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Chapter

Conversion of Cellulose into Value-Added Products

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Abstract

Rapid industrialization has led to development of various platform chemicals and fossil fuel refinery is one of the mainstays for their production. However continuous depletion of fossil fuels reserves has led an urge to look for alternate source of feedstocks. Among various renewable sources, biomass is found to be most sustainable as it is replaced naturally. Biomass by virtue of its nature is comprised of various recalcitrant polymers and cellulose is one of them, which can be used for the generation of various platform chemicals. This chapter gives a background of cellulose and illustrate platform chemicals that can be produced from cellulose.

Keywords: cellulose, renewable resources, biomass, glucose, value-added products

1. Introduction

Till date most platform chemicals were produced from petroleum refinery where crude oil is converted into fuels and chemicals that provide multiple products and revenue streams but depletion of fossil fuels and global warming are the two major concerns in chemical manufacturing. These concerns have led to the utilization of renewable resources such as biomass as an alternative feedstock for the development of Biorefinery [1]. Biorefineries are facilities that convert renewable biomass into biofuels, chemicals, and materials such as plastics and polymers [2]. The products developed in biorefinery are usually called bioproducts because it is based on the sustainable manufacture of products from biomass, this encompasses all processes, from feedstock production and supply to processing stages and end product manufacturing, as well as research, development, and commercialization procedures. The advantage of renewable resources over fossil fuels is that renewable resources can be replaced over time by natural processes. Moreover, complying green chemistry principle these renewable resources-based methods are also environmental benign. The global market for biorefinery products, by application includes industrial, manufacturing, transportation, flame retardants, safe food and supply, environment, communication, construction/housing, recreation, agriculture, health and hygiene, and energy. As an estimate, the worldwide market for biorefinery products would increase from $586.8 billion in 2020 to $867.7 billion by 2025, with a compound annual growth rate (CAGR) of 8.1 percent from 2020 to 2025 [3].

The main raw material for a biorefinery is the plant biomass, whose structural carbohydrate polymers (cellulose and hemicellulose) can be used to develop a
variety of platform chemicals. The Department of Energy (DOE) of the United States (US) discovered 12 platform chemicals that can be produced from cellulose in biomass: 1,4-diacids (succinic, fumaric, and malic), 2,5-furan dicarboxylic acid, 3-hydroxypropionic acid, aspartic acid, glucaric acid, glutamic acid, itaconic acid, levulinic acid, hydroxybutyrolactone, glycerol, sorbitol, and xylitol/arabinitol [4]. A brief outline of the platform chemicals that are produced from biomass (cellulose) is illustrated in Figure 1.

The application of platform chemicals which makes them very interesting for industrial purpose. In the following section the focus will be on the cellulose, various pre-treatment used in biomass, more information about the platform chemicals and how they are produced from biomass, examples of platform chemicals produced from biomass.

2. Cellulose

Cellulose (C₆H₁₀O₅)n is a stable crystalline homopolysaccharide polymer composed of the anhydroglucopyranose monomers formed via β-1,4-D-glucose linkage of carbohydroglucose units and contains several intra and intramolecular hydrogen bonds as well as intermolecular van der Waals forces [5]. Cellulose is insoluble in water and unlikely to be dissolve in most organic solvents due to the strong intramolecular and intermolecular hydrogen bonds which increases the rigidity [6]. Natural cellulose molecules are bundle of microfibrils (i.e, crystalline and amorphous regions) that are stabilized by hydrogen bonds between hydroxyl groups [7]. Figure 2 shows the structure of one chain of the polymer cellulose [8].
3. Pre-treatment of plant biomass

Cellulose, the most abundant natural polymer, is predominantly found in plant cell wall. However, besides cellulose, plant cell wall also comprised of hemicellulose and lignin, which make an interwoven structure in plant cell wall to make it recalcitrant for microbial and/or enzymatic attack [9]. Table 1 illustrate how the compositional analysis of different type of biomass differs.

