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Optimization of hydrothermal pretreatment of wheat straw for production of bioethanol at low water consumption without addition of chemicals

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ABSTRACT

In the IBUS process (Integrated Biomass Utilization System) lignocellulosic biomass is converted into ethanol at high dry matter content without addition of chemicals and with a strong focus on energy efficiency. This study describes optimization of continuous hydrothermal pretreatment of wheat straw at pilot scale (up to 100 kg h^{-1}) where six different pretreatment conditions have been investigated; all pretreatment conditions have been evaluated with regards to recovery of sugars after pretreatment (both C5 and C6) and convertibility of the cellulosic part of the fibers into ethanol.

The experiments show that the optimum pretreatment parameters are 195 °C for 6–12 min. At these conditions, a total of app. 70% of the hemicellulose is recovered, 93–94% of the cellulose is recovered in the fibers and app. 89% of the cellulose in the fibers can be converted into ethanol by commercial cellulase mixtures – increasing to 92% when adding a commercial xylanase.

1. Introduction

The process of converting lignocellulose into ethanol consists of four main parts: pretreatment, enzymatic hydrolysis, fermentation (the last two can be combined in simultaneous saccharification and fermentation – SSF) and separation [1].

A central part of the lignocellulose-to-ethanol process is the physical/chemical pretreatment. This process is energy demanding but also necessary to enable enzymatic breakdown of the polysaccharides in the lignocellulosic material. Furthermore, the pretreatment affects the costs of most of the other unit operations in the process [2]. Worldwide, there are many different approaches to pretreatment e.g. hydrothermal, wet-oxidation, steam explosion and AFEX [1–4]. Common for all types of pretreatment is that they should minimize loss of sugars, consume a minimum of energy, and increase enzymatic digestibility. This is done by removing or rearranging the lignin structure, partial or full removal of hemicellulose from the fibers, and possibly altering the cellulose structure [1,4]. Pretreatment is, however, always a compromise between sugar recovery (primarily hemicellulose) and substrate accessibility [5].

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In Denmark, the IBUS process (Integrated Biomass Utilization System) has been developed. The focus in the development of this process has been on high dry matter content in all process steps from pretreatment via prehydrolysis and SSF to distillation, integration into a power plant, and no use of added chemicals – the straw is converted into ethanol by the use of only hot water and steam, enzymes and yeast [6–8].

One of the advantages of hydrothermal pretreatment is the fact that hemicellulose is mainly solubilized as oligomers not monomers, which reduces the risk of degradation of the soluble hemicellulose fraction [9]. However, xylan oligomers from pretreatment with a degree of polymerization of 15 or more can be adsorbed onto cellulose and thereby hinder hydrolysis of cellulose by cellulases [10]. Other advantages include avoided addition of chemicals resulting in reduced consumption of chemicals for pH adjustment, and reduced risk of corrosion of the pretreatment reactor.

It is well known that parts of the carbohydrates in lignocellulosic biomass are degraded and/or dissolved during pretreatment and thereby removed from the fibers. Especially hemicellulose is degraded into soluble oligomers and free sugars. A part of the hemicellulose is also degraded into acetic acid, formic acid, 2-furfural and other degradation products. During pretreatment, small amounts of the cellulose are also solubilized as oligomers and free glucose and some are degraded into e.g. 5-hydroxy-methylfurfural [11,12].

Initial experiments with two-step pretreatment in the IBUS pilot plant showed that an optimum pretreatment without addition of chemicals was achieved using a large water flow; experiments showed that at 190 °C for 12 min it was possible to reach a hemicellulose recovery of app. 80%. However conversion of the yield of glucose from the fibers after enzymatic hydrolysis was only app. 55% [7]. After the system was rebuilt as a three-step pretreatment system, Thomsen et al. [8] showed that total hemicellulose yields above 80% could be achieved using a relatively large total water-to-biomass ratio of 10:1, where water usage includes steam condensation in the reactor, measured as liquid flow from the reactor.

However, a commercial process using these large flows of water is not economically feasible due to the energy required for heating. Therefore, the current work has focused on minimizing water consumption while maintaining high hemicellulose recoveries by reducing the residence time of the process water which contains the dissolved and degraded hemicelluloses in the reactor. To increase energy efficiency, the three-step pretreatment described in previous publications has been reduced to a two-step pretreatment.

Several researchers have reported that most of the xylan should be removed during pretreatment for the fibers to be fully degradable with commercial cellulase mixtures [10,13]. When water consumption is lowered, hemicellulose solubilization is decreased leaving a larger part of the hemicellulose in the fiber fraction after pretreatment. This is confirmed by Jacobsen and Wyman, who have shown that there might be a trend towards larger yields of solubilized hemicellulose at low solid concentrations in experiments with up to 10% solids [14], indicating that a conflict might exist between removal of hemicellulose and low water consumption in hydrothermal pretreatment. When some of the hemicellulose is left in the fibers used for enzymatic hydrolysis and fermentation, it is very important that the enzyme mixture used in the process not only contains cellulase activity but also hemicellulase activity as supplementation of commercial cellulase mixtures with xylanases has been demonstrated to improve hydrolysis yields [15,16].

This paper describes optimization of hydrothermal pretreatment of wheat straw in pilot scale. This is done by evaluation of different pretreatment parameters with respect to sugar recoveries (both cellulose and hemicellulose) and enzymatic conversion of the fiber fraction after pretreatment. The aim of the pretreatment is to reach a cellulose recovery in the fiber fraction of more than 90% and total recovery of more than 95%; furthermore, it is important for the process economics also to reach a hemicellulose recovery of at least 70%. High sugar recoveries, however, is not the sole success criterion; it is equally important that the fibers can be degraded into cellulose by the cellulase enzymes, and subsequently converted into ethanol by *Saccharomyces cerevisiae*. Therefore, the analytical results from the pretreatment experiments are supplemented with SSF experiments evaluating the potential conversion of the cellulosic part of the fibers into ethanol.

2. Materials and methods

2.1. Raw materials

Wheat straw (*Triticum aestivum* L.) was grown and harvested after a drying period in Denmark during summer 2006. The straw was cut into 1–6 cm pieces on the field and stored in containers at ambient temperature. The chemical composition of the untreated wheat straw can be seen in Table 1.

2.2. Pretreatment

Six experiments have been performed with continuous pretreatment of wheat straw at 100 kg h^{-1} scale under different pretreatment conditions. An overview of the experimental conditions can be seen in Table 2.

For these experiments, a pretreatment pilot plant with a maximum capacity of up to 100 kg h^{-1} was used (Fig. 1). The straw is fed continuously with a flow rate of 50 kg h^{-1} to a soaking reactor where it is soaked at 80 °C for 5–10 min. After soaking, the excess water is removed from the straw, and the straw is fed to the pretreatment reactor by a particle pump. In the pretreatment reactor, the straw is moved through the

Table 1 – Chemical composition of untreated wheat straw used in this experiment.

	Composition straw [g per 100 g DM]
Cellulose	35.0
Xylan	19.6
Arabinan	2.7
Total hemicellulose	22.3
Klason lignin	15.6
Ash	6.5
Residual	20.9