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Feasible process development and techno-economic evaluation of paper sludge to bioethanol conversion: South African paper mills scenario

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ABSTRACT

Paper sludge samples collected from recycling mills exhibited high ash content in the range of 54.59%–65.50% and glucose concentrations between 21.97% and 31.11%. Washing the sludge reduced the total ash content to between 10.7% and 19.31% and increased the concentration of glucose, xylose and lignin. Samples were screened for ethanol production and fed-batch simultaneous saccharification and fermentation (SSF) was optimised for the washed samples that resulted in highest and lowest ethanol concentrations. Maximum ethanol concentrations of 57.31 g/L and 47.72 g/L (94.07% and 85.34% of the maximum theoretical yield, respectively) was predicted for high and low fermentative potential samples, respectively, and was experimentally achieved with 1% deviation. A generic set of process conditions were established for the conversion of high ash-containing paper sludge to ethanol. Techno-economic analysis based on three different revenue scenarios, together with Monte Carlo analysis revealed 95% probability of achieving IRR values in excess of 25% at a paper sludge feed rate of 15 t/d. Feed rates of 30 t/d and 50 t/d exhibited a cumulative probability of 100%. This study presents the technical feasibility and economic viability of paper mills expansion towards bioethanol production from paper sludge.

1. Introduction

Bioethanol gained significant attention as a sustainable source of liquid fuel for the transport sector. Given food security issues and environmental concerns associated with 1st generation biofuels [1], lignocellulosic biomass shows much promise as an alternative and abundant feedstock source [2–4]. However, several challenges remain to ensure feasible conversion of lignocellulosic biomass to ethanol [5]. The complex structure of lignocellulose and the inhibitors generated during its conversion to sugars are major hindrances for the process to be technically and economically feasible [6]. Effective pretreatment of biomass is necessary to make cellulose digestible by enzymes, but severity of the treatment should be restricted due to the inhibitors generation, making optimisation of pretreatment conditions challenging. Hence, focus on renewable fuel expanded towards alternative feed stocks, such as industry wastes and by-products, where pretreatment can be avoided or reduced. Utilisation of industrial waste for on-site fuel production will reduce the energy dependence of industry, environmental issues, including water wastage, and costs associated with waste disposal.

Paper and pulp mills generate significant quantities of paper sludge, which is currently land filled. Due to the emission of greenhouse gases and ground water contamination, there are several legal constraints leading to associated costs with the disposal of paper sludge. Paper sludge is mainly comprised of short fibres (cellulose), fillers and ash. The cellulosic content of the paper sludge, low quantities of lignin and no requirement for thermochemical pretreatment makes it a promising substrate for ethanol production [7–9]. Utilisation of paper sludge for ethanol production is a waste-to-energy conversion process with the advantage of no harvesting and transportation costs generally associated with lignocellulosic biomass to ethanol conversion process. However, high viscosity of paper sludge at high solid concentrations leading to mass transfer limitations represents a main disadvantage of this feed stock. A minimum concentration of 40 g/L ethanol is considered as the threshold to off-set costs associated with energy-intensive distillation recovery processes [10], which signifies the importance of high solids loading. High solids loadings in separate

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hydrolysis and fermentation (SHF) would require increased enzyme dosages due to accumulation of sugars and feed-back inhibition of enzymes, which in turn will impact on process economics due to expenditure on enzymes. During hydrolysis at high solids loading, oligosaccharides could accumulate to 18%–25% of total soluble sugars. Accumulation of low degree of polymerisation (DP) oligosaccharides is inhibitory to commercial cellulases and monomeric sugars accumulation further intensify the inhibition [11]. Hence, producing ethanol from paper sludge through fed-batch simultaneous saccharification and fermentation (SSF) could overcome the sugar accumulation and subsequent enzyme inhibition, which will eventually benefit overall operational profitability. Enzyme dosage, fermentation type and fermentation parameters should thus be studied to achieve the required solids loading, while ensuring the economic attractiveness of the process by maximizing the ethanol yield.

Once the optimum process is established, for an investor or organization to consider an investment in paper sludge to ethanol conversion project by expansion of current operations in the paper mill, the economic viability of the project should be demonstrated. Economic viability can be determined by using economic modelling techniques that are based on financial statements and the resulting cash flows. Key economic indicators, such as the internal rate of return (IRR), net present value (NPV), payback period and discounted payback period are often used as measures of economic viability [12].

