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# Economic evaluation of the conversion of industrial paper sludge to ethanol



ABSTRACT

The conversion of industrial paper sludge to ethanol was simulated using engineering process simulation software loaded with laboratory generated conversion data and financially analyzed. In one scenario, sludge is fractionated to remove ash, generating a higher concentration carbohydrate stream for separate hydrolysis and fermentation (SHF). In a second scenario, non-fractionated sludge is processed with only pH adjustment. Four primary sludges from mills producing either virgin or recycled paper were analyzed and the experimental conversion results used to inform the simulations. Financial analysis was conducted assuming ethanol wholesale price of US\$ 0.608 per liter. The most profitable case was fractionated virgin sludge (from a virgin paper mill) to ethanol (F-VK1) with a net present value (NPV) of US\$ 11.4 million, internal rate of return (IRR) of 28%, payback period of 4.4 years and minimum ethanol revenue (MER) of US\$ 0.32 per liter. Risk analysis showed that the F-VK1 case obtained a near 100% probability of business success with both central and bearish (pessimistic) assumptions.

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#### 1. Introduction

The level of research for ethanol production from lignocellulosic biomass (second generation biofuels) has become unprecedented with substantial government and private investment, aiming to overcome food-fuel dispute and environmental limitations of first generation biofuels from cereals, grains, sugar crops and oil seeds. The expansion of traditional ethanol production from corn grain results in an increase of corn crop prices. Over 99% of the world's total biofuel production is from first generation processes in 2010, which accounts only for 0.5% of global energy consumption (Schenk et al., 2008). Despite special interest and extensive efforts of government, research institutions, enterprises and universities to improve the competitivity of cellulosic biofuels, several barriers to second generation biofuels still remain. These include: i) lignocellulosic biomass natural recalcitrance that prevents high ethanol yield within tolerable capital expenditure (CAPEX) and ii) high production costs. Also, delivered biomass cost and

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availability, high pretreatment and chemical cost, intensive CAPEX and overall production costs have been identified as the major obstacles for commercializing cellulosic ethanol with competitive financial returns (Gonzalez et al., 2011a; Hess et al., 2007).

Industrial paper sludge is composed of short pulp fibers from a fraction of the paper making feedstock removed along with clavs, fillers and other contaminants (Jeffries and Schartman, 1999; McGovern et al., 1983). As a potential candidate raw material for producing ethanol, paper sludge has certain advantages over other feedstocks such as agricultural residues or wood sources: i) paper sludge is produced at a concentrated site and permanent production location, making the sourcing of sludge easy at practically no cost; ii) the utilization of sludge for ethanol diverts material going to landfill (avoiding truck hauling costs and landfill investments) and iii) paper sludge is composed of carbohydrate materials in the form of very fine fibers with high specific surface area and often with little lignin present. Since industrial paper sludge has already been subjected to an extensive mechanical and chemical processing (previously imposed during pulping and papermaking processes like cooking, refining and bleaching), polysaccharides in recycled paper sludge are more amenable to enzymatic hydrolysis compared to raw wood or plant material (Keating et al., 2006; Lynd et al., 2001; Wingren et al., 2003). This avoids costly pretreatment to open up the







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lignocellulosic structure or remove lignin to make it more amenable for enzymatic hydrolysis (Yu et al., 2011).

The process development of sludge to ethanol via a biochemical pathway has been studied over the last few decades (Duff et al., 1994; Fan and Lynd, 2007a; Fan et al., 2003; Lark et al., 1997; Lynd et al., 2001). Previous studies (Kang et al., 2011; Nikolov et al., 2000) show that enzymatic hydrolysis of paper sludge has been inefficient in separate hydrolysis and fermentation (SHF) due to the interference of large amount of ash in the sludges during enzymatic reaction. Our previous work (Chen et al., 2012b) showed that acid soluble ash like CaCO<sub>3</sub> not only buffers the pH level (usually 2-3 units higher than the optimum pH) making pH adjustment with acid required for enzymatic hydrolysis, but also adsorbs cellulase with a higher affinity than cellulosic fiber. Acid insoluble ash like clay presents inactive binding with cellulase thereby decreasing enzyme digestibility of fiber in sludge. By fractionating recycled copy paper it was determined that the enzyme hydrolysis of the fractionated material with 0.6% ash content produced approximately 40% higher sugar than unfractionated material at an enzyme dosage of 4 FPU/OD g substrate. In another study (Kang et al., 2011), the fractionation of sludge by using floatation and screening with air and 100 mesh screens, showed a 10% improvement in ethanol yields at an enzyme dosage of 10 FPU/g glucan in a simultaneous saccharification and co-fermentation (SSCF) process. Therefore, in order to achieve higher efficiency in enzymatic hydrolysis (lower enzyme dosage and higher sugar output), a mechanical fractionation approach prior to enzymatic hydrolysis was further investigated in this study. Fractionation of paper sludge lowers total inlet flow into the SHF process thus reducing the reactor sizes needed, and it improves saccharification efficiency by removing a high proportion of ash and impurities that can interfere with enzymatic hydrolysis and fermentation (Chen et al., 2012a). Sugar concentration into the fermentation unit is also higher and thus can result in higher ethanol concentrations in the beer, reducing steam demand for distillation.

