

Upgrading of Postconsumer Absorbent Hygiene Products for Bioethanol Production

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Supporting Information

ABSTRACT: Postconsumer absorbent hygiene products (AHPs), one of the most intractable municipal solid waste (MSW) streams, are generally subjected to landfills, causing urban waste management difficulties and environmental pollution. This study investigated the viability of recycling the cellulose component from postconsumer AHPs, demonstrated the fermentability of obtained cellulosic sugars for bioethanol production, and evaluated the technical feasibility of the process through scale-up. A set of integrated unit operations was developed to effectively upcycle the recovered AHPs into bioproducts. The processes were further optimized and scaled up 50-fold. Up to 83.5% glucose and 46.0% xylose yields were achieved during enzymatic saccharification. The sugars were readily fermentable through separate hydrolysis and fermentation (SHF) as well as simultaneous saccharification and fermentation (SSF) with maximum 89.9% ethanol theoretical yield.

KEYWORDS: Municipal solid waste recycling, Biological conversion, Scale up



INTRODUCTION

As population grows and daily product consumption increases worldwide, the recycling and reuse of municipal solid waste (MSW) is becoming increasingly important. Disposable absorbent hygiene products (AHPs) contribute significantly to the lives of people around the world by providing the convenience of single-use AHPs such as disposable baby diapers, incontinence pads, feminine care products, wipes, and so forth. However, as one of the most intractable components of MSW, postconsumer AHPs have also led to urban waste management issues in some countries. Postconsumer AHPs are traditionally collected together with the unsorted MSW and disposed by landfill or incineration, as only limited recycling processes are being implemented in certain regions of Europe.¹ With economic growth, more countries have increased consumption of AHPs, but their recycling is rarely reported. The production and recovery mechanisms are generally influenced by economy, culture, and social traditions of each country.²

Early stage research and application of disposable AHPs originated in the United States and Europe. Disposable diapers have a market share of 96% in the United States, similar to Europe and Japan.³ It was reported that approximately 3.6 million tons of disposable diapers were generated in the United States in 2013.⁴ In municipal cities worldwide, where a high level of separate waste collection is required, postconsumer AHPs such as diapers constitute one of the main difficulties in increasing recycling levels.⁵ This is due to the complicated

compositions of AHPs, including cellulose pulp, super absorbent polymer (SAP), polypropylene, polystyrene, tape, elastic, and adhesive materials and the fact that some of them are not naturally biodegradable. Traditional incineration or landfill disposal is not an ecological practice in the long run. On one hand, a lot of research is being done in AHP production and design to improve environmental performance.⁶ On the other hand, recycling and upgrading the postconsumer AHPs is becoming equally important. With most of the used AHPs ending up in landfill, a variety of collecting and tipping fees are derived and charged to households, nurseries, waste management companies, etc., and the potential economic benefit of recycling AHPs has been neglected.

Several recent studies reported that the composition of later generation AHPs is compatible with biological treatments such as composting for waste reduction.^{2,5,7} Due to high cellulose content, they could be recycled,⁷ and the cellulose fraction could be reclaimed regardless of manufacturer and then be upgraded into high-value products such as chemicals or fuels. Arena et al. reported an integrated syngas production process using the cellulose fraction of AHPs to replace fossil fuels.¹ It was also reported that used disposable diapers can be degraded by certain fungi producing a high protein content food product free from pathogenic organisms.¹ To date, the feasibility of

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using AHP-derived cellulose for biofuel production through a biological pathway has not been reported. It is unknown how the absorbent materials could affect convertibility or whether it is possible to reuse the cellulose fraction to substitute plant-derived feedstocks in biorefineries.

For the first time, to our knowledge, we have conducted benchmarking studies to assess the technical feasibility and identify the key unit operations for converting cellulose component from postconsumer AHPs into biofuel (e.g., bioethanol). The recycling processes to recover valuable raw materials from postconsumer AHPs have been implemented and commercialized at FATER S.p.A, a P&G and Angelini joint venture, and other institutes.⁸ This work aims at understanding the characteristics of postconsumer AHPs and evaluation of their conversion into fermentable sugars and biofuels. Different pretreatment routes, enzymatic hydrolysis, SAP separation, and fermentation processes were tested and optimized at milliliter to liter scales.

