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Comparative techno-economic analysis of steam explosion, dilute sulfuric acid, ammonia fiber explosion and biological pretreatments of corn stover

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HIGHLIGHTS

- Techno-economics of four different pretreatment processes were compared.
- Biological pretreatment required 2 fold more feedstock than sulfuric acid.
- Ammonia fiber explosion required 4 fold more external energy than sulfuric acid.
- Biological pretreatment required 5 fold less external energy than sulfuric acid.
- Sulfuric acid and steam explosion were cost competitive pretreatment methods.

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ABSTRACT

Pretreatment is required to destroy recalcitrant structure of lignocelluloses and then transform into fermentable sugars. This study assessed techno-economics of steam explosion, dilute sulfuric acid, ammonia fiber explosion and biological pretreatments, and identified bottlenecks and operational targets for process improvement. Techno-economic models of these pretreatment processes for a cellulosic biorefinery of 113.5 million liters butanol per year excluding fermentation and wastewater treatment sections were developed using a modelling software-SuperPro Designer. Experimental data of the selected pretreatment processes based on corn stover were gathered from recent publications, and used for this analysis. Estimated sugar production costs ($/kg) via steam explosion, dilute sulfuric acid, ammonia fiber explosion and biological methods were 0.43, 0.42, 0.65 and 1.41, respectively. The results suggest steam explosion and sulfuric acid pretreatment methods might be good alternatives at present state of technology and other pretreatment methods require research and development efforts to be competitive with these pretreatment methods.

1. Introduction

Butanol is a potential next generation liquid biofuel that can be produced from lignocellulosic biomass, including forest residues, agricultural residues and energy crops. Among these lignocelluloses, corn stover is a viable feedstock due to its immediate availability, high residue yield rate and ease of hydrolysis (DOE, 2011). Despite widespread effort in commercializing butanol production and availability of corn stover, lignin is a key problem in corn stover utilization as a feedstock. The lignin tightly binds cellulose and hemicellulose, and, at the same time cannot be used by the *Clostridium* species to produce butanol (Qureshi et al., 2008). Thus, lignocellulosic biomass requires pretreatment and enzymatic hydrolysis to transform the complex structure of cellulose and hemicellulose into simple sugars, which can be further transformed into biofuels.

Over the years, several pretreatment techniques are being investigated to disrupt the lignin matrix of a lignocellulosic biomass, which can be broadly divided into three categories: (1) physical; (2) chemical; and (3) biological (Jurgens et al., 2012; Kumar et al., 2009; Mood et al., 2013). Additionally, combinations of these pretreatment methods are also available. Generally, physical pretreatment reduces size of particle using mechanical energy and increases porosity of material without adding chemicals, such as

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acid or base (Cheng, 2010; Kumar et al., 2009). Some common examples of physical pretreatment methods are mechanical comminution, steam explosion, carbon dioxide explosion and hydrothermal pretreatment. In contrast to physical pretreatment, chemical pretreatment processes use chemical either acid or base for the pretreatment of lignocellulosic biomass at room temperature to elevated temperature for few minutes to several hours (Cheng, 2010). Sulfuric acid, hydrochloric acid, phosphoric acid, oxalic acid, tartaric acid, citric acid and acetic acid are used for pretreatment of lignocellulosic biomass. Additionally, some commonly used chemicals for alkaline pretreatment are sodium hydroxide, potassium hydroxide, lime, urea, ammonia, sodium carbonate, calcium hydroxide and methylamine. Apart from physical and chemical pretreatment methods, interests on biological pretreatment are also growing in recent years due to less pretreatment energy and simple reactor requirements as well as unnecessity of chemicals (Kumar et al., 2009; Wan and Li, 2010). Biological pretreatment uses bacteria, and brown, white and soft rot fungi to degrade lignocellulosic biomass (Wan and Li, 2010). Regardless of the method, the main goal of pretreatment is to destroy the structure of biomass and increase accessibility of enzyme by increasing porosity or surface area.

