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Status of Canada's lignocellulosic ethanol: Part I: Pretreatment technologies

Edmund Mupondwa[a,b,*], Xue Li[a], Lope Tabil[b], Shahab Sokhansanj[c,d], Phani Adapa[e]

[a] Bioproducts and Bioprocesses National Science Program, Science and Technology Branch, Agriculture and Agri-Food Canada, Government of Canada, Saskatoon Research and Development Centre, 107 Science Place, Saskatoon, SK, Canada S7N0X2

[b] Department of Chemical and Biological Engineering, University of Saskatchewan, 57 Campus Drive, Saskatoon, SK, Canada S7N5A9

[c] Department of Chemical and Biological Engineering, University of British Columbia, Vancouver, BC, Canada V6T1Z3

[d] Environmental Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA

[e] Global Institute for Water Security, University of Saskatchewan, National Hydrology Research Centre,11 Innovation Boulevard, Saskatoon, SK, Canada S7N 3H5

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ABSTRACT

Canada is endowed with abundant lignocellulosic biomass from agriculture and forestry. These sources provide a foundation for the development of Canada's cellulosic ethanol biorefinery concept which is supported by government renewable energy policy initiatives. However, the chemical structure of lignocellulosic biomass comprising carbohydrate polymers and lignin makes the structure recalcitrant to deconstruction, thereby constraining the ability of enzymes to convert these polymers into fermentable sugars without expensive and highly capital intensive pretreatment processes. The challenges are further compounded by the diversity of lignocellulosic biomass available in Canada, which typically necessitates commercial pretreatment pathways optimized for each feedstock type. In turn, these conditions constrain the development of viable business models for the commercialization of Canada's cellulosic ethanol biorefinery concept. In order to address these challenges, Canadian researchers have continued to undertake research to develop pretreatment technologies applicable to several Canadian lignocellulosic biomass sources. The objective of this paper is to review contributions by Canadian researchers vis-à-vis the development of bioconversion pretreatment technologies needed to advance the commercialization of Canada's cellulosic biorefinery concept. These pretreatment technologies include physical, physico-chemical, biological, and processes that combine these methods. This paper also highlights the role of multi-institutional science and innovation collaborative approaches for advancing Canada's cellulosic ethanol biorefinery concept further downstream.

1. Introduction

Canada's potential to be a major player in the development of a cellulosic biorefinery industry is well recognized, especially given its abundant supply of agricultural-based lignocellulosic biomass [1,2]. This available supply capacity is augmented by Canadian research efforts to develop and adapt dedicated lignocellulosic bioenergy crops such as perennial forages and grasses (alfalfa, wheatgrass, switchgrass, bluestern, needlegrass) in various Canadian agronomic regions [3–6]. In spite of this recognized potential, there are concerns about the pace of full-scale commercialization of cellulosic ethanol which was expected to represent the next industry milestone in the evolution of Canada's cellulosic biorefinery concept.

A key challenge in the advancement and commercialization of cellulosic ethanol is associated with the chemical structure of lignocellulosic biomass (Fig. 1) whose carbohydrate polymers and lignins make the structure recalcitrant to deconstruction, thereby entailing highly capital intensive pretreatment processes to make cellulose more accessible to enzymes that convert the carbohydrate polymers into fermentable sugars followed by hydrolysis and fermentation, two additional steps in the bioconversion of lignocellulosic feedstocks to ethanol. Among these steps, pretreatment has been identified as the most challenging both technically and economically because of its potential to constrain a biorefinery's upstream and downstream processes including rates of hydrolysis, fermentation efficiency, and co-product yield, including generation of waste streams.

The objective of this paper is to review recent efforts by Canadian researchers in addressing challenges associated with pretreatment of lignocellulosic biomass in Canada. This paper represents Part I of a related review (Part II) by the authors [7] which provided an integrated

* Corresponding author at: Science and Technology Branch, Saskatoon Research and Development Centre; and Department of Chemical and Biological Engineering, University of Saskatchewan, 57 Campus Drive, Saskatoon, SK, Canada S7N5A9.

E-mail addresses: Edmund.Mupondwa@agr.gc.ca, Edmund.Mupondwa@usask.ca (E. Mupondwa).

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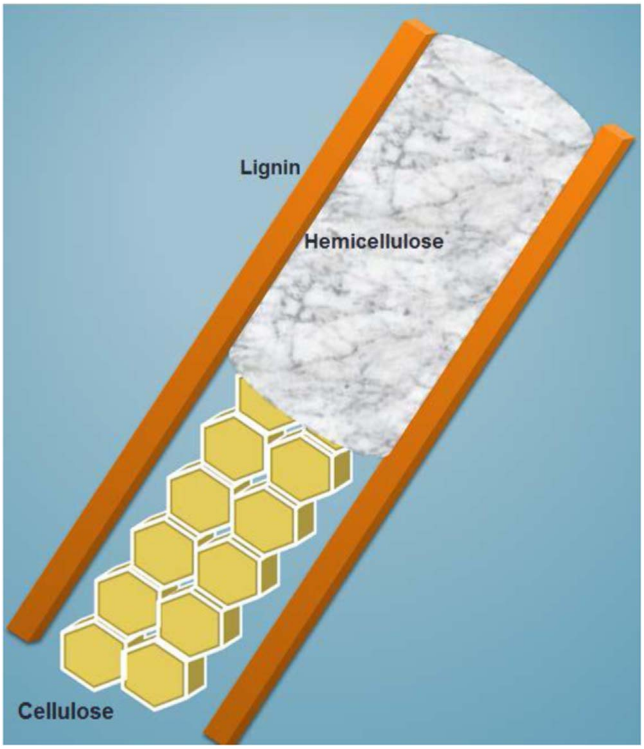