EXPERIMENTAL SECTION

Deconstruction Routes and Mechanical Separation. As shown in Figure 1, six different deconstruction routes have been

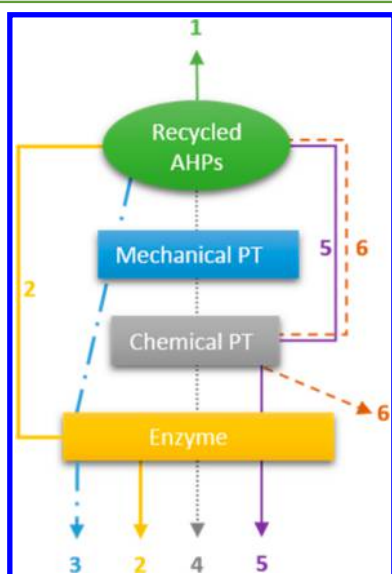


Figure 1. Six different deconstruction routes. Refer to experimental section. Route 1: used as received. Route 2: used directly for enzymatic saccharification (ES). Route 3: mechanical pretreatment (PT) + ES. Route 4: mechanical PT + chemical PT + ES. Route 5: chemical PT + ES. Route 6: used directly for chemical PT.

developed. No treatment was applied in route 1. Route 2 is direct enzymatic saccharification (ES). Mechanical PT is employed in route 3 and route 4 to increase the material surface area. Thermochemical PT is employed in routes 4, 5, and 6. For mechanical PT, a Thomas Wiley Mill (Model 4, 0.2 cm sieve, Thomas Scientific, Swedesboro, NJ, USA) was used to reduce particle size. Milled materials were transferred to a multilayer vibratory sieve shaker (Retch AS 200, Newtown, PA, USA) for separation. The throwing motion can settle particles of different sizes into corresponding sieve levels vertically with sieve sizes of 0.18 mm, 0.8 mm, 1.0 mm, and 2.0 mm. The feasibility of mechanical removal of the SAP was also tested to improve downstream processes and reduce the influence of SAP in aqueous solutions. Cellulose-rich materials after SAP separation were collected from the top and bottom layers of the vibratory sieves and used in the process optimization and scale-up studies. The middle two layers were collected as the SAP-rich fraction.

Thermochemical Pretreatment. Dilute acid pretreatment (1 wt % sulfuric acid based on dry material, 120 °C, 15 min) and hydrothermal pretreatment (DI water, 120 °C, 15 min) were carried out in Incoloy tube reactors (25 mL, Alloy Metal and Tubes, Houston, TX, USA), with a fluidized sand bath (Omega FSB-4, Stamford, CT, USA) for rapid heat transfer. Working volume was 10 mL with a 10 wt % biomass loading. Typically, dry material (1 g) was loaded into the tube reactor after thorough mixing with sulfuric acid or DI water and soaked overnight before heating in the sand bath. After pretreatment, tube reactors were immediately immersed in an ice bath to quench the reaction.

Shake Flask Enzymatic Saccharification. Enzymatic saccharification was conducted as the last step in routes 2, 3, 4, and 5 (Figure 1). Experiments were carried out in glass Erlenmeyer flasks (50 mL) with a 20 mL working volume. Cellulase (Cellic CTec2 with protein concentration: 190 mg/mL) and hemicellulase (Cellic HTec2 with protein concentration: 174 mg protein/mL) were provided by Novozymes, Inc. (Davis, CA, USA). In the initial base condition, enzymes were loaded at a fixed ratio (9 mg protein/g cellulose, CTec2/HTec2 = 9:1). Recycled AHPs and raw cellulose fluff were conditioned by citrate buffer (50 mM) with sodium azide (1 wt %). Two solid loadings (5 and 10 wt %) were compared. Experiments were carried out in duplicates at 50 °C for 72 h with an agitation rate of 250 rpm in MaxQ 8000 Orbital Shakers (Model 443, Thermo Fisher Scientific, Marietta, OH, USA).