Economic analyses that use deterministic estimates in calculating key economic indicators might neglect uncertainty and risk in the model [13]. The use of probability distributions quantifies the possibility of the economic success and risk of failure [14]. A probabilistic method, such as Monte Carlo analysis provides a powerful tool to model uncertainty in the input by accounting for the uncertainties characterized by probability distributions. The response from the Monte Carlo analysis results in a probability distribution of key economic indicators, thus providing an assessment of the probability of success [15]. To date, there is no information available in the literature, where Monte Carlo analysis was used to estimate the economic viability of the production of ethanol from paper sludge. In fact, such analyses are quite scarce for lignocellulosic material in general, especially in the South Africa setting.

The aim of this investigation was four-fold, namely, (1) to optimize the process for high solid loadings, low cellulase loadings and high ethanol conversions, (2) to model the process *in silico* to obtain the necessary mass and energy balances required for the estimation of the capital investment, (3) to assess key economic indicators forthcoming from the resultant discounted cash flows and (4) to assess the degree of uncertainty associated with these outputs.

2. Materials and methods

2.1. Experimental methods

2.1.1. Culture maintenance and growth

Saccharomyces cerevisiae MH1000 was maintained as glycerol stock cultures in the yeast culture collection of the Department of microbiology at Stellenbosch University, South Africa. Revived cultures were grown in YPD (g/L: 10, yeast extract, 20, peptone and 20, glucose) at 37 °C for ethanol fermentation experiments.

2.1.2. Feed stock and enzymes

Nine paper sludge samples (Samples 1–9) were collected from three paper mills owned by Nampak Tissue (Pty) Ltd., which has paper mills located throughout South Africa. Samples named 1–3 were collected from one mill, but at different time points. Samples 4–6 and sample 7–9 were from the second and third mill respectively, at different time points. Nampak Tissue (Pty) Ltd., produces both low and high grade tissue paper from various grades of recycled paper. Various grades of tissue paper are produced from recycled fibre, which are sourced from waste office paper, news print and magazines. In the mill, the waste paper is pulped to obtain individual fibres, which undergoes various stages of contaminant removal including different screening stages, hydrocyclone banks, deinking, washing and bleaching for tissue paper production.

Ash removal from the samples was done by a method applied for Kraft mill sludge [8]. Paper sludge at 20 g/L solids was disintegrated at 37,500 rpm in a laboratory mixer (British pulp evaluation apparatus, Mavis Engineering, London), based on the method described by Tappi [16]. The paper sludge slurry was then washed over a 200 μm screen and mechanically pressed to 35% (w/w) solids.

An enzyme cocktail comprising of Optiflow RC 2.0 cellulase (Genencor, Cedar Rapids, IA, USA) and Novozyme 188 β-glucosidase (Novozymes, Denmark) was used to hydrolyse the feed stock during fermentation. Enzyme activities were determined according to the method described by Ghose [17].

2.1.3. Simultaneous saccharification and fermentation

2.1.3.1. Batch fermentation. Screening experiments were performed in the batch mode in 100 ml rubber capped serum bottles with hypodermic syringes as bubbling CO_2 exhaust. Sampling was done at regular intervals by using sterile hypodermic syringes, without removing rubber caps. The inoculum for fermentation was prepared by growing *S. cerevisiae* MH1000 in YPD medium for 18 h at 200 rpm. A low cost, corn steep liquor-based medium (3 g/L, corn steep liquor and 2.5 mmol/L, $MgSO_4.7H_2O$) was used as a source of nitrogen and additional nutrients. The solid loading of paper sludge was fixed at 20 g/L in the corn steep liquor medium and sterilized for 15 min at 121 °C. The medium was supplemented with filtered cellulase and β-glucosidase at the loadings of 15 FPU/gds and 60 IU/gds, respectively, and was inoculated with 5% (v/v) YPD-grown culture. Considering the buffering effect of $CaCO_3$ present in the ash of paper sludge, pH of the medium was left unadjusted at close to neutrality [8]. Fermentation was continued for 168 h at 35 °C and 200 rpm.

2.1.3.2. Fed-batch SSF. All fed-batch experiments for the central composite experimental design (CCD) as well as for the model validation runs were conducted in 1.3 L bioreactors (BioFlo 110 Modular Benchtop Fermenters, New Brunswick Scientific, Enfield, CT, USA) at 200 rpm and 35 °C temperature without pH and dissolved oxygen control. For the inoculum and medium preparation, the same procedure as described above for batch SSF cultivations was followed, except that initial paper sludge loading was 3% (w/w). