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Trends in harnessing energy from waste biomass: pathways & future potential

Neelu Raina[1,*], *Malinee* Sriariyanun[2], *Sukanya* Areeya[2], *Atin Kumar* Pathak[3], *Garima* Slathia[1], and *Himanshu* Saraswat[4]

[1]School of Biotechnology, Shri Mata Vaishno Devi University, Katra, J&K-182320, India
[2]Biorefinery and Process Automation Engineering Center, Department of Mechanical and Process Engineering, Sirindhorn International Thai-German Graduate School of Engineering, King Mongkut's University of Technology North Bangkok, Bangkok, Thailand
[3]School of Energy Management, Faculty of Engineering, Shri Mata Vaishno Devi University, Katra, J&K-182320, India
[4] Centre for Systems Biology and Bioinformatics, Panjab University, Chandigarh-160015, India

Abstract. Demand for fuel energy is continually on the rise. There is also a constant challenge involved to ensure that all our energy needs are fulfilled. Persistent overconsumption of conventional fossil fuels due to the rise in global population aided by economic expansion has resulted in reduction of fossil fuel reserves. This has fuelled the need to boost research efforts on renewable and sustainable bioenergy feedstocks. Since bioenergy utilizes organic matter; therefore, it is an economically viable and clean solution, which can minimize our reliance on non-renewable resources. The bioprocessing of lignocellulosic biomass to produce bio-based products under biorefinery setup is gaining global attention. The main challenge however remains to strike a balance between energy harvesting and economic viability with minimum environmental impacts. The development of zero-waste lignocellulosic biorefinery aligns completely with the idea of sustainable development without increasing carbon footprint. This concept is self-sustainable. It also advocates re-usage or recycling of waste; of which using lignocellulosic biomass waste is a major thrust. Improving the techno-economic efficiency of currently employed pretreatment methods and looking for combined pretreatment strategies will prove to be a stepping stone in the commercialization of zero-waste lignocellulosic biorefineries. This review investigates the most widespread pretreatment types, highlighs their advantages/disadvantages, and reviews the current status and technological advances in the bioconversion process of LCB into bioenergy in a biorefinery set-up.

Keyword. Biorefinery, Bioethanol, Lignocellulose, Pretreatment, Bioconversion

1 Introduction

The rising of global population is expected to reach 9.2 billion by the year 2050 [1]. Simultaneously, by the year 2030, the predicted enhancement in world energy demand is likely to reach around 53% [2]. Coal, oil and natural gas, the major forms of energy that we use today, are non-renewable and thus bound by limited availability. Gobbling up of non-renewable resources by industrial evolution has eventually brought fossil fuel reservoirs to the verge of exhaustion. Their consumption at this pace has severely degraded the environment as well. Therefore, the sharp rise in the demand for energy resources is far beyond the handling capacity of fossil-based resources. The extravagant use of fossil fuels as the sole source of energy has given rise to the dire need for gradual energy shift. This shift is driving us away from the traditional sources of energy towards alternative resources which are sustainable in the long run.

1.1 Is bioenergy the ultimate answer to our rising energy needs?

Biomass as a resource is present in large quantities. Its sustainable utilization offers a variety of benefits ranging from sequestration of carbon to generation of bioenergy. It has the capacity to serve diverse purposes as part of the integrated biorefinery system. In addition, it yields numerous benefits by strengthening the socio-economic structure of the society. Production of renewable and sustainable biofuels is thus gaining overwhelming interest globally. Biofuels are important from an economic

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[*]Corresponding author: ancient99india@gmail.com

point of view as they can support a stable, secure and low-cost energy supply. Bioethanol is currently the front runner in the race among all the biofuels. It presents promising opportunities in terms of a green, carbon-neutral, environmentally safe, and clean alternative to fossil-derived fuels. The utilization of bioethanol blended gasoline in the transportation sector reduces the consumption of fossil fuels and the concomitant release of greenhouse gases (GHGs) in the environment. Its chemical-physical properties (evaporation, enthalpy and octane number) are similar to that of gasoline. In terms of chemical production, ethanol can also be used as a platform chemical itself. Its bioconversion *via* microbial fermentation can utilize a wide array of biomass types (simple sugars, starchy biomass, and lignocellulosic biomass) with the potential co-production of several industrially significant compounds. The presence of many macromolecular organic compounds in a biorefinery process acts as platform chemical/s for molecules of greater interest; besides decreasing the cost of associated biofuels.