Shake Flask Fermentation. The hydrolysates obtained from shake flask saccharification were further carried out for ethanol fermentation using *Saccharomyces cerevisiae* ATCC 201389. The first stage seed was prepared by transferring *S. cerevisiae* glycerol stock (1 mL) into a Yeast Extract–Peptone–Dextrose (YPD) medium (49 mL: 20 g/L glucose, 10 g/L yeast extract, 20 g/L peptone, Sigma-Aldrich, St. Louis, MO, USA). First stage seed was incubated at 30 °C for 12 h at 250 rpm in the MaxQ 8000 Orbital Shakers. Second stage seed was prepared by transferring seed one cells (10 mL) into fresh YPD media (190 mL) and incubating for another 8 h. Fermentation experiments were then performed in Erlenmeyer flasks (500 mL) by adding hydrolysate (72 g), yeast extract (1 g), peptone (2 g), and DI water (20 g) and 5 v/v% cell inoculum and then incubating for 72 h. YPD medium (40 g/L synthetic glucose, 10 g/L yeast extract, and 20 g/L peptone) was used as the control to compare with hydrolysate fermentation.

Process Optimization and Scale-Up. The optimization of enzymatic saccharification was conducted using mock material after SAP removal. In addition to the 1× enzyme loading (9 mg protein/g cellulose), 2×, 4×, and 8× enzyme loadings were tested and compared. Different solid loadings were also attempted (5, 10, and 15 wt %). The optimized conditions were chosen for scale-up at 1 L working volume in a glass reactor (2 L, IKA LR-2.ST, IKA-Works, Wilmington, NC, USA) equipped with an anchor impeller and flow breaker for efficient mixing.

Separate hydrolysis and fermentation (SHF) was then conducted in fermenters (Bioengineering Inc., Wald, Switzerland) at 1 L working scale. Hydrolysate slurries (720 g, without solid/liquid separation) were used as the fermentation carbon source. The hydrolysate was obtained from the saccharification scale-up experiment with 10 wt % solid loading and 36 mg protein/g cellulose as the enzyme loading. The same ratios of yeast extract and peptone supplement as well as 5 v/v% cell inoculum were applied as in the small scale fermentation. Incubation was controlled at 37 °C with 250 rpm agitation for 72 h. Sterile airflow (100 NL/h, normal liters per hour) was supplied continually to improve mixing and provide oxygen. The consumption of monomeric sugars such as glucose and xylose was tracked in relation to ethanol production.

Furthermore, the feasibility of ethanol production through simultaneous saccharification and fermentation (SSF) was also evaluated. First, the cellulose-rich material went through enzymatic saccharification at 50 °C in the fermenter at 15 wt % solid loading with 72 mg protein/g cellulose enzyme loading. During the first 8 h of saccharification, maximum air flow was supplied to the fermenter to boost the stirring efficiency at a rate of 300 NL/h. The fermenter was

Table 1. Major Chemical Composition (wt%) of the Materials

material	moisture	glucan	xylan	SAP (+ lignin)	ash	other ^a
raw cellulose fluff	7.8 ± 0.2	68.6 ± 2.6	9.5 ± 0.5	1.5 ± 1.0	7.8 ± 0.1	12.6
recycled AHPs	14.9 ± 0.2	26.1 ± 4.2	3.8 ± 0.2	41.2 ± 0.1	1.4 ± 0.1	12.6
cellulose-rich	7.6 ± 0.1	59.8 ± 2.6	9.0 ± 0.7	19.1 ± 2.5	4.8 ± 2.7	0.0
SAP-rich	15.1 ± 0.2	15.7 ± 3.0	2.0 ± 0.7	50.5 ± 5.2	4.4 ± 1.2	12.3

^aAll values are based on the weight of materials.

reset to 37 °C after 8 h of saccharification. When the temperature reached 37 °C, the medium was inoculated with *S. cerevisiae* and supplied with sterile yeast extract and peptone in a similar concentration with a shake flask study for SSF incubation. Raw cellulose fluff was used in parallel as the control substrate.

