies for biomass conversion have been conducted in recent years. Although some reviews are available, most did not comprehensively analyze microwave–physicochemical pretreatment techniques for biomass conversion. The study of these techniques is crucial for sustainable biofuel generation. Therefore, the biomass pretreatment process that combines the physicochemical method with microwave-assisted irradiation is reviewed in this paper. The effects of this pretreatment process on lignocellulosic structure and the ratio of achieved components were also discussed in detail. Pretreatment processes for biomass conversion were substantially affected by temperature, irradiation time, initial feedstock components, catalyst loading, and microwave power. Consequently, neoteric technologies utilizing high efficiency-based green and sustainable solutions should receive further focus. In addition, methodologies for quantifying and evaluating effects and relevant trade-offs should be develop to facilitate the take-off of the biofuel industry with clean and sustainable goals.

1. Introduction

The world has pursued a linear economic system since the industrial revolution under the assumption that natural resources are abundant and cheap. Energy is a key driver of a country's socioeconomic growth (Nižetić et al., 2019). A considerable portion of the current global energy supply, i.e., 78.4%, still originates from fossil fuels, such as petroleum, coal, and natural gas (McGlade and Ekins, 2015; Hoang, 2019), whereas renewable resources, including solar, hydroelectric, wind, and biomass, account for only 19% of the total world energy mix (Kumar et al., 2020). Fossil fuels remain a controversial source of energy given their nonrenewable nature and limited reserves (Kama, 2020), price volatility, and higher greenhouse gas emissions than renewable sources (Haites, 2018; Hoang et al., 2021a). Considering these limiting factors, finding alternative energy sources that will meet future needs is an urgent matter (Asomaning et al., 2018; Hoang, 2019). For these reasons, the new biobased economy model presents a huge opportunity for the 21st-century global economic system (Balakrishnan et al., 2019; Hoang et al., 2021b) because it promotes the sustainable consumption of natural resources through resource recovery and reclamation to meet current and future economic, social, and environmental sustainability goals (Hoang et al., 2018, 2021c,d). In this context, biomass energy derived from various renewable and readily available organic-based resources, for example, nonfood energy crops, agricultural residues, and food waste,

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represents a valuable opportunity for achieving sustainable economic growth and a well-balanced global economy (Fan et al., 2018). Examples of potential biomass-based feedstock for biorefinery operation strategies are given in Table 1.

Biomass resources are organic materials containing carbon, hydrogen, and oxygen atoms within their structures (Shahni et al., 2021). The consumption of biomass-derived energy sources, such as biofuels, that originate from a wide range of organic-based matter types has the potential to address the current energy–climate change dilemma (Nguyen et al., 2021). Moreover, the process of biofuel production and consumption results in zero net CO_2 emissions (Rosillo-Calle and Woods, 2012) because the amount of CO_2 released during combustion equals the amount of CO_2 that is captured by trees and plants through photosynthesis (Mandley et al., 2020). However, further research into advanced production bioenergy technologies is warranted to improve energy efficiency, optimize yield, and minimize environmental pollution (Bhatia et al., 2017; Lee et al., 2020). Among all the available technologies currently utilized in the production of bioenergy from biomass, the pretreatment of lignocellulosic biomasses is a crucial step in exposing cellulose and hemicellulose contents to enzymatic or chemical hydrolysis (Roy and Ray, 2019). Many technologies for lignocellulosic biomass pretreatment have been developed to solve this issue (Ong et al., 2019). Therefore, the present review considered the use of lignocellulosic biomasses to produce sustainable biofuel.

Pretreatment processes for lignocellulosic biomasses are categorized into physical (Sankaran et al., 2020), chemical (Behera et al., 2014), physicochemical (Sankaran et al., 2020), and biological methods (Sindhu et al., 2016). They include extrusion, sonication, milling, steam explosion, organosolv, ozonolysis, liquid hot water (LHW), ammonia fiber explosion, alkali pretreatment, acid pretreatment, wet oxidation, supercritical CO_2 explosion, ionic liquid, and microwave-assisted methods. Biomass pretreatment offers an effective and clean process for surface area expansion and improves accessibility to enzyme-binding sites (Karthikeyan et al., 2018). Pretreatment tends to split lignocellulosic biomasses down into their components and to overcome the natural recalcitrance of lignocellulosic biomasses by disordering lignin (Ong et al., 2020). It thereby increases the susceptibility of cellulose and hemicellulose to enzymatic hydrolysis for the production of simple sugars that are then fermented to produce biofuels (Sun et al., 2016). Microwave-based technology is believed to allow for redefining a large number of reactions wherein thermal effects are considered as a core factor. Moreover, the microwave-based heating method is a potential alternative technology to traditional heating because of its efficiency and easy operation (Milano et al., 2018). Its energy consumption is reduced because its processing time is 10 times shorter than that of other heating systems. It does not require the use of separation agents, solvents, or other auxiliary chemicals and does not generate wastes or smoke.

Table 1
Estimated biomass feedstock for biofuel production.

Feedstock	Major biomass producer	Estimated biomass (billion tons/year)	References
Forest-originated residues	Sweden, Canada, USA, Brazil, China	6.00 by 2050	Food and Agriculture Organization of the United Nations (2015)
Industrial wood residue	France, Canada, Sweden	2.28 by 2050	Ruth et al. (2013)
Municipal solid waste	USA, Japan, Germany China, Brazil,	2.60 by 2025	Institute (2012)
Sugarcane bagasse	Brazil, Thailand, Pakistan India, China	0.67 by 2022	OECD-FAO (2012)
Corn stover	USA, Argentina,	1.64 by 2022	(Food and Agriculture
Sorghum stover	Ukraine Brazil, China, India, Nigeria, Mexico	0.11 by 2022	Organization of the United Nations, 2015; OECD-FAO, 2012)
Commercial wood residue	France, Canada, Sweden	7.58 by 2050	Ruth et al. (2013)
Rice straw	Bangladesh, Vietnam, India, Indonesia	1.27 by 2025	(Food and Agriculture Organization of the United Nations, 2015; Gadde et al., 2009; Hoang et al., 2018)
Wheat straw	China, Russia, USA, India, France	1.10 by 2025	(Food and Agriculture Organization of the United Nations, 2015; Agricultural Biomass Sources, 2020; Shahbandeh, 2019)
Industrial corn	USA, Argentina, China, Brazil, Canada, Mexico	4.82 by 2050	Ruth et al. (2013)

Among the available methods for the conversion of lignocellulosic biomass into biofuels, the microwave-assisted physicochemical pretreatment has garnered special interest among researchers. Given its numerous advantages, several studies (Kostas et al., 2017; B. Kumar et al., 2020; Bichot et al., 2020; Rezania et al., 2020) have focused on microwave-based pretreatment technology for biomass conversion in the last several years. However, these studies did not comprehensively discuss or analyze microwave application or physicochemical pretreatment despite the importance of assessing these methods for bioenergy conversion. Therefore, this review aims to address the issues identified in the combined application of microwave heating and physicochemical methods in biomass pretreatment as reported in the current scientific literature. It further illustrates how combined microwave–physicochemical pretreatment resolves the challenges faced by each bioenergy conversion pathway. The utilization of the microwave–physicochemical method in the biomass energy industry is also mentioned in detail. This review presents special interests and benefits to specialists who already have a good understanding of the technology and underlying principles of the microwave–physicochemical method and nonexperts who may be interested in microwave pretreatment. The production of biofuels from lignocellulosic biomasses is believed to bring a bright future to developing countries by providing self-reliant energy supplies. In addition, ecological, socioeconomic, and energy security benefits can be also ensured by using biomasses for drop-in biofuel production. However, the balance in the production–consumption cycle of biomasses should be carefully considered.

2. Methodology for the collection, selection, and review of relevant data

This review aims to study the effect of microwave irradiation on physicochemical biomass pretreatment by reviewing high-quality papers in the literature. Databases, such as Google Scholar, Scopus, and Web of Science, were searched to retrieve relevant papers. Keywords, including “lignocellulosic biomass,” “microwave”, “physical pretreatment,” “chemical pretreatment,” and “physicochemical pretreatment,” were used to search for relevant publications that were suitable for this work. Subsequently, the bibliographies and references of the above-mentioned publications were filtered and collected to find additional pertinent papers. The main purpose of the current study is to outline the advantages and disadvantages and evaluate the economic aspects and the challenges of the microwave-assisted physicochemical method in biomass pretreatment.

3. Lignocellulose composition

Lignocellulosic biomasses are any plant-derived biomass that can be broadly classified into the following four categories: hardwood, softwood, farm waste, and grasses (Ostergren et al., 2014). Although

billions of tons of agricultural residues are generated annually worldwide, the majority of these residues are either burned in open fields or disposed of as wastes (Hoang et al., 2018). Lignocellulosic biomasses are organic materials that are naturally recalcitrant to biodegradation and contain cellulose, lignin, and hemicellulose as shown in Fig. 1 (Ethaib et al., 2015).

Cellulose is a linear polymer, which is unbranched and comprises linked β-(1 bis4)-D-glucopyranose units. The enzymatic digestibility of lignocellulosic biomasses is affected by cellulose crystallinity because highly ordered regions are structurally compact and hence naturally resistant to enzymes, acids, and swelling underwater. Analyses have shown that in comparison with crystalline cellulose (Ravindran and Jaiswal, 2016), amorphous cellulose can quickly be hydrolyzed at a higher rate (Kong-Win Chang et al., 2018). The polymerization degree of cellulose indicates the recalcitrance of biomass. Short cellulose chains are associated with rapid cellulose hydrolysis. Decreasing the polymerization degree can enhance the level of enzymatic hydrolysis; however, these outcomes must be carefully interpreted because changes in the polymerization degree always involve changes in other variables, such as crystallinity (Zhao et al., 2012).

Among the main components of biomasses, lignin is the most recalcitrant constituent of plant cell walls, which form a natural barrier that protects against the chemical and enzymatic degradation of plant cells. In contrast to cellulose and hemicellulose, lignin is not a carbohydrate and plays an important role in connecting the other two components and preventing the penetration of other solutions or enzymes (Bichot et al., 2018). Lignin is a three-dimensional polymer that contains phenolic groups with a structural and cross-linked design that provides structural integrity and rigidity to plant cell walls. The important aspects that are expected to affect the level of the biological degradation of lignocellulose by enzymes are the crystallinity and accessible surface area of cellulose, the polymerization degree of cellulose, and the degree of hemicellulose acetylation (Philippidis, 2018).

Hemicellulose possesses linear and branched linear polymers that originate from a class of different anhydrous sugars. Given its poor level of polymerization, hemicellulose has the poorest resistance to mechanical and chemical degradation (Chong et al., 2019). Hemicellulose is always present with cellulose and lignin in plant cell walls and is rarely found existing by itself in nature (Schutyser et al., 2017). It is often regarded as a natural barrier to enzymatic hydrolysis, although cellulose with its inherent crystallinity and lignin with its complex molecular structure play a much more effective role in protecting plant cell walls against degradation and decomposition. Enzymatic hydrolysis has been shown to achieve greater efficiency than either the acid hydrolysis or LHW methods in the pretreatment of biomass (Mohan et al., 2018). The fractions of the main components, such as cellulose, hemicellulose, and lignin, in lignocellulosic biomasses are given in Table 2.

4. Microwave mechanisms and reactors

4.1. Microwave process and mechanisms

Microwaves are electromagnetic energy waves with wavelengths and frequencies ranging from 1 m to 1 mm and 0.3 GHz–300 GHz, respectively (Bayat et al., 2021). During transmission, microwaves are transformed into suitable frequencies from which energy can be absorbed by the material being heated (Puligundla et al., 2016). Constant changes in electrical fields cause dipoles in polar liquids to undergo continuous alignment and realignment (Bundhoo, 2018). The continuous changes in dipoles, coupled with the transfer of ions, generate friction within the material. Friction then generates internal energy in the form of dispersed heat in the material being heated (Surati et al., 2012). In contrast to that in conventional heating, energy conversion in microwave irradiation results directly in volumetric heat generation within the target material and not through the material surface (Mohan et al., 2018). A flow diagram of microwave irradiation pretreatment is illustrated in Fig. 2.

Microwave irradiation models have been applied in organic synthesis (Isikgor and Becer, 2015), chemical catalysis, and solid-state reactions (Pandey et al., 2014). Microwave irradiation application models have a beneficial effect on biomass metabolism. Several investigations have thus been performed to examine the appropriate operating parameters of microwave pretreatment to optimize the conditions for biomass pretreatment with increased efficiency (Zheng et al., 2014). Many experts have referred to energy savings as a potential benefit of microwave heating (Wei et al., 2019). Microwaves directly penetrate heating container walls and effectively heat materials (Shah and Mohanraj, 2014). Microwave-induced thermal energy is created at the atomic level through the direct conversion of electromagnetic energy into heat; energy is thus uniformly dissipated across the material as illustrated in Fig. 3. The temperature of materials irradiated by microwaves is drastically higher than that of the surrounding environment (Mushtaq et al., 2014).