The conversion of biomass into value added chemicals involves the hydrolysis of plant biomass to fermentable sugars. However, to access cellulose a pre-treatment step to remove hemicellulose and lignin is required [20]. Biomass recalcitrance is due to various factors such as the degree of lignification, crystalline structure of cellulose, and the structural heterogeneity and complexity of cell-wall elements and it needs to be overcome for valuable exploitation of lignocellulosic feedstocks [21]. The recalcitrant structure of lignocellulose is disrupted during the pre-treatment process, resulting in lignin sheath breakage, hemicellulose degradation, and a reduction in crystallinity and degree of polymerization of cellulose [22, 23].

There are various types of pre-treatments such as physical methods (such as milling, grinding, irradiation, and sonication); chemical methods (such as alkali, acid, oxidizing agents, organic solvents, ionic liquids, and deep-eutectic solvents); physico-chemical methods (such as steam pretreatment with/without a catalyst, wet-oxidation, and hydrothermalysis); or biological methods (Table 2).

The alkali pre-treatment involves the addition of bases to biomass, which increase the internal surface by swelling, and it decreases the polymerization degree and crystallinity, destruction of links between lignin and other polymers, and lignin breakdown [27, 28]. While, acid treatments are known to extract hemicellulose by disrupting the Van der Waals force, hydrogen bonds, and covalent bonds that hold the biomass components, which consequently causes the solubilization of hemicellulose and the reduction of cellulose [27]. Steam explosion is another important pretreatment method. Steam explosion is carried out by treating biomass with high-pressure saturated steam, and then the pressure is abruptly lowered, causing the

| Biomass                | Cellulose (%) | Hemicellulose (%) | Lignin (%) | Reference |
|------------------------|---------------|-------------------|------------|-----------|
| Sugarcane bagasse      | 44            | 28                | 24         | [10]      |
| Wheat straw            | 30            | 50                | 15         | [11]      |
| Rice straw             | 28–36         | 23–28             | 15–23      | [12]      |
| Oil palm fronds        | 45            | 20                | 11         | [13]      |
| Cotton straw           | 43            | 27                | 18         | [14]      |
| Empty fruit bunch      | 41.1          | 13.3              | 25         | [15]      |
| Kenaf                  | 32            | 9.9               | 40         | [15]      |
| Corn stover            | 34            | 21                | 15         | [16]      |
| Switch grass           | 31–38         | 25–32             | 17–18      | [17]      |
| Sorghum                | 34            | 22                | 19         | [16]      |
| Miscanthus             | 39            | 20                | 21         | [16]      |
| Elephant grass         | 36            | 24                | 28         | [18]      |
| Bamboo                 | 45            | 24                | 20         | [19]      |
| Corn hobs              | 45            | 35                | 15         | [11]      |

Table 1.
Various feedstock composition analysis.
Steam explosion is generally initiated at a temperature of 160–260°C (corresponding pressure, 0.69–4.83 MPa) for few seconds to a few minutes before the material is subjected to air pressure [29]. It facilitates the removal of hemicelluloses from the microfibrils exposing the cellulose surface and increasing the enzyme accessibility. Lignin is only partially eliminated during the pretreatment, but it is redistributed on the fiber surfaces as a result of melting and depolymerization/repolymerization processes. Liquid hot water (LHW) treatment is similar to steam explosion, except instead of steam, LHW uses water at extreme temperatures (170–230°C) and pressure (up to 5 MPa). LHW hydrolyzes hemicellulose by releasing its acetyl groups and eliminates lignin, exposing cellulose fibers [30]. Figure 3 illustrates the necessity of pre-treatments in biomass [31].

These pre-treatments methods can be combined depending on the requirement of the pre-treated material. The pre-treatment method selection is not only based on the yields of the cellulose, hemicellulose, or lignin it was able to extract but there are other factors also to consider, such as the effectiveness of the pre-treatment for a certain post-processing step. For instance, for ethanol production besides sugars, low level of toxic inhibitor compounds such as aldehydes, organic acid,
ketones, phenolic compounds, in the hydrolysate is one of the prerequisite factors [32]. While in case of production of polymers, gels or binders from plant biomass, the mechanical strength and swelling properties of the polymers should be kept in consideration while selecting the appropriate pretreatment method.

