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Temperature profiling to maximize energy yield with reduced water input in a lignocellulosic ethanol biorefinery

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HIGHLIGHTS

- Synergetic effects affecting PSSF for whole biomass conversion was resolved.
- Highest ever ethanol titer of 82.6 g/L for softwood biorefinery was achieved.
- Energy and water footprint was evaluated for the first time among related techniques.
- Total energy yield is 2410 MJ per ton dried wood conversion.
- Total water consumption is 3.65 tons per ton dried wood conversion.

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ABSTRACT

Softwood biomass is an attractive renewable feedstock for bioethanol production. The net energy yield of the related biorefinery processes has been limited, however, by its high lignin content, which is recalcitrant to hydrolysis and fermentation. New understanding of the causes of the inhibiting effects is critical to approach the optimal energy/water nexus in a biorefinery. This paper introduces a new prehydrolysis simultaneous saccharification and fermentation process to convert sulfite pretreated Monterey pine into bioethanol, resulting in an extremely high titer of 82.6 g/L or 10 vol%. The new process was carried out at a solid content of 25% by using a commercial enzyme and *Saccharomyces cerevisiae*. Sugars in the pretreatment spent liquor were concentrated and mixed into the fermentation broth for complete utilization of the sugars, but it was found that this liquor can also affect the ethanol titer during fermentation. Control experiments suggested that sugar-based pretreatment by-products were not the major contributors to the inhibition, while small molecular weight compounds played a major role in affecting the fermentability of the slurry under high temperature. Cell viability tests showed that reducing fermentation temperature from 35 °C to 28 °C can overcome the impacts of pretreatment by-products on cell growth and ethanol production. Without the need of detoxification, the resulting ethanol titer is approaching the theoretical yield and is currently among the highest in softwood conversion. The net energy yield of the new process was 2410 MJ per ton oven-dried wood, which is approximately 730–1690 MJ higher than that of the other biorefinery processes. The water input before reclamation was 3.65 tons per ton dried wood, which is 25.8–51.2% lower than most of the other processes.

1. Introduction

Global climate change and increased energy demand have boosted tremendous efforts in developing innovative technologies to extract biofuel and chemicals from renewable sources. Cellulosic ethanol has been regarded as an attractive alternative to first-generation biofuels and fossil fuels [1–3]. Woody biomass generated from the forest industry and municipal wastes have been considered the most promising feedstock for cellulosic ethanol production, which benefits the communities in the social, economic, and environmental perspectives [4,5]. Woody biomass, in comparison with other lignocellulosic biomass (*i.e.*, corn stover or rice straw), can be generated year-round and with specific physiochemical features such as high density, low moisture contents, and low inorganic contents, which benefit its logistics and transportation when serving as a feedstock for biofuel production [5,6].

The major economic barrier to a wider application of woody biomass in biorefineries is its well-known high recalcitrance to

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bioconversion. Woody biomass is a complex microstructure composed of cellulose, hemicelluloses, and lignin. This structure needs to be decomposed to enhance the accessibility of cellulase to cellulose before being converted into fermentable sugars and the related products [7]. The recent pretreatment techniques for woody biomass conversion, such as steam explosion [8], dilute acid [9], co-solvent pretreatment using tetrahydrofuran (THF) [10], γ-valerolactone (GVL) [11], and sulfite pretreatment to overcome recalcitrance of lignocellulose (SPORL) [12,13], have been successfully developed to achieve remarkable sugar yields ($>$ 90% conversion of glucan to glucose) at reasonable enzyme loadings (*i.e.*, 5–15 filter paper unit, or FPU, per g glucan in biomass). These innovative pretreatment techniques have different mechanisms, which aim to mitigate (*i.e.*, steam explosion, dilute acid), modify (*i.e.*, sulfite treatment), and/or decompose the structural lignin and hemicellulose (*i.e.*, co-organosolv) in the plant cell wall for different applications. For example, sulfite pretreatment can enhance the sugar yield of the overall processes. Organosolv process aims to recover the value-added aromatic monomers. Acid catalysts are typically applied to remove hemicelluloses and has been applied in all the stated pretreatment processes [14].

Following the pretreatment process, the simultaneous saccharification and fermentation (SSF) or prehydrolysis SSF (PSSF) processes have been applied to convert cellulose/hemicelluloses into fermentable sugars and to produce the fermentation products at the same time [15]. As increasing ethanol titer can significantly reduce the distillation energy, one major target of PSSF is to increase the solids loading of the substrate in the fermentation broth while reducing the impacts of growth-inhibiting factors on the fermentation microorganisms. Optimization of the PSSF process is complex as the operating temperatures differ significantly between enzymatic hydrolysis (50 °C) and fermentation (30–45 °C) [15,16]. The pretreatment spent liquor may contain significant amounts of pretreatment by-products, *i.e.*, 5-(Hydroxymethyl) furfural (HMF), furfural, and phenolic compounds, which can seriously affect the efficiency of either enzymatic hydrolysis, yeast fermentation, or both [17,18]. Washing or detoxification has been applied to improve the digestibility and fermentability of the substrate, but both of those approaches result in significant losses of sugars and lowering the final ethanol yield. Energy, water, and chemical inputs may be increased due to the toxic impacts of pretreatment by-products or accumulation of lignin in the fermentation broth.

