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Lignocellulose in future biorefineries: Strategies for cost-effective production of biomaterials and bioenergy

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Abstract

Lignocellulosic biomass has been emerging as a biorefinery precursor for variety of biofuels, platform chemicals and biomaterials because of its specific surface morphology, exceptional physical, chemical and biological characteristics. The selection of proper raw materials, integration of nano biotechnological aspects, and designing of viable processes are important to attain a cost-effective route for the development of valuable end products. Lignocellulose-based materials can prove to be outstanding in terms of techno-economic viability, as well as being environmentally friendly and reducing effluent load. This review should facilitate the identification of better lignocellulosic sources, advanced pretreatments, and production of value-added products in order to boost the future industries in a cleaner and safer way.

Keywords: Biorefinery; Lignocellulosic biomass; Biofuel; Bioammonia; Biomaterial; Platform chemicals

1. Introduction

Biorefinery has proved to be a viable option for replacing petroleum-based goods with bio-derived alternatives to develop a cost efficient long-term economy (Cho et al., 2019; Yu et al., 2020). Lignocellulosic biomass (LCB) has the ability to partially substitute petroleum as a feedstock in the production of a wide range of products tailored to consumer demands. This could be accomplished by implementing biorefineries, which allows biomass to be converted into a variety of products like as biofuels, energy and high value-added products (Forde et al., 2016; Sindhu et al., 2020).

The lignocellulose is a viable substitute because of the availability and variety of raw materials, as well as the favourable market position of conversion products. Currently, starch is the most popular feedstock for biorefineries. The most commonly used technologies in the bioethanol industry are based on the fermentation of carbohydrates derived from sugar and starch crops that are all advanced processes with minimal potential for growth. However, since it uses food as a feedstock, the use of sugar and starch crops to manufacture bioethanol has been challenged. Cellulose is a more viable renewable resource because it is easily accessible and does not compete with feed. It is a renewable energy source that holds solar energy in its chemical bonds, making it a promising candidate for the development of affordable bioethanol (Lu, 2019).

Pretreatment of LCB is crucial in optimizing conversion to precursors for value-added products due to the complex composition and form of the biomass cell wall. Several pretreatment procedures have been developed and effectively used to enhance the technology throughout the years (Sankaran et al., 2020). Lignocellulosic components can be extracted from LCB using top-down approaches that intend to disintegrate cost effective feedstocks for the production of different value-added commodities with a wide range of applications

(Squinca et al., 2020). The potential for numerous modifications to improve cost-effective production is often a focus criterion in biorefinery (Choi et al., 2019). This can be a valuable step in techno-economical analysis for start-up firms before investing in the bio-economy field (Bondancia et al., 2020).

A biorefinery's economic performance is influenced by a number of design factors, including location, raw material, conversion technology, and end products. Firstly, because of the increased demand for the particular feedstocks used during process, the deployment of large-capacity biorefineries would have an effect on a variety of feedstock prices. Secondly, the greater competition for a given raw material would cause the biomass market to reallocate other raw materials, resulting in a price signal. This would make market formation for the materials in the system more difficult, which means that the raw material prices estimated in the supply chain evaluation model will no longer be reliable by the time the system is expected to be applied commercially (Yang et al., 2020; Zetterholm et al., 2020).

By focusing on specific lignocellulosic linkages, nanotechnological techniques can help to build enzymes for tailored utilization of very specific substrates. These do not really generate any harmful residue, and the hydrolysis is carried out in a safe environment (Low et al., 2021). There is an increasing awareness of the benefits of nanotechnological treatments. The use of nanotechnological advancements for nanocellulose manufacture of biomaterials and biofuels has sparked attention. In this article, various aspects of lignocellulosic biorefinery including selection of sources and advanced pretreatments for extracting various types of cellulose in the development of cost-effective biorefineries are discussed. This review also focused on different biofuels such as bioethanol, biohydrogen, bioammonia, and so on along with a short description about major platform chemicals and biomaterials from the biorefinery for future industry application.

2. General aspects of Lignocellulose based biorefinery

Biorefineries are becoming more popular in society, despite society's concerns about plants for bio-based materials, products, and liquid fuel substitutes. There have been three major phases of biorefineries identified. Phase I biorefineries use only one type of raw material, a single-step process, and only one major product; Phase II biorefineries use a single type of raw material, but multiple processes and major products; and Phase III biorefineries use multiple raw materials, processes, and final products (Takkellapati et al., 2018). The lignocellulosic biorefinery is one of the so-called phase-III biorefinery concepts, which are defined by their potential to manufacture multiple products from a wide range of resources through various routes (Fig.1). The main feedstocks in a lignocellulosic biorefinery are biomass such as agrowastes, forest residues, aquatic residues and industrial wastes (Naik et al., 2010).

A two-platform biorefinery design was recently implemented by National Renewable Energy Laboratory (NREL), USA. This form of biorefinery is made up of two platforms: sugar and syngas platforms. Biochemical conversion methodologies are used on the sugar platform; while thermo-chemical methods are used on the syngas platform together with generate valuable biomaterials and energy. The plants stabilize about 90 billion tonnes of CO_2 per year, the majority of which is in the form of wood (Okolie et al., 2020). Forestry and agricultural residues, yard waste, animal wastes, wood products and aquatic residues can be used in a cellulosic biorefinery. Chemical digestion or enzymatic hydrolysis are used to clean and break down feedstock materials into the three major fractions (Galbe and Wallberg, 2019). The alkaline and acid treatment can also create hemicellulose and cellulose. Further lignin can be decomposed with enzymes. The carbohydrate polymers hemicellulose and cellulose together can be hydrolyzed to obtain their constituent sugars. The high development costs for bioenergy products are the biggest impediment to a big breakthrough. When compared to fossil-based products, cellulose-derived products can dramatically decrease greenhouse gas (GHG) emissions. A major concern for cellulosic biorefinery is the efficiency and profitability of the conversion processes and logistics.

The advancement of several biomass fractionation approaches and the conceptualization of many biorefinery technologies has been facilitated by the interest in these biomass resources for biomaterials, biofuels and biochemicals development (Popa, 2018; Reshmy et al., 2022, 2021b). In the field of biorefineries, the production system is categorized based on four key characteristics: (1) raw materials (2) pretreatments (3) platforms and (4) products. These four characteristics can be combined in a diversity of processes to create a variety of biorefinery concepts with various processing arrangements. They are divided into two types based on the products they produce: energy-driven and material-driven biorefinery systems (Takkellapati et al., 2018).

In an energy-driven biorefinery system, biomass is primarily used to produce biofuels, energy, and heat. A material-driven biorefinery device, on the other hand, produces bio-based products like as platform chemicals, and other biomaterials. In both systems, process residues can be used to generate electricity, reducing waste generation. Within such numerous complex possible combinations of cellulosic process chains, two particular phases, biomass supply and fractionation; have received a lot of attention because of their significant impact on the performance of biorefineries. For the sustainable and responsible implementation and advancement of these biorefinery concepts, it is important to be aware of the possible positive and negative implications of biomass processing works.

3. Significance of nanotechnology

Nanotechnology is a tool for fabricating and applying materials with molecular precision at 100 nm or smaller dimensions (Malik and Sangwan, 2012). They have many times more surface area to volume fraction than other materials of the same weight. The physicochemical characteristics of many materials have been found to alter at the nanoscales; in case, the physical characteristics of NC vary greatly from those of plain cellulose. For the same substance at different dimensions, distinctive characteristics such as electrical, chemical, magnetic, mechanical, and optical properties are unique (Reshmy et al., 2021a, 2020a). The quantum effects and specific surface area are responsible for the changes in these properties. Quantum effects are important at the nanoscale, as evidenced by the nonmagnetic nature in silver at the macroscale but its magnetic character at the nanoscale due to electronic interactions (Singh et al., 2014).