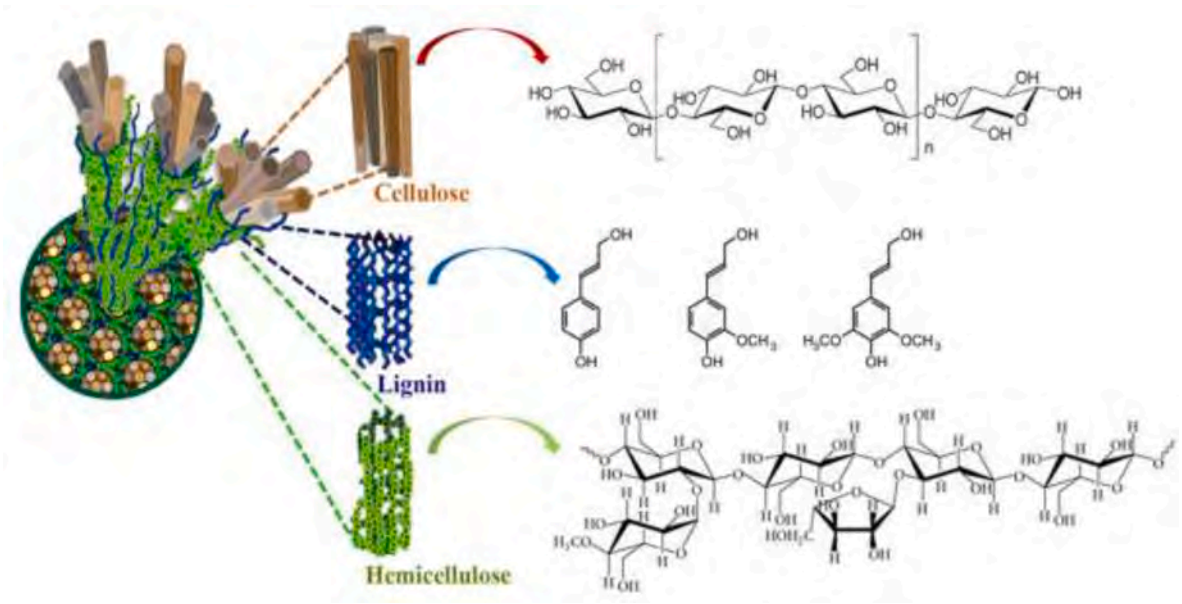

Fig. 1. Structure of lignocellulosic biomass along with cellulose, hemicelluloses, and lignin.

Table 2
The fraction of the main components in lignocellulosic biomass.

Biomass name	Composition of lignocellulosic biomass, %			References
	Cellulose	Lignin	Hemicellulose	
Oak	43.20	35.40	21.90	Yu et al. (2017)
Spruce	47.10	29.20	22.30	
Rubberwood	39.56	27.58	28.43	Khan et al. (2018)
Palm oil frond	37.32	26.05	31.89	
Japanese cedar	33.50	52.70	13.80	Muranaka et al. (2017)
Hybrid poplar	15.50–16.30	40.30–47.30	16.60–22.60	Mussatto & Dragone (2016)
Pinus armandii Franch	24.10	48.40	17.80	Wang et al. (2016)
Barley straw	36.00–43.00	6.30–13.10	24.00–33.00	Liu et al. (2017)
Rice husks	15.50–26.00	12.00–29.30	28.70–40.00	Serna et al. (2016)
Sugar cane bagasse	42.00	20.00	25.00	Zadeh et al. (2020)
Hybrid poplar	17.00	52.00	25.00	Kim et al. (2016)
Corn stalk	34.85	8.16	29.87	Wang et al. (2019)
Corn stovers	32.70	25.40	20.90	Gong et al. (2019)
Miscanthus	52.10	18.60	21.30	Kashcheyeva et al. (2019)
Olive pomace	19.00	40.00	22.00	Álvarez et al. (2018)
Hazelnut shell	30.00	38.00	23.00	
Bamboo	46.50	25.70	18.80	Chen et al. (2017)
Banana waste	13.20	14.00	14.80	Kumar & Sharma (2017)
Bamboo leaves	35.00	34.14	25.60	Huang et al. (2016)
Walnut shell	23.30	53.50	20.40	de Caprariis et al. (2017)
Groundnut shell	37.00	28.00	18.70	Subhedar et al. (2018)
Pistachio shell	15.20	29.40	38.20	
Cocoa pods	32.30	21.44	27.70	(Yetri1 et al.)
Sago palm bark	42.60	19.20	24.30	Ethaib et al. (2020)
Waste cotton	36.00	16.70	18.00	Ramamoorthy et al. (2020)
Coconut coir	32.80	22.10	44.20	Subhedar et al. (2015)

Materials that are used for microwave heating are categorized into three groups, namely, insulators, absorbers, and conductors. For example, transparent materials, such as glass and ceramics, used for microwave heating are known as insulators; highly conductive materials, including metal, are considered as conductors; and materials that can absorb and convert microwave energy into heat are considered as absorbers (Hassan et al., 2018). Materials that are subjected to microwave radiation have higher core temperatures than outside surface temperatures (Mohan et al., 2018). The majority of biomasses is typically regarded as low-loss materials, and the use of materials, such as graphite, charcoal, activated carbons, and pyrites, can help accelerate heating. Salema et al., (2017) examined the dielectric characteristics of various biomasses in agricultural and wood-processing industries and identified them as low-loss dielectric materials. They also reported that the utilization of microwaves effectively disrupts lignin and cell walls, thus facilitating enzymatic hydrolysis. Thermal effects occur when microwaves cause dipoles to align with the oscillating electric field and thereby break hydrogen bonds (Bichot et al., 2020). The rupture of hydrogen bonds can lead to the destruction of cell walls and the fractioning of cellulose chains. As a result, the crystal configuration of cellulose components is disrupted, therefore promoting hydrolysis. This effect is illustrated in Fig. 4.

Selective heating by microwaves often permits direct biomass heating without the use of hot gases or ablative surfaces in a cool setting. Moreover, volumetric heating in the microwave method enables overcoming traditional heat transfer limits, leading to considerably increased heating rates, small processing device sizes, and large particle size processing capability (Motasemi and Afzal, 2013). The total processing system can exploit volumetric heating because the general workflow could be substantially simplified with reduced requirements for raw material, equipment size, and hot gas handling and recuperation systems. Although its energy requirements are similar to those of traditional processes, microwave heating provides great potential for decreasing the operating costs of processing systems and increasing production capacity. These advantages cannot be obtained with traditional systems. Improvements in microwave irradiation heating mechanisms have resulted in the many other merits of microwave heating over traditional heating as tabulated in Table 3.

4.2. Microwave reactors

Microwave reactors are mainly designed to improve the capability to perform chemical reactions on a laboratory scale under controlled conditions. Reactor design plays an important role in modeling and construction. Microwave equipment includes some primary

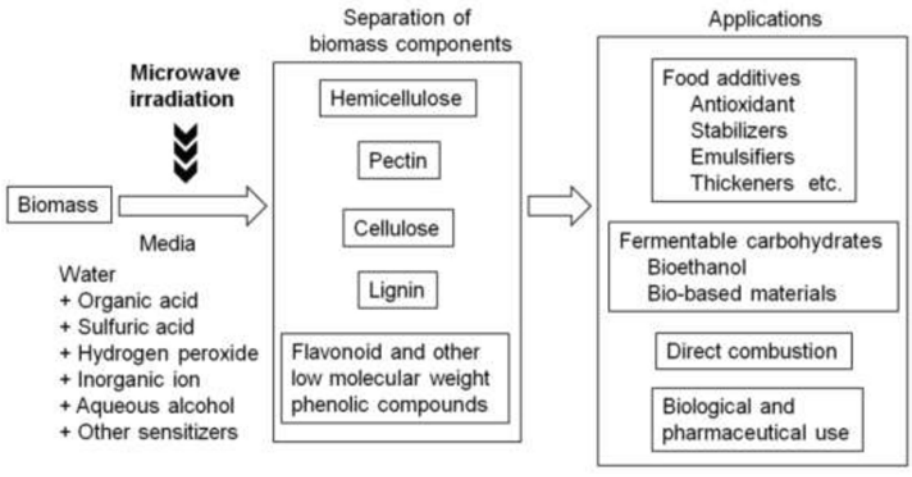

Fig. 2. Microwave pretreatment process for biomass refinery (Tsubaki and Azuma, 2011).

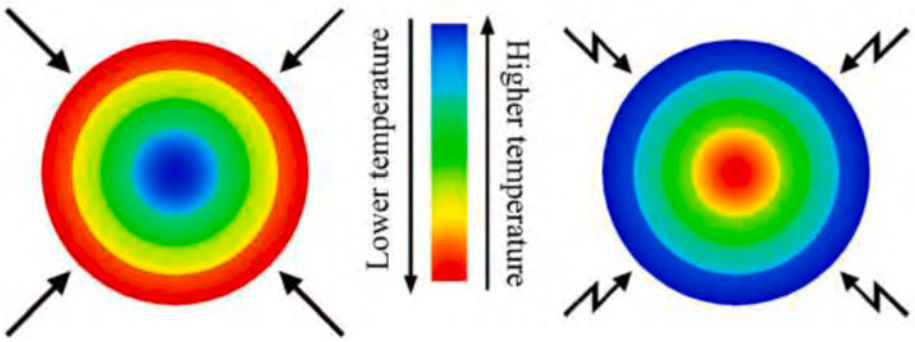

Fig. 3. The difference in the distribution mechanism of temperature between conventional and microwave heating (Mushtaq et al., 2014).

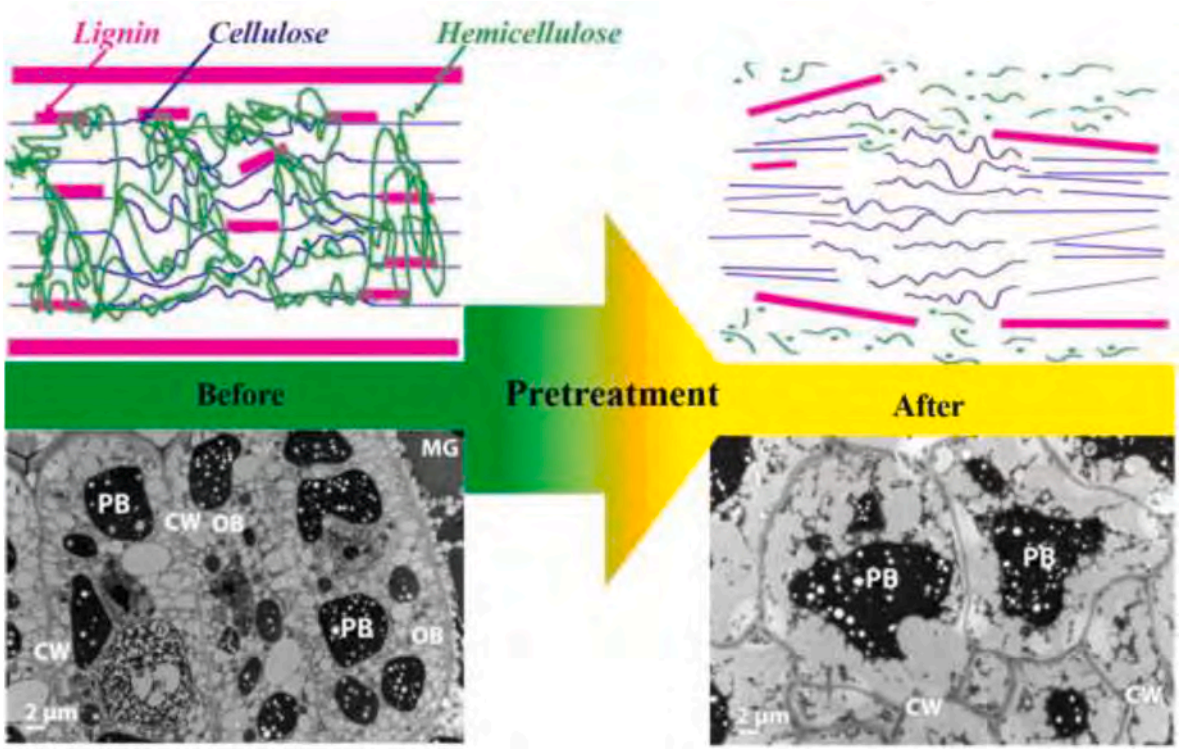

Fig. 4. Pretreatment process of lignocellulosic-based materials by microwave (Adapted from reference (Puligundla et al., 2016) (Cantero et al., 2019).

Table 3
Advantages and disadvantages of microwave heating compared to conventional heating (Saini et al., 2015).

Merits:
The heating rate is rapid and efficient, the time for reaching the required temperature is only seconds to minutes.
Selective efficiency is high for polar substances.
The in-core volumetric heating is conducted uniformly.
The overheating phenomenon on the surfaces could be avoided due to indirect contact with the heat source.
Local temperatures in the material are much higher than that measured in the bulk because of the inhomogeneity effect of MW, causing hot spots.
The working efficiency is much dependent on the material properties.
The heating system is controlled precisely.
Demerits:
Selective efficiency is low for apolar substances.
There are no hot spots.
The working efficiency is less dependent on material properties.
The heating system is less controllable.

components, such as a generator, cavity, waveguide, applicator, and resonator (Fig. 5a) (Mohan et al., 2018). Parameters, including type (batch/flow), mode (mono/multi), size, materials used, microwave frequency, homogeneity, and penetration depth (shape), should be considered and are interconnected in many cases. Microwave instruments are often divided into two categories: monomode reactors and multimode reactors (Dąbrowska et al., 2018). Mode type influences cavity size and shape, penetration depth, field homogeneity, and frequency. Monomode indicates that microwave distribution in the cavity or a specific spot is very well defined. Typically, small cavities are used, and the reactor is placed directly inside the waveguide with the highest concentration of microwaves. Monomode reactors can irradiate a single reactor vessel only, whereas multimode reactors can irradiate multiple vessels concurrently. A monomode instrument generates a highly homogeneous energy field with high power density, resulting in rapid heating.

