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A Technical and Economic Analysis of Acid-Catalyzed Steam Explosion and Dilute Sulfuric Acid Pretreatments Using Wheat Straw or Aspen Wood Chips

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ABSTRACT

Lignocellulosic biomass is one of the most plentiful and potentially cheapest feedstocks for ethanol production. The cellulose component can be broken down into glucose by enzymes and then converted to ethanol by yeast. However, hydrolysis of cellulose to glucose is difficult, and some form of pretreatment is necessary to increase the susceptibility of cellulose to enzymatic attack. An analysis has been completed of two pretreatment options, dilute sulfuric acid hydrolysis and sulfur dioxide impregnated steam explosion, for two feedstocks, wheat straw and aspen wood chips. Detailed process flow sheets and material and energy balances were used to generate equipment cost information. A technical and economic analysis compared the two feedstocks for each of the two pretreatments. For the same pretreatment, sugars produced from aspen wood hydrolysis were cheaper because of the higher carbohydrate content of aspen, whereas dilute acid pretreatment is favored over acid-catalyzed steam explosion.

Index Entries: Pretreatment; sulfuric acid; steam explosion; aspen wood; wheat straw.

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INTRODUCTION

Of the many biomass feedstocks available for ethanol production, one of the most abundant and cheapest is cellulosic biomass. Cellulose, a polymer of glucose, can be broken down into glucose by enzymes and then converted to ethanol by yeast. However, hydrolysis of cellulose in raw cellulosic biomass is difficult. This has been attributed to the crystalline nature of cellulose and the lignin–hemicellulose sheath that surrounds the cellulose. Thus, some form of pretreatment is necessary to disrupt the lignin–hemicellulose sheath and increase the susceptibility of the cellulose to enzymatic attack. Pretreatment can also hydrolyze hemicellulose to its individual sugar components. In the case of hardwoods and wheat straw, the hemicellulose is composed primarily of the five-carbon sugar xylose, a sugar that can also be converted to ethanol. The conversion of xylose to ethanol improves the overall economics of the cellulosic biomass-to-ethanol process (*1*).

Several processes can be used for pretreatment of biomass, including autohydrolysis steam explosion, steam explosion with an acid catalyst, dilute sulfuric acid hydrolysis, and the "organosolv" process. The dilute-acid process uses low concentrations of sulfuric acid at relatively low temperatures (160°C for 10 min) to achieve nearly complete conversion of the hemicellulose xylans to xylose (2,3). However, prior to this step, particle size is reduced to about 1.0 mm, which requires significant amounts of energy. The two steam-explosion processes use high-pressure steam and rapid depressurization to both reduce the size of the biomass particle and partially hydrolyze the hemicellulose fraction. Both require operationally complex steam-explosion guns. With autohydrolysis steam explosion, the yield of xylose is low (30–50%) (*4*). However, the yield can be improved by using a catalyst, such as SO_2 (*5*), which is the basis for the acid-catalyzed steam-explosion process. The organosolv process uses an organic solvent to dissolve the lignin and hemicellulose fractions from the cellulose. The lignin is then precipitated from solution, leaving the xylose in the liquid stream. This process is complicated and expensive, but produces a high-quality lignin stream that could be converted to high-value products.

Because of the low xylose yields and their negative impact on ethanol-production economics, a detailed analysis of autohydrolysis steam explosion was not carried out in this study. Furthermore, because of the expense and complexity of the organosolv process, and because currently there are no large markets for high-quality lignin, this process was also not considered further. The two remaining pretreatment options, steam explosion with an acid catalyst and dilute sulfuric acid pretreatment, were evaluated with two different feedstocks: wheat straw and aspen wood chips. These materials represent two of the most abundant categories of cellulosic biomass, herbaceous, and wood energy crops.