Nanomaterials such as nanorods, nanoparticles, nanopores, nanofibers, nanocrystals, nanotubes, nanosheets and nanocomposites are currently used in industrial biotechnological applications. Their large surface area allows for a greater amount of enzyme to be immobilised, resulting in improved biocatalytic stability and activity. In contrast to macroscale matrices, an enzyme immobilised on nanoparticle has a low resistance to mass transfer, resulting in increased enzyme stability and operation. Furthermore, magnetic nanoparticles also allow for simple and effective separation using a magnet, allowing for the rapid removal of enzymes from a substance. Various nano biotechnological aspects of nanocellulosic biorefinery are depicted in Fig.2.

Unlike other traditional approaches, which cause mechanical shearing causes enzyme instability, nanoparticles provide a quick and cost-effective solution for this method. This is a multidisciplinary area of study in which nanotechnology could be used to recognize properties of materials at the nanoscale level and to develop novel applications and products in order to develop high-value eco-friendly materials (Nizami and Rehan, 2018).

Enzyme immobilization during lipase-catalyzed cellulosic ethanol and biodiesel synthesis processes is another interesting application of nanobiotechnology in the biofuel sector. The advantages of nanostructures that might lower the operating cost of large-scale biofuel production facilities. Nano-encapsulation, self-entrapment, and host-guest interactions are examples of nanotechnology-based immobilization methods to enhance the yield of value-added products. Fast catalytic pyrolysis and syngas conversion, biopower, advanced biomaterials for biofuel cells, biochar, carbon capture, storage, gasification, and hydrothermal liquefaction are some of the potential areas of agro-residue utilization for value-added products where nanotechnology could deliver more breakthroughs. Precision farming based on nanotechnology might increase crop output and feedstock creation. To overcome both engineering and scientific restrictions, nanotechnology might make a substantial contribution to energy efficiency and storage, transformation, transportation, and end-product usage.

The overall techno-economic efficiency of biorefinery is also dependent on many nanotechnological aspects such as supply chain aspect ratio specifications starting from the biomass feedstock and ending with the finished product. To distinguish between methods for determining plant-level efficiency, this is related to the design and operating parameters used at each site and methods for evaluating performance of the supply chain (Zetterholm et al., 2020). The conversion of raw materials into nano dimensions during processing enables the development of more value-added products in reduced cost that comes with manufacturing biomass resources in order to build economically feasible biorefineries (Rebello et al., 2020). The by-products productivity can also increased in nanosized materials that make more number of value added products from biorefinery. As a result, simple nanotechnological methods with low energy or material costs profit the most in the end. In terms of economic growth, the choice of sources and pretreatments is critical in deciding the cost-effectiveness of biorefinery production.

4. Major Lignocellulosic sources and Pretreatments

Many biomass sources have also shown potential as product precursors in a biorefinery environment, including sugarcane bagasse, coconut, soy, corn, sorghum, kenaf, grasses, plantain, hemp and a variety of others (Maheshwari, 2018; Reshmy et al., 2021c). However, extracting and processing the materials to obtain precursors is difficult due to the high pressure and temperature used to process components like lignin, hemicellulose and cellulose (Pandiyan et al., 2019). The utilization of cellulosic biomass from industrial and agricultural activities has proved to be an effective way to manage and valorize these wastes. Major sources and its pretreatment techniques are listed in Table 1. Though several pretreatment strategies are available, alternative strategies seems to be promising for better hemicelluloses and lignin removal (Sindhu et al., 2016a; Sindhu et al 2016b; Sindhu et al., 2013; Sindhu et al; 2011).

Some examples of the alternative strategies include microwave-assisted ultrasonication (Sindhu et al., 2018), sono assisted acid pretreatments (Sindhu et al., 2016), detoxification of lignocelluloses hydrolysates (Chandel et al., 2013), intracellular cellobiose assimilation (Parisutham et al., 2017), and so on. The cellulose nanofiber (CNF) and cellulose nanocrystal (CNC) manufacturing processes appear to be simple to implement into the core product manufacturing line using well-proven technologies (Maheshwari, 2018; Scopel and Rezende, 2021). Different bioconversion stages including major sources and pretreatments are depicted in Fig.3.

4.1. Agricultural residues

Agricultural residues are mainly composed of hemicellulose, cellulose, pectin, lignin, and wax (Deepa et al., 2011). Agricultural residues have substantially lower cellulose content, but higher lignin content compared to forest wastes. Agricultural cellulosic residues include sugarcane bagasse, rice straws, corn stover, corn cobs, rice husk, and wheat straws among other plant parts other than harvested crops or fruits (Ponnusamy et al., 2018). At the moment, the use of waste materials from agricultural processing is still in its early stages, with less important purposes such as combustion agents or in compost conversion for agriculture. But, in terms of environmental and cost-effective advantages, these applications have yet to reach their full potential or importance from a content conversion perspective (Arevalo-Gallegos et al., 2017). In reality, LCB can be utilized to produce various forms of cellulose, which can be used in a variety of material production fields such as biomedical, cosmetics, automotive, pharmaceuticals or many other industries (Machado et al., 2020).

Phantong et al. used a particular buffer solution to extract the lignin from a raw apple stem. An alkali treatment was used to remove the hemicellulose, allowing the cellulose to be isolated. Finally, an acid processing phase yielded around 52% of the highly crystalline and thermally stable NC with diameter in the range of 10-20 nm (Phanthong et al., 2015). Suopajarvi et al. used deep eutectic solvents treatments for delignification and produced nanopapers from different raw materials such as corn stalk, wheat straw, and rapeseed stem (Suopajärvi et al., 2020). These nanopapers were found to have a competitive potential for lead adsorption. Abraham et al. used steam explosion to isolate cellulose from different agro-residues such as jute fibres and pineapple leaves for NC preparation. These NC possessed a diameter of 4-40 nm and a wide aspect ratio, making it suitable for use as a polymer reinforcing agent (Abraham et al., 2011). Another pathway for NC extraction based on a biological approach involving microorganisms or a specific enzyme act as a catalyst on cellulose moieties, such as bacterial degradation or decomposition by a specific cellulolytic enzyme (Fouzi et al., 2019). The benefit of a biological cellulose degradation process is that the structural properties of NC can be precisely regulated, making it simple to produce high-end quality NC. The disadvantages of this approach include a long-standing reaction time and the cost associated with lower yield (Agbor et al., 2011).

A composite route became one of the most widely recommended methods for improving the process by increasing hemicellulose conversion from agricultural wastes. Kang et al. reported the extraction of NC from corn cob cellulose using a mechano-chemical esterification process, with particle diameters ranging from 1.5 to 3.0 nm. The nanopapers developed from this NC have higher optical property with a transparency of 90% at 550 nm. These can be used to enhance electronic and optical parameters of final products (Kang et al., 2017). Reshmy et al. extracted cellulose fibres from jack fruit peel using a combination of alkaline treatment and acid hydrolysis after a green chemical bleaching using soap nut extract. These CNCs could be used as a reinforcement to strengthen biocomposites that have great potentials in food packaging (Reshmy et al., 2021c). A mild TEMPO-oxidation process followed by ultrasonic treatment was suggested to extract an NC from agro wastes in another recent combination approach (Cheng et al., 2017). Due to its conversion efficiency, product consistency and flexibility for diverse nanocomposites in various forms, the combination method for isolating NC from agricultural wastes is significantly more sustainable and productive.

4.2. Forest residues

Charcoal, firewood, agglomerates, fillers, and wood boards are all examples of low-value-added products made from wood residues (Guo et al., 2015). However, China's government policy on biomass recycling is currently focusing on high-end commodities, which has inexorable significance in some associated sectors. In general, there are four types of NC isolation methods for waste wood material: physical, chemical, biological, and a hybrid approach that combines two techniques for a complementary advantage. Biological methods to extract NC from forest wastes have received very little attention due to its cost-effectiveness or production issues (Agbor et al., 2011).