Among several physical, chemical and biological pretreatment methods, steam explosion, sulfuric acid, ammonia fiber explosion (AFEX) and biological with white rot fungi are commonly used pretreatment methods for lignocellulosic biomass (Jurgens et al., 2012; Kumar et al., 2009; Mood et al., 2013; Wan and Li, 2010). Overall comparison of some common pretreatment processes is available elsewhere (Jurgens et al., 2012; Kumar et al., 2009; Mood et al., 2013). All the above common pretreatment processes destroy structure of lignocellulosic biomass and partially convert cellulose and hemicelluloses into the fermentable sugars such as glucose, xylose, arabinose, galactose, and mannose. And then, a subsequent enzymatic hydrolysis process transforms remaining polymers into fermentable sugars. Depending upon the type of pretreatment and the process parameters, such as temperature and time, lignin is degraded into phenolics and the fermentable sugars mainly into hydroxymethylfurfural (HMF), furfural, acetic, levulinic, ferulic and glucuronic acids (Baral and Shah, 2014; Jurgens et al., 2012). The presence of these degradation products, commonly referred to as microbial inhibitors, results in low biofuel yield during fermentation. These inhibitory compounds either halt the fermentation process or slow down the reaction rates. Detailed discussions on microbial inhibitors formation from different pretreatment methods is found elsewhere (Baral and Shah, 2014).

While various potential pretreatment processes are available, they have different operating conditions and product yield rates. However, very few techno-economic studies, such as Baral and Shah, 2015; Eggeman and Elander, 2005; Kazi et al., 2010; Klein-Marcuschamer et al., 2011; Kumar and Murthy, 2011, are available for different pretreatment processes, such as sulfuric acid, hot water, ionic liquid, and ammonia fiber explosion. Apart from Baral and Shah, 2015, other studies did not consider impact of variation in process parameters, such as temperature and time, of a particular pretreatment. However, this study is also limited to ionic liquid pretreatment only. Recent review (Baral and Shah, 2014) found that the process parameters, specifically temperature and time, significantly impact unit sugar production and microbial inhibitors formations. Another recent techno-economic analysis of ionic liquid pretreatment process (Baral and Shah, 2015) found that pretreatment temperature and time not only significantly affect sugar production cost but also impact on quantity of process equipment requirement. A detail techno-economic analysis considering variations in process parameters is essential for sustainable butanol production. Additionally, all of the above mentioned techno-economic studies are based on ethanol fermentation only. However, for the same quantity of biofuel production, ethanol and butanol fermentation require different levels of fermentable sugars requiring different levels of process equipment, feedstocks and other input materials.

In this regard, choice of a single pretreatment process for commercial butanol production is difficult at this stage. A thorough techno-economic analysis of each of these common pretreatment methods is required to identify the most cost effective pretreatment method for butanol production through ABE fermentation from corn stover feedstock. Thus, four common pretreatment methods, including steam explosion, dilute sulfuric acid, ammonia fiber explosion and biological pretreatment, are considered for techno-economic analysis in this study based on total sugars yield, microbial inhibitors formation, current research interests and general comparison of different pretreatment process available from literatures (Baral and Shah, 2015; Jurgens et al., 2012; Mood et al., 2013). This study also provides possible optimization techniques for each pretreatment method.

2. Materials and methods

2.1. Overview of modelling process

Techno-economic process models for the selected pretreatment methods for a cellulosic biorefinery with a capacity of 113.5 million liters (i.e., 30 million gallons) butanol per year were developed using SuperPro Designer software v9.0009.2100 (Intelligen, 2014). Since the scope of the study was to compare different pretreatment technologies for fermentable sugars production, fermentation and wastewater treatment were excluded from the analysis. Annual capacity of the first generation cellulosic biorefineries is in the range of 75.7–133.5 million liters (20–30 million gallons) (Shah and Darr, 2016), thus, this biorefinery capacity of 113.5 million liters butanol per year was chosen as a model biorefinery. Fig. 1 depicts the overview of process model with some major process equipment. The process model considered all unit operations required to transform the corn stover into biofuels. These unit operations were grouped into four major discrete units, namely corn stover preparation, pretreatment, detoxification and enzymatic hydrolysis.

2.2. Distinct modelling unit and data sources

Modelling assumptions were based on the data gathered from recently published studies on the selected pretreatment methods. After completion of the process flow diagram for all discrete units, necessary expressions and data required to accomplish material and energy balance, equipment size and purchasing price, maintenance and installation expenses for different operations were assigned to the models. Material and energy balance analysis results were further used to determine the equipment size and quantity, utilities and raw materials quantity, as well as other direct and indirect costs associated with capital and operating costs. The governing expressions to estimate purchasing price of equipment are available elsewhere (Intelligen, 2014; NREL, 2011, 2002).

2.2.1. Corn stover preparation unit

Based on the literature data on different ABE fermentation and recovery methods (Ezeji et al., 2003; Mariano et al., 2012; Qureshi et al., 2010), average butanol production from fermentable sugars is about 24 wt%. Thus, about 386,924 metric ton (t) annual total sugars are required to produce 113.5 million liters butanol. In this study, corn stover was used as a model feedstock. Depending on type of pretreatment methods and their corn stover to sugar