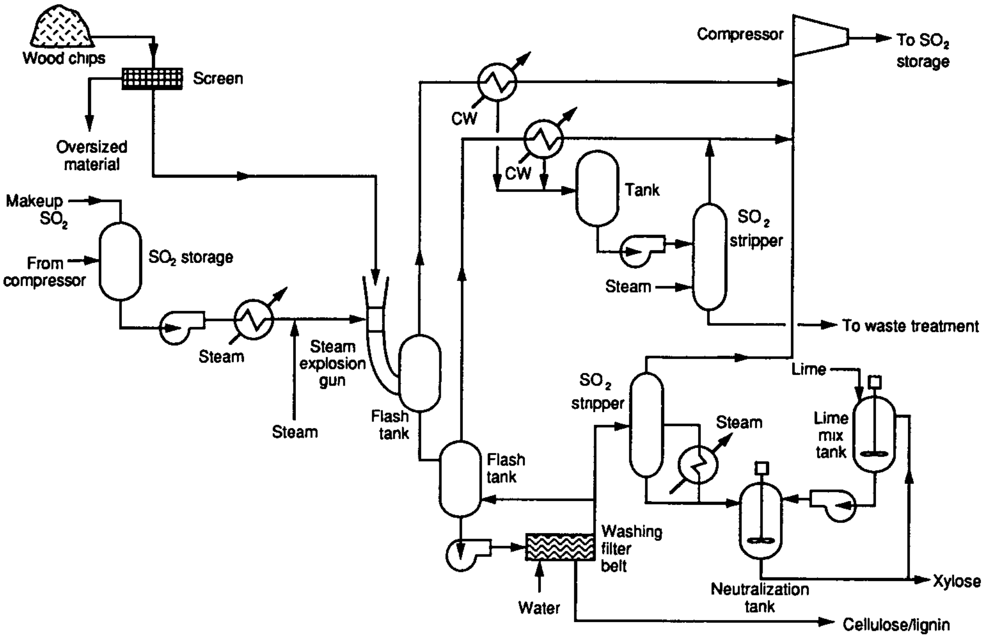

Fig. 1. Flow sheet for acid-catalyzed steam explosion of aspen wood chips.

METHODOLOGY

Each of the four feedstock/pretreatment combinations is analyzed on the basis of total sugar (glucose as unconverted cellulose and xylose) produced, which could then be converted to ethanol. The economic information is summarized on the basis of total sugars, assuming 90% of the cellulose leaving the pretreatment process can be converted to glucose. However, the analysis does not consider conversion of cellulose to glucose or subsequent conversion of sugars to ethanol, but considers only the pretreatment processes. Assuming a different number for conversion of cellulose to glucose will change the absolute cost of the processes, but not the relative comparison between processes.

Process Description

Acid-Catalyzed Steam Explosion with Aspen Wood

A flow sheet of the acid-catalyzed steam-explosion process for aspen wood chips is shown in Fig. 1. The design for the steam-explosion system is adopted from a design by Stone and Webster Engineering Corp. (Boston, MA) (*6*), and the design of the SO_2 recovery section is based on the work of several groups (*5,7–9*). Finally, the design of the lime slurry preparation system section is adopted from a study by Badger Engineers, Inc. (Cambridge, MA) (*10*).

Aspen wood chips are stored in an open pile and delivered by front-end loaders to a screen that removes oversized material, which is sent to a rechipper. The screened material is loaded into a stainless steel steam-explosion gun. Each stainless steel gun is a 3.5-ft (1.07-m) diameter pipe, 14.5 ft (4.45 m) long, designed for 650 psig (4480 kPa), and sealed on each end by quick-opening, full-port plug or ball valves. Once the chips are sealed inside the gun, steam and vaporized sulfur dioxide are added and the chips are cooked for 2 min. Sulfur dioxide added to the gun is taken as a liquid from a carbon steel storage vessel, pumped to 600 psig (4236 kPa), vaporized, and mixed with the steam feed stream. After cooking, the material is blown down into a 316 stainless steel flash vessel operating at 60 psig (515 kPa). The steam-exploded wood then flows to a second 316 stainless steel atmospheric flash tank, where final cooling takes place.

Flash vapor from each of the flash tanks, containing water vapor and SO_2 is sent to partial condensers (304 stainless steel tubes/carbon steel shell). The condensate, still containing a small amount of SO_2, is collected in a condensate receiver, then pumped to a 316 stainless steel open-steam stripping column. The SO_2 removed from the top of the column is combined with the uncondensed SO_2 from the partial condensers, compressed to a liquid, and recycled back to the liquid-SO_2 storage tank. The water from the bottom of the column is sent to waste treatment. This extensive SO_2 recovery system is employed to reduce significantly any discharge to the environment.

Exploded wood in the bottom of the atmospheric flash tank is combined with a fraction of the liquid steam from the downstream washing filter belt to obtain a pumpable slurry. The slurry is pumped to the washing filter belt to remove the SO_2, xylose, and other soluble material from the solid cellulose/lignin slurry. The cellulose/lignin material can then be sent to an ethanol production unit. The liquid stream from the filter belt is sent to a stripping column and heated by a steam reboiler. The SO_2 removed by this column is compressed and condensed, and then recycled back to the liquid-SO_2 storage tank.

