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Optimization of switchgrass and extruder parameters for enzymatic hydrolysis using response surface methodology

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ABSTRACT

Switchgrass is an important biomass that can be hydrolyzed to yield fermentable sugars through pretreatment, which is the primary and expensive step in conversion of biomass to bio-ethanol. Most of the pretreatment operates in batch mode, which is energy intensive, requires high capital, results in decomposition of hemicellulose, and formation of inhibitors. Considering these shortcomings, a novel biomass pretreatment method using a high shear bioreactor could be a viable continuous one. The current study was undertaken to determine the effect of biomass parameters such as moisture content (10, 20, 30, 40, and 50% wb) and particle size (2, 4, 6, 8, and 10 mm) over a range of barrel temperature and screw speed (45–225 °C and 20–200 rpm). Statistical analyses revealed that among the independent variables considered temperature, screw speed, and moisture content had significant effect on sugar recoveries. Proposed quadratic model to predict glucose, xylose, and combined sugar recoveries from switchgrass had a high F and R^2 values indicating that the model has the ability to represent the relationship among the independent variables studied. The optimum pretreatment condition of barrel temperature 176 °C, screw speed 155 rpm, moisture content 20% wb, and particle size 8 mm resulted in maximum glucose, xylose, and combined sugar recoveries of 41.4, 62.2 and 47.4%, respectively. The optimum pretreated switchgrass had 50% higher surface area than that of the control.

1. Introduction

In the last three decades researchers have been focusing on alternate fuel resources to meet the ever-increasing energy demand and to avoid the dependence on crude oil. Wood et al. (2003) estimated the peak-oil situation within the next two decades; studies have predicted to have a steady decline in crude oil production in coming decades. According to National Research Council (2000), the goal of biobased industry is to provide at least 10% of liquid fuels by the year 2020, and to provide 50% of liquid fuels by the year 2050. Due to supply limitation and matured corn ethanol technology, there is a need to develop alternate resources for biofuel and products. Lignocellulosic biomass provides a low cost material for the production of fuel and chemicals, which also offers economical and environmental benefits. The US Department of Energy identified switchgrass as a model herbaceous energy crop (McLaughlin, 1993) for intensive study. Switchgrass has many advantages such as high productivity, suitability for marginal land quality, low water and nutritional requirements, environmental benefits, and flexibility for multipurpose uses (McLaughlin et al., 1999).

Biomass generally contains 60–70% of cellulose and hemicellulose, which are not readily available for enzymatic hydrolysis; hence, the pretreatment step becomes inevitable. The purpose of pretreatment is to open up the biomass structure, to reduce the cellulose crystallinity, to increase the surface area and porosity. Physical, chemical and biological principles are being used in biomass pretreatment. Each pretreatment method has its own advantages and disadvantages. For example, dilute acid pretreatment achieves more than 90% sugar yield but it requires corrosion resistant material, neutralization prior to biological steps. The elevated temperature in hydrothermal pretreatment leads to sugar degradation. Alkalis are too expensive, difficult to recover and reuse them during biomass pretreatment. The cost of ammonia and its recovery are important factors in AFEX pretreatment. A few studies have shown that extrusion can address most of the above listed issues (Dale et al., 1999; de Vrije et al., 2002; Muthukumarappan and Julson, 2007; Karunanithy et al., 2008; Lee et al., 2009; Karunanithy and Muthukumarappan, 2009, in press, 2010a,b).

Extruder has the ability to provide high shear, rapid heat transfer, effective and rapid mixing, and adaptability to many different processes – all in a continuous process. The foremost advantage of the extrusion over other pretreatment methods such as hydrothermal (190–210 °C), compressed liquid/hot water

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(180–230 °C), steam pretreatment (190–210 °C, and steam explosion (193–230 °C), is the use of low barrel temperature. It prevents the carbohydrates degradation and oxidation of lignin, which otherwise would result in potential fermentation inhibitors (de Vrije et al., 2002). Extrusion does not produce any effluent thereby no effluent disposal cost, no solid loss, no safety issues is involved; moreover, scale up and adaption at industry level is also easy are few more advantages to mention. Karunanithy et al. (2008) results have shown that screw speed and barrel temperature had a significant influence on sugar recovery for switchgrass. Recently, the authors have reported that glucose, xylose, and combined sugar recovery of 45.3, 43.9 and 45.4% for the switchgrass pretreated at a screw speed of 50 rpm and barrel temperature of 150 °C with a moisture content of 15% and a particle size of 4 mm using an extruder screw compression ratio of 3:1 (Karunanithy and Muthukumarappan, 2010b). These extrusion pretreatments yielded encouraging results and however, the extrusion factors such as barrel temperature, screw speed, moisture content, and feedstock size were not optimized and it is the focus of this study.