Chemical treatments are most commonly used for obtaining NC from this source. Moriana et al. used alkaline pretreatments, bleaching and acid hydrolysis to extract CNCs from logging residues. The CNCs with the highest crystallinity and aspect ratio have enormous potentials for use in composite materials reinforcement (Moriana et al., 2016). However, this form has many drawbacks, including a lengthy reaction time, high energy, low yield and corrosive chemicals consumption.

Several studies have looked into the possibility of using other mineral or organic acids, such as hydrochloric acid, phosphoric acid, a blend of phosphoric and sulphuric acids, a blend of hydrochloric and acetic acid, oxalic acid, malonic acid, maleic acid, citric acid combined with ultrasound, and citric acid blended with ultrasound, to produce CNC as an alternative to the traditional sulphuric acid method (Ahmad et al., 2018; Trache et al., 2020). Physical methods are often widely used to extract NC from forest residues, and yields of NC are significantly higher. Phanthong et al. obtained NC with a 93.1% yield using a planetary ball mill. The obtained NC had a diameter of 10-25 nm and was more thermally stable (Phanthong et al., 2018). Vallejos et al. fruitfully isolated NC from Eucalyptus sawdust (Vallejos et al., 2016) and Carvalho et al. prepared CNFs with spruce and birch sawdust (Carvalho et al., 2019). A chemical alteration through bleaching, followed by a solvent casting process, can be used to make the NC thin films. The crystallisation index of the NC thin film will improve with better thermal stability after an alkali pretreatments and bleaching process (Reshmy et al., 2021a, 2020b). It has the ability to be utilized to fabricate nanocomposites in a variety of industries for innovative materials (Al-ahmed and Inammudin, 2020; Reshmy et al., 2020a). The combination approaches are significantly more reliable than pure chemical or physical methods (Hassan et al., 2018; Mascheroni et al., 2016; Xie et al., 2016). Acid/alkali pretreatments are a relatively simple operation, making waste liquid generation simple. As a result, modern and more eco-friendlier and reliable methods for processing NC from wood waste must be explored. Although biological methods may be useful, there are very few reports on the isolation of NC from woody biomass, meaning that our current methods should be improved.

4.3. Aquatic residue

Aquatic residues are plentiful, can grow in a variety of habitats, are inexpensive and simple to produce in the natural environment, and can be harvested all year. Utilization of aquatic plants has been recommended for fertilizer, compost, papermaking, biofuel generation, and other uses since at least the early twentieth century (Wilkie and Evans, 2010). A primary point for attempting to use such plants, aside from the need to limit overgrowth, is that they frequently demonstrate primary productivity rates that are substantially higher than conventional bioenergy feedstock alternatives. In natural lakes, water hyacinths, for example, have been proven to produce yearly crop yields of 100 dry metric tonnes (Gaurav et al., 2020). But, the highest yields for switch grass have been in the range of 25 dry metric tonnes in the USA.

According to research study, the sugar contents of water lettuce hydrolysate is 1.8 times greater than those of water hyacinth, implying that water lettuce may be an even better bioethanol feedstock than water hyacinth. The fast rise of the bioenergy industry appears to present a clear opportunity for the introduction of initiatives that can channel aquatic plant productivity into beneficial uses. The production of value-added bioproducts from non-edible aquatic species residue is receiving more attention (Fouzi et al., 2019).

To address the multiple issues of global warming and eutrophication, the use of aquatic systems as feedstocks for cellulosic biorefineries is also becoming a viable option. Using China as an example, the 2019 Bulletin of China Marine Disaster reports that aquatic residue is a very useful feedstocks or special biological resources because of its high polysaccharides and protein content. Some non-edible green algae as well as enveloping green algae species that cause algae blooms, have been reported as good sources of NC (Fouzi et al., 2019). Algae NC is primarily prepared using chemical and combination processes, similar to how NC is isolated from forestry and agricultural wastes. NC made from algae has higher crystallinity compared to forest or agricultural residues. The resulting NC with a large aspect ratio had a high antibiotic adsorption capability, which could be useful in drug delivery. Correspondingly, Bhutiya et al. isolated NC from *Chaetomorpha antennina* using a bleaching, acid hydrolysis and ultrasonic assisted treatments, yielding 34-85% cellulose crystallinity. The subsequent composites showed excellent thermostability, tensile strength, and some antibacterial properties after being combined with Cu_2O, indicating that they have a lot of potential in eco-friendly products (Bhutiya et al., 2018). An acid hydrolysis method has also been used to isolate NC from brown and red algae residues, such as *Gelidium elegans*, *Gelidium sesquipedale*, and *Laminaria japonica*. A combination of pretreatments, such as Soxhlet extraction, pretreatment, and bleaching can be used to remove polysaccharides, fatty acids, and lipids. In comparison to forest wastes and agriculture residues, NC extraction methods from algae residues are relatively simple due to the noncoherent cellular structure and lack of lignin.

Biocatalyst functions as a promoter of hydrolysis process of fibre components in the biological system (Wahlström and Suurnäkki, 2015). Endoglucanase, cellobiohydrolase, and also hemicelluloses have all been used as biocatalysts in the bioconversion of agrowastes to cellulose. Enzymatic pretreatment has a much prolonged retention period than chemical pretreatment. According to several studies, enzyme hydrolysis combined with homogenization of softwood cellulose and the resulting NC has a higher aspect ratio and is less aggressive than acidic therapy (Phanthong et al., 2018). Another study reports the preparation of bacterial nanocellulose (BNC) production using rice husk using solid acids as catalyst with high yield of reducing sugars. As a result, the bioconversion of carbohydrates to cellulose is a multifaceted process that requires multiple biocatalysts (Chen et al., 2019). In addition to conventional pretreatment, cellulose enzymes were discovered in the eco-friendly method. Many researchers claim that the synergistic effect of enzymes has a positive impact on cellulose biomass surface modification. Cellulases A and B, also known as cellobiohydrolases, act on crystalline cellulose, while endoglucanases of the C- and D-type act on amorphous cellulose. Cellobiohydrolase I and II, which are derived from *Trichoderma reesei* and have high polymerization properties when combined with cellulose-degrading catalyst (Yarbrough et al., 2017).

4.4. Industrial by-products

Another form of valuable recyclable biomass resource for NC extraction is industrial residues or by-products generated from food processing, printing, furniture manufacturing, pulping and papermaking (Carrillo-nieves et al., 2019). Based on differences in the structural and chemical composition of the feedstock, the extraction process for NC from industrial residues is complex and highly dependent on the residue form. Sugarcane bagasse, which has a high lignocellulose content and a loose structure, can be acid hydrolyzed directly to extract NC

(Chandel, 2018). The isolation of NC from furniture industry manufacturing residues was reported using steam explosion and acid hydrolysis procedures for developing hydrophobic NC aerogels to absorb oil. Gibril et al. reported CNCs preparation from pulp and paper mill sludge through ammonium persulfate under oxidising conditions (Gibril et al., 2013). The enzymatic hydrolysis is a novel approach for the extraction of NC from juicing industrial residues with a loose and soft cellular structure. BNC cultivation in nutrient-rich industrial processing water, in addition to extracting NC from industrial solid biomass residues, is another promising method for generating NC from low-value biomass.

Dubey et al. used the *Komagataeibacter europaeus* SGP37 to turn sweet lime pulp waste into high-quality BNC under static intermittent fed-batch cultivation, demonstrating a new route to convert wastes into high-end products. Furthermore, wastes from other factories and households could be used to obtain NC. Ogundare et al. reported the extraction of NCC using ethanol, followed by bleaching, alkali pretreatment, and acid hydrolysis from discarded cigarette filters. In the field of biomedical composites, high purity NC might have some potential application. CNFs with a width of 5-30 nm and a crystallinity of 45% were developed, and they demonstrated superior water redispersibility, implying that they could be used in packaging (Yu et al., 2021).