Fig. 1. Simplified internal structure of lignocellulosic biomass.

review of hydrolysis and fermentation as critical components required to advance Canada's cellulosic ethanol biorefinery concept to a commercial stage. Hence, Part I and Part II represent a unifying theme of critical processing steps in the bioconversion of lignocellulosic feedstocks to ethanol. This paper is part of ongoing joint endeavours to create a common platform for communicating knowledge and advancing this sector. Since the biochemical conversion pathway characterizes a majority of Canada's current industrial level commercialization initiatives for cellulosic ethanol and co-products [2], this paper only considers Canadian research initiatives related to the biochemical conversion pathway. It is however appreciated that the technology for the conversion of lignocellulosic feedstocks is founded on both the sugar platform (involving biochemical conversion processes emphasizing fermentation of sugars extracted from the biomass) and the synthesis gas (syngas) platform (which involves thermochemical conversion processes based on gasification of biomass and by-products to produce syngas comprising hydrogen and carbon monoxide) [8].

2. Canadian research into pretreatment technology

Pretreatment is the first key processing step. As a business operation, the challenge lies in dealing with heterogeneous sources of lignocellulosic biomass and then converting them into desired uniform components. The wide diversity in lignocellulosic biomass explains why researchers have come up with numerous methods of biomass pretreatment in an attempt to create optimal conditions tailored to each feedstock type [9–13]. Approximately 20% of the total cost of cellulosic ethanol production is attributable to the pretreatment step, with the enzyme production and enzymatic hydrolysis steps accounting for another 20% [14]. Pretreatment methods (Fig. 2) can be grouped into physical, physico-chemical, chemical, biological, and processes that combine these methods [10–13,15]. Physical methods in general involve biomass size reduction to reduce cellulose crystallinity or degree of polymerization and increase available surface area [11]. Physico-chemical pretreatment combines mechanical and chemical processes. The range of methods includes catalyzed steam explosion such as carbon dioxide (CO_2) and Sulfur dioxide (SO_2) explosion, ammonia fibre expansion (AFEX), liquid hot water, and ammonia recycle percolation (ARP), a variant to AFEX [12]. The literature often does not distinguish between ARP and AFEX technologies which, according to Kim and Lee [16], are differentiated by the fact that AFEX is accomplished with liquid ammonia while ARP is conducted using aqueous solution. Chemical pretreatment involves the use of diverse chemicals including acids, alkali, oxidizing agents, organic solvents, and ionic liquids [17–19]. Biological pretreatment involves the use of natural microorganisms (e.g., brown-, white-, and soft-rot fungi, and bacteria) possessing enzymes capable of deconstructing the cell wall of lignocellulosic biomass [20].

Canadian research initiatives in pretreatment and lignocellulosic bioconversion technologies involve collaborations spanning researchers from universities, Agriculture and Agri-Food Canada (AAFC), Environment Canada, Natural Resources Canada, National Research Council, and several industrial organizations like FPInnovations, Lignol technologies, and Iogen. These entities all have an interest in advancing the science and technology of biomass conversion. A number of these Canadian research initiatives have also involved collaboration with the US Department of Energy, in particular Oak Ridge National Laboratory. Research networks have included the Bioconversion Network and the Canadian Cellulosic Biorefinery Network. In the next sections, a description of Canadian research in pretreatment is provided.

2.1. Biomass size reduction

The overall objective of mechanical or physical pretreatment is to reduce cellulose crystallinity, resulting in an increase in available surface area and reduction in the degree of polymerization [11]. In this case, pre-processing steps such as biomass size reduction, e.g hammer mill grinding, is an important step. Size reduction is energy intensive but necessary for lignocellulosic biomass conversion processes in order to lower transportation costs associated with such low bulk density feedstocks [21]. The energy requirements of mechanical comminution of lignocellulosic materials depend on the final particle size and agricultural biomass characteristics [22].

There has been significant Canadian research on biomass size reduction, especially as it relates to challenges associated with characteristics of lignocellulosic biomass such as high moisture content, irregular shape and sizes, and low bulk density which make the biomass very difficult to handle, transport, store, and utilize in its original form in biochemical conversion without expensive material transformation systems. These factors have been analysed in a number of Canadian studies [23–29]. According to Mani et al. [24,26], raw cellulosic biomass possesses a low bulk density of 30 $kg\,m^3$ and moisture content of 10–70% (weight basis). Densification via pelletization increases specific biomass density to over 1000 $kg\,m^{-3}$ [24,30], thereby rendering itself more amenable for efficient and cost-effective handling and storage [25,28,31].

In recent Canadian research, Adapa et al. [23] analysed biomass comminution, including grinding performance of a chopper and hammer mill, and physical characteristics of non-treated and steam exploded barley, canola, oat and wheat straw. They reported that the bulk and particle density of four types of agricultural straw significantly increased with a decrease in hammer mill screen size, and the particle size of pre-treated straw was shown to be smaller than non-treated straw. Sokhansanj et al. [32] and Adapa et al. [33] found that pretreatment disintegrates biomass lignocellulosic structure, making it easier to grind due to a lower shear strength.

In general, studies on the economics of comminution are varied. A number of studies suggest that milling prior to or following other pretreatments like chemical result in several benefits, including: a) decrease in milling energy consumption; b) decrease in the cost of