The acidified liquid (from conversion of SO_2 to H_2SO_4) from the bottom of the stripping column is sent to a 304 stainless steel vessel for neutralization by a lime slurry, producing a neutral xylose-containing stream, which can be sent to an ethanol production unit. A small amount of this stream is taken and mixed with lime in a carbon-steel vessel to produce the lime slurry.

Acid-Catalyzed Steam Explosion with Wheat Straw

The wheat-straw plant accepts bales that are managed by a crane system and stored in a pile. After the bales are broken apart in a bale shredder, the material is screened and sent to a separator bin that removes dirt, dust, and grains from the fibers. The fibers are then sent to the steam-explosion guns. The remainder of the process is shown in Fig. 1.

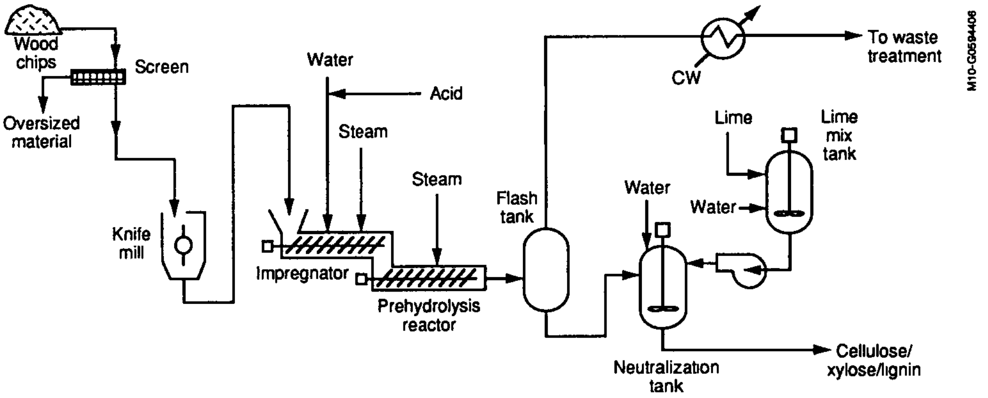

Fig. 2. Flow sheet for dilute sulfuric acid pretreatment of aspen wood chips.

Dilute Sulfuric Acid Pretreatment with Aspen Wood

A flow sheet for the dilute sulfuric acid process with aspen wood chips is shown in Fig. 2. The pretreatment part of this process is based on the design of Torget et al. (*3*). Wood chips are screened to remove the oversize material, which is sent to a rechipper. The acceptable material is sent to a knife mill. Here the chips are milled to reduce the particle size to approx 1.0 mm. Following this, the milled wood enters the steam-heated impregnator, where water and acid are added. This reactor ensures that acid thoroughly diffuses into the wood particles. The acidified slurry is then fed to the prehydrolysis reactor, which is steam heated to 160°C. Both the impregnation and prehydrolysis reactors are constructed of Carpenter-20 alloy for corrosion resistance. After prehydrolysis, the wood slurry is cooled by flashing to atmospheric pressure and subsequently conveyed to the neutralization tank. The vapors from the flash tank are condensed and sent to waste treatment. The sulfuric acid in the wood slurry is neutralized in the neutralization tank by a lime slurry, and additional water is added to make a pumpable wood slurry.

Dilute Sulfuric Acid Pretreatment with Wheat Straw

The design of the feed-handling/storage and preparation areas is the same as that described for acid-catalyzed steam explosion with wheat straw. After screening, the straw is sent to a knife mill, and the remainder of the process is as described above for dilute sulfuric acid pretreatment with aspen wood.

Plant Capacity and Feed Composition

Process flow diagrams for each feedstock were developed based on a delivered flow rate of 160,000 lb/h (72,700 kg/h) of dry feedstock to the pretreatment section of the plant. Aspen wood enters the plant at 50% moisture, and wheat straw enters the plant at 12% moisture (*11*). The chemical composition of each feedstock is given in Table 1.

Table 1
Chemical Composition of Aspen Wood and Wheat Straw

	Aspen Wood (*10*) (%)	Wheat Straw (*2*) (%)
Cellulose	46.2	40.8
Xylan	24.0	27.0
Lignin	24.0	18.4
Ash	0.2	11.2
Other	5.6	2.6

Design Basis

Acid-Catalyzed Steam Explosion

The chips are held in the steam-explosion guns for 2.0 min at 240°C (*7*). An additional 0.5 min is needed for loading and preheating and another 0.5 min for blowdown and cleaning. The total cycle time per gun is 3 min, although this has not been demonstrated at the large scale assumed for this plant.