4. Platform chemicals

Most chemicals produced in biorefinery are platform chemicals. A platform chemical is a building block which can be converted to a wide range of chemicals or materials. Bomtempo et al. [33] listed ways on how to determine if a compound is a platform chemical:

a. Act as a link in the value chain.

b. Have a flexible chemical structure with functional groups that allow them to produce a variety of derivatives.

c. Have interfaces with varying degrees of openness that allow them to be transformed into a diverse range of derivatives at a reasonable cost. These interfaces are chemical and physical transformation processes that can be integrated or performed by external agents.

d. Possess a flexible chemical structure with functional groups that enable them to synthesize a variety of derivatives.

e. Relate to innovation agents with varying levels of competence and diverse interests which is located at different points along the value chain and are interested in different final products, so that they are structured in innovation ecosystems formed by a set of production chains.

f. Are governed by control and command mechanisms that vary depending on the organizational context. The governance mechanisms should enable the platform leader to capture value in a favorable position.

g. Add value by leveraging scope and scale economies.

Various processes have been used for the production of platform chemicals from cellulose in the biomass based biorefinery. Table 3 summarize different methods used to produce the platform chemicals from plant biomass.
5. Cellulose derived platform chemicals

Among various platform chemicals produced from cellulose, ethanols and acids have already been explored more hence in this chapter the focus is on the less explored platform chemicals which may have more enormous potential.

**Levulinic acid (LA):** Levulinic acid (LA) also known as 4-oxopentanoic acid with a molecular formula of C$_5$H$_8$O$_3$ is one of the twelve valued added chemicals that can be produced from biomass. LA is a ketoacid because it consists of one ketone group (CO) and one carboxylic group (COOH) in its structure [36–38]. Owing to its highly reactive nature LA serves as a versatile platform molecule for fuel additives, resin, herbicides, pharmaceuticals, flavor substances and chemical intermediates with wide potential industrial applications [39–41]. The US Department of Energy selected LA as one of the “12 top value-added compounds from biomass” that may be utilized to produce a variety of high-volume organic chemicals with a wide range of potential industrial uses [39, 40]. Figure 4 illustrate levulinic acid derivatives applications.

Table 3.
Biorefinery platform chemicals and production processes [34, 35].

| Building blocks | Processes |
|----------------|-----------|
| 1. 5-Hydroxymethyl furfural | Dehydration of fructose |
| 2. Levulinic acid | Acid-catalyzed hydrolysis of glucose |
| 3. Furfural | Dehydration of xylose |
| 4. Succinic acid | Fermentation of glucose |
| 5. Sorbitol | Enzymatic hydrolysis and hydrogenation of glucose |
| 6. Glycerol | Propylene is synthesized in a variety of ways. |
| 7. Ethanol | Thermal gasification with chemical catalytic method, or biochemical fermentation process |
| 8. Lactic acid | Fermentation of glucose |
| 9. Xylitol | Hydrogenation of xylose |
| 10. 2,5-furandicarboxylic acid | HMF oxidation |
| 11. Isoprene | Fermentation |
| 12. 3-hydroxypropionic acid | Fermentation of glycerol and oxidation |

Undergoing condensation reaction, LA produces diphenolic acid (DPA), which is a replacement of bisphenol-A (BPA) used in food containers and consumer products [34]. $\gamma$-valerolactone can be produced by hydrogenation of LA, which subsequently can be used as a solvent for lacquers, insecticides, liquid fuel, food additive and adhesives, and brake fluid. Another LA based product 5-amino levulinic acid (5-ALA), a porphyrin precursor, is produced via two ways through C4 and C5 pathways. The C4 process, which is present in photosynthetic bacteria, yeast, and human cells, involves the pyridoxal-phosphate-dependent condensation of succinyl-CoA and glycine by ALA synthase (EC 2.3.1.37). The C5 process, found in photosynthetic algae and cyanobacteria, uses glutamate as a co-substratum with ATP and NADPH, and consists of three stages catalyzed by glutamyl-tRNA synthetase, glutamyl-tRNA reductase, and glutamate-1-semialdehyde aminotransferase [42]. 5-ALA is used to
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Treat actinic keratosis of the face, scalp, and upper extremities, as well as to visualize a glioma, esterification of LA results levulinate esters which are used in the flavoring and fragrance, solvents, plasticizers, or as a blending component in biodiesel [43]. GF Biochemicals located in Caserta, Italy commercialized the production of LA from biomass in 2015 [44].