Biomass particle size is an important factor, which affects the diffusion kinetics and effectiveness of pretreatment (Kim and Lee, 2002; Chundawat et al., 2007), sugar yield (Chang et al., 2001; Hu et al., 2008; Yang et al., 2008), lignin removal (Hu et al., 2008), hydrolysis rate, rheological properties (Chundawat et al., 2007; Desari and Bersin, 2007), and acetic acid formation (Guo et al., 2008). Smaller particle sizes are more readily hydrolyzed by cellulase due to higher surface to mass ratio. According to an US patent 5677154 (Draanen and Mello, 1997), the ethanol production process needs a particle size ranging from 1 to 6 mm of ground biomass. The power requirement for size reduction of biomass depends on the final particle size and characteristics of biomass (Cadoche and Lopez, 1989). Mani et al. (2004) recorded the specific energy consumption of switchgrass (27.6 kWh/t) for 3.2 mm screen size and Jannasch et al. (2005) reported 55:9 kW h/t for the hammer mill screen sizes of 5.6 and 2.8 mm for switchgrass. Recently, Bitra et al. (2009) reported optimum total specific energy of 114.4 MJ/Mg for switchgrass when size reduced at 2000 rpm for mass flow rate of 2.5 kg/min and 3.2 mm screen with 90°-hammers (6.4 mm-thick); total specific energy per unit size reduction was 14.9 MJ/Mg mm. Hence, the biomass size has a significant impact on size reduction energy requirement as well as economics. Thus, it was decided to investigate the influence of switchgrass particle size on sugar recovery in extrusion pretreatment.

Optimization of pretreatment conditions is one of the most important stages in the development of an efficient and economic pretreatment method. The traditional 'one-factor-at-a-time approach' is time consuming and moreover the interactions between independent variables are not considered. Response surface methodology (RSM) is an effective optimization tool wherein many factors and their interactions affect the response can be identified with fewer experimental trials. RSM mainly includes central composite design, Box-Behnken design, one-factor design, D-optimal design, used-defined design, and historical data design. RSM has been widely used in various fields ranging from food process operations including extrusion (Jorge et al., 2006; Altan et al., 2008), new product development, biotechnology- media composition, to bioprocessing such as enzymatic hydrolysis and fermentation. Recently, RSM has been successfully applied to biomass pretreatment by many researchers (Neureiter et al., 2002; Rahman et al., 2007; Lu et al., 2007; Canettieri et al., 2007; Kim and Mazza, 2008; Xin and Saka, 2008). In the present study, response surface methodology with central composite rotatable design (CCRD) adopted to optimize the extrusion parameters for maximum sugar recovery from switchgrass. Earlier extrusion pretreatment studies conducted by the authors have shown encouraging results, but extrusion parameters were not optimized. Therefore, the present study was undertaken: (1) to evaluate and optimize the effect of extruder parameters such as barrel temperature and screw speed; biomass parameters such as moisture content and particle size for maximum sugar recovery and (2) to develop a statistical model to predict glucose, xylose, and combined sugar recovery from switchgrass.

2. Materials and methods

2.1. Experimental design

A CCRD with four variables was used to study the response pattern and to determine the optimum combination of temperature, screw speed, moisture content and particle size for maximizing the sugar recovery from switchgrass. The CCRD combines the vertices of the hypercube whose coordinates are given by a 2^n factorial design with star points. The star points provide the estimation of curvature of the nonlinear response surface. The experimental design was developed using Design Expert 7.1.6 (Statease, Minneapolis, MN), which resulted in 30 runs, in addition 6 more center points were added to allow for the estimation of the pure error sum of squares. The 36 experiments (16 factorial, 8 star, and 12 center points) were randomized to maximize the effects of unexplained variability in the observed responses due to extraneous factors. Independent variable levels were selected based on one factor at a time experiments and previous studies. The independent variables were coded according to the following equation:

$$x_i = \frac{(X_i - X_0)}{\Delta X_i} \qquad (1)$$

where x_i and X_i are the dimensionless and actual values of the independent variable, respectively. X_0 is the actual value of the independent variable at the center point, and ΔX_i is the step change of X_i corresponding to a unit variation of the dimensionless value. The variables optimized included: barrel temperature (45–225 °C), screw speed (20–200 rpm), moisture content (10–50%), and particle size (2–10 mm) each at five levels: −2, −1, 0, 1, and 2 based on earlier studies and one-factor-at-a-time, as shown in Table 1.

2.2. Biomass preparation and characterization

Switchgrass is a native perennial grass designated as an energy crop by the U.S. Department of Energy for its high biomass production capability from which renewable sources of fuel and electricity can be generated (Missaoui et al., 2005; McLaughlin and Kszos, 2005). Switchgrass obtained from a local farm was ground using a hammer mill (Speedy King, Winona Attrition Mill Co., MN) using 2, 4, 6, 8, and 10 mm sieves to understand the influence of particle size on sugar recovery. Moisture content of the biomass samples was determined as described by Sluiter et al. (2008a). The moisture content of ground biomass was adjusted to 10, 20, 30, 40, and 50% (wb) by adding water and equilibrated overnight to determine the effect of moisture content on sugar recovery. The compositional analysis of switchgrass was carried out as outlined by Sluiter et al. (2008b,c). The raw switchgrass and optimum pretreated switchgrass samples were subjected to particle size distribution and surface area measurement using LS 13 320 particle size analyzer and SA 3100 surface area and pore size analyzer (Beckman Coulter, CA, USA), respectively.

2.3. Extrusion pretreatment

Extrusion was performed using a single screw extruder (Brabender Plasti-corder Extruder Model PL 2000, Hackensack, NJ), which had a barrel length to screw diameter ratio (l/d) of 20:1. In order to have a smooth biomass (plug) flow into the die section, the