Analytical Methods. The compositional analysis was carried out following the two-step sulfuric acid hydrolysis of the Laboratory Analytical Procedures (LAPs) developed by the National Renewable Energy Laboratory.⁹ Collected materials were first dried at 42 °C until their moisture content was below 10 wt %. Dried samples were hydrolyzed with sulfuric acid (72 wt %) for 1 h in a 30 °C water bath. The hydrolysates were then diluted to 4 wt % sulfuric acid with DI water, and a second hydrolysis was carried out in an autoclave (121 °C for 1 h). The mixture was filtered through a porcelain crucible, and the dried residue was used to determine Klason lignin and ash content. The weight of SAP was jointly measured and reported with the Klason lignin. The acid insoluble materials were burned in a muffle furnace (575 ± 25 °C for 4 h) to measure the ash content. Acid soluble lignin content was determined by a UV–vis spectrophotometer (Shimadzu UV-2401, Kyoto, Japan) at 205 nm wavelength. Quantification of carbohydrate and ethanol was conducted using a High Performance Liquid Chromatograph (Thermo Fisher Scientific, Ultimate 3000, Waltham, MA, USA), which was equipped with an Aminex HPX-87H column (Bio-Rad, 300 × 7.8 mm, Hercules, CA, USA) and a refractive index detector. The mobile phase was 5 mM sulfuric acid with a flow rate at 0.6 mL/min and column oven temperature at 65 °C. The RI detector was heated at 50 °C. The samples were filtered using a 0.45 μm centrifuge filter and then diluted with 5 mM sulfuric acid for injection.

RESULTS AND DISCUSSION

Postconsumer AHPs were collected from households in Italy by FATER S.p.A. Collected AHPs were sanitized in an autoclave (121 °C for 1 h) and dried at FATER's facility. Such treatment ensures the biosafety in the process of handling, transporting, and storing. The dried material was then sent to a separator, which separates the cellulosic fraction (containing SAP) and the plastic fraction. The resulting cellulose fraction mainly contains cellulose fluff, polyacrylate, paper, and organic matter (urine, excrement, etc.). Polyacrylate is one of the SAPs commonly used in new AHP production and can retain at least 200 times its mass in water. To facilitate the experimental discussion, the postconsumer AHPs after sanitizing, drying, and plastic removal are named "recycled AHPs." The original cellulose fluff used to manufacture the AHPs is referred to as the control and named "raw cellulose fluff" in this study.

Chemical Composition of Materials. As shown in Table 1, raw cellulose fluff contains about 78.1% carbohydrate (glucan + xylan), 7.8% ash, and 1.5% lignin on a dry weight basis. The unquantified 12.6% mass may include oils, waxes, pectins, proteins, mineral matter, organic acids, etc.¹⁰ On average in this study, recycled AHP samples have about 29.9% carbohydrate and 41.2% combined acid soluble lignin, Klason lignin, and SAP. Though lignin and SAP are reported jointly in this study, SAP is the main component while lignin only accounts for a very small percentage. The data reflect a market average, as the recycled AHPs come from different commercial brands in the

collection process. Each manufacturer may have their own formulation with certain cellulose, SAP, plastics, and polymer ratios, but the variations in AHPs' composition arise from the proportion of the constituents rather than differences in the materials used.¹¹ Therefore, the carbohydrate fraction in recycled AHPs should have similar quality. After SAP separation, the carbohydrate content was greatly increased in the cellulose-rich material (68.8%), with SAP content reduced to half (41.2% to 19.1%). The SAP-rich fraction is enriched with 50% SAP, and the carbohydrate content is reduced to 17.7%.

Evaluation of the Deconstruction Routes. To evaluate the different deconstruction routes as shown in Figure 1, a mock material (mixture of raw cellulose fluff and SAP at 7:3 ratio) was prepared and immersed in water followed by autoclaving under the same conditions as the actual recycled AHPs. The results for mock material are summarized in the Supporting Information. It has been shown that knife milling is effective in size reduction of the raw cellulose fluff, mock material, and recycled AHPs. Milled samples had a noticeable volume decrease due to a change in density. By comparing the conversions in routes 3 and 4 that include a milling step with route 2 (direct enzymatic saccharification), it was found that the sugar yields after milling did not improve. The milling step is indeed helpful in the material handling process in small-scale experiments and compositional analysis, but it does not appear necessary to increase sugar yields at larger scales. After autoclave treatment and drying at FATER's facility, the fluffy material particle size was in the inch range, and the material can be easily accessed by enzymes.