The conversion of the lignocellulosic biomass to LA requires pre-treatment to remove the lignin and hemicellulose and be left with cellulose which is where LA is produced. To demonstrate effective polymerization of biomass carbohydrate, the catalytic conversion of biomass to LA needs the presence of both Bronsted and Lewis acids [45, 46]. Current commercial production of levulinic acid utilizes sulfuric acid which is corrosive and not environmentally friendly hence more research now focuses on utilizing ionic liquids because they are environmentally friendly. Ionic liquids, which are frequently fluid at room temperature and composed of ionic species, are known as “designer solvents” because their unique characteristics for a given demand may be obtained by suitable alteration of cations or anions. For example, the addition of sulfonic acid (SO$_3$H) groups and carboxylic acid groups clearly increased their acidities and water solubility, allowing for the development of ecologically acceptable acidic catalysts [47, 48]. Another approach to synthesize LA is one-pot biomass conversion in ionic liquid [49, 50]. However, compared to biomass feed without separation, separated pre-treated biomass will still produce more LA in the following reaction [51].

**Succinic acid (SA):** Succinic acid (SA) is another important cellulose derived platform chemical. It is member of the C4-dicarboxylic acid family. Succinic acid is also known as butanedioic acid or amber acid, occurs naturally in humans, animals, plants, and microorganisms [52, 53]. Succinic acid is a platform chemical hence it can be used as a precursor to produce variess chemicals namely 1,4-butanediol, tetrahydrofuran, γ-butyrolactone and other bulk chemicals [54]. Figure 5 illustrate succinic acid applications.

Various chemicals can be produced from SA. To mention few, succinate ester is produced via esterification reaction, which is used as a precursor for...
1,4-butanedion, γ-butyrolactone, and tetrahydrofuran [26, 27, 34, 55, 56]. Succinic acid anhydride produced via dehydrogenative cyclization reaction is utilized as a starting material to produce fumaric acid and maleic acid [55]. Coatings, surfactants, dyes, detergents, green solvents, and biodegradable polymers are common SA uses [57]. The worldwide succinic market is predicted to be USD132 million in 2018 and is expected to grow to USD183 million by 2023 [57].

Succinic acid is produced commercially by chemical processes from maleic anhydride, a petroleum-based raw material. Although chemical hydrogenation produces a high yield, purity, and selectivity of succinic acid, it is a complex and costly process with environmental concerns [58]. A more viable method is to use microorganisms to generate succinic acid. Furthermore, bio-based succinic acid has additional environmental benefit using CO₂, a greenhouse gas, as a substrate [59]. Many research on bio-based SA synthesis use pure sugars as substrates. High yields have been produced in these cases using Anaerobiospirillum succiniciproducens, Asuccinogenes, modified strains of Escherichia coli, and Mannheimia succiniciproducens. The biological synthesis of SA is presently being researched as well using lignocellulosic sugars. Succinic acid has been produced by fermentation of corn fiber and sugarcane bagasse [60]. Kuglarz et al. [61] reported on the succinic acid production from rapeseed straw after dilute-acid pre-treatment.

Several companies, such as Myriant, Reverdia, BASF, and BioAmber have commercialized production of succinic acid from glucose [62–64] but SA production from lignocellulosic hydrolysate has yet to be realized at commercial scale [60].