The potential to homogenize rotating equipment with the electromagnetic field provides considerable advantages to reactors with multimode applicators. Various microwave inlet ports can be set up within multimode reactors, which are more beneficial than reactors with

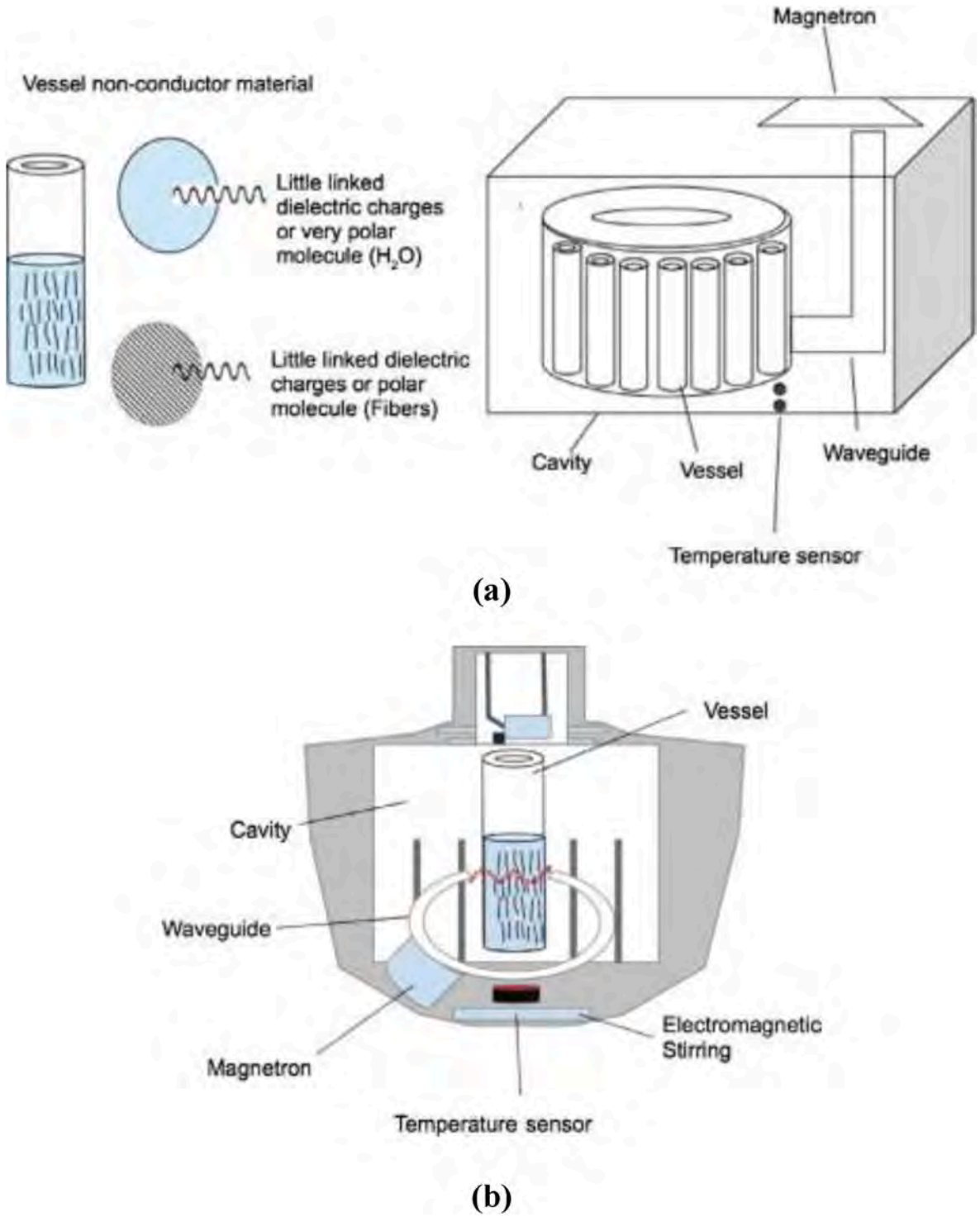

Fig. 5. (a) Operating diagram of multimode microwave reactor, (b) mono mode microwave reactor (Aguilar-Reynosa et al., 2017).

a monomode applicator. Moreover, their large dimensions enable multimode reactors to work with high capacity. Monomode microwaves (Fig. 5b) specifically target microwaves by using a waveguide to penetrate samples; given that they satisfy wavelength requirements by directing waves into the material, they boost radiation efficiency. Furthermore, designing and presenting analytical solutions for monomode reactors are easy. Nonetheless, monomode reactors possess limited dimensions that result in considerably high manufacturing expenses per load volume (Cherbański and Rudniak, 2013).

A typical domestic microwave is an example of a multimode nonstirred batch system. The majority of laboratory-scale systems are of either mono- or multimode batch construction with implemented stirring and an option for performing chemistry in an open vessel or pressurized reactor. In the case of focused reactors, power and temperature can be continuously regulated with consistent radiation dispersion because it is uniform in the cavity; thus, additional reproducible outcomes are produced. However, further study related to the performance of a continuous microwave reactor is necessary. Peng et al., (2014b) developed an original continuous flow microwave reactor for lignocellulosic biomass pretreatment on a pilot scale. The reactor was 3 m in

size, 0.76 m in length, and 1.75 m in width with the maximum working temperature of 300 °C and the maximum pretreatment capacity of 5 kg/h. Périno et al., (2016) reported the pilot-scale extraction of polyphenols in lettuce by using a MAC-75 multimode microwave reactor outfitted with four magnetrons. The reactor had a capacity of 150 L and was coupled with a rotating PTFE drum, thus allowing for the maximum load of up to 75 L of biomass feedstock.

5. Effects of microwave irradiation on the pretreatment process of biomass

The purpose of microwave pretreatment is to break up lignocellulosic materials and monitor lignin, hemicellulose, and cellulose contents (Mohan et al., 2018). The decomposition of polymers and the disintegration of the crystalline arrangements of cellulose molecules (Mohan et al., 2018), as well as the solubilization of other organics, such as proteins, carbohydrates, and lipids, are monitored (Passos et al., 2015). Several follow-up studies investigated the effect of single or combined microwave irradiation on hydrolysis prior to biofuel conversion.

5.1. Effects of microwave irradiation on particle size and surface area

Microwave-based biomass pretreatment causes the rupture of cell walls by increasing pressure and temperature (Mohan et al., 2018). The fragmentation of the lignocellulosic structure reduces particle size and increases outer surface area (Hassan et al., 2018). The surface area of microcrystalline cellulose (180–200 μm) (Peng et al., 2014a) increased by 56% after 20 min of microwave pretreatment at 800 W. Surface area was further increased by 75% when NaOH-supported microwave irradiation was used. In the microwave pretreatment of sugarcane bagasse, particle size decreased and surface area increased with the increase in temperatures and time (Chen et al., 2011). The SEM of raw and pretreated sugarcane bagasse is shown in Fig. 6. After microwave pretreatment at various solution temperatures, the bagasse structure was damaged as demonstrated by the appearance of several cracks on its surface (Fig. 6b). The disruption of the lignocellulosic structure became increasingly pronounced at 160 °C (Fig. 6c). As surface area increases, biochemical reaction frequencies tend to increase with the enhanced accessibility of cellulose molecules to hydrolysis, thereby leading to an improvement in biofuel production (Cheng and Liu, 2010).

5.2. Effects of microwave irradiation on lignin, hemicellulose, and cellulose contents

A large number of studies have reported on delignification related to the fragmentation of lignocellulosic structure because of the thermal effects of microwave irradiation in various materials, i.e., sorghum biomass (Chen et al., 2012a; Choudhary et al., 2012), beechwood (Verma et al., 2011), kans grass and giant reed (Komolwanich et al., 2014), triticale straw (Monteil-Rivera et al., 2012), and oil palm empty

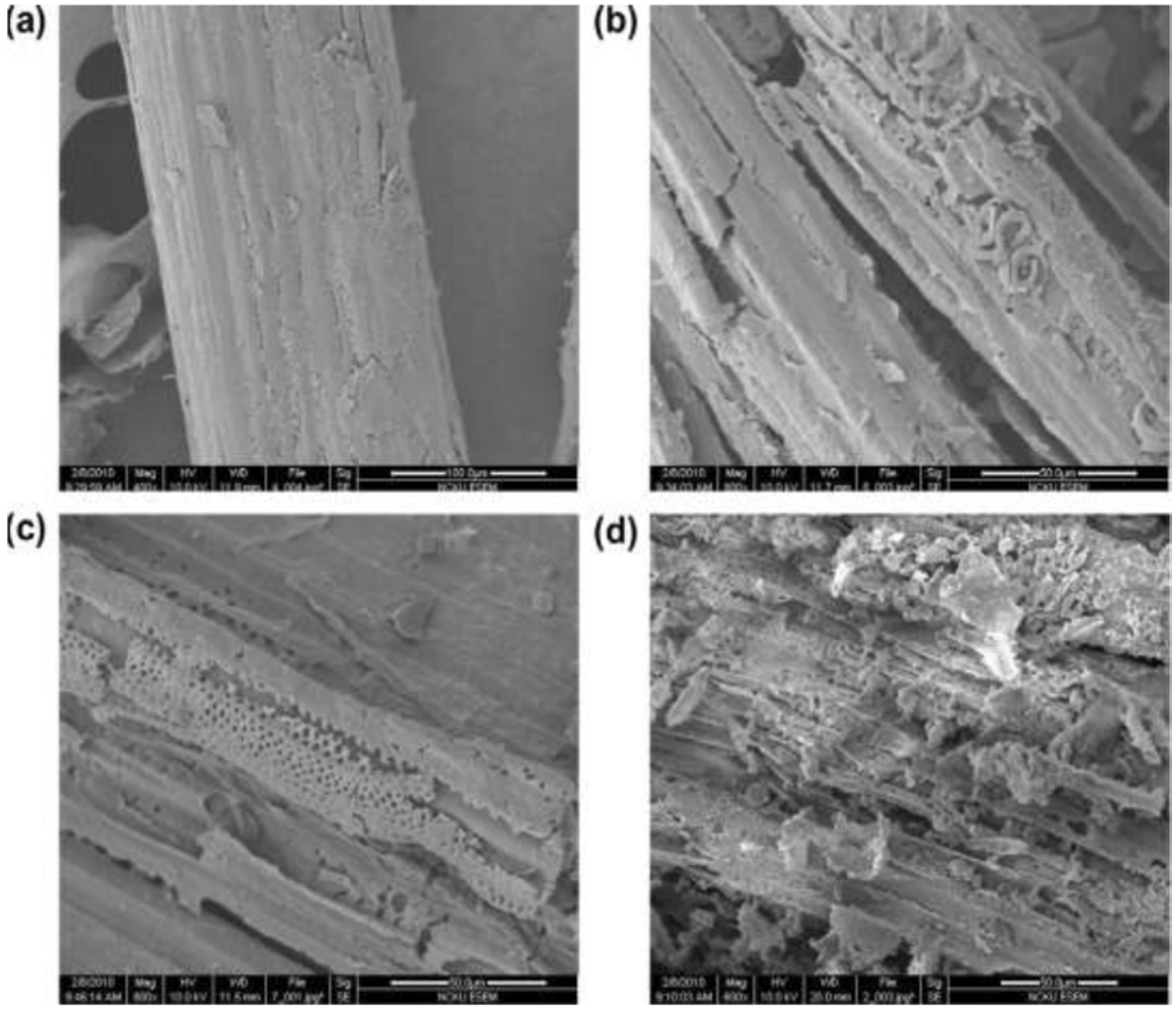

Fig. 6. SEM of raw bagasse (a) and pretreated bagasse by microwave at various reaction temperatures 130° (b), 160° (c), 190° (d) after 5 min of heating time (Chen et al., 2011).

fruit bunch (Akhtar et al., 2015). Other studies demonstrated that hemicellulose was removed from wheat straw (Fan et al., 2006), and cornstalk (Li et al., 2014) after pretreatment. Hu and Wen (2008) examined the effect of the duration and temperature of microwave irradiation in biomass pretreatment on lignin removal from switchgrass and found that delignification improved with the prolongation of pretreatment time and the increase in temperature. In comparison with alkaline pretreatment methods, microwave-assisted acid pretreatment methods have a greater impact on the removal of hemicellulose (Karungi et al., 2020), whereas microwave-assisted alkaline pretreatments have more significant effects on lignin extraction.