Homogeneous feedstock, because of their seasonality, has less utilization rate. The advantages of selecting homogeneous feedstock are easy to store, predictable yield, and fixed consumable and enzyme usage. But, in order to improve value-added products generation a better option will be the selection of heterogeneous feedstock such as fruit and vegetable waste, agro-residue mixed with industrial waste, and so on. Heterogeneous feedstock provides composition variations of percentage of cellulose, hemicelluloses and lignin, conversion efficiency, and easy downstream processing in the biorefinery. Therefore, in nearby future, the biorefinery will shift into utilization of heterogeneous feedstock to improve output of processing.

5. Value added products from lignocellulosic biorefineries

Cellulose is an excellent substrate for the production of biopower since it contains about 70% sugar. The separation of lignin from feedstocks is a crucial step in the process. Several pretreatments as discussed above are applied for this: physical, chemical, physicochemical and biological processes. Since separating carbohydrates from cellulosic residue is such an essential step in the process, many available commercially enzymes, such as cellulase from *Aspergillus fumigatus, Aspergillus niger* and *Penicilium purpurogenum* can be used. Commercial status of current lignocellulosic biorefinery is provided in Table 2.

5.1. Energy-driven biorefinery

The growing demand on renewable biomass fuels instead of petroleum is widely regarded as a critical component of the development of a sustainable industrial civilization and successful GHG emission control. Future biorefinery processes will begin by extracting high-value compounds present in biomass, such as perfumes, food-related products, flavoring agents, and high-value health supplements. The biorefinery will emphasis on processing lignin and polysaccharides for bio-derived products and fuels once these comparatively valued compounds have been extracted. Some of the major energy-driven biorefinery products are bioethanol, biomethane, biohydrogen and bioammonia. A pictorial representation of different biofuels produced from biorefinery is depicted in Fig.4.

5.1.1. Bioethanol

Cellulosic ethanol processing entails a number of steps, including enzymatic digestion after physico-chemical treatment, hydrolysate fermentation, and ethanol separation. Since lignocellulosic biomass is recalcitrant to bioconversion due to its structural complexity, to remove biomass intransigence and make it agreeable to microbial and enzymatic attack, a prerequisite physico-chemical processing or treatment phase is performed (Manmai et al., 2020). The pretreated substrate is then depolymerized by enzymes, which are the most expensive process due to the expensive of enzymes. The resulting hydrolysate includes sugars with five and six carbons, which can be fermented to create bioethanol (Raghavi et al., 2016; Sindhu et al., 2017).

Pentose fermenting microbes are uncommon in nature and produce significantly less bioethanol than C-6 fermenting microbes. The bioethanol developed after fermentation is extracted and concentrated in the final step, either by membrane separation or by distilling the medium. All of these issues prompted a push to grow bioethanol-based biorefineries in order to offset the expense of bioethanol with high value-added products. Recent years have seen biotechnological advancements in the generation of bioethanol from aquatic plants, with a special concentration on water hyacinth.

The most common method for producing bioethanol from aquatic residues, like in other cellulosic feedstocks, involves acid hydrolysis of raw materials followed by inoculation of the hydrolyzed material with a suitable fermenting microorganism. From dry water hyacinth residue, a bioethanol yield of roughly 0.05 g/g was produced using *Pichia stipitis* as the fermenting bacterium. Other bacteria, such as *Saccharomyces cervisiae*, produced a higher yield of 0.17 g/g, while *Candida shehatae* produced an even higher yield of 0.19 g/g from dry water hyacinth. The residual unreacted cellulosic residues can be used to manufacture various value-added materials after bioethanol yields fractions such hemicelluloses and cellulose. This could be a worthwhile consideration to minimize the cost of bioethanol (Carrillo-nieves et al., 2019; Gavahian et al., 2018).

5.1.2. Biomethane

A variety of crop residues, such as rye, wheat, rice straw, maize and other grains, could be used as a biomethane substrate (Abraham et al., 2016, 2020). According to estimates the annual cereal and maize wastes have the capacity to generate 2000–4500 MT of methane per hectare (Bazaluk et al., 2020). Biomethanation, like bioethanol processing, is a multistep process. Hydrolysis, acetogenesis, acidogenesis and methanation are all stages in the methane fermentation of cellulosic biomass (Bazaluk et al., 2020). Each phase necessitates a different mix of microorganisms. The microorganisms hydrolyze the undissolved complex structural polymers of cellulose into monomers after passing through different phases. Other species use the monomeric sugars that result from hydrolysis to create various organic acids, C_1–C_5 molecules, CO_2, alcohols, and short chains of hydrogen. During the acetogenic process, organisms convert organic acids and alcohols to acetate. Finally, in the methanogenesis phase, obligate anaerobes convert these carbon sources to methane. A number of products generated in the presence can be retrieved and used to offset the expense of the process. Furthermore, the remaining biomass could be composted, and the method could have a waste management solution.

5.1.3. Biohydrogen

The agriculture waste, kitchen waste, industrial waste, stillage, fibre waste and other cellulosic feedstocks are all viable options for H_2 generation. The method involves only one stage is normally the most cost-competitive of the biohydrogen manufacturing processes feasible. Generally, the cellulose is pretreated and hydrolyzed, and then the hydrolysate is dark fermented for hydrogen output. The pretreatments could have the disadvantage of producing undesirable by-products, which could threaten the hydrolyzate's fermentability (Bru et al., 2012). Sustainable developments are being implemented in this regard, and there has lately been a substantial shift in research interest. Phototrophic microalgal strains are utilized to develop a novel biohydrogen production process that involves two stages with alternating light and dark phases. The procedure also emphasises the importance of combining all procedures in order to generate several products at the same time(Cheng et al., 2012; Nagarajan et al., 2021).

5.1.4. Bioammonia

Bioammonia is gaining attention as hydrogen replacement in biofuel industry. It is a non-petroleum, high-density, ecologically friendly liquid fuel that releases energy quickly when combusted. Although bioammonia has half the fuel economy of gasoline, it emits no CO_2. It is a remarkable substitute for conventional gasoline due to the accessibility of a distribution system, restricted range of fire risk, fast dissipation in air, and strong distinctive smell at an even small concentrations enabling simple detection. The full burning of ammonia produces only nitrogen gas and water vapor (Panahi et al., 2019).

The production of ammonia from biomass feedstock can have a major influence on decreasing GHG footprints due to the potential reduction in the amount of natural gas and other conventional hydrocarbons presently used for its manufacture. To generate various quantities of bioammonia, nowadays about 24 bacterial species have been utilized, that were mostly isolated from the digestive tracts of swine and ruminants dung (Selvaraj et al., 2020). *Eubacterium*, *Clostridium*, *Peptostreptococcus*, *Pseudomonas*, and *Fusobacterium* are the most common hyper ammonia-producing bacteria, with productivities as high up to 680 mg/L/d when employed (Sankaran et al., 2020). Babson et al. described a technique for increasing ammonia output during anaerobic digestion by shifting CH_4 production to NH_3 synthesis. Approximately 61% N flow was converted to NH_3 by changing the C:N ratio in the digesting feedstock. It has been observed that adding a separate degradation fermenter upstream of such anaerobic digester reduces NH_3 toxicity in fermenting microorganisms and improves digester stability (Babson et al., 2013).

Another new technique incorporated enzymatic hydrolysis and microbial submerged fermentation of the food industry residues using *Clostridium* sp., were reported. Total ammonia recovered using NH_3 gas capture technologies, followed by stripping to convert the NH_4^+ ionic form to NH_3 (Chai et al., 2021). Because of the zero carbon emission features of bioammonia-based vehicles, the development of suitable technologies for manufacturing, transportation, and preservation may bring global ammonia-based vehicular applications appealing.