Zhu et al. [19] recently introduced the “whole slurry” SSF process to directly apply sulfite pretreated Lodgepole pine chips with the pretreatment spent liquor to produce ethanol without detoxification. As sulfite pretreatment (SP) aimed to modify the surface of residual and dissolved lignin to satisfy enzymatic cellulose conversion without complete removal of lignin [14], ethanol titer was expected to be significantly improved by completely utilizing the hexoses for fermentation. However, the maximum ethanol titer was still at 52.8 g/L [20]. The solutions needed to overcome the possible impacts of the components in the pretreatment spent liquor have not yet been identified.

This study aims to clarify three essential knowledge gaps toward the economical application of the softwood-to-bioethanol biorefinery processes for maximizing the energy yield of the pretreated substrate. Complete utilization of woody biomass is an economically and environmentally attractive targets for process development, but the key mechanism to improve the ethanol titer to achieve similar levels as in other low-lignin-content biomass [21,22] is still unclear. The energy consumption platform of different operating conditions, especially the relationships among the SSF process, ethanol titer, and distillation energy has not been well-developed. The water-and-energy nexus for the most recent bioconversion processes for woody biomass conversion has never been reported in previous research works.

In this study, proper configurations of the SSF processes were investigated through changing the operating conditions to facilitate the performances of the commercial cellulase and yeast. The whole slurry substrate was prehydrolyzed under different operating conditions (*i.e.*, prehydrolysis times, solid contents, and concentrations of the mixed spent liquor); and then different fed-batch approaches were applied to maximize the rates of hydrolysis. Water-soluble contents in the pretreatment spent liquor were fractionated and purified to study the possible components inhibiting the metabolisms of the yeast, while the tolerance of yeast to the inhibitors was investigated by changing the fermentation temperatures. Numerical analysis using both experimental and literature data on energy and water footprints were conducted and compared with the other recent biorefinery techniques. The findings of this study provide an opportunity to complete utilization of various feedstocks, such as energy crops, agriculture residues, and woody biomass.

2. Materials and methods

2.1. Setup of the biorefinery platform

The conceptual diagram of the proposed biorefinery process and the related energy/water footprints of each unit process are shown in Fig. 1. The woody biomass is hammer-milled before installing into the pretreatment vessel. The wood chips are cooked under designed pretreatment conditions [13] and then transferred to the solid-liquid separator. The solid fraction of the pretreated wood is disc-milled together with a portion of the pretreatment spent liquid, and the remainder of the spent liquor is concentrated by using an evaporator. The mixture of the milled substrate and concentrated spent liquor is then discharged into a fermenter for PSSF and then the fermentation

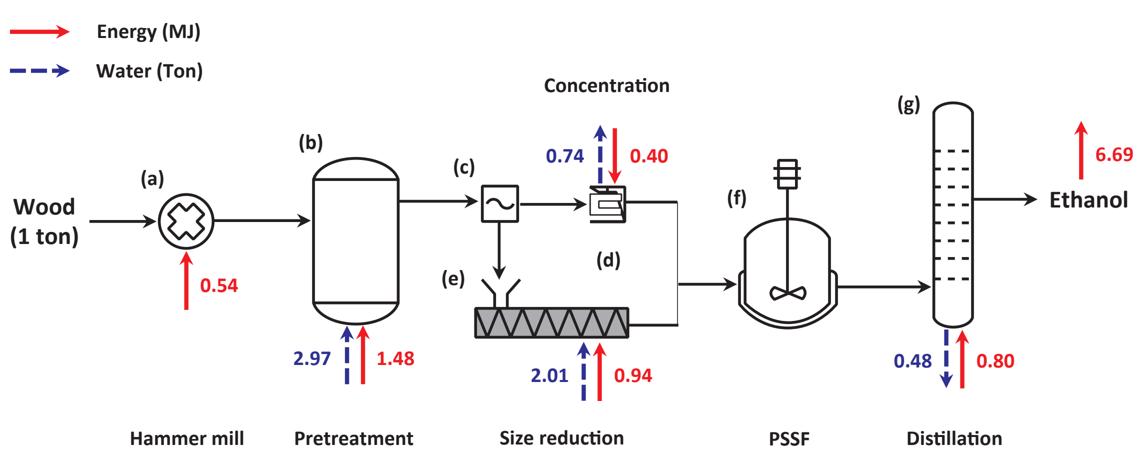

Fig. 1. Flow diagram of the water and energy balances of different unit processes in the proposed biorefinery for conversion of 1 ton softwood feedstock in ethanol.