In addition, chemical pretreatment is not required prior to enzymatic saccharification (route 4, 5, and 6). Both dilute acid and hydrothermal pretreatment failed to improve the sugar yields of this substrate under the tested conditions. Route 6 (thermochemical PT only) had the lowest sugar yields. Generally, pretreatment of lignocellulosic biomass results in structural changes of lignin and cellulose as well as solubilization of hemicellulose, which in turn contributes to the reduction of biomass recalcitrance.¹² The feedstock cell wall needs to be disrupted to some extent in order to improve substrate accessibility to the enzymes.¹³ However, it was found that such treatment is unnecessary for the recycled AHPs. FATER's autoclave and drying process as described above might have already acted as a mild pretreatment step, which helped to break down the cellulosic matrix. The data listed in the Supporting Information demonstrate that direct enzymatic saccharification yields the highest titer of sugars.

Table 2 shows the sugar conversion data for recycled AHPs with comparison to raw cellulose fluff, as the control material. Totals of 89.1% glucan and 40.1% xylan of raw cellulose fluff were converted to glucose and xylose at a moderate enzyme loading (9 mg protein/g cellulose). Recycled AHPs had relatively lower glucose yield (68.0%) due to the presence of SAP and other noncellulosic components. SAP or other

Table 2. Sugar Conversions and Titters after 120 h Direct Enzymatic Saccharification

samples	glucose (g/L)	xylose (g/L)	glucose yield (%)	xylose yield (%)
raw cellulose fluff, 5% ^a	36.6 ± 0.8	3.5 ± 0.1	89.1 ± 1.9	40.1 ± 1.4
recycled AHPs, 5% ^a	9.4 ± 1.2	1.1 ± 0.2	68.0 ± 9.0	41.5 ± 7.6
recycled AHPs, 10% ^a	19.1 ± 1.4	2.1 ± 0.2	68.9 ± 5.1	38.9 ± 4.0

^aSample solid loading percentage is by weight, wt%

polymers may have hindered mass transfer, thus affecting the accessibility of cellulosic substrate to the cellulase (Cellic CTec2). The xylose yields for raw cellulose fluff and recycled AHPs were similar in the range of 40–42%, indicating a limited inhibitory effect of xylan conversion. Moreover, while the recycled AHP solid loading was increased from 5 wt % to 10 wt %, the titters of glucose and xylose in the hydrolysate doubled with similar sugar yields (68.9% glucose, 38.9% xylose at 10 wt % solid loading vs 68.0% glucose, 41.5% xylose at 5 wt % solid loading). This indicates a possibility to further increase the solid loading to achieve hydrolysates with concentrated sugars.

Mechanical Removal of SAP. SAP's strong water absorption capacity poses a challenge for cellulose recovery and conversion since cellulose is well-mixed with SAP during AHP manufacturing. In a simple water absorption test, deionized (DI) water was mixed with SAP (white granules). The SAP-water mixture exhibited as a semisolid gel even when 200 times the mass of water/SAP was added. The presence of SAP limits the cellulose loading in the saccharification experiment and also reduces the mobility of the substrate in downstream processes such as fermentation and product recovery. It is reported that chemical methods can eliminate the effects of SAP. For example calcium chloride (CaCl₂) has been reported to be helpful in a new recycling system to reverse the water absorbance of SAP.¹⁴ However, an overdose of salts may also cause problems in the following saccharification and fermentation processes.¹⁵ Salts at high concentration may have a solvent effect on proteins-breaking hydrogen bonds and denaturing enzymes or dissolving the cell membrane of microorganism.