The most promising molecular substrate for the production of bioethanol is lignocellulose. It is composed of cellulose, hemicellulose and lignin. Cellulose and hemicellulose are the reservoirs of sugar polymers, which are fermentable in nature. These sugars can be converted into bioethanol by the use of microorganisms. On the other hand, lignin is an aromatic polymer. The transition from fossil fuels towards biomass-based fuels has presented lignocellulosic biomass (LCB) as the favorable biomass feedstock. Since the annual worldwide production of LCB is estimated to be around 1×10^{10} metric tons (MT); LCB thus accounts for approximately half of the total biomass reserves present on planet earth [3]. LCB generated from various sectors (forestry, agriculture, agro-industrial and industrial) acts as precursors for the generation of cellulosic bioethanol (2nd generation) accompanied by a wide array of biochemicals. Also, bioethanol thus generated does not contribute to the price surge of food when compared to the conventional sugar/starch-based bioethanol (1st generation).

1.2 Lignocellulosic biomass waste as a source of bioenergy

Biorefinery approach presents an opportunity to integrate production of biofuels and industrially useful bio-based platform chemicals from LCB. It assimilates many conversion techniques over a single platform. The major focus of the lignocellulose biorefinery that tends to enhance its economic viability is the maximum utilization of the majority of LCB (*i.e.* cellulose, hemicellulose and lignin). It is accompanied by minimum or zero wastage from the system thus supporting the concept of circular bioeconomy (CB). However, the recalcitrant nature (presence of lignin inhibits LCB solubilization *via* enzymatic saccharification) of LCB is the main obstacle in its valorization to bioethanol and other value-added products. The bioconversion of LCB thus depends on many steps involved- a) pretreatment, b) hydrolysis, and c) fermentation. The present review entails the different steps involved in the valorization of LCB for bioethanol production along with various value-added products generated. Also, it brings forward future perspectives, recent technological advances (biotechnological) and significant challenges encountered in bioethanol production that need to be highlighted. A carbon-neutral biorefinery system using LCB as feedstock opens the door for a transition from linear production of biomass to a closed-loop system emphasizing zero-waste bioeconomy [4]. However, the scientific fraternity has explored several bottlenecks keeping in mind the “zero-waste” concept using LCB as substrate.

Waste forms an inherent and unavoidable constituent of any process that can be ascribed to various factors *i.e.* process inefficiencies, usability of resources and discarding of non-essential parts of the feedstock. The use of residual biomass available locally contributes to enhanced energy productivity and sustainable growth. Residual materials generated from forestry, agriculture and agro-industrial sectors amount to millions of tons of waste every year. These agro-industrial transformations need to be managed and have a disposal cost associated with them, therefore raising serious economic and environmental concerns. Also, their underutilization limits all possibilities of maximum utilization of the biomass. Therefore, the utilization of agricultural waste is an excellent choice for biofuel and biochemical production. Converting unwanted agricultural waste streams into valuable resources by their bioconversion requires improved technologies, which support practical applications for sustainable development. A total of 174.1 million tons of residues are generated per year in Thailand and the projected capacity for bioethanol production is estimated to be around 20,213.5 million liters per year. Nevertheless, only 31.2% of the fuel demand from the transportation sector can be met using this volume [5]. This calls for the need to improve bioenergy production. Looking for alternative energy sources can aid in the mitigation of GHGs in the environment.