5.3. Effects of microwave irradiation on the depolymerization and crystalline structure of cellulose

Biomass pretreatment aims to reduce cellulose polymerization and crystallinity (Chen et al., 2011). The thermal effects of microwave irradiation can help depolymerize cellulose by breaking hydrogen bonds and can continue to increase the fracture of the cellulose molecules' crystalline arrangement. Ha et al., (2011) observed that in the pretreatment of cellulose in ionic liquids, polymerization declined when the microwave power was increased to 50 W. Likewise, Hu et al., (2012) found a 9.2% reduction in the crystallinity index (CI) in microwave pretreatment. The CI is a complex parameter for investigating pretreatment productivity.

Reductions in the CI reflect the breakdown of the crystalline cellulose structure and demonstrates pretreatment efficiency, whereas increments in the CI indicate the disruption of lignin structure and increased accessibility to cellulose molecules, again demonstrating the efficacy of the pretreatment (Li et al., 2014). The CI increased after the microwave-supported steam explosion of maize stover (Pang et al. 2012, 2013). The more noticeable effect of microwave irradiation on the amorphous area than on the crystalline area of cellulose was observed to contribute to increased crystallinity and decreased lignin and hemicellulose contents (Q. Li, Guo, and Liu, 2014; Binod et al., 2012). Mosier et al., (2005) argued that hydrolysis could not be solely limited by crystallinity, whereas polymerization level was found to have a more significant effect than crystallinity in hydrolysis (Ha et al., 2011). The influences of microwave irradiation on crystallinity are presented in Table 4.

5.4. Effects of microwave irradiation on organic matter solubilization

The disturbance, breakdown of cell membranes and walls, and organic transfer from the particle to the soluble phase occur due to the thermal effects of microwave irradiation (Kang et al., 2020). Most studies on microwave pretreatment for the enhanced solubilization of organic material focused on sludge (Yi et al., 2014; Park and Ahn, 2011; Chang et al., 2011; Ebenezer et al., 2015; Yang et al., 2013). Microwave irradiation increased the solubilization of kitchen waste by 40% (Marin et al., 2011). Similarly, the solubilization of waste (Shahriari et al., 2011) and organic fraction of municipal solid waste (Shahriari et al., 2013) improved after microwave pretreatment.

5.5. Effects of microwave power

In contrast to the conventional heating pretreatment method, microwave-assisted pretreatment is believed to produce high power densities, thus resulting in the increase in production rate and the reduction of costs. Xu et al., (2011) reported that microwave power had a positive effect on the recovered glucose yield from the alkali-based pretreatment of wheat straw. However, increasing microwave power in the pretreatment of sugarcane bagasse through the combination of microwave–acid methods changed sugar yields insignificantly (Fig. 7).

Fig. 7 shows that the increase in microwave power and the decrement in sugar yield under microwave-assisted acid pretreatment are inversely related. The change levels of reducing sugar yields at various microwave power are different. Moreover, pretreatment time decreases with increasing microwave power. The polymerization degree of cellulose decreases due to the collapse of long-cellulose chains into short molecular groups (Ling et al., 2019). The optimal microwave power has been reported to be 180 W, which allows sufficient time for biomass pretreatment with the negligible volumetric losses of the liquid phase (Nomanbhay et al., 2013). Nonthermal and thermal microwave effects have been investigated via comparative experiments conducted in a glass reactor and an in-house-built jacketed glass reactor that was constructed to prevent sample heating (Bichot et al., 2020). Only thermal microwave effects were observed under the test conditions. Microwave pretreatment with enhanced pressure and power density was also evaluated to maximize the biosolubilization of biomass components. Cell walls were greatly affected by the solubilization of 33% of hemicellulose content. These results pave the way for industrial applications.

5.6. Effects of microwave irradiation time

Microwave irradiation time or reaction time is the main factor that affects the pretreatment process (Gan et al., 2020). Biomass digestibility is remarkably affected by the interactive effect between microwave power and reaction time through the enhancement of cellulose

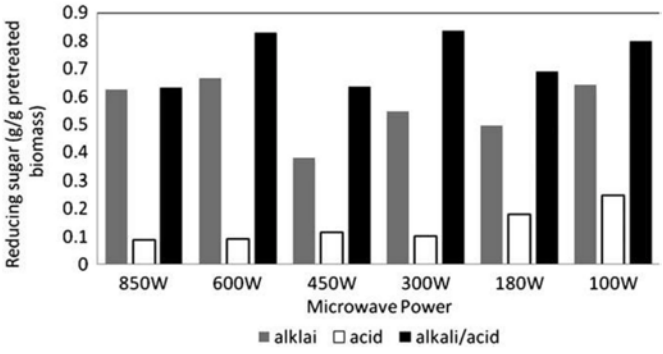

Fig. 7. Effects of microwave power on reducing sugar yield for biomass pretreatment process based on methods of acid, alkali, and alkali/acid (Binod et al., 2012).

Table 4
The crystallinity index of biomass after and before pretreatment by microwave.

Biomass type	Pretreatment methods	CI before pretreatment, %	CI after pretreatment, %	Ref.
Microcrystalline cellulose	Microwave (P = 800 W, τ = 20 min)	76.04	73.34	Peng et al. (2014a)
	Microwave (P = 800 W, τ = 20 min) +1% NaOH		70.70	
Cornstalk	Microwave time = 90 min + 0.12 g NaOH	38.70	52.3	Li et al. (2014)
Water hyacinth	Microwave+ 0.2% NaOH	16.00	13.00	Lin et al. (2015)
Rice straw	Microwave + 0.5% NaOH at 140 °C temperature, 15 min	44.03	56.54	Boonsombuti et al. (2019)
	Microwave + 2% HNO_3 at 100 °C temperature, 7 min		50.99	
Cassava stem	Dilute acid pretreatment	35.4	56.8	Martín et al. (2017)
	Microwave + alkali pretreatment	39.56	47.15	Kamalini et al. (2018)
Cassava rhizome	Microwave + alkali pretreatment	30.39	42.72	Sombatpraiwan et al. (2019)

digestibility and hemicellulose removal. However, increasing the reaction time and power of microwave irradiation can lead to excessively high temperatures in the used materials, resulting in the decomposition of released sugar. Karunanithy et al., (2014) carried out the comparative experiments on the pretreatment of switchgrass and big bluestem via the extrusion–microwave and extrusion pretreatment methods. Compared with the extrusion pretreatment method, the extrusion–microwave method increased sugar recovery from big bluestem and switchgrass by 14.2% and 15.2%, respectively. However, increasing the microwave exposure time to 10 min had a modest influence on sugar recovery. Komolwanich et al., (2014) revealed that the increase in temperature could result in a reduction in monomeric sugar yields. Notably, they identified reaction temperature as the most critical parameter in the release of monomeric sugars. Their finding was also consistent with previously reported results (Jackowiak et al., 2011) for the microwave pretreatment of switchgrass.

6. Microwave-assisted physicochemical pretreatment of biomasses

6.1. Combined mechanical and microwave methods

Decreased cellulose crystallinity and increased delignification are considered as the important results of physical pretreatment. Grinding the biomass into small particles has a positive effect on pretreatment because it increases the available specific surface area and enhances accessibility by the enzyme for subsequent hydrolysis (Bian et al., 2014). Biomass pretreatment by combining ball milling with microwave irradiation is innovative, ecofriendly, and productive (Peng et al., 2013). The glucose yield of the combination of 1 h of ball milling and 20 min of microwave irradiation was the same or higher than that of BM3 (ball milling for 3 h) and BM6 (ball milling for 6 h). In addition, the energy consumption of the combination of ball milling and microwave irradiation was 54.8% and 77.4% lower than that of BM3 and BM6, respectively. Furthermore, this method did not involve the use of any chemicals.

The necessary particle downsizing should be performed with the thermochemical pretreatment used for lignocellulose conversion instead of only with mechanical pretreatment to achieve economically feasible biomass alteration (Vidal et al., 2011). The ideal disposal of lignocellulose with the highest delignification rate and lowest loss of sugar was conducted with particle sizes of 1–2 mm (Chen et al., 2012a). Hu and Wen (Hu and Wen, 2008) reported disposal rates of 63%–70% with particle sizes of 1.0–2.0, 0–0.5, 0.5–0.25, and <0.25 mm for switchgrass pretreated through the microwave-supported alkaline method. However, this process was energy intensive, therefore increasing its associated expense. Karunanithy et al., (2014) milled switchgrass and bluestem into particles with sizes ranging from 0.3 mm to 1.2 mm for switchgrass and 0.4 mm–0.8 mm for bluestem. Extrusion coupled with microwave pretreatment enhanced the recovery of xylose, total sugar, and glucose from switchgrass by 16.7%, 21.4%, and 27.0% respectively.

In contrast to those from specimens subjected to microwave pretreatment, the recoveries of glucose, xylose, and total sugar from big bluestem pretreated through enzymatic hydrolysis were 24.9%, 19.7%, and 17.3% respectively. Camani et al., (2020) subjected eucalyptus waste to microwave pretreatment after milling (Fig. 8). They compared the efficiency of the combined microwave pretreatment method with that of the pretreatment method without microwave heating. The pretreatment method combined with microwave heating significantly shortened the duration of the pretreatment process and yielded cellulose with 99.1% purity. Although microwave heating showed high interactive efficiency for enhancing the recovered sugar rate from powdered samples, concerns regarding energy consumption persist. The utilization of controlled conditions for microwave pretreatment will minimize energy requirements and boost product yield by eliminating unwanted excess energy via selective target heating.

6.2. Combined microwave and ultrasound methods

Microwave and ultrasound pretreatment methods for biomass pretreatment reduce particle size and increase the exposed surface area and availability of cellulose, hemicellulose, and oligosaccharides (Trobe and Burke, 2018). Microwave and ultrasound pretreatment methods accelerate hydrolysis and the biodegradation of agricultural residues and sewer sediments adopted for biogas production. Ultrasonication has been proven to be a more efficient pretreatment than microwave heating only (Alagöz et al., 2018). Several patents (Olsen, 2013; Augustin et al., 2013; Gjermansen, 2014) have asserted the efficiency of the combination of microwave heating and ultrasonication for biomass pretreatment. This combined method has been proven to diminish waxes and lignin selectively, and microwaving strips the waxy coating from the biomass surface to boost the utilizable area for enzyme activity. The combination of ultrasound and microwave energy represented an additional treatment method for heating biomass internally that enhanced the hydrolysis of hemicellulose (North, 2015).

The use of ultrasonication (20 and 60 kHz for 10 min for the former and 20 min for the latter) in combination with microwave heating (400 and 600 W for 100 and 130 min, respectively) in the hydrothermal pretreatment of corncobs generated high yields of hydrolyzated maize xylose (Junli et al., 2016). Patent innovators have recently documented the superimposed dual power of ultrasonication and microwave assistance in ionic liquids. The combination of microwaving at 15–1000 W microwave power and 1500–3000 MHz frequency and ultrasonication at 200–1000 W and 15–30 KHz could effectively eliminate lignin, improve

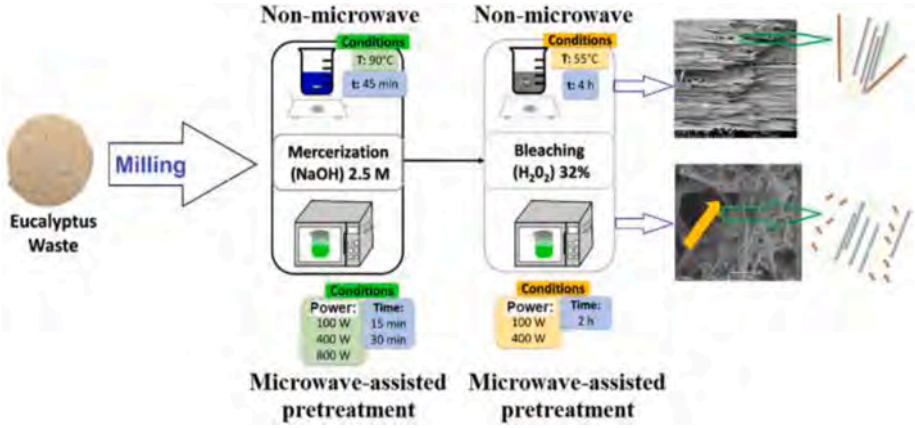

Fig. 8. Efficiency comparison in lignocellulosic biomass by the mechanical pretreatment method with microwave and non-microwave (Camani et al., 2020).

cellulose enzyme hydrolysis, and remarkably boost fermentable sugar (glucose and xylose) output (Xing et al., 2017). Chowdhury and Abd Hamid (Chowdhury and Abd Hamid, 2016) carried out a series of experiments on dried jute stalk to synthesize nanocrystalline cellulose with a high CI through the combination of microwaving and ultrasonication. They found that the CI for the H_2SO_4-based hydrolysis process could reach 88.32% and that for the $[EMIM]^+Cl^-$-assisted hydrolysis process could reach 83.42%.