Sorbitol: Sorbitol (D-glucitol, D-sorbitol, D-glucohexane-1,2,3,4,5,6-hexol) is a sugar polyol that is widely utilized in nutrition, cosmetics, and medicinal and industrial purposes. Sorbitol is utilized as a low-calorie sweetener, as a humectant in cosmetics and medicinal goods, and as an intermediate platform for the production of value-added compounds such as 1,4-sorbitan, isosorbide, glycols, L-ascorbic acid, and so on [65]. Sorbitol is one of the most promising platform molecules included in the list of the twelve building block chemicals of highest potential derived from biomass [66, 67]. Figure 6 illustrate the sorbitol and derivatives applications. Because of sorbitol’s rising industrial relevance, there is a lot of interest in upgrading manufacturing methods and looking into novel methods.
The direct conversion of cellulose to sorbitol includes two primary reactions: cellulose hydrolysis to glucose, which is facilitated by the presence of acidic sites, and subsequent hydrogenation of glucose to sorbitol over metal catalysts [68]. One of the major problems in the manufacture of sorbitol is the high cost of processing the raw materials, notably the conversion of cellulose to glucose and the separation process; hence, attempts have been made to accomplish direct conversion of cellulose into sorbitol [69, 70]. Efforts have been undertaken to create a one-pot conversion of cellulose into sorbitol in order to minimize current manufacturing costs. Fukuoka and Dhepe. [71] evaluated the effectiveness of various supporting metal (platinum, Pt, and ruthenium, Ru) catalysts in the conversion of cellulose into sugar alcohols, namely sorbitol and mannitol Pt/g-Al\textsubscript{2}O\textsubscript{3} produced the greatest yield (31%) of sugar alcohols with a molar ratio of sorbitol/mannitol of 4:1 or higher.

Zhang et al. [72] produced sorbitol directly from cellulose with the Cu/Al/Fe catalyst in an exceptionally low concentration of phosphoric acid (0.08 percent, w/w), yielding 68.07 percent sorbitol When compared to the fresh catalyst, the catalytic activity of Cu/Al/Fe dropped by 29% in the fourth run. A typical difficulty with direct cellulose-to-sorbitol conversion is the poor sorbitol yield when compared to glucose-to-sorbitol conversion in a commercial process [73]. Zhu et al. [74] also produced sorbitol from cellulose using a sulfonic acid-functionalized silica-supported ruthenium catalyst (Ru/SiO\textsubscript{2}–SO\textsubscript{3}H), the catalyst was reused five times with a slight decrease in sorbitol yield (up to 61.2%). Sorbitol is commonly industrially produced from the hydrogenation reaction of glucose using metal catalysts, and it global market size is projected to reach USD 2918.1 million by 2026, from USD 2400 million in 2019, at a CAGR of 2.8% during the forecast period 2021–2026 [68, 75–77]. However, the noble metal catalyst is extremely costly for industrial use [74].

Furans: Biorefineries can also produce the sugar degradation products such as furans (furfurals and hydroxymethylfurfurals), which is another important platform chemical. Figure 7 illustrates furfural and 5-hydroxymethylfurfural applications. Furfural is a natural dehydration product of xylose, a monosaccharide often present in high quantities in the hemicellulose portion of lignocellulosic biomass, from which it is almost entirely generated. In theory, any substance having...
a significant quantity of the pentose (five carbon) sugars arabinose and xylose can be used as a raw material for furfural synthesis [78]. It is an aldehyde that consists of heteroaromatic ring. The US Department of Energy [36], which was later revised by Bozell and Petersen [37] identify furfural as one of the top 30 added-value compounds from biomass due to the factors such as manufacturing cost, market price, and function as an intermediary in the manufacture of other important chemicals [36]. These factors are increasing demand, which is anticipated to double between 2014 and 2022 [79]. Furfural is a renewable platform chemical with a diverse chemistry that has the potential to generate new families of bio-based, sustainable chemicals.

These bio-based, sustainable chemicals can be produced via selective hydrogennolysis, reduction, ring opening, aldol condensation reactions [80]. Furfural is a substantial component of bio-oil and is widely used in the production of pharmaceuticals, resins, food additives, fuel additives, and other specialty chemicals [81, 82]. It is a significant component of bio-oil and widely applied in the manufacture of medicines, resins, food additives, fuel additives and other special chemicals [81, 82]. Tetrahydrofurfuryl alcohol, tetrahydrofuran, dihydropyran, acetylfuran, furfurylamine, and furoic acid are other significant furfural-derived compounds. Furfuryl alcohol synthesis followed by acid hydrolysis can also be used to produce levulinic acid from furfural. Furfural is also used in the production of medicines, cosmetics, perfumes, flavors, and resins (the latter produced by condensation with phenol, formaldehyde, acetone, or urea to make thermosetting resins with exceptional physical strength), as well as household cleansers and detergents [79]. While, 5-(hydroxymethyl)furfural (5-HMF) consists of a furan ring with two functional groups namely aldehyde and alcohol group. 5-HMF is utilized for the production of value-added fuels and chemicals (biofuel, solvents, polymers, adhesives, plastic, pesticides, and organic compounds) that were only produced from petroleum-based feedstock [57, 83, 84] 5-HMF is a precursor for the production of levulinic acid,
2,5-furan dicarboxylic acid (FDCA), 2,5-diformylfuran, dihydroxymethylfuran, and 5-hydroxy-4-keto-2-pentanoic acid [85].