The strategy to systematically utilize LB waste for bioethanol production strongly depends on its composition. Lignin, the aromatic polymer is a major impediment in the fermentation of cellulose and hemicellulose and the subsequent release of sugars and valuable chemicals. Therefore, it becomes necessary to loosen the lignin shield from the accompanying organic matrix. Recent studies have shown that the primary aim of the researchers during pretreatment is reducing cellulose crystallinity, cleavage of H-bonds and Van der Waals forces and solubilization of the hemicellulose. Many pretreatment methods have been used to separate the lignin polymer from the rest of the polysaccharide pool. Their primary focus is on the release of maximum concentration of fermentable sugars. This is accompanied by the production of platform chemicals which are of importance from an economic point of view. Pretreatment studies using acids, alkali, organosolv, deep eutectic solvents and combined pretreatment methods among others have been developed. Acids result in deconstruction of lignin, hemicellulose and cellulose solubilization, and destruction of β(1-4) glycosidic bonds. On the other hand, alkali solubilizes the recalcitrant layer of lignin and hemicellulose depending upon the chemical concentration, temperature and pressure conditions [6]. Ionic liquid pretreatment (imidazolium, choline and chloride acetate, pyridinium) depends on the selection of anions and cations in the reaction mixture. β-O-4 bonds are converted into ion dipole bonds thus dissolving the cellulose layer and resulting in the separation of lignin and hemicellulose components. Organosolv pretreatment dissolves β-O-aryl, 4-O-methylglucuronic acid, α-O-aryl in between lignin and hemicellulose. Recently the effect of more than two solvents has been explored during pretreatment studies usually operated at a high temperature of 200 °C [7]. Deep eutectic solvents are considered green, non-toxic solvents comprising of a hydrogen bond acceptor (HBA) and a hydrogen bond donor (HBD) at varied molar concentrations. During the pretreatment process, a new H-bond is formed between the hydroxyl group present in lignin and the carbohydrate complex. This results in hydrolysis of lignin-carbohydrate complex (LCC) thereby causing cleavage of ether bonds between lignin and hemicellulose. Combined pretreatment studies have been successful in terms of product recovery at multiple stages. For example, studies have shown that hot water and deep eutectic solvents have resulted in hemicellulose and lignin recovery from LCB [8].

Also, pretreatment methods have been designed to minimize the loss of polysaccharides and the formation of sugar derivatives from the substrate. This helps in obtaining near theoretical sugar yield from the biomass and decreases enzyme load to be used in the enzymatic hydrolysis step. However, until now pretreatment has proven to be challenging and a costly step with only a few reaching the commercial scale. An effective pretreatment method plays a prime role in increasing the digestibility of LCB. Pretreatment technology has proven crucial for decreasing the cost of production involved and making the overall process more sustainable. Table 1 highlights key features of some of the major pretreatment types studied at lab, pilot and full-level scales.

The subsequent step that follows after pretreatment is hydrolysis. Hydrolysis is required for the conversion of polymeric carbohydrates such as starch, cellulose and hemicellulose into reducing/fermentable sugars, since yeast cannot directly utilize carbohydrates as substrate. Enzymes can specifically hydrolyze both cellulose and hemicellulose polymers in order to produce soluble sugars at low temperatures (45-50 °C). Nowadays "*de-novo* enzyme designing models" and a series of "omics" based approaches (metatranscriptomics, metagenomics, metaproteomics and metabolomics) are serving as enzyme discovery tools that aid in designing highly useful enzyme cocktails. Recombinant DNA technology and site-directed mutagenesis have also helped in the advancement of the enzyme development sector [1]. Efficient bioconversion of carbohydrates present in the lignocellulosic residue into bioethanol can be attained by optimizing certain operational parameters of the hydrolysis process, such as biomass loading, additives in the hydrolysate medium, enzyme loading, shaking speed and the incubation time.