6.3. Combined microwave and plasma methods

Plasma applications are commonly studied as methods for biomass transformation. Nonthermal plasma was applied under atmospheric pressure to modify the surfaces and structural characteristics of various biomasses (Puligundla et al., 2016). Flowing afterglow plasma pretreatment was introduced to replace discharge plasma for the isolation of cellulose from the lignin–hemicellulose complex housed in biomasses. Such a change expanded the exposed surface area and improved biofuel production (Raud et al., 2019). Pretreating sugarcane bagasse with a 2.45 GHz microwave plasma torch at atmospheric pressure in mixed Ar steam–air and pure-air plasmas resulted in substantial surface alteration and lignin layer destruction resulting from the etching effect of plasma (Bundaleska et al., 2012).

Zanini et al., (2005) found a considerable modification in lignin chemical structure when cold Ar-plasma pretreatments were used to alter the structure of lignocellulosic fibers. Plasmas generated by microwaves at atmospheric pressure possess many advantages over those generated by dc and RF electric fields; these advantages include good energy transmission to the plasma, near-100% coupling efficiency between the microwave and plasma, low power consumption, and throughput atmospheric processing (Henry et al., 2017). Delikonstantis et al., (2019) presented the potential of utilizing microwave plasma in biomass gasification (Fig. 9). Surprisingly, up to 89% of carbon conversion efficiency was reported. The recent application of a plasma source established in a microwave tornado-type air–water configuration operating at 2.45 GHz demonstrated that the high reactivity of plasma in an air–water environment offers a variety of durable functional active species that are capable of breaking down cellulosic wrapping (Bundaleska et al., 2013). Reactive species containing oxygen, including HNO_2, single-delta molecules, OH radicals, and NO_2, were reported to have a significant influence on the pretreatment process.

6.4. Microwave-induced torrefaction of biomass

Torrefaction is known as a biomass pretreatment method for improving and enhancing the quality of lignocellulosic biomasses via thermo–chemical treatment in an inert environment (Dai et al., 2019; Olugbade and Ojo, 2020). The many advantages of torrefied biomass over raw biomass include decreased mass, improved grindability, increased heating value, and biodegradation resistance (Bach and Skreiberg, 2016; Q. Hu et al., 2018). Torrefaction, which is also regarded as hydrothermal carbonization, can be categorized as dry or wet (Wnukowski and Owczarek, 2015; Heikkinen et al., 2019). The working temperature of dry torrefaction (DT) ranges from 200 °C to 300 °C (Acharya et al., 2012; Gan et al., 2020) and that of hydrothermal carbonization is between 180 °C and 260 °C (Lynam et al., 2011). Torrefaction produces a small amount of liquid and gas, in addition to the primary products of char (Bach and Skreiberg, 2016).

6.4.1. Microwave-induced DT of biomasses

DT, which is deemed to be the most benign biomass pretreatment method among the thermo–chemical process (Hossain et al., 2020). It is a low-temperature (200 °C-300 °C) method that typically occurs for approximately 1 h or less in the absence of oxygen (Ciolkosz and Wallace, 2011). The products obtained from this method are solid products with enhanced combustible elements and greater energy density than unpretreated biomass (Chen et al., 2020). DT causes adjustments in biomass as a consequence of hemicellulose thermal decomposition and

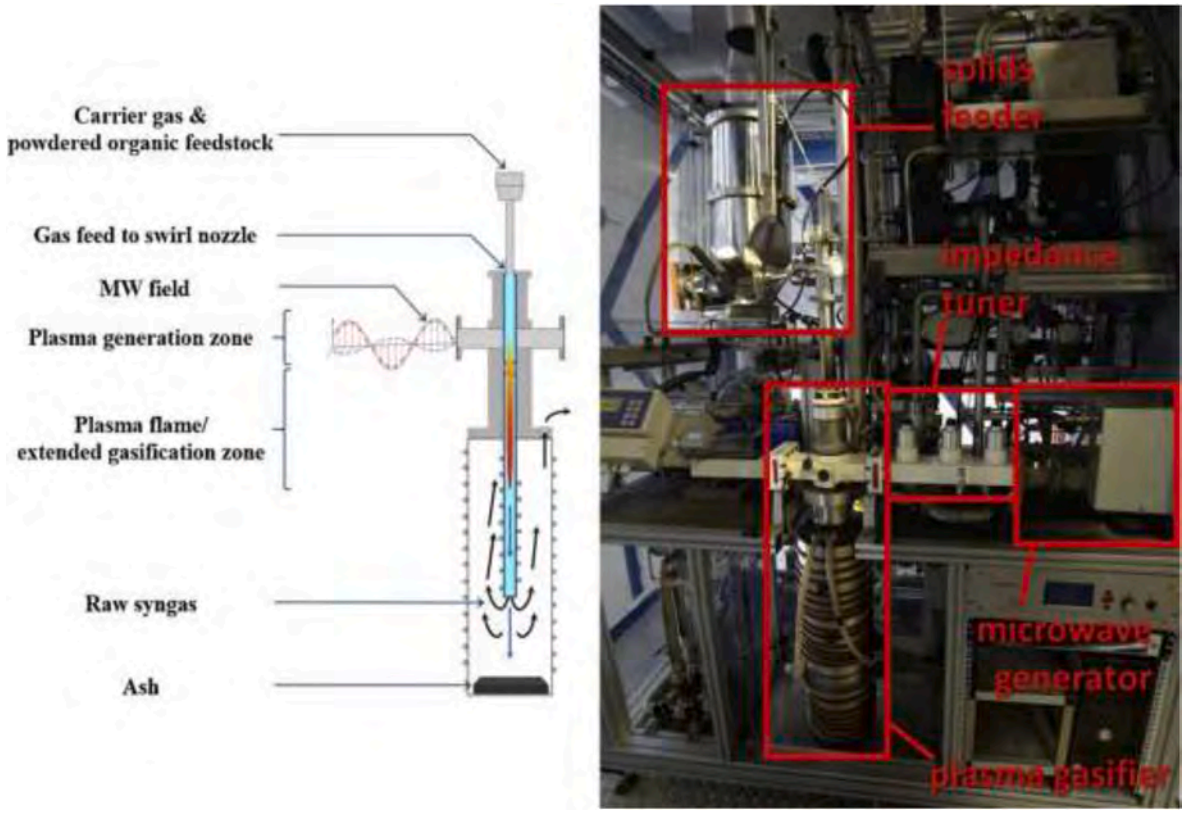

Fig. 9. Diagram for biomass pretreatment based on microwave plasma (Bundaleska et al., 2013).

partial lignin and cellulose depolymerization, thus providing torrified biomass that is rich in carbon and high in calorific value but low in overall mass (Paoluccio, 2011).

Most of the investigations on DT biomass used the traditional heating method as a heating process and analyzed the effect of temperatures and durations on the efficiency of DT biomass pretreatment (Kostas et al., 2017). The influence of various experimental test conditions on the characteristics of torrified biomass had been assessed in a good number of studies. The investigated conditions included microwave power, biomass moisture content, processing time, and particle size. The elemental investigation of torrefied biomasses demonstrated the effect of the microwave drying method on proximate and ultimate analyses (Amer et al., 2019). At all torrefaction temperatures, any biomass, except for cotton stalks, torrefied via traditional oven drying had higher carbon and lower oxygen contents than their corresponding microwave-dried samples.

Most of the relevant studies demonstrated the considerable influences of microwave power and reaction time, as well as the negligible effects of biomass water content on torrefaction (Satpathy et al., 2014). Given that water is strongly susceptible to microwave heating, biomass torrefaction could be conducted without a predrying phase (He et al., 2018). The mass and energy yields obtained through microwave torrefaction were comparable with those obtained through conventional heating methods at lower power consumption and shorter reaction times, indicating the low energy input of microwave-based torrefaction (Huang et al., 2012). Microwave-induced DT also greatly improved fuel properties, such as the energy intensity of the torrefied biomass, the ratios of hydrogen/carbon and oxygen/carbon, and higher heating value (HHV) (Ren et al., 2012).

Some studies had compared the HHV of microwave-torrefied biomass with that of raw biomass (Ren et al. 2013, 2014). Microwave-assisted torrefaction experiments mainly evaluated the influences of microwave energy intensity, retention times, and working temperature on the HHV of torrefied biomasses (Satpathy et al., 2014; Y.-L. Lin, 2015). Microwave torrefaction dramatically enhanced the grindability and hydrophobicity of the torrified biomass, thus demonstrating the unique selective mechanism of microwave-based heating. Satpathy et al., (2014) reported that low-power microwave heating with short reaction times heated biomass unevenly and thus produced hot spots in torrefied biomasses. Microwave heating produces hot spots for a combination of reasons, including the nonuniform nature of the biomass, the selective microwave heating process, the unequal distribution of the electric field, and the utilization of nonoptimal microwave irradiation. Generally, shortfalls related to the microwave-based DT of biomasses, for example, the previously mentioned hot spots, must be resolved via the electromagnetic design of the torrefaction reactor to turn this pretreatment method into a competitive technology.

6.4.2. *Microwave-induced wet torrefaction of biomass*

Wet torrefaction (WT) can be often considered as hydrothermal carbonization or hydrothermal pretreatment. Normally, this method involves the heating of the biomass suspension with subcritical water for reaction times of 30 min to several hours at 180 °C-250 °C under saturation pressure (Bach and Skreiberg, 2016). The exothermic process yields three products, namely, gases, aqueous chemicals, and hydrochar (Gan et al., 2020; Li et al., 2015). These products represent 88% of the weight and nearly 90% of the energy of the raw biomass (Bach et al., 2013). The mechanisms underlying WT are usually different from those underlying DT. Specifically, the following reactions are found in DT: condensation, aromatization, decarboxylation, decarbonization, dehydration, intermolecular reconstruction, and demethoxylation. Conversely, in WT, hydrolysis occurs because of the existence of compressed water; this process splits ester and ether molecular links and thus lowers the energies required for activating the depolymerization reactions of the biomass polymers (Libra et al., 2011). Therefore, WT degrades the hemicellulose component in biomass more than DT.

Give that biomass, water, and inert gas are the only inputs required for biomass pretreatment based on the WT method, the WT method is thus believed to be less toxic and more ecofriendly than the DT method. The attributes, such as HHV and energy density, shown by fuel from WT were higher than those shown by fuel from DT (Yan et al., 2009). Indeed, because biomass consists of fairly high amounts of moisture (normally >50%), the use of WT has become highly preferable (Acharya et al., 2015). Given that DT and WT do not require the predrying of the raw lignocellulosic biomass, they are cost-effective pretreatment methods. Moreover, this characteristic implies that a wide range of lignocellulosic biomass materials may be appropriate for WT pretreatment. The generated solid fuels have low ash content, high calorific value, and great grindability and pelletability (Iroba et al., 2017).

Microwave-supported heating for biomass WT is rare, and only a limited number of published works are available within current academic circles. Nevertheless, a growing number of researchers have focused on evaluating the applicability of microwave–WT for a variety of various lignocellulosic biomasses and the corresponding assessment of the energy characteristics of the produced hydrochars. Chen et al., (2012b) reported that microwave-torrefied sugarcane bagasse exhibited an improvement of 20.3% in energy density and heating value over DT sugarcane bagasse. Elaigwu et al. (Elaigwu and Greenway, 2016a) analyzed the energy output of hydrochars that were generated by heating rapeseed husks through a combination of microwaving and WT. In this approach, hydrochars were produced via the microwave heating of the biomass in deionized water for a specific duration at 150 °C-200 °C. The experiment revealed that the high temperature of 200 °C and long residence time of 30 min increased the HHV by nearly 32%.

In a different test, bamboo was exposed to microwave–WT in diluted HCl solutions at 180 °C for 5–30 min (Li et al., 2015). After 20 min of microwave–WT with 0.2 M HCl, the HHV of hydrochars (24.86 MJ/kg) had increased compared with that of the converse school-sub coal (21.67 MJ/kg). However, the HHV of hydrochars was only equal to that of German braunkohole lignite (25.10 MJ/kg). The capacity of generating hydrochars with either equivalent or even higher HHV than industrially used coals through the industrial application of microwave–WT was demonstrated clearly. Elaigwu and Greenway (Elaigwu and Greenway, 2016b) reported that the microwave-supported pretreatment of *Prosopis africana* husk required significantly less time to generate equivalent or superior HHV than traditional hydrothermal carbonization.

Several studies (W.-H. Chen, Ye, and Sheen, 2012b; Guiotoku et al., 2014) have revealed the rise in the HHV of microwave-supported biomass hydrothermal carbonization (HTC) with the increase in microwave temperature and pretreatment time. In addition to improving HHV, WT decreased the yields of mass and energy under high microwave power and temperature and long residence times (Y.-L. Lin, 2015; Elaigwu and Greenway, 2016b; Bach et al., 2017). The reduction in mass yield might be due to the loss of humidity and volatiles and the conversion of biomass from the solid-state into liquid and gas under pretreatment conditions (Huang et al., 2012) (Huang et al., 2012) (Huang et al., 2012) (Huang et al., 2012) (Huang et al., 2012) (Huang et al., 2012) (Huang et al., 2012) (Huang et al., 2012) (Huang et al., 2012) (Huang et al., 2012) (Huang et al., 2012) (Huang et al., 2012) (Y. F. Huang et al., 2012) (Y. F. Huang et al., 2012) (Y. F. Huang et al., 2012; Y. F. Huang et al., 2016). Afolabi et al., (2015) found that microwave temperature and time were two variables influencing the yield of solid char from the HTC of human biowaste.