In this study, mechanical removal of SAP from recycled AHPs has been shown to be efficient. Physical separation of SAP does not affect the quality of cellulose; thus no downstream inhibition is observed. Up to 77 wt % of SAP was successfully removed by an integrated process using a knife mill and vibrating sieves as shown in Figure 2. In a 10 kg test, a

3.8 kg mass was collected as the “cellulose-rich” material, and the remaining 6.2 kg was separated as “SAP-rich.” After SAP removal, the material quality was significantly improved. Table 1 shows the chemical composition data of the cellulose-rich material and SAP-rich material. Total carbohydrate content of cellulose-rich material largely increased to 68.8% (59.8% glucan and 9.0% xylan). The total SAP/lignin fraction decreased to 19.1%, with 4.8% ash and 7.6% moisture. The Supporting Information shows the material morphologies of recycled AHPs in four different layers of the vibrating sieves, where the SAP-rich fraction was captured in the middle two sieves (0.18–1 mm) and cellulose-rich material was collected from the top (>1 mm) and bottom (<0.18 mm) sieves. From there, the recovered cellulose-rich material is used in the subsequent process optimization and scale-up studies.

Saccharification Process Optimization and Scale-Up.

On the basis of the sugar conversion results using mock material (Supporting Information), experiments of actual recycled AHPs were carried out under two selected conditions with different cellulose and enzyme loadings as shown in Table 3. It was found that higher cellulose loading resulted in higher

Table 3. Comparison of Sugar Conversions and Titters of Recycled AHPs in Flask and Scale-up Saccharification

conditions	glucose (g/L)	xylose (g/L)	glucose yield (%)	xylose yield (%)
condition 1 ^a at 20 mL	45.6 ± 1.1	4.5 ± 0.0	67.8 ± 1.6	38.1 ± 0.4
condition 2 ^b at 20 mL	75.4 ± 1.6	7.5 ± 0.2	74.8 ± 1.6	42.4 ± 1.0
condition 1 at 1 L scale	56.1	5.4	83.5	46.0
condition 2 at 1 L scale	83.2	7.8	82.4	44.2

^a10 wt % solid, 36 mg protein/g cellulose. ^b15 wt % solid, 72 mg protein/g cellulose.

sugar titters in the hydrolysate. Higher enzyme dosage increased both sugar yields and reaction rates. In condition 1 (10 wt % solid loading, 36 mg enzyme protein/g cellulose), 67.8% glucose and 38.1% xylose yields were achieved after 48 h of saccharification, with 45.6 g/L glucose and 4.5 g/L xylose in the hydrolysate. Furthermore, with increased solid loading and enzyme dosage in condition 2 (15 wt % solid loading, 72 mg protein/g cellulose), glucose and xylose yields increased by 7% and 4% separately with more concentrated sugars obtained in the hydrolysate (75.4 g/L glucose and 7.5 g/L xylose). It was also observed that condition 2 hydrolysate was more liquefied

**Figure 2.** Mechanical removal of SAP from AHPs.

during the transfer of material with less solid residue and lower viscosity.

The two selected conditions were successfully scaled up 50-fold (from 20 mL to 1 L) in IKA glass reactor vessels. Effective mixing to enable good mass transfer is essential in the scale-up process. With the assistance of an anchor impeller and a flow breaker in the IKA vessels, sugar conversions were further increased. A maximum 83.5% glucose and 46.0% xylose yields were achieved under condition 1 scale-up. The sugar yields under condition 2 scale-up are similar, but more concentrated hydrolysate was produced with higher sugar titers (glucose, 83.2 g/L; xylose, 7.8 g/L).

Microbial Fermentation and Scale-Up. The carbon source used for separate hydrolysis and fermentation (SHF) at shake flask scale (100 mL) was from the condition 1 hydrolysate, prepared without solid/liquid separation. The sugars derived from cellulose-rich material are readily fermentable to ethanol by *Saccharomyces cerevisiae* ATCC 201389. As shown in Figure 3, similar sugar and ethanol

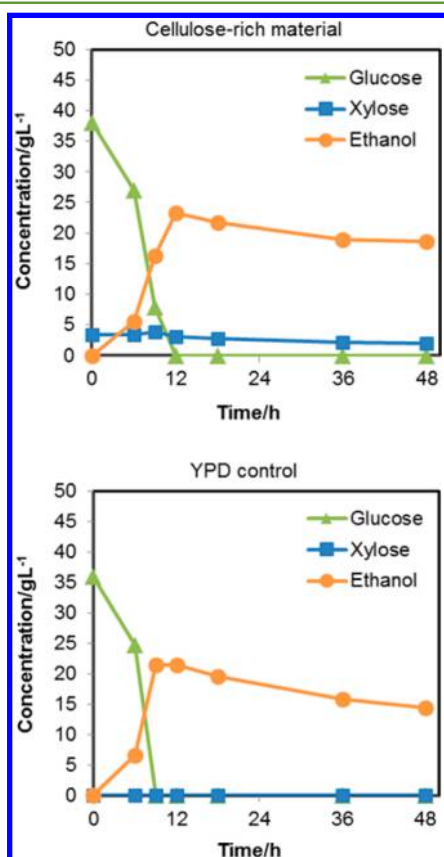


Figure 3. Ethanol fermentation (SHF) in shake flasks (100 mL) using the hydrolysate produced from condition 1 at 1 L scale (Table 3).

variation trends were observed between cellulose-rich material fermentation and control fermentation (using YPD media as a substrate: Yeast extract–Peptone–Dextrose). As expected, glucose was the preferred sugar and was consumed within 12 h. The xylose metabolism was fairly slow compared to glucose. Ethanol production peaked at 12 h with a titer of 23.2 g/L in hydrolysate fermentation and 21.5 g/L in YPD fermentation. Similar ethanol titers indicate no or very limited inhibition by the hydrolysate to yeast metabolism. The small difference may be due to sampling and ethanol evaporation from shake flasks.

In addition, common inhibitors from lignocellulosic feedstock hydrolysate such as furfural (FF), hydroxymethylfurfural (HMF), acetic acid, formic acid, etc. were not detected.

The above SHF experiments have shown that the hydrolysate is nontoxic to *S. cerevisiae* and is readily fermentable without solid/liquid separation. In order to further evaluate the sugars derived from cellulose-rich material, the more concentrated hydrolysate of condition 2 was subjected to an SHF test (100 mL) and then was scaled up in a bioengineering fermenter (1 L). As shown in Figure 4, a similar ethanol titer was obtained in

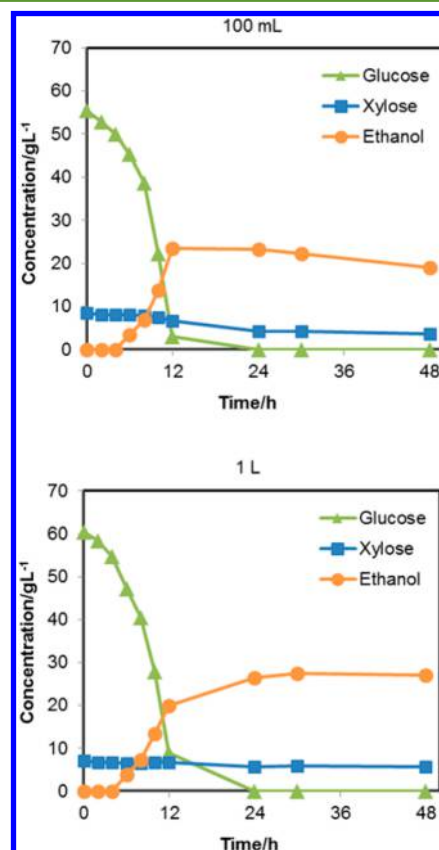


Figure 4. Ethanol fermentation (SHF) in shake flasks (100 mL) and fermenters (1L) using the hydrolysate produced from condition 2 at 1 L scale (Table 3).

shake flask studies (23.4 g/L vs 23.2 g/L using condition 1 hydrolysate). A maximum ethanol titer (27.5 g/L) was obtained in the 1 L SHF, which corresponds to 89.9% of the theoretical glucose to ethanol conversion yield. It is worth noting that the maximum ethanol titer in the fermenter trial is higher than that of the shake flask (27.5 g/L vs 23.4 g/L). This can be attributed to the better control of ethanol evaporation in the fermenter than the semiclosed flask. The yeast performed better when the temperature was controlled at a constant value and was not interrupted during sampling. Additionally, the supply of air-accelerated agitation in the fermenter also facilitated the mass transfer.