Paper mill sludges vary widely because of different feedstocks (e.g., recycled paper, tissue paper, hardwood, softwood) and processes used at different mills. Even among similar mills using similar processes and feedstocks, sludges can vary due to different operating conditions within thermomechanical and chemical unit operations (Mahmood and Elliott, 2006). Therefore, candidate sludges for bioethanol production must be characterized and analyzed before consideration for bioconversion (Lynd et al., 2001). In this research, four sludges from different mills producing either virgin or recycled paper products were evaluated with both non-fractionated and fractionated scenarios. Process simulations (mass and energy balances) of industrial paper sludge to ethanol processes were constructed based on lab data of fractionation and enzyme conversion efficiency and paper mill data of sludge generation rates. Financial evaluation and sensitivity analysis for both scenarios were presented to identify potential business models for the different sludge conversion processes. This paper fills a gap in the literature regarding a rigorous process economics analysis of using paper sludge for ethanol production as well as the mechanical separation of ash from sludge and its economic impact. This research should therefore provide practical information to researchers, paper industry managers, and investors by considering paper sludge to ethanol as a short term commercial pathway for cellulosic ethanol production.

#### 2. Materials and methods

#### 2.1. Laboratory experiments

#### 2.1.1. Feedstock

Four types of primary sludge (the primary treatment residue captured in the primary clarifier) were donated from four paper mills in North and South Carolina (USA). Sludges were collected from each paper mill during the summer of 2011 and stored in sealed buckets in a cold room. Candidate sludges were selected from well mixed sludge in the buckets, fluffed and stored in separate sealed plastic zip-lock bags for at least 24 h prior to solid content measurement and subsequent treatments. Composition analyses of all paper sludges were conducted using the NREL standard procedure (Sluiter et al., 2004). Three parallel tests were conducted for all samples. Ash content of sludge was measured using the Tappi standard method, T 211 om-85 with a furnace temperature of 575 °C (TAPPI, 1993a) and CaCO<sub>3</sub> content was measured per Tappi standard method, T 211 om-93 using furnace temperatures of 575 °C and 950 °C (TAPPI, 1993b). Composition of the sludges is reported in Table 1.

#### 2.1.2. Paper sludge fractionation

Fractionation of paper sludge was conducted using a Pulmac Masterscreen (Pul-mac International, Montpelier, VT, USA) with a 0.2 mm hole size opening screen. For individual paper sludge, 30 OD (oven dry) g of sludge sample was first presoaked overnight at 1.5% solid content and dispersed using a disintegrator for 5 min (15,000 revolutions). Program C was used for the Masterscreen fractionation, this program allows for faster cleaning and screening of sludge. A quantity of 500 mL (7.5 OD grams) of sludge suspension was loaded for each run in the Pulmac Masterscreen. Two portions of retained fiber and fine rich streams were obtained after fractionation: (1) primary fiber that would not pass through the 0.2 mm hole screen and were retained on the reject tray and (2) secondary fiber that passed through the 0.2 mm hole screen but was retained on a 200 mesh (0.074 mm) screen and were collected. Compositions of original sludges, total recovered material, organic material recovery, and percent ash removal are listed in Table 1. Sludges from virgin wood kraft pulping processes were named VK; while sludges from recycled paper deinking process were named RD. Primary and secondary fibers were mixed proportionally according to their yield for enzymatic hydrolysis testing. In general, sludges from virgin paper mills had higher carbohydrate (glucan and xylan) content and less ash content than sludges from recycled paper mills, since the feedstock of virgin paper mills are virgin wood containing high fiber content and no fillers which is in contrast with recycled office type papers with high ash content, Table 1. Also, sludge from recycled paper production is expected to have a higher concentration of short fibers and fines relative to sludges from virgin paper production and thus also contribute to lower overall yields.

#### 2.1.3. Enzymatic hydrolysis

Cellic® CTec2 (cellulase complex blended with high level of cellulases, beta-glucosidase, and hemicellulase) and Cellic® HTec2 (endoxylanase with cellulase background) were kindly supplied by Novozymes (Franklinton, NC, USA). Total cellulase activity of 136 Filter Paper Unit (FPU)/mL was confirmed for Cellic® CTec2 using the filter paper assay as described in the International Union of Pure and Applied Chemistry standard method (Ghose, 1987). CTec2 protein content was 160 mg/mL and HTec2 was 40 mg/mL as provided by the enzyme manufacturer. Enzyme dosage was 4 FPU/OD g sludge and the mixing ratio of CTec2 and HTec2 was 9:1 by volume. A total weight of 2 OD g was immersed in 50 mM sodium citrate buffer (pH of 4.8) and 98% sulfuric acid was added if necessary to adjust the pH to 4.8  $\pm$  0.2. Enzymatic hydrolysis experiments of paper sludge were carried out in 50 mL plastic centrifuge tubes with a solid content of 5% (w/v) at 50 °C for 48 h and 180 rpm in an environmental incubator shaker (New Brunswick Scientific, Edison, NJ, USA). Sugar concentrations (glucose, xylose, galactose, arabinose and mannose) for composition analysis and enzymatic hydrolysis results were quantified using a high-performance liquid chromatography (HPLC) system (Agilent 1200, Agilent, Santa Clara, CA, USA). The HPLC system was equipped with a deashing filter (Bio-Rad 125-0118, Bio-Rad, Hercules, CA, USA) and a Shodex SP0810 column ( $8 \times 300$  mm, Showa Denko, Tokyo, Japan). The mobile phase was Milli-Q water at a flow rate of 0.5 mL/min and temperature of 80 °C.

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