Elaigwu and Greenway (Elaigwu and Greenway, 2016a) reported that the processing time of the microwave-assisted HTC of glucose was faster than that of the conventional process. Bach et al., (2017) examined the influences of microwave–WT on the characteristics of hydrochar generated from microalgal species. The calorific value of the microalgae increased by 21%, and 61.5% of the algae's energy was retained after pretreatment through the microwave–WT method. These

results emphasized the possibility of using microwave–WT as an efficient pretreatment approach for biomass feedstocks with a substantial amount of ash to mitigate cash-related risks when using microalgae as solid fuels. Seaweeds (macroalgae) are another ash-enriched biomass source, with variants, such as *Palmaria*, having an ash content of nearly 25.7% (Kostas et al., 2016), which is considerably greater than the ash contents present in untreated land-dwelling plants due to the seaweed's capability to absorb inorganic compounds from their natural habitat easily (Yanik et al., 2013).

Only one study (Yuan and Macquarrie, 2015) had analyzed the HHV of the brown seaweed *Ascophyllum nodosum* pretreated via microwave-supported acid hydrolysis. The capability of the microwave treatment of seaweed with an extra acid catalyst to effectively generate a sugar-rich liquid product and hydrochar with more than 50% energy yield in one process was proven. Furthermore, a hydrochar with an HHV of 24 MJ/kg that was generated under optimal pretreatment specifications represented a promising solid fuel alternative. Investigating the extensive use of this pretreatment method with different species of seaweed and other forms of biomasses with high water and ash is necessary given that additional technical advancements may be introduced to enhance process yields. The characteristics and working conditions of microwave-assisted torrified biomass pretreatment are presented in Table 5.

6.5. Microwave-assisted pyrolysis

In conventional heating methods, lignocellulosic biomass is commonly milled into small particles to minimize the presence of high thermal gradients and then subsequently heated via indirect thermal conduction or high-pressure steam injection at temperatures of 160 °C-250 °C. Therefore, the degradation of hemicellulose into furfural or humic acids may considerably influence the recovery and conversion of fermentable sugar (Li et al., 2016a). Alternatively, the rapid heating and internal uniformity of large biomass particles that result in fiber swelling and fragmentation under the support of microwave heating are reported to enhance enzymatic saccharification (Diaz et al., 2015). However, virtually no effect is seen on plant fiber content under microwave processing at temperatures equivalent to or less than 100 °C (Chen et al., 2017).

Microwave performance depends on the dielectric characteristics of the biomass and thus enables the stockpiling of the material and the conversion of electromagnetic power into heat. Although biomass is typically a low microwave absorber, its heat absorbency may increase due to the existence of fairly significant humidity levels and inorganic compounds (Li et al., 2016b). The pretreatment of lignocellulose may be correlated with the ever-increasing economic accessibility of flow-through microwave systems. In an assessment utilizing microwave radiation for the pretreatment of sweet sorghum bagasse (SSB), Choudhary et al., (2012) found that approximately 65% of the gross total sugars were recuperated when 1 g of SSB mixed in 10 mL of water was subjected to microwave pretreatment at 1000 W power for 4 min,.

In another experiment wherein corn straw and rice husk immersed in alkaline glycerol were subjected to microwave-supported pretreatment, scanning electron microscope evaluation revealed substantial ruptures in the structure of the plant cells (Diaz et al., 2015). Ravindran et al., (2018) found that for brewer's spent grain, microwave-assisted alkaline pretreatment was superior over dilute acid hydrolysis, steam explosion, ammonia fiber explosion, organosolv, and ferric chloride pretreatment. This result was obtained by pretreating 1 g of brewer's spent grain in 10 mL of NaOH 0.5% at 400 W microwave power for 1 min. Low upfront investment, easy use, and high energy savings are among the benefits of microwave radiation for lignocellulosic pretreatment (Kostas et al., 2017; Pandey et al., 2019; Iliopoulou et al., 2019). Pretreatment through the organosolv method is performed primarily to address the lignin portions of lignocellulosic biomasses and to stimulate delignification by severing ether bonds within the lignin structure.

The end products of this process, namely, bio-oil, biochar, and gas, are formed as the result of the quick dousing of discharged volatiles (Yu et al., 2020). Frequently, three main chemical substances, including (i) the cellulose-rich fraction; (ii) sugar degradation outputs and hemicellulosic monosaccharides, such as galactose, xylose, and arabinose; and (iii) an organosolv lignin substance along with a water-soluble fraction that mainly consists of acid-soluble lignin, are produced following the organosolv pretreatment of lignocellulosic biomass (Zhang et al., 2016). A wide range of organic substances (primarily bulk chemicals), including ethanol, methanol, acetic acid, formic acid, ethylene glycol, and glycerol, can be used as organosolv pretreatment solvents (Li et al., 2012b). The microwave-supported organosolv pretreatment of biomass before rapid pyrolysis and the potential ethanol production path from levoglucosan can be seen in Fig. 10.

The potential for utilizing the fast pyrolysis of cellulose for the indirect production of levoglucosan has been confirmed in several studies (Kuzhiyil, 2013). However, the inconsequential and inadequate amount of levoglucosan measured in bio-oil is typically deemed financially unsustainable for the production of the required level of ethanol because the cellulosic component in lignocellulosic biomass is trapped inside a matrix network comprising chemically bounded hemicellulose and lignin. Nonetheless, microwave-assisted organosolv pretreatment proved to yield greater energy-saving benefits than conventional organosolv pretreatment due to the capability for identifying and selecting a

Table 5
Effects of microwave-based dry torrefaction on characteristics of pretreated biomass.

Biomass type	Microwave irradiation parameters			Findings	References
	Power, W	Time, minutes	Temperature, oC		
Sugar cane residue	900	30	250	68.88% yield of mass; 82.5% yield of energy	Amer et al. (2019)
	900	30	300	42.96% yield of mass; 63% yield of energy	
Waste straw	350	30	220	68% yield of mass; 88% yield of energy	Lin (2015)
Rice husk	900	30	250	71.37% yield of mass; 82% yield of energy	Amer et al. (2019)
	900	30	300	44.82% yield of mass; 61.2% yield of energy	
Eucalyptus regnans sawdus	–	30	180	↑ 3.5 times for sugar	Amini et al. (2018)
Sugarcane leaf waste	700	3.5	–	↑ 40.6% times for sugar	Moodley & Kana (2017)
Rice straw	900	30	250	63.8% yield of mass; 66.3% yield of energy	Amer et al. (2019)
	900	30	300	41.63% yield of mass; 51.1% yield of energy	
	850	3	150	60.1% Cellulose; 4.5% Ligin; 14.9% Hemicellulose	Akhtar et al. (2017)
Corn straw	1300	2	180	22.6% Cellulose; 13.4% Ligin; 11.5% Hemicellulose	Diaz et al. (2015)
Cotton stalk	900	30	250	66.75% yield of mass; 82.1% yield of energy	Amer et al. (2019)
	900	30	300		
Walnut shell	–	20	250	84.32% yield of mass; 91.45% yield of energy; Oxygen content (15%)	Zhang et al. (2019)
	–	20	300	65.57% yield of mass; 82.94% yield of energy; Oxygen content (15%)	
Leucaena	250	25	360	20.29% yield of mass; 32.42% yield of energy	Huang et al. (2017)
	250	30	360	17.25% yield of mass; 27.87% yield of energy	

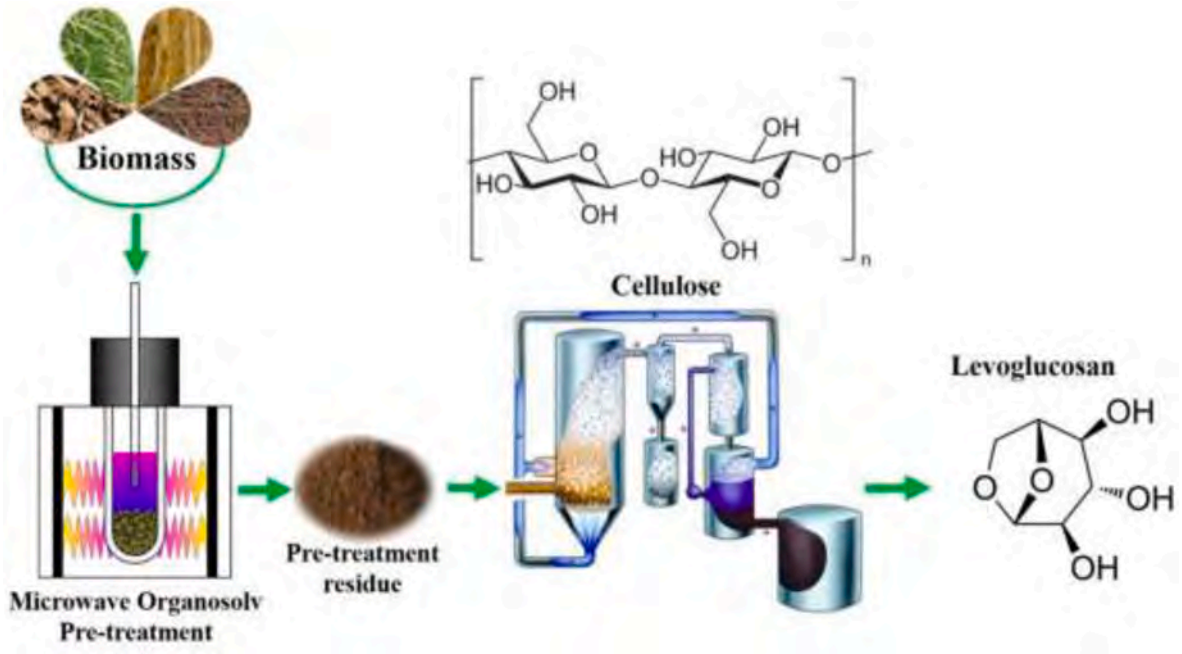

Fig. 10. Scheme of biomass pretreatment based on microwave-assisted solvolysis for the biofuel production process (Kostas et al., 2017).

predetermined lignin portion of biomass to be extracted. In the experiment conducted by Zheng et al., (2015), an atmospheric microwave reactor was applied in the microwave-supported organosolv pretreatment of corncobs, pine, eucalyptus, and straw with glycerol. This step was followed by the subsequent pyrolysis of the resulting residues. The anhydrosugar yields of pretreated pine, eucalyptus, and straw each microwaved at 150 W for 20 min were 33.2%, 59.4%, and 36.0%, respectively (Zheng et al., 2016). Notably, in the microwave pretreatment of corncobs at 150 W for 18 min, the subsequent amount of product yielded by the fast pyrolysis of the pretreated biomass was approximately 189 times higher than that of the unpretreated biomass (Zheng et al., 2015).

Most related studies demonstrated that microwave-assisted organosolv pretreatment with glycerol is an effective approach for enhancing levoglucosan output from the fast pyrolysis of lignocellulosic biomass-based agricultural residues. In addition to the judicious elimination of lignin and hemicellulose, the pretreatment of corncobs via the microwave-supported organosolv method aids in the removal of earth-metal-based alkali and alkaline compounds from biomass materials. Biomass pretreatment by using the microwave-assisted organosolv method is optimal preceding fast pyrolysis. Therefore, a wide range of organic chemicals (besides sole glycerol) should be tested with a variety of biomasses to determine the ideal combination of pretreatments for certain agricultural materials and particular organic catalysts.

6.6. *Microwave-steam explosion-assisted pretreatment method*

Steam explosion is another alternative biomass pretreatment method that relies on exposure to high and low pressures in quick succession. Materials are subjected to rapid and volatile decompression due to the sudden change in pressure (Auxenfans et al., 2017). Pang et al., (2013) explored an innovative method combining steam explosion and microwave irradiation (SE–microwave). The authors applied both methods in the pretreatment of corn stover at 540 W microwave power for 3 min. Both treatments occurred over the temperature range of 170 °C-210 °C for a residence time of 3–15 min. The SE–microwave process for biomass pretreatment is shown in Fig. 11.