On the basis of the successful scale-up of the SHF process, simultaneous saccharification and fermentation (SSF) tests were then also performed in the bioengineering fermenters with the same working volume, 1 L. SSF is more challenging because both hydrolysis and fermentation are typically carried out in a fermenter, and the fermenter cannot provide mixing as thorough as the IKA vessels. However, SSF may be more

economically viable than SHF because it reduces total reaction time from 120 to 48 h. As shown in Figure 5, the first 8 h is the

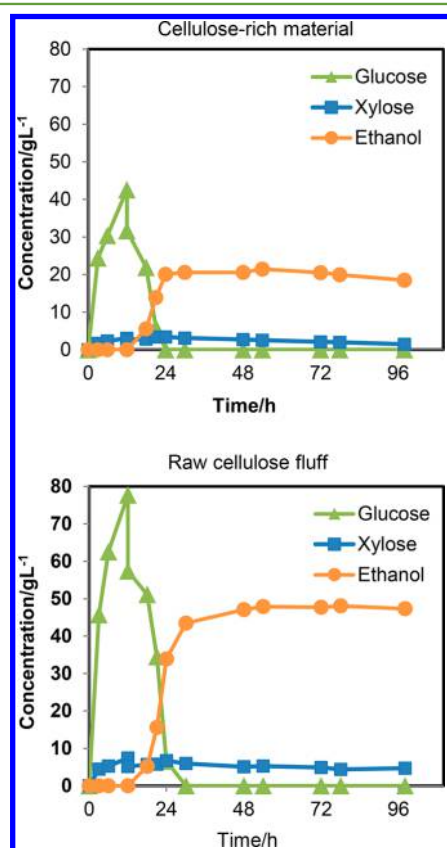


Figure 5. SSF of cellulose-rich material and raw cellulose fluff at 1 L scale (15 wt % solid, 72 mg protein/g cellulose).

stage of enzymatic saccharification and sugar accumulation. Right after the start of fermentation by cell inoculation (time = 8 h), glucose in the broth dropped sharply and was consumed completely within 24 h. Ethanol began to accumulate right after the inoculation. After 24 h, the glucose concentration remained at zero, but ethanol gradually increased until 54 h and then stayed constant. This may be due to faster fermentation kinetics relative to saccharification,¹⁶ so the accumulation of glucose became undetectable. Also, the activity of the enzymes (Ctec2, Htec2) was optimized at 50 °C, while the yeast grew best around 32 °C. Additionally, xylose only had a slight decline from 24 to 96 h, which means that xylose is not efficiently metabolized by the *S. cerevisiae*, as expected.

The ethanol titer in SSF is slightly lower than that in SHF (22.3 g/L vs 27.5 g/L). For the control (raw cellulose fluff at the same solid loading, but with higher cellulose concentration), a higher titer of ethanol (48.6 g/L) was obtained. This method has been proved feasible and largely reduced operation time. Further optimization such as upgrading of the fermenter's agitation system, phase control of saccharification to fermentation, and material import and export design would be helpful to improve the SSF process.

CONCLUSION

This work established an integrated process that converts postconsumer AHPs to intermediate sugars and bioethanol. For the first time, recycled AHPs have been demonstrated as an alternative feedstock for value-added biofuel production

through effective plastic and SAP removal, cellulose recovery, enzymatic saccharification, and microbial fermentation. The recycled AHPs can potentially benefit fuel biorefineries and related biomaterials as they are a cellulose-rich, low or negative cost, and abundant feedstock. Relieving or eliminating AHP landfills will benefit waste management facilities as well as create a positive impact on our environment.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acssuschemeng.7b03931.

Material morphologies of recycled AHPs in sieves and sugar conversions of mock materials (PDF)

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Notes

The authors declare no competing financial interest.

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