5.2. Material-driven biorefinery

In today's globalized world concerns associated with the widespread usage of petrochemicals, there has been an increase in interest to researchers in the importance of bio-sourced materials over last decade. Biomass derived cellulose in nano dimensions can be very effective as an alternate material for the development of different platform chemicals. Many agro-industrial cellulosic materials have received a lot of attention because they are carbon neutral, renewable, organic in nature, and recyclable, and therefore have a lot of advances in the development of high-value bio-products (Choudhury et al., 2020). The major products from the material-driven biorefinery are shown in Fig.5. From these platform chemicals and biomaterials, numerous other valuable materials can be manufactured (Reshmy et al., 2022).

5.2.1. Platform chemicals

The development of bio-refineries to manufacture platform chemical is a fantastic chance to reduce our inevitable reliance on petroleum-based chemicals. According to recent research, producing platform chemicals from feedstocks necessitates a variety of pretreatment methods, as well as single or multiple pretreatments with the purpose of initially separating cellulose, hemicellulose and lignin fractions (Takkellapati et al., 2018). The early pretreatment systems have the changes depending on the raw biomass existence and technical growth. The cellulose and hemicellulose components are hydrolyzed to sugar monomers after separation, while the lignin fraction is mainly used to manufacture aromatic compounds. The sugar monomers, either C_5 or C_6, are used in the sugar platform of a biorefinery to produce mainly propylene, xylose, galactose, ethylene, acetic acid and mannose (Kumar et al., 2018). The cellulosic biomass converted into platform chemicals such as toluene, glucaric acid, syringols xylitol, aspartic acid, styrene, 3-Hydroxypropionic acid (3-HPA), phenols, glutamic acid, xylene, eugenol, and others has been reported (Arevalo-gallegos et al., 2017; Bilal and Iqbal, 2019). While considering platform chemicals production from water hyacinth, several possibilities are involved. One route is decomposition of hydrolyzed cellulose into fructose and glucose at a temperature above 200ºC. Also, crystalline cellulose converted into polymorph or amorphous polymorph via gelatinization (Gaurav et al., 2020).

Butanol production using lignocellulosic feedstocks are nowadays in a rapid growing face (Vivek et al., 2019; Sindhu et al., 2016). *Clostridium acetobutylicum* was used to manufacture butanol in a biorefinery with a fluffy hemicellulosic wood hydrolysate and alfalfa juice fermentation process. The production of 2,3-Butanediol was reported simultaneous saccharification and fermentation of oil palm residues (Hazeena et al., 2019). *Basfia succiniproducens* was used to manufacture succinic acid from corn stover, yielding 0.43 g/L/h. *Actinobacillus succinogenes* and *Basfia succiniproducens* were used to study the synthesis of succinic acid using depleted sulphite liquor. After fractionation, up to 39 g/L succinic acid, nearly 32.4 g/L LS, and 1.15 g of a phenolic-rich extract were obtained (Kohli et al., 2019). Microwave assisted delignifications were also reported for effective processing using deep eutectic solvents (Kohli et al., 2020).

5.2.2. Biomaterials

Future biorefinery operations will begin by production of high value-added chemicals currently present in biomass, like fragrances, flavouring agents, food-related products, and high-value nutraceuticals with health and medical advantages. Fermentation, as well as enzymatic and chemical transformations, can transform sugars into building-block chemicals in the biorefinery process (Mautner et al., 2016; Mongkhonsiri et al., 2020).

The fermentation products from cellulosic biorefinery processes can be transformed directly into a variety of useful chemicals that can be used to produce biomaterials such as polyethylene, polylactic acid, polyhydroxybutyrate and other biopolymers (Kaur et al., 2017; Sirohi et al., 2020). Methylcellulose, carboxymethylcellulose hydroxypropylcellulose and hydroxypropyl-methylcellulose are examples of some common cellulose derivatives manufactured by surface modification of cellulose in cellulosic biorefinery. In addition, cellulose acetate, cellulose acetate butyrate, cellulose acetate phthalate, hydroxypropyl methylcellulose phthalate and cellulose acetate trimellitate are also derived from cellulose that have been used in commercial products (Abdul Khalil et al., 2017).

The use of ionic solutions, which totally dissolve cellulose and facilitate hydrolytic depolymerization, is a newer method (Usmani et al., 2020). However, a few of the large biomaterials that can be manufactured using cellulosic biorefinery are bioplastics made from plant oils and carbohydrates, biofoams and biorubber made from plant oils and latex, and biocomposites made from agricultural and forestry biofibers, which are used in the manufacture of vehicle door panels and components. Rather than petroleum feedstocks, bioplastics are polymers made from starch crops and vegetable oils. For example, corn starch can be biochemically converted to lactic acid, which can then be chemically converted to polylactic acid for use in production of bioplastic. The biodegradable bioplastics are used in a variety of sectors, including packaging, gardening, cutlery and pharmaceuticals. Plastics prices are directly linked to the price of crude oil, which is fuelling interest in bioplastics production. The bacteria can produce cellulose, which can be extracted and used. BNC is a straight-chain polysaccharide obtained from plants that has the same chemical structure as cellulose. The absence of lignin, pectin, and hemicelluloses in BNC are a benefit. BNC is a high-purity material that has found uses in wound dressings, tissue engineering, and nanocomposite.

6. Future perspective

The potential improvements in nanocellulosic biorefinery outcomes are connected with many steps such as selection of agroresidue, pretreatments, enzyme viability and regeneration, targeted industries, and techno-economical factors. The analysis of the chemical compositions and extraction processes is dependent on the raw material. Heterogeneous feedstock can be utilized to improve the yield of production. But the selection of different biomass residues to use as feedstock should consider the composition of cellulose, hemicelluloses and lignin in them. The acid to fibre ratio, temperature, hydrolysis duration, as well as the applied chemical, mechanical, or enzymatic pretreatments are the factors influencing the type and nano dimension of biorefinery products. The use of collective cellulosic residues from industrial and agricultural activities emerges as the most cost-effective and energy-efficient choice when developing high value-added products in a nanocellulosic biorefinery. In future biorefinery, heterogeneous feedstock utilization is suggested for improving the production of different value-added materials.

Enzyme selection and its regeneration processes also will have a crucial role in economic control of biorefinery processes. If the biochemical approach is to become commercially feasible, new bioagents, modified enzymes with improved activity, reactor designs with improved mass transfer of the substrates into the end products, and better liquid and gas separation procedures are required. The use of novel engineered cellulolytic enzymes for cellulose synthesis allows for the coproduction of C5 and C6 sugars, resulting in a more energy- and material-driven biorefinery. Further researches are needed to determine the process conditions that will enable it to be implemented in upcoming biorefineries. Along with the technological advancements, an evaluation of the technology's cost is needed in order to assess the significant cost generation and competitiveness during manufacturing.

GHG emissions linked to the biorefinery supply network may be monitored during the process to attribute the various flows with the relevant emissions, as this is the main political demand for innovative biorefinery concepts. Extending the system to monitor any further external environmental impacts is necessary for a more comprehensive and cost effective large-scale biorefinery. The economic output of a biorefinery may be influenced differently by changing biomass market values depending on the device boundaries and that should be chosen in the techno-economic assessment of the modern technologies. Future study focusing on bioammonia, biokerosene, and biocrude will be more beneficial. Bioammonia production yields that are currently not commercially viable, platforms may be created based on novel advancements to bring the commercialization of bioammonia as a non-carbon fuel to reality. To improve the lignocellulosic biorefinery strategy and provide more economic strength to biorefinery technologies, simultaneous development of a wider range of material-driven and energy-driven products is required.

7. Conclusion

Agricultural residue is a commonly available, inexpensive and underutilised source of cellulose that could be used to manufacture various platform chemicals and biofuels on a large scale. The integration of locally generated raw materials and industrial residues to form heterogeneous feedstock will open up the processes that generate an intriguing avenue for their commercialization. The development of novel energy-driven or material-driven biorefinery based on lignocellulosic feedstocks will emerge in the next nanogeneration for the development of value-added products. The utmost challenge still remaining is the fine-optimization of pretreatment strategies for diverse biomass varieties to achieve supreme techno-economic feasibility.