Glycerol: Glycerol is another important platform chemical that is produced from biomass-based refineries. Figure 8 illustrates chemicals produced from glycerol and their applications. Glycerol is a versatile carbon source and used as an important raw material for food, pharmaceutical, and cosmetic manufacturing process [86–89].

Glycerol is a common energy-producing food that is widely distributed in food, both naturally and as a GRAS (generally recognized as safe) additive. Glycerol is a solvent that is used in the production of flavors and food colors. It’s also used in low-fat food products like cookies as a humectant, plasticizer, emollient, sweetener, and filler.

Glycerol is used in the production of dynamite and propellants (nitroglycerol), cosmetics, candy, liqueurs, printing and copying inks, lubricants, pharmaceuticals (suppositories, cough syrups, elixirs, expectorants, and cardiac medications), personal care products (toothpaste, mouthwashes, skin care products, hair care products, and soaps), and antifreeze. Glycerol can also be utilized to treat glaucoma-related intraocular pressure and cerebral edema [90]. Glycerol is also used to keep textiles malleable, as well as cellophane and high-quality papers flexible and durable. Furthermore, glycerol derivatives include glycerol carbonate, which has several uses in the production of industrial compounds such as glycidol and polymers, coatings, adhesives, and lubricants [34].

Isoprene: Isoprene is a C5 platform chemical that is mostly used for the synthesis of polymers, is another platform chemical to be produced from biomass-based refineries. Figure 9 illustrate isoprene and derivatives applications. Conventionally, most of the isoprene manufactured is converted into polyisoprene polymer, which is utilized in a wide range of items including footwear, mechanical instruments, medical equipment, rubber tires, and sporting goods [34].

Isoprene is an important platform chemical for synthesizing pesticides, medicines, oil additives, fragrances, and more and is especially important in the rubber production industry. Isoprene is still commercially produced from petroleum-based feedstocks because the utilization of biomass as the feedstock is under investigation.
Biologically isoprene is produced by fermentation of glucose. Since, the conventional production processes of isoprene is unsustainable, and the chemical synthesis processes could cause serious environmental problems [91].
Lactic Acid: Lactic acid is another platform chemical which is produced from biorefinery. It is commercially produced via the fermentation of various sugars namely glucose, sucrose, or lactose [92]. Figure 10 shows lactic acid and derivatives applications. Direvo industrial biotechnology produces lactic acid from biomass in a single step using consolidated bioprocessing technology [93]. Lactic acid as a platform chemical, produces chemicals such as lactate esters by esterification which is used as a green solvent. The reduction of lactic acid produces propylene glycol, which is used in food industry as a humectant, solvent, and preservative. Besides, lactic acid can also be used in medicine as a drug stabilizer and a solvent, in cosmetics as a humectant and can also be used in E-cigarettes [94].

6. Conclusions

Biomass based biorefineries are the best sustainable alternative to the fossil fuel crisis and environment protection. There are varies types of biomass with varying cellulose, hemicellulose, and lignin content. To extract cellulose from biomass a pre-treatment is necessary due to biomass components being recalcitrant. The common pre-treatments have disadvantages and limitations, and it is necessary to understand that the pre-treatment cannot be selected based on the yield of cellulose only because there are other effect caused by the pre-treatment which may be in conflict with the next reaction step to value-added products. This chapter have made an attempt to throw some light on the significance of the use of cellulose as a potential feedstock for various platform chemicals. Although the production of such platform chemicals using green chemistry approach is in very nascent phase however these processes have shown immense potential. Both government and industries should switch their focus towards the development of environmentally friendly and efficient process for such platform chemicals.

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Conflict of interest

The authors declare no conflict of interest.
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