Compared with the steam explosion method, the SE–microwave process resulted in higher glucose and xylose contents and slightly

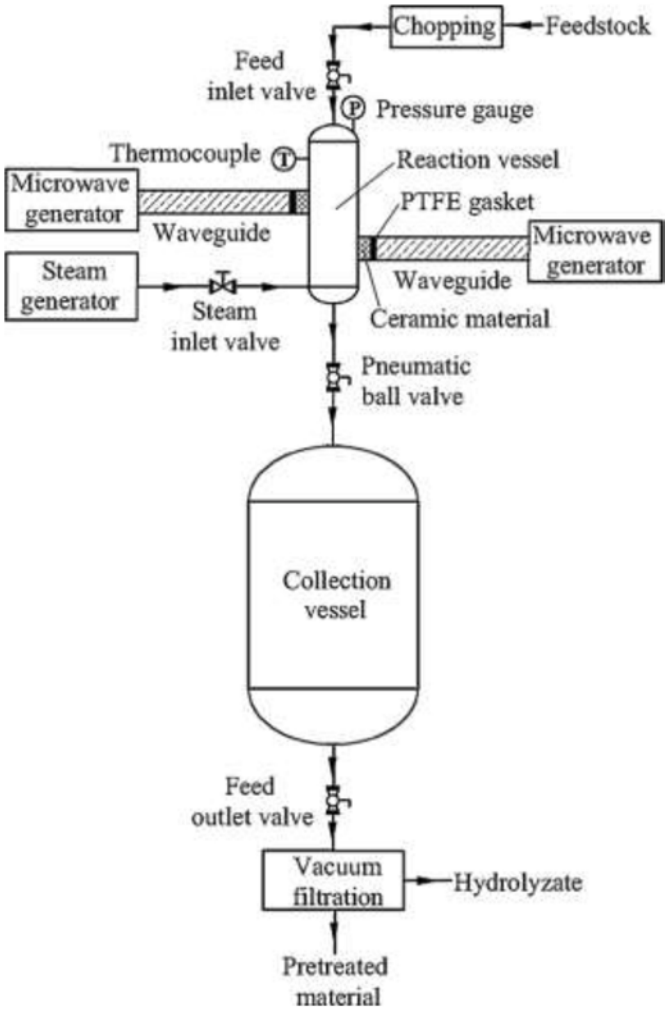

Fig. 11. The scheme of SE–ME process used for biomass pretreatment (Pang et al., 2013).

higher overall sugar yield (i.e., 57.4%, 17.8%, and 75.2%). The actual weights of each product were 28, 8.7, and 36.7 g per 100 g of raw biomass at 200 °C and 5 min of residence time. Additionally, the SE–microwave technique was proven to be highly effective in limiting biomass crystallinity. Biomass crystallinity under this technique was 19% lower than that under steam explosion pretreatment with the same operating conditions.

6.7. Microwave–chemical-assisted pretreatment methods

6.7.1. One-stage microwave–chemical-assisted pretreatment

The one-stage microwave-assisted chemical pretreatment of lignocellulosic materials is often performed by adding a chemical compound into the biomass mixture during heating by microwave energy. Liquid NaOH and H_2SO_4 are the chemical substances that are commonly used as additives in this process.

6.7.1.1. One-stage microwave–alkali-assisted pretreatment. Microwave-assisted alkali pretreatment is one of the promising methods for the bioconversion of biomass into a useful energy commodity. The benefits of microwave heating and alkaline pretreatment include reduced process energy consumption, quick heating, and low toxic compound formation. The integrated microwave energy and NaOH method was used to pretreat two different biomass species, namely *Saccharum spontaneum* and *Arundo donax* (Komolwanich et al., 2014). A positive influence on the cellulose fraction of biomass was observed along with measurable reductions in the amounts of hemicellulose and lignin in the treated biomass. Additionally, the use of 5% NaOH solution resulted in the highest sugar monomer yield of 6.8 g per 100 g of biomass. Another study used the microwave–NaOH pretreatment technique to treat oil palm empty fruit bunch (Binod et al., 2012), which is a type of biomass residue generated during palm oil production. The study revealed that the use of the optimal parameters (i.e., 3% NaOH solution, 180 W microwave power, and 12 min duration) resulted in 74% and 24.5% reductions in lignin and hemicellulose contents, respectively (Nomanbhay et al., 2013). The application of 1% NaOH solution combined with 600 W microwave power for 4 min in the enzymatic hydrolysis of cellulose provided a yield of 0.665 g of reducing sugar per gram of dry biomass materials.

In another experiment conducted by Vani et al., (2012), residue from harvested cotton plants was pretreated through a combined microwave–alkali method at 300 W for 6 min. The microwave–alkali pretreatment method required 108 kJ of power to produce 0.495 g of reducing sugar per gram of biomass, whereas the high-pressure reactor method required 540 kJ of power. In the pretreatment of biomass, the NaOH solution effectively removed hemicellulose and lignin contents (Lai and Idris, 2016) and facilitated the breakage of the lignin structure and its chemical bonds. The resulting enhancement in the accessibility of celluloses and their subsequent enzymatic hydrolysis could be due to an increase in the available contact area and substrate pore size for enzymatic reactions. Zhao et al., (2010) drew similar conclusions. The pretreatment of rice hulls with NaOH solution heated by microwave energy enhanced the surface accessibility of the cellulose substrates as the result of the destruction of the molecular structure. The resulting reducing sugar yield was 13% higher in the case of microwave-supported pretreatment than that in the case without microwave assistance.

6.7.1.2. One-stage microwave–acid-assisted pretreatment. Palmarola-Adrados et al., (2004) utilized the microwave-supported method combined with dilute sulfuric acid solution to pretreat modified wheat fiber for the removal of starch content. The authors observed that the glucose yield obtained from this method was more optimal than that obtained through steam explosion. Combined treatment with microwave and dilute sulfuric acid (2% concentration) was highly effective in the removal of the hemicellulose component of biomass (i.e., up to 29% compared with just 10.1% in distilled water treatment). Importantly, hemicellulose content had an inverse relationship with acid concentration. High acidity resulted in the low hemicellulose content of the post-treated solid biomass fraction. Once the acid level reached 0.015 M or higher, hemicellulose content decreased to approximately 1%. Particularly, the treatment of raw bagasse with 0.02 M sulfuric acid enabled 97.8% of hemicellulose to be hydrolyzed in this method. By contrast, only 79.8% of hemicellulose was hydrolyzed under pretreatment with distilled water only. Similarly, cellulose degradation was enhanced by an increase in acid concentration. Considering the effect of acid concentration on the lignin content of biomass, similar results were obtained when the acid concentration was increased from 0.015 M to 0.02 M (Chen et al., 2012c).

The results of recent studies confirmed the positive effects of dilute acid pretreatment on the synthesis of hemicellulose into soluble sugars, which further allowed for the ensuing enzymatic hydrolysis of cellulose (Zhu et al., 2016). Binod et al., (2012) utilized 1% sulfuric acid and 600 W of microwave power for 4 min to pretreat sugarcane bagasse. The subsequent enzymatic hydrolysis returned a reducing sugar yield of 0.091 g for each gram of treated biomass. Changing the chemical additive from sulfuric acid to sodium hydroxide at the same 1% concentration increased the sugar yield of the pretreatment process to 0.665 g/g under the same operating conditions. Alkali-based compounds, such as NaOH, were found to be highly effective in penetrating the lignin structure in lignocellulosic materials, thus enhancing hydrolysis efficiency (Lin et al., 2015).

6.7.2. Two-stage microwave–chemical-assisted pretreatment

The one-step microwave-chemical pretreatment has several shortcomings as mentioned earlier. The use of alkali compounds in combination with microwave-supported treatment has been shown to be highly effective in solubilizing lignin and enhancing the subsequent enzymatic hydrolysis through which cellulose is converted into various sugar forms (i.e., hexose and pentose). These sugar monomers are precursors that can further undergo additional fermentation processes in biofuel production. By contrast, the poor performance of acid–base compounds in removing lignin limits enzymatic hydrolysis and lowers overall efficiency and the resulting sugar production. The advantages of the two-step microwave–chemical pretreatment of biomass over the single-step method were reported in previous studies (Pedersen and Meyer, 2010). For example, Binod et al., (2012) utilized a two-step method to pretreat sugarcane bagasse with NaOH and sulfuric acid solution at a concentration of 1%. At 600 W of power for 4 min, the two-step approach resulted in a higher overall fermentable sugar yield of 0.83 g/g of dry biomass than the single-step microwave–alkali method. The former pretreatment was only able to obtain a reducing sugar yield of 0.665 g/g dry biomass.

Utilizing alkaline and acidic compounds in microwave-supported pretreatment with fast residence time can produce a high amount of reducing sugars as shown by previous studies. Zhu et al., (2006a) compared the performance of the microwave–alkali method with that of the microwave–acid/alkali pretreatment method in terms of xylose content resulting from enzymatic hydrolysis. While no trace of xylose could be detected in the post-treatment liquid fraction from the microwave/alkali method, the latter pretreatment approach could result in the production of crystalline xylose likely because of the presence of contaminants from lignin in the pretreatment mixture. The findings for the methods combining microwave heating with physicochemical biomass pretreatment methods are summarized in Table 6.

7. Environmental and economic aspects of microwave-assisted physicochemical pretreatment

The overall cost of biofuel manufacturing depends greatly on pretreatment energy expenses because they are a crucial component of biorefinery processes (Ubando et al., 2020). Optimal pretreatment

Table 6
The efficiency of applying the combined methods for biomass-based on microwave-physicochemical pretreatment.

Biomass name	Pretreatment methods	microwave irradiation parameters			Findings		References
		Power, W	Time, minutes	Temperature, °C	Before pretreatment	After pretreatment	
Corn stover	Microwave – H_2SO_4 (1%)	26	20	140	Gn = 35.2% Xn = 20.6% L = 22.6%	Gn = 31.7% Xn = 1.0% L = 14.6%	Chen et al. (2011)
Sugarcane bagasse	Microwave - H2SO4 (0.2 M)	900	10	190	C = 52.5% L = 12.7% H = 26.0%	C = 68.4% L = 14.1% H = 0.6%	
Herbal residues	Microwave - NaOH (0.08 g)	700	15	–	C = 21.2% L = 10.7% H = 26.8%	C = 21.0% L = 5.6% H = 22.8%	Cheng & Liu (2010)
Beechwood	Microwave - ammonium molybdate (1 mM) - H_2O_2 (0.88 M)	400	30	140	C = 49.9% L = 23%	C = 73.2% L = 37%	Verma et al. (2011)
Winter wheat	Microwave - Alkali	300	15	120	C = 42.7% L = 17.4% H = 31.3%	C = 53.1% L = 12.2% H = 32.9%	Boonmanumsin et al. (2012)
	Mmicrowave - Acid				C = 42.7% L = 17.4% H = 31.3%	C = 61.5% L = 15.8% H = 20.7%	
	Microwave- Alkali - Acid				C = 42.7% L = 17.4% H = 31.3%	C = 69.7% L = 10.4% H = 18.4%	
Corn stover	SE-microwave	540	5	–	Gn = 57% Xn = 9.9% L = 25.8%	Gn = 30.3% Xn = 13.3% L = 17.2%	Pang et al. (2012)
Corn stover	SE-microwave, SE at pressure of 1.45 MPa	540	5	200	Gn = 57% Xn = 9.9% L = 25.8%	Gn = 23.8.3% Xn = 3.2% L = 10.1%	Pang et al. (2013)
Sugarcane bagasse	Microwave - NaOH (1%)	600	3	–	C = 34% L = 18% H = 27%	C = 66.6% L = 4.9% H = 26.5%	Binod et al. (2012)
Oil palm trunks	Microwave - NaOH (2.5 M)	700	60	80	C = 50.8% L = 17.9% H = 30.4%	C = 71.9% L = 15.1% H = 11.8%	Lai & Idris (2013)
Oil palm fronds	Microwave - NaOH (2.5 M)				C = 41.9% L = 20.7% H = 33.6%	C = 68.9% L = 16.9% H = 11.8%	
Garden biomass	Microwave – H_2SO_4 (4%)	700	5	200	C = 39.57% L = 24.46% H = 26.65%	C = 53.95% L = 27.03% H = 11.62%	Gabhane et al. (2011)
Rice straw	Microwave - water	680	24	–	C = 33.4% L = 6.8% H = 16.2%	C = 41.8% L = 6.9% H = 23.6%	Ma et al. (2009)
Switchgrass	Microwave – alkali (0.1 g/g)	1000	30	190	Gn = 33.6% Xn = 19.3% L = 21.4%	Gn = 1.8% Xn = 11.8% L = 3.85%	Hu et al. (2007)
Corn straw	Microwave - water	1300	2	180	C = 27.9% L = 19% H = 14.8%	C = 28.9% L = 21.1% H = 13.3%	Diaz et al. (2015)
	Microwave - glycerol				C = 27.9% L = 19% H = 14.8%	C = 22.6% L = 13.4% H = 11.5%	
	Microwave - alkaline - glycerol				C = 27.9% L = 19% H = 14.8%	C = 23.1% L = 14.7% H = 12.8%	
Softwood	Microwave – Organosolv – C_2H_5COOH (25%) – HCl (0.1%)	–	6	180	C = 47.8% L = 31% H = 20.3%	L ≈ 25.6%	Liu et al. (2010)
Cotton plant	Microwave – alkali (0.05%)	300	6	180	C = 38.26% L = 29.95% H = 13.7%	C = 42.37% L = 2.12% H = 2.81%	Vani et al. (2012)
Wheat straw	Microwave – NaOH (1%)	700	25	–	C = 41.2% L = 21.3% H = 25.8%	C = 79.6% L = 5.7% H = 7.8%	Zhu et al. (2006a)
Rice hulls	Microwave – NaOH (4%)	450	3	–	Gn = 37.49% Xn = 15.68% L = 23.11%	Gn = 49.77% Xn = 22.93% L = 20.76%	Zhao et al. (2010)
Sugarcane bagasse	Microwave – H_2SO_4 (0.2 M)	900	5	190	C = 52.45% L = 12.72% H = 25.97%	C = 67.31% L = 15.67% H = 0.8%	Chen et al. (2012c)
Sugarcane bagasse	Microwave - H_2O	900	30	180	C = 48.5% L = 17.1% H = 29.9%	C = 58.5% L = 21.4% H = 10.1%	
	MicrowaveAH	–	30	180	↑ 3.5 times for sugar		Amini et al. (2018)