The SO_2 used for steam explosion is 1.6 kg/100 kg of dry wood, and the high-pressure steam use is 0.85 kg/kg dry wood (*7*). For wheat straw, the SO_2 use is the same, and the high-pressure steam use is calculated to be approx 0.45 kg/kg dry straw because of the lower water content. The xylan conversion from a SO_2-catalyzed steam explosion is 75% conversion to xylose, 15% conversion to furfural, 5% unchanged, and 5% degraded to solid products (*7*). Cellulose is assumed unchanged. The SO_2 is converted as follows: 9.2% to sulfuric acid, 9.7% to lignin sulfonic acids, 74.2% unconverted and available for recycle, and 6.5% retained with the lignin (*7*). The washing filter belt system is a five-stage washing operation, similar to a paper pulp stock washer, in which 99% of the solubles is recovered (*6*).

Dilute Sulfuric Acid Pretreatment

The milling step requires 85 hp/ton/h (64 kW/ton) of electrical power for wood and 8.5 hp/ton/h (6.4 kW/ton) for wheat straw (*12*). The impregnator operates at 100°C with a 10-min residence time (*3*). The prehydrolysis reactor operates at 160°C for a 10-min residence time and with an acid concentration of 1 wt% after addition of steam and water (*3*). Xylan is assumed converted as follows: 80% to xylose, 13% to furfural, and 7% unconverted (*2*). Also, during prehydrolysis, 4% of the cellulose is converted to glucose (*13*); the rest is assumed unchanged.

Capital Cost Estimate and Economic Analysis

Heat and material balances were developed and used to specify equipment sizes. Purchased equipment cost is estimated using information from Chemcost (*14*), Icarus (*15*), Guthrie (*16*), Stone and Webster Engineering Corp. (*6*), and Badger Engineers, Inc. (*10*). The total capital invested (fixed plus working capital excluding service utilities) is estimated as 4.0 times the purchased equipment cost (*17*), and the annual capital charge (depreciation, taxes, insurance, and rate of return) is total capital invested times a fixed charge rate (FCR) of 0.13 (*18*) or 0.20, typical for these types of plants (*19*). Chemical costs are taken from the *Chemical Marketing Reporter* (*20*). Utility cost for process water, cooling water, and steam are estimated from Peters and Timmerhaus (*17*), and electricity is assumed to cost $.04/kWh. Labor is estimated as 0.001 man-h/y per dollar of total capital, invested at an average rate of $22.00/h. Maintenance is 4% of total capital invested, and overhead is 60% of labor plus maintenance. Byproduct credit is taken for lignin sent to the boiler and is estimated as the heating value of lignin divided by the total heating value of the feedstock times the feedstock cost.

RESULTS

An economic summary of acid-catalyzed steam explosion for both aspen wood and wheat straw is given in Table 2 for a feedstock cost of $42/dry ton and a FCR of 0.13 (*21*). The selling price of sugar (glucose and xylose) as a function of feedstock cost and FCR is shown in Fig. 3 for both feedstocks. It is somewhat cheaper to produce sugars from aspen wood for the same feedstock cost. This is because of the greater amount of sugars contained in aspen wood (70% cellulose and xylans) than contained in wheat straw (68% cellulose and xylans). Also, aspen wood has a larger byproduct credit because of its larger lignin content.

An economic summary of dilute sulfuric acid pretreatment for both aspen wood and wheat straw is given in Table 3 for a feedstock cost of $42/dry ton and a FCR of 0.13. The selling price of sugar as a function of feedstock cost and FCR is shown in Fig. 4. In this case, aspen wood also results in a lower selling price of sugar for the same feedstock cost for the same reasons given above.

DISCUSSION

The choice of feedstocks for sugar production will probably be governed by plant location. Plants situated in food-crop agricultural areas will probably use agricultural residues, such as wheat straw. In areas where agricultural residues are not readily available, it will probably be

Table 2
Economic Summary of the Steam Explosion Process

Grassroots plant, 1st quarter, 1990
Plant Capacity: 160,000 dry lb/h
Total Capital Investment:
Aspen wood plant $74.2 MM
Wheat straw plant $74.8 MM

	Aspen	Wheat
	¢/lb sugars	
Raw Materials		
Feed	3.28	3.44
SO_2	0.07	0.08
Lime	0.01	0.01
Utilities		
Process Water	0.07	0.08
Cooling Water	0.09	0.10
Steam-60 psig	0.28	0.26
Steam-600 psig	0.30	0.16
Electricity	0.03	0.05
Labor	0.20	0.21
Maintenance	0.36	0.38
Overhead	0.34	0.35
By-Product Credits		
Lignin	1.06	0.96
Capital Charges	1.18	1.24
TOTALS	5.15	5.40

Feedstock Cost: $42/dry ton
FCR: 0.13

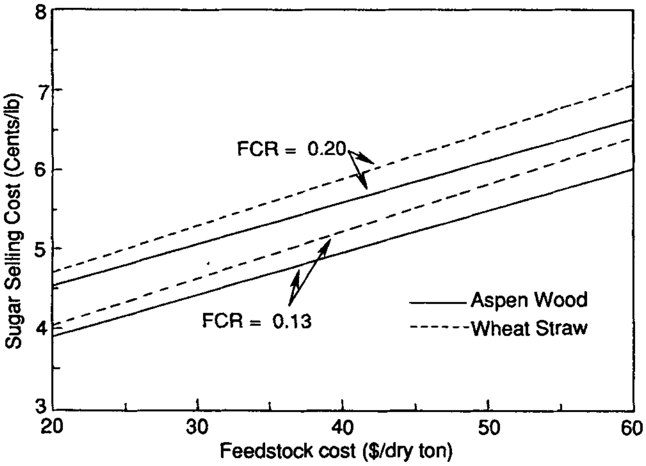

Fig. 3. Selling price of sugar as a function of feedstock cost and fixed charge rate (FCR) for acid-catalyzed steam explosion.

Table 3
Economic Summary of the Dilute Acid Process

Grassroots plant, 1st quarter, 1990
Plant Capacity: 160,000 dry lb/h
Total Capital Investment:
  Aspen wood plant $28.7 MM
  Wheat straw plant $31.2 MM

	Aspen	Wheat
	¢/lb sugars	
Raw Materials		
Feed	3.09	3.21
Acid	0.07	0.08
Lime	0.05	0.06
Utilities		
Process Water	0.03	0.04
Cooling Water	0.02	0.02
Steam-60 psig	0.21	0.15
Steam-600 psig	0.0	0.0
Electricity	0.25	0.08
Labor	0.08	0.09
Maintenance	0.13	0.15
Overhead	0.13	0.14
By-Product Credits		
Lignin	1.00	0.90
Capital Charges	0.43	0.48
TOTALS	3.49	3.54

Feedstock Cost: $42/dry ton
FCR: 0.13

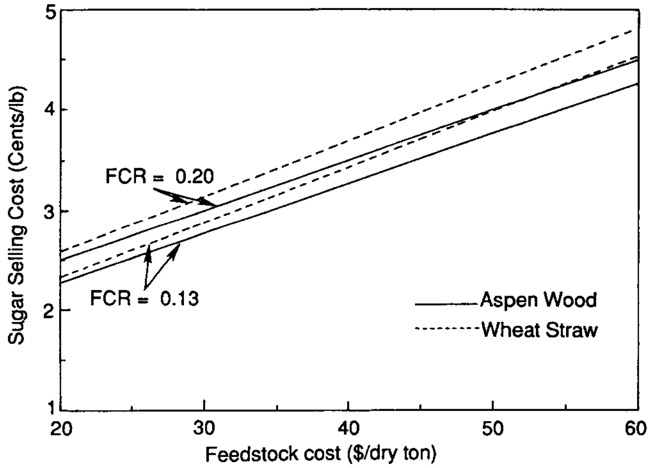

Fig. 4. Selling price of sugar as a function of feedstock cost and fixed charge rate (FCR) for dilute sulfuric acid pretreatment.

more advantageous to use wood energy crops, as represented by aspen. There is not an overwhelming advantage to either feedstock, particularly for dilute sulfuric acid, if they are obtained at the same price. However, feedstock cost does have a significant effect on the selling price of sugar. A decrease in feedstock cost from $60 to $20/dry ton decreases the selling price by approx 35% for steam explosion and 45% for dilute sulfuric acid. Similarly, the FCR also has an effect. A drop from 0.20 to 0.13 lowers the selling price of sugar by approx $0.006/lb for steam explosion and $0.0025/lb for dilute sulfuric acid.

The data in Tables 2 and 3 indicate that a dilute sulfuric acid pretreatment is approx 30% cheaper than acid-catalyzed steam explosion using the process configuration and yields assumed in this study. The higher cost for steam explosion is primarily a result of the higher capital cost associated with SO_2 recovery. These results are based on our current understanding of each of these processes. Future improvements and changes to the process configuration could alter these results.

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