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1 **Pretreatment of Lignocellulosic Feedstocks**

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12 **Abstract**

13 With the aim of reducing greenhouse gas (GHG) emissions and with our dependence
14 on non-renewable fossil fuels, lignocellulose has been proposed to be an alternative
15 sugar-rich raw material for renewable biofuel production, especially for the
16 transportation sector. Biorefineries can efficiently convert lignocellulosic biomass
17 into fuels, value-added chemicals, and other forms of energy. When lignocellulose is
18 transformed using biochemical routes, via sugar conversion platform the highly
19 recalcitrant structure of lignocellulosic materials hampers the conversion process by
20 limiting the accessibility of the chemical building blocks. It is therefore imperative to
21 include a pretreatment step to reach high overall yields and productivity in subsequent
22 hydrolysis and fermentation steps. The main purpose of pretreatment is to break down
23 the structure of lignin and/or to solubilize hemicellulose in order to improve the
24 accessibility of cellulose towards hydrolytic enzymes. The effects of pretreatment on
25 the different lignocellulosic polymers depends on the type of pretreatment itself and

26 the conditions used. This chapter outlines the most common physical, chemical,
27 physicochemical, and biological pretreatment technologies that have been developed
28 and optimized over last four decades for disruption and/or fractionation of
29 lignocellulosic feedstocks. The mechanisms of action and the potential benefits and
30 drawbacks of each pretreatment are listed and discussed. Furthermore, the
31 pretreatment technologies are also analyzed from an economic and environmental
32 point of view to evaluate their sustainability.

33 **Keywords:** Sugar biorefinery platform; Biomass fractionation; Biofuels.

34

35 **1. Introduction**

36 After the first oil crisis in the mid-late twentieth century, the need for a more
37 sustainable and renewable energy system became clear. Today, 40 years later, a
38 continuous increase in energy demand and the urgency in reducing carbon emissions
39 are still good reasons for us to give up our dependence on non-renewable fossil
40 resources. Consequently, the scientific community is in the process of helping to
41 develop and implement novel technologies to establish the basis of a new bio-based
42 economy that will bring relevant products for the energy, chemical, material, and food
43 sectors. In this context, lignocellulose derived from plant biomass is a great source of
44 renewable raw material in a bio-base economy because it is widely available,
45 relatively inexpensive and do not compete with food production (FitzPatrick et al.
46 2010).

47 Lignocellulose is considered the most abundant renewable organic matter in nature,
48 with an estimated annual production of more than 10^{10} MT worldwide (Sánchez and
49 Cardona 2008). It includes agricultural waste, forest products, and energy crops, and
50 its chemical composition varies depending on the raw material (Table 1). The main

51 components of lignocellulosic biomass are cellulose, hemicellulose, and lignin. From
52 a biochemical point of view, cellulose and hemicellulose consist of sugar units
53 (glucose, xylose, mannose, arabinose, etc.), while lignin incorporates phenol-derived
54 monomers (*p*-coumaryl, coniferyl, and sinapyl alcohol). Due to their different
55 properties, each polymer has a specific role in the lignocellulosic material. Cellulose
56 fibers interact to each other by hydrogen bonds, forming a compact crystal structure
57 that confers rigidity and stability. Hemicellulose, on the other hand, acts as a link
58 between cellulose and lignin, while lignin provides a recalcitrant matrix together with
59 cellulose and hemicellulose and protects these two components from chemical and
60 biological degradation (Brett and Waldron 1996). Furthermore, lignin is able to form
61 ether- and ester-type covalent bonds with hemicellulose, forming lignin-carbohydrate
62 complexes (LCCs) (Jørgensen et al. 2007). These intrinsic properties of lignocellulose
63 give a three-dimensional structure that is difficult to disrupt. For optimal use of
64 lignocellulosic feedstocks as an energy source, a pretreatment step to improve its
65 digestibility is therefore essential.

66 *1.1. Purpose of pretreatment*

67 In a sugar biorefinery platform, lignocellulose is converted to several value-added
68 compounds such as ethanol, organic acids, or lipids, by enzymatic hydrolysis and
69 microbial/chemical catalysis processes (Fig. 1). Enzymatic hydrolysis of cellulose is
70 carried out by cellulases, which major components are endo-1,4- β -D-glucanases (EC
71 3.2.1.4., attack cellulose regions with low crystallinity, creating new free chain ends),
72 exo-1,4- β -D-glucanases (EC 3.2.1.91., degrade the cellulose molecule by releasing
73 cellobiose units from the free chain ends), and 1,4- β -D-glucosidases (EC 3.2.1.21.,
74 hydrolyze cellobiose to produce glucose) (Jørgensen et al. 2007).

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