After enzymatic hydrolysis the next step is fermentation. It is the process of converting glucose (C_6) and other reducing sugars (obtained from the cellulosic/lignocellulosic biomass) using microorganisms such as bacteria or yeast in order to produce bioethanol. Some yeasts and bacteria can metabolize monosaccharides, such as glucose and fructose as well as disaccharides, such as sucrose and maltose (in the absence of oxygen) resulting in the production of bioethanol (C_2H_5OH) and carbon dioxide (CO_2). Efficient utilization of glucose and xylose using fermentation is one of the bottlenecks in the efficient production of bioethanol from LB [17]. The process of Direct Microbial Conversion (DMC) or Consolidated Bioprocessing (CBP) encompasses all the reactions that are needed for the transformation of LCB into bioethanol. This approach varies from other bioconversion studies as it makes use of only a single microbial community that can effectively carry out both production of enzymes and fermentation process (*i.e.* enzyme production; enzymatic hydrolysis as well as fermentation) in a single step. This process holds many advantages since no capital is required for enzyme production and complete substrate utilization takes place for the optimal release of reducing sugars.

2 Bioconversion of lignocellulosic biomass waste into biofuels and other valuable by-products

A wide range of established pretreatment technologies along with abundant availability of LCB waste is a boon that can be used to suffice the energy demands of mankind. LCB can be converted *via* physical, chemical, or biological processes for generating energy and a wide range of products (lactic acid, xylitol, succinic acid, itaconic acid, furfural, *etc.*) at a competitive price. Around 50

Table 1 Comparative assessment of key parameters of various pretreatment methods

Pretreatment Type	**Operational conditions**	**Advantages**	**Disadvantages**	**Developmental stage**
Milling/Mechanical Comminution	1. Particle size: ~0.003-30 mm	1. Decreases cellulose crystallinity 2. Does not produce inhibitory compounds	1. Requires high energy and power [9,10]	Commercial [11]
Freezing	1. Freezing temperature = ~ -20 °C	1. Utilizes negligible quantity of chemicals 2. Decreased negative effects on the environment	1. Expensive [12]	Laboratory scale
Alkaline	1. Temperature range= 15-210 °C 2. Alkali concentration= 0.5–15% 3. Solid to liquid ratio-1:5–1: 20	1. Low inhibitor formation 2. High xylose and glucose yield 3. Improves accessible surface area 4. Removes hemicelluloses and lignin content	1. The residence time required is high 2. Salts formed are irrevocable and incorporated into the biomass 3. High percentage of water is required for salt removal	Commercial [13]
Acidic	1. Temperature= 130–210 °C 2. Acid=0.2–2.5% (w/w)	1. Removes hemicellulose 2. Yields high glucose content 3. Production of xylose without the use of enzymes	1. Solvents need to be recycled 2. Corrosion issues 3. Inhibitor formation 4. Formation of degradation products	Commercial [13]
Steam explosion	1. Temperature=160–260 °C 2. Pressure=0.7–5 MPa	1. Hemicellulose solubilization and transformation of lignin 2. Higher glucose yield 3. Cost-effective	1. Toxic compound generation 2. Partial degradation of hemicellulose	Commercial [13]
Ammonia Fiber Explosion (AFEX)	For AFEX 1. Weight of ammonia=1-2 kg 2. Temperature=60–100 °C 3. Time=5–30 min For ARP (Ammonia Recycled Percolation) 1. 10–15 wt% aqueous ammonia 2. Temperature=150–170 °C 3. Time=10–12 min	1. Increased accessible surface area 2. Inhibitor concentration formed is low	1. Not efficient for biomass containing high lignin content 2. The cost of ammonia is high	Commercial: Method operationalized to DuPont's cellulosic ethanol biorefinery in the US [13,14]
Biological	1. Depends on the type of fungus, bacteria or enzyme used or their combination (crude or concentrated)	1. Degrades hemicellulose and lignin 2. Consumption of energy is low	1. Rate of hydrolysis/biodegradation is low 2. Costly due to extended pretreatment time [15]	Laboratory scale
Liquid hot water	1. Temperature= 160-220 °C 2. Time= Minutes to hours	1. Chemicals are not required therefore no need for neutralization making this process cost-efficient	1. Water consumption and energy required are high 2. The rise in temperature leads to the generation of inhibitory compounds	Commercial scale [13]

Deep Eutectic Solvents	1. Temperature=60–200 °C 2. Time=0.5–24 h 3. Biomass loading =1:5–1:3	1. Biodegradable 2. Cheap 3. Synthesized easily 4. Biocompatible	1. Economic viability is to be maintained by recycling/reusing the chemicals	Laboratory scale [16]
Organosolv treatment	1. Temperature= 180–200 °C 2. Biomass loading=4:1–10:1 3. pH =2–4 4. Time=30–90 min	1. Disruption of hemicellulose and lignin bonds	1. Expensive process 2. Removal of solvents is necessary	Laboratory scale
Combined pre-treatment	The operational conditions affect the pretreatment process	Usually more effective in comparison to other pretreatment methods	The difficulties vary according to the combination type	-

billion kilograms of total renewable biochemical (apart from biofuels) are produced every year on a global scale [14]. The main aim is to focus on progress in pretreatment technologies paving the way toward improved biofuel and biogas production (Figure 1).

2.1 Production of Biogas

A considerable increase in biogas (methane) production has been reported by anaerobic digestion of LCB after the pretreatment process. 6% w/w NaOH pretreatment of *Asparagus* stem yielded 242.3 (mL/g VS) biogas after 18 days of digestion in comparison to that produced by untreated biomass substrate (175.1 mL/g VS). In another study, the digestion time of corn straw was reduced by 34.6% using yeast and cellulolytic bacteria [18]. The induction of an upgraded biogas system into LCB biorefinery will be able to promote a green economy and induce environmental susceptibility.

2.2 Liquid biofuel production

Liquid biofuels like bioethanol, biobutanol and biodiesel can be generated from LCB. A wide range of research and studies have been carried out to enhance the production of biofuels primarily targeting pretreatment techniques. An ionic liquid [Emim][OAc] pretreatment of spruce and oak sawdust (forest residues) at a low temperature of 45 °C resulted in approximately 2.5-4 times increase in the ethanol yield as compared to the native samples [19]. A few notable LCB biorefineries include the Raizen plant in Brazil and the POET-DSM Advanced Biofuels plant in South Dakota. They generate 8 ml/yr and 80 ml/year of bioethanol from bagasse and corn stover, respectively, by fermentation [20].

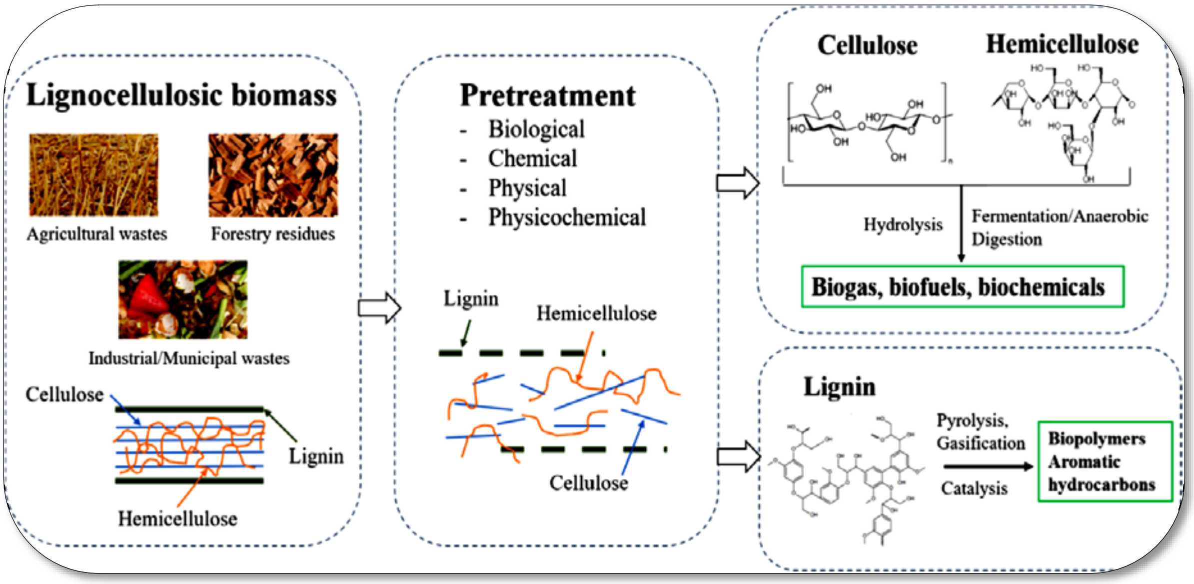

Figure 1. Process of bioconversion of lignocellulosic biomass residues into bio-chemicals and bio-products *via* pretreatment [18]

2.3 Biochemicals and Bioproducts

LCB can be used to generate organic acids (lactic acid, acetic acid), essential chemicals (phenolic compounds, phenyl propanoids), enzymes (pectinase, amylase, and cellulase), and chemicals for biopolymer production. USA and Asia Pacific currently dominate the lactic acid market. The current valuation of lactic acid market (a biochemical) was placed at 0.69 billion USD in 2017 and by the year 2023, it is estimated to reach 2.09 billion USD. Similarly, the production of xylitol, which finds application in chewing gum industry is estimated to reach 130 million USD by the year 2025. In the case of vanillin, the global market is expected to cross 700 million USD approximately [1]. These biobased compounds can be produced using the process of Separate Hydrolysis and Fermentation (SHF) and Simultaneous Saccharification and Fermentation (SSF). The recalcitrance of LCB does play a major role in affecting the production of these bioprocesses as they do in the case of biofuels. This again calls for the development of a suitable pretreatment process keeping in view that the bio-based market is anticipated to remain around 50 billion EURO by the year 2030 [21].

3 Research gaps and future research directions

Due to wide variability in the composition of LCB in our surroundings, it becomes really difficult to optimize a specific type of pretreatment process for optimum recovery of biofuels and other by-products. It varies with the biomass type and the desired end-product. However; these factors are intertwined with the need for environmental sustainability and cost-effectiveness making the process more challenging. Efforts directed at developing advancements in recycling/recovering of enzymes, chemicals and by-products from pretreated hydrolysate could greatly improve cost benefits. In addition, the effects of removal of inhibitors such as acetic acid, furfurals, hydroxymethyl furfurals (HMFs) and phenolics could further improve process economics. Advancement in the detoxification process can also facilitate the commercialization of LCB biorefinery. Combining different pretreatment techniques has shown promising results by synergizing their effects in terms of sugar yields, biofuel production and many various by-products. However, the cost of equipments and chemicals needs to be regulated.

4 Conclusions and future recommendations

The depletion of petroleum reserves and rising energy demands have shifted our research focus towards the potential of LCB waste for the generation of biofuels, biochemicals, biopolymers, food/feed supplements and bioactive compounds *etc*. It is clear that the use of LCB waste can be an optimal solution to our energy demands. Pretreatment being a vital step still requires advancement in terms of cost-cutting, sugar yields and therefore overall process feasibility. Many pretreatment methods consume a large amount of water, energy, chemicals and equipments in addition to release of inhibitory and harmful waste into the environment. This needs to be thoroughly addressed. LCB-based biorefineries could be the ‘Zero-waste’ based biomass biorefineries but they face a series of challenges, which hinder their advancement towards the production of a sustainable and eco-friendly economy. Full-scale conversion of LCB into fuel, chemicals and by-products is challenged by factors such as sustainability, policy initiatives, social, socio-economic issues and market challenges. On the part of industries and research sectors, a collaborative approach is the need of the hour that can address all these challenges in a holistic way. Suitable integration of pretreatment methods with minimum input of resources and energy will contribute to the complete commercialization of ‘zero-waste’ biorefinery processes using LCB.

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