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Table 1: Major sources and pretreatments in biorefinery

Category	Source	Pretreatment	Properties	Reference
Agro residues	Rice husk	Chemical treatments /ultrasonication	Diameter: 6-20 nm; Crystallinity:65%; Improved aspect ratios and thermal stability	(Dilamian and Noroozi, 2019)
	Sugarcane bagasse	Acid treatments /ultrasonication	Diameter:18.17-32.84 nm; Crystalline nature: 93%	(Antti et al., 2014)
	Corncob	One-pot esterification	Diameter: 1.5-3.0 nm; Transparency:90%; Tensile strength: 110 -125 MPa	(Kang et al., 2017)
	Jute dried stalks	Steam explosion/ acid hydrolysis	Diameter: 45-50 nm; Young's moduli: 138 GPa; Higher crystallinity	(Martin George et al., 2015)
	Pineapple stalks	Steam explosion/acid treatments	Diameter: 10-40 nm; High surface area	(Abraham et al., 2011)

Forest residues	Pine needles	Chemical pretreatment/ Ultrasonication	Diameter: 25-70 nm; Crystallinity: 66%; High flexiblility and thermal stability	(Xiao et al., 2014)
	Eucalyptus sawdust	TEMPO oxidation	Diameter: 41 nm; Surface area: 60 m^2 g^{-1}	(Vallejos et al., 2016)
	Logging residues	Chemical treatments	High aspect ratios >10; Excellent thermo stability	(Moriana et al., 2016)
	Birch and Spruce sawdust	Chemical treatments	Tensile strain: 80-200 MPa; Young's moduli : 4.8-8.5 GPa	(Achaby et al., 2016)
	Pinecone biomass	Chemical treatment/mechanical grinding.	Diameter: 5-20 nm; Crystallinity: 70%; Tensile strength: 273 MPa	(Rambabu et al., 2015)
Industrial wastes	Cotton linter wastes	Acid treatments	Width: 12 nm and length: 177 nm; Crystallinity: 90.5%; Enhanced hydrophilicity	(Oliveira et al., 2018)
	Paper mill pulp	Treatments using Ammonium persulfate	Width: 10-20 nm and length:150-500 nm.	(Gibril et al., 2018)
	Discarded	Chemical treatments	Width: 8 nm and length: 143 nm;	(Ogundare et

cigarette filters		Crystallinity: 97%.	al., 2017)
Wood furniture waste	Acid treatments/ steam explosion	Diameter: 20-40 nm	(Shahabi-ghahafarrokhi et al., 2015)
Lime residues	Steam explosion/ homogenization	Diameter: 5-30 nm; Crystallinity: 46%; Improved aspect ratio; Better redispersibility	(Jongaroontaprangsee et al., 2018)
Gelidium sesquipedale.	Chemical treatment/acid hydrolysis.	Diameter: 5-40 nm; Crystallinity: 69.8%; Length: 80-430 nm; Improved aspect ratio	(Oliveira et al., 2018)
Cladophorales	Acid treatments	Width: 25-30 nm; Pure crystals with high aspect ratios	(Mihranyan, 2011)
Chaetomorpha antennina	Chemical treatment/acid hydrolysis	Crystallinity:85%; Improved tensile strength and thermal stability	(Bhutiya et al., 2018)
Ulva lactuca.	Chemical treatment/acid hydrolysis	Good absorption capacity	(Rathod et al., 2015)

Table 2: Commercial status of energy-driven and material-driven biorefineries

Company	Country	Source	Pretreatment	Production capacity	Product
Abengoa	USA	Agricultural residues	Biochemical	325,000tons/year, 25 Mgal/year power	Cellulosic ethanol, Bioenergy
Borregard	Norway	Agricultural residues	Modified neutral/acidic sulfite treatment	Pilot plant, 50 kg/h,	Bioethanol
INEOS New Planet Bioenergy LLC	Florida	Industrial by product	Hybrid	Demoplant, 8MG/year	Bioethanol
Chempolis	Finland	Agricultural residues	Organosolv	Demo scale plant, 25,000 tons/year	Bioethanol, Platform chemical
Beta Renewables	Italy	Agricultural residues	Chemical treatments	75 ML/year	Cellulosic ethanol
DuPont	USA	Agricultural residues	AFEX/enzymatic hydrolysis	750 tons/year	Platform chemical
Iogen	Canada	Agricultural residues	Modified steam explosion, enzymatic hydrolysis	70,000 tons/year	Cellulosic ethanol
Inbicon (Dong Energy)	Denmark	Agricultural residues	Liquid hot water, autocatalyzed	4000 tons/year	Cellulosic ethanol

Clariant	Germany	Agricultural residues	Thermal pretreatment/enzymatic hydrolysis	1000 tons/year ethanol	Biothanol, Platform chemical
Verenium Proces	USA	Agricultural residues and forest residues	Mild acid hydrolysis and steam explosion	4200 tons/year	Cellulosic ethanol
Weyland A	Norway	Agricultural residues and forest residues	Concentrated acids	Pilot plant, 75 kg/h	Cellulosic ethanol
CIMV	France	Agricultural residues	Concentrated organic acid solvolysis	Pilot plant, in operation since 200	Bioethanol
Blue Sugars Corporation	USA	Agricultural residues and Industrial byproduct	Thermo-mechanical	4500 tons/year	Cellulosic ethanol, Bioenergy
Lignol	Canada	Agricultural residues and forest residues	Organosolv	Pilot plant, 1tons/day	Biothanol, Platform chemical
Virdia Inc.	USA	Agricultural residues	Concentrated HCl	Demo scale plant	Platform chemical, Biofuel

Figure captions

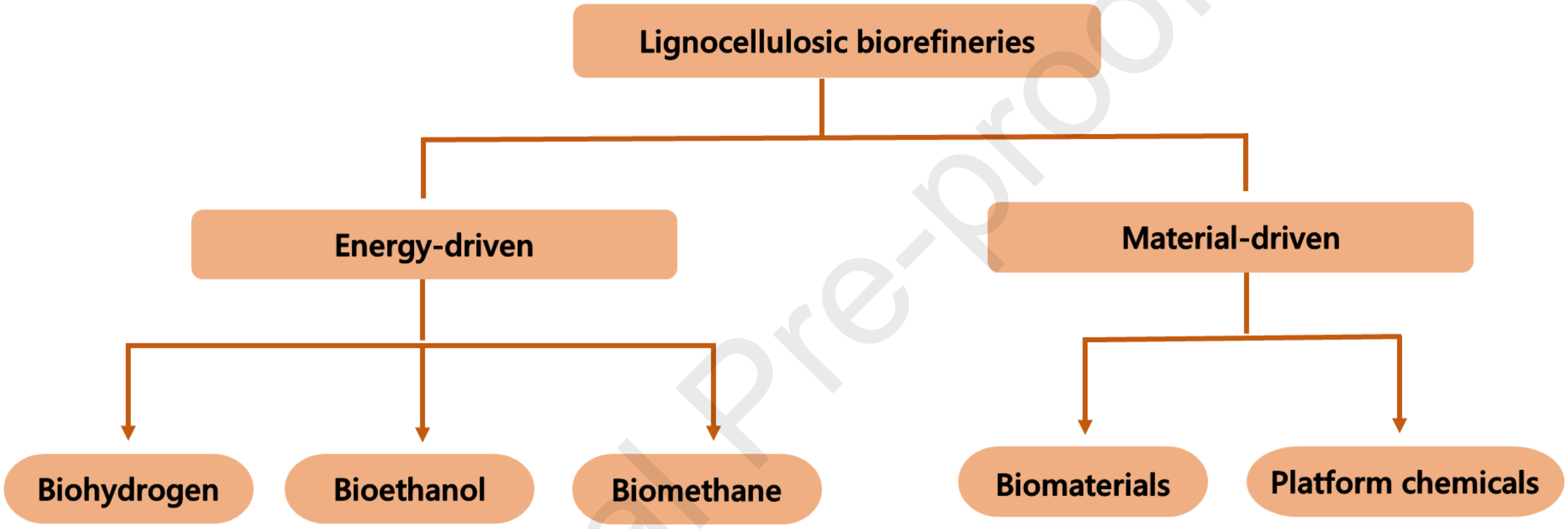
Fig.1. Classification of lignocellulosic biorefinery