(*continued on next page*)

Table 6 (*continued*)

Biomass name	Pretreatment methods	microwave irradiation parameters			Findings		References
		Power, W	Time, minutes	Temperature, °C	Before pretreatment	After pretreatment	
Eucalyptus regnans sawdus							
Sugarcane leaf waste	Microwave - $FeCl_3$ (2 M)	700	3.5	–	↑ 40.6% times for sugar		Moodley & Kana (2017)
Corncob	Microwave - Phosphate dodecahydrate (11.55%)	700	6	–	↑ 76% times for sugar		Sewsynker-Sukai & Kana (2018)
Green tea residue	Microwave – NaOH (1%)	–	5	200	↑ 89% times for sugar		Tsubaki & Azuma (2013)
Rice straw	Microwave – NaOH (1%)	700	30	–	C = 38.6% L = 13.6% H = 19.7%	C = 69.2% L = 4.9% H = 10.2%	Zhu et al. (2005)
Sugarcane bagasse	Microwave – H_2SO_4 (0.005 M)	900	30	180	C = 48.45% L = 17.12% H = 29.92%	C = 59.66% L = 27.72% H = 3.84%	Shi et al. (2011)
Corn stover	Microwave – $CaCl_2$	800	12	162	C = 23.93%	C = 47.10%	
Rice Straw	Microwave – CH_3COOH (25%)	230	5	–	C = 41% L = 17.8% H = 20%	L ≈ 8.5%	Gong et al. (2010)
Rice straw	Microwave - Alkali - Acid	850	3	150	C = 42.5% L = 9.2% H = 24.5%	C = 60.1% L = 4.5% H = 14.9%	Akhtar et al. (2017)
Palm fiber	Microwave - NaOH (3.5%)	700	–	–	C = 35.4% L = 27.3% H = 19.9%	C = 56.7% L = 12.3% H = 15.5%	Laghari et al. (2016)
	Microwave - H2SO4 (3.5%)				C = 35. % L = 27.3% H = 19.9%	C = 44.4% L = 20.7% H = 14.5%	
	Microwave - Na_2CO_3 (2.0%)				C = 35.4% L = 27.3% H = 19.9%	C = 44.4% L = 20.3% H = 17.4%	
	Microwave -H_2O_2 (5.0%)				C = 35.4% L = 27.3% H = 19.9%	C = 51.3% L = 15% H = 15.8%	
Corn stover	Microwave - H_2SO_4 (0.2 M)	700	90	–	C = 36.5% L = 11.9% H = 31.3%	C = 38.5% L = 12.2% H = 15.6%	Liu et al. (2010)
Rice straw	Microwave - NaOH (1%)	800	60	–	C = 30.5% L = 19.7% H = 21.1%	C = 46.8% L = 8.3% H = 23.3%	Li et al. (2012a)
Rape straw	Microwave - H_2SO_4 (2% (v/v))	900	1	–	C = 37.0% L = 18% H = 19.6%	C = 42.3% L = 15.4% H = 23.6%	Lu et al. (2011)
Wheat straw	Microwave - NaOH (2.75% (w/v))	800	22.5	100	C = 45.0% L = 14.8% H = 28.0%	C = 51.0% L = 5.8% H = 31.0%	Singh & Bishnoi (2012)
Rice straw	Microwave - NaOH (1%) - H_2SO_4 (2%) - H_2O_2 (0.3%)	300	60	–	C = 38.6% L = 13.6% H = 19.7%	C = 80.6% L = 3.8% H = 3.2%	Zhu et al. (2006b)
Sugarcane bagasse	Microwave - H_2O	–	5	–	C = 46.9% L = 27.1% H = 16.3%	C = 46.5% L = 23.5% H = 15.9%	de Souza Moretti et al. (2014)
	Microwave - H_3PO_4				C = 46.9% L = 27.1% H = 16.3%	C = 47.3% L = 22.4% H = 16.4%	
	Microwave - Glycerol				C = 46.9% L = 27.1% H = 16.3%	C = 59.5% L = 15.8% H = 10.9%	

C stands for Cellulose; L stands for Lignin; H stands for Hemicellulose; Gn stands for Glucan; Xn stands for Xylan.

methods guarantee low manufacturing expenses on a large scale. Microwaving with an energy-efficient technology is considered as an effective approach to achieving low power input (Darji et al., 2015). Power consumption by microwave heating during water separation is 22.5-fold better than that by traditional heating (Cheng et al., 2008). Xia et al., (2013) showed that pretreatment time dramatically reduced by over 10-fold after using microwaving heating. The energy input of the pretreatment method combining microwave and acid treatment with 20 g/L water hyacinth and 1% H_2SO_4 heated at 140 °C for 15 min had an energy waste of 11.8%. Therefore, the construction of a continuous microwave system to reduce the pretreatment energy consumption of large-scale operation by reusing waste heat has been proposed for large-scale application.

Wang et al., (2013) reported that microwave pretreatment in ionic liquids for wood delignification consumed 192 kJ of energy, whereas oil-bath heating consumed 560 kJ. Compared with the oil-bath technique, microwave pretreatment generated a significantly lower amount of lignin and had lower energy consumption. Lu et al., (2011) calculated that rape straw pretreatment with 10% solid loading at a power of 700 W for 3 and 6 min and a power of 900 W for 10 min required 67.2, 153.9, and 169.2 kJ to generate 1 g of glucose. Water evaporation required the highest amount of energy and required 2.5 kJ of energy to evaporate 1 g water at 25 °C. Therefore, water evaporation accounted for approximately 48%–64% of the overall energy consumption.

Vani et al., (2012) compared energy consumption by cotton plant residue pretreatment based on alkaline treatment assisted by microwave heating with that by pretreatment under high pressures. Microwave pretreatment with the loading of 17.5% w/v at 300 W microwave power for 6 min was compared with high-pressure pretreatment with the loading of 5% w/v at 180 °C and 100 bar pressure for 45 min. Energy consumption by microwave pretreatment was 108 kJ and was five times lower than that by high-pressure pretreatment (540 kJ).

Limayem et al. (Limayem and Ricke, 2012) evaluated the effectiveness of energy consumption during pretreatment on the basis of the ratio of total sugar recovered to the overall energy consumed. Steam explosion was found least efficient with 0.26 kg sugar/MJ, followed by organosolv with 0.31–0.40 kg sugar/MJ and sulfite pretreatment with 0.35–043 kg sugar/MJ. The efficiency in energy consumption by microwave treatment can be measured and used to establish a reference pattern. The loading of biomass feedstock in reactors is an important element in decreasing overall energy consumption. High solid loading is a practical approach that enables increasing product yields while reducing operating expenses. However, appropriate pretreatment criteria are required because of the agitation issues associated with high substrate loading (Erdei et al., 2013) (Romaní et al., 2014).

Vani et al., (2012) stated that the biomass feedstock in the microwave reactor must exceed 20% to be economically viable. The lowest energy usage of 33 kJ (1100 W for 30 s) utilizing 40% (w/w) of biomass feedstock was demonstrated by Kannan et al., (2013). Throughout this process, the production of 1 g of sugar after enzymatic hydrolysis and 1 g of fermented ethanol required 1.27 and 1.76 kJ of energy, respectively. Therefore, compared with the aforementioned microwave pretreatment of rape straw, microwave pretreatment could save 80% of energy consumption during bioethanol production. The cost of bioethanol production by using microwave-assisted pretreatment was also calculated by considering the cost of instruments, chemicals, and operating expenses (Sondhi et al., 2020). The overall cost of bioethanol production was approximately $0.143 per L of ethanol. The price of produced ethanol was reduced by 8.32 times relative to the market price of ethanol, thus increasing the economic and industrial viability of the developed pretreatment process. Pang et al., (2013) performed the economic assessment of the microwave-assisted steam explosion-based pretreatment of corn stover with consideration of the effects of pretreatment time. The cost involved in producing 1 kg of sugar was $0.093. Therefore, this approach was deemed to be a financially feasible pretreatment method.

Microwave pretreatment has a high capital turnover ratio despite its large initial investment (Vani et al., 2012). Hasna (2011) assessed the cost–benefit of the use of microwave-assisted drying compared with that of conventional steam platen method in manufacturing corrugated paperboard. The capital cost of microwave pretreatment was $7000; however, this cost could be off-set by $128–380/h due to the power savings of this method over the conventional steam platen method. Indeed, under the assumption of the operation of $6000 h/year, the capital cost could be recovered in less than 1 year. Importantly, some benefits of microwave use, such as improvements in quality, reductions in wastage, and the minimization of starch consumption, over conventional methods were also evaluated. Low maintenance costs are believed to enhance the process in industrial-scale operations. Given its strong economic and energy efficiency potential, microwave pretreatment is seen as a viable and potentially promising technology.

The concept of regional biomass processing centers was first described by Carolan et al., (2007). This concept involves a network of processing facilities that are responsible for the pretreatment and conversion of biomass into intermediate products. Regional biomass processing depots (RBPDs) are another potential concept that focuses on the decoupling of pretreatment and preprocessing from the main production activities. After arriving at RBPDs, the raw biomass materials undergo pretreatment and densification before being transported to different biorefinery sites (Bals and Dale, 2012). This step in the supply chain is of critical importance in addressing the logistical issues associated with biomass, including low bulk density and seasonality. After pretreatment, the raw biomass materials become highly compact and easy to transport. The increase in energy density per transport load not only improves the efficiency of the supply chain but also alleviates potential regional congestion problems (Sultana & Kumar 2011). An increase in potential utilization and investment are some of the positive benefits that RBPDs can provide to the rural communities wherein they are located. These biomass pretreatment facilities are capable of processing the raw biomass materials collected from surrounding farms into products that can be further converted into biofuels, animal feed, and other biomaterials.

The overall pretreatment process performed at each RBPD must be simple and low in operational cost but should still be capable of delivering the necessary intermediate products for later processing stages. Examples of commonly used preprocessing techniques include microwave-assisted diluted acid application and microwave-assisted hot water explosion (Digman et al., 2010). The decentralized pretreatment of biomass has inherent disadvantages in achieving its full economic potential. In addition to the high energy requirements of the densification and size reduction of biomass and the lack of economy of scale, compound mass loss during pretreatment, storage, and transportation can exert a negative effect on the operation's profitability. Specifically, 10%–15% of total mass loss is an important threshold at which potentially significant loss of income occurs. Therefore, further research into the model of biomass processing depots is warranted to ensure financial and economic viability as examined in recent technoeconomic models.

8. Future perspectives of microwave-assisted pretreatment technologies

The development of industrial microwave heating applications and the integration of microwaves into the physicochemical method for biomass pretreatment are fairly limited despite the remarkable benefits of microwave irradiation. Furthermore, the analysis of the nature of the electromagnetic interaction with lignocellulosic biomass has seldom been mentioned to provide a complete contribution to the knowledge of the scientific community. In addition, operating parameters should be optimized to obtain the maximum cellulose component. The optimization of operating parameters should target reducing energy requirements and chemical use to improve overall efficiency and environmental sustainability. Although the use of microwave heating to support physicochemical methods has been concluded to have many incentives for incorporation into biomass pretreatment, several challenges have not been thoroughly addressed and fully understood yet. Therefore, challenges to lignocellulosic biomass pretreatment for biofuel production on the commercial and industrial scale continue to exist and are based on the factors of sustainability, energy savings, and capital cost. These challenges may include issues related to energy consumption, the clarification of fundamentals with respect to biomass heating induced by temperature gradients, and the dielectric properties of various biomasses.

Further studies must be carried out to identify the most efficient microwave-based technology for application in commercial biorefineries to attain critical strategies for clean energy, high efficiency, and the sustainable exploitation of available renewable resources. Studies on energy balance and the development of products close to the commercial market should be performed. Moving the biomass pretreatment processes from the lab to commercial and industrial facilities must be investigated in detail. Furthermore, energy integration should be closely considered for the design and development of advanced techniques and processes. The success of biorefineries is undeniably highly dependent on attractive and complete technoeconomic analyses. Therefore, the economic balance of biorefineries can be improved and enhanced if other values from coproducts, such as proteins, oils, and lignin, are achieved.

9. Conclusion

The characteristics and fundamentals of microwaves for the support of physicochemical methods in pretreating lignocellulosic biomasses were comprehensively introduced in this review. Compared with that of conventional heating, the use of the microwave-assisted process provide many benefits to biomass pretreatment. In addition, the influences of microwave irradiation on the particle size; surface area; lignin, hemicellulose, and cellulose contents; depolymerization; and crystalline structure of cellulose were thoroughly reviewed. Combined microwave-assisted methods significantly increased the cellulose content and efficiency of the obtained products. The microwave–physicochemical pretreatment of biomass was noticeably influenced by temperature, initial feedstock components, irradiation time, microwave power, and catalyst loading. Among the analyzed combined methods, microwave-induced torrefaction showed massive potential for industrial-scale application due to its definitive link between microwave application and physicochemical processes. The future perspectives and the recommendations drawn in this study on the basis of the above findings will assist researchers, scientists, energy personnel, and energy executives to utilize effectively microwave-assisted pretreatment technology for the conversion of lignocellulosic biomass into sustainable bioenergy, including biofuels.

Credit author statement

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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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