Fig.2. Nanotechnological aspects of lignocellulosic biorefinery

Fig.3. Bioconversion steps in lignocellulosic biorefinery

Fig.4. Major value-added products from energy-driven biorefinery

Fig.5. Major value-added products from material-driven biorefinery



Lignocellulosic biorefineries
Energy-driven
Material-driven
Biohydrogen
Bioethanol
Biomethane
Biomaterials
Platform chemicals

Fig.1

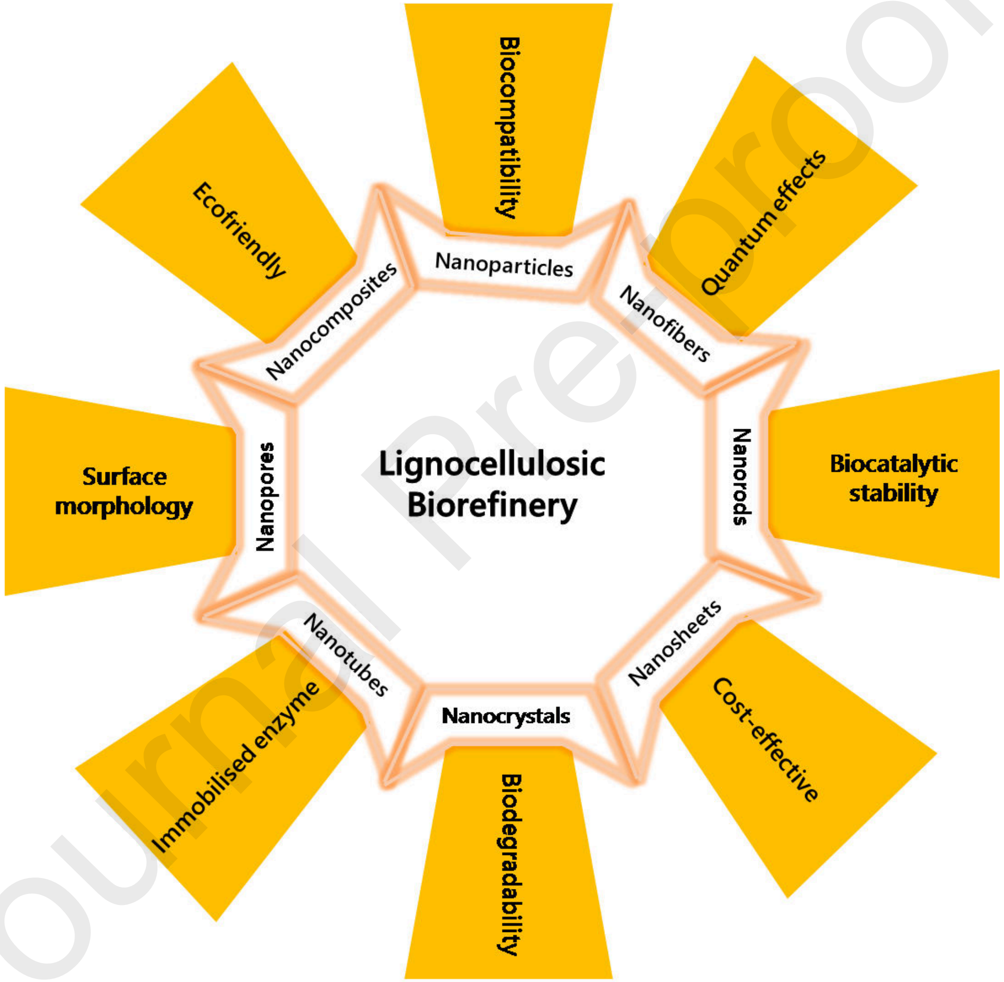

Biocompatibility
Ecofriendly
Quantum effects
Nanoparticles
Nanocomposites
Nanofibers
Surface morphology
Nanopores
Lignocellulosic Biorefinery
Nanorods
Biocatalytic stability
Nanotubes
Nanosheets
Nanocrystals
Immobilised enzyme
Cost-effective
Biodegradability

Fig.2

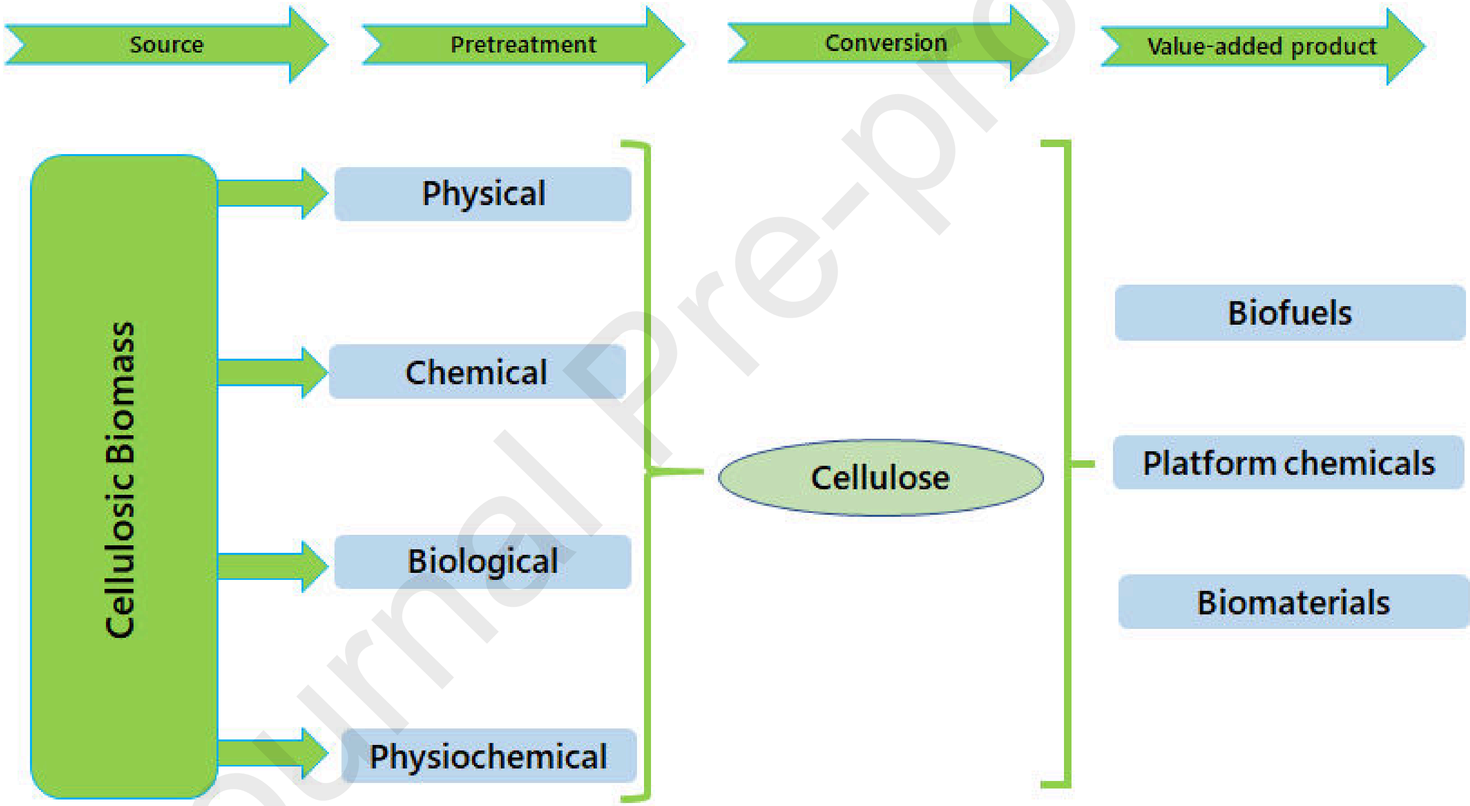

Source
Pretreatment
Conversion
Value-added product
Cellulosic Biomass
Physical
Chemical
Biological
Physiochemical
Cellulose
Biofuels
Platform chemicals
Biomaterials

Fig.3

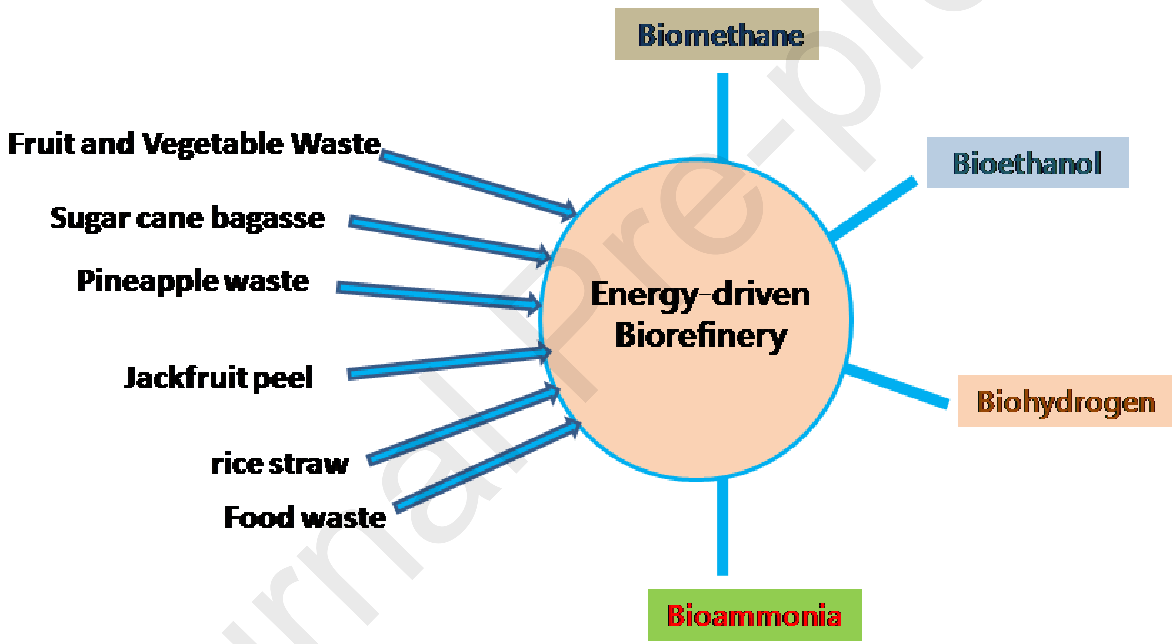

Biomethane
Bioethanol
Fruit and Vegetable Waste
Sugar cane bagasse
Pineapple waste
Energy-driven
Biorefinery
Jackfruit peel
Biohydrogen
rice straw
Food waste
Bioammonia

Fig.4

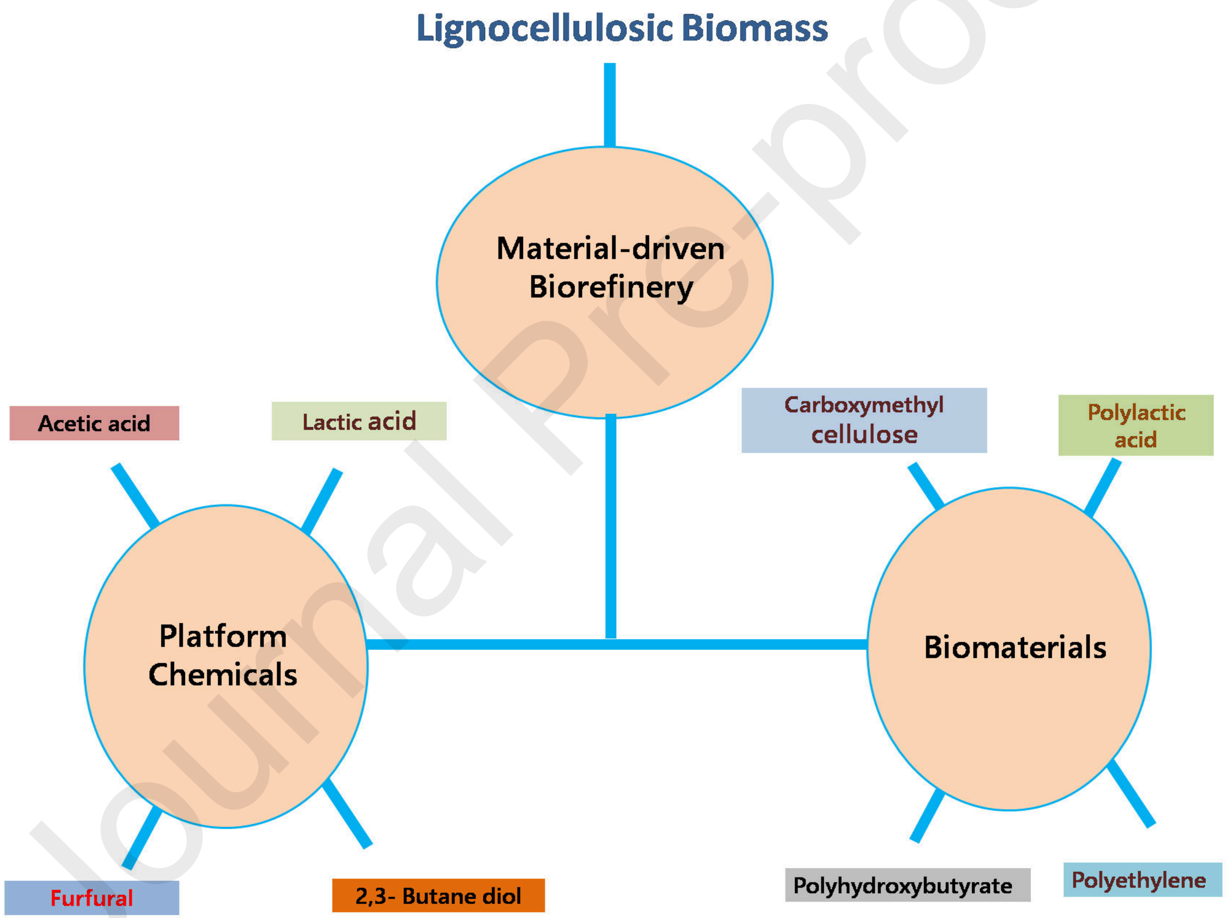

Lignocellulosic Biomass
Material-driven Biorefinery
Acetic acid
Lactic acid
Carboxymethyl cellulose
Polylactic acid
Platform Chemicals
Biomaterials
Furfural
2,3- Butane diol
Polyhydroxybutyrate
Polyethylene

Fig.5

CRediT Author Statement

Reshmy R, Eapen Philip and Aravind Madhavan: Conceptualization, Literature survey, Writing original draft and revision

Ranjna Sirohi, Arivalagan Pugazhendhi, Narisetty Vivek and Mukesh Kumar Awasthi: Writing original draft and editing

Parameswaran Binod and Vinod Kumar: Reviewing and Editing

Raveendran Sindhu: Project Administration, Conceptualization and Visualization

Declaration of interests

☐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. X

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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.

Highlights

- An overview of general aspects of lignocellulose based biorefinery.
- Significance of nanotechnology aspects in developing value-added products.
- Importance of selection of biomass for techno-economical feasibility discussed.
- Exploitation of high yield energy-driven and material-driven concepts.

Lignocellulose in future biorefineries: strategies for cost-effective production of biomaterials and bioenergy

Reshmy, R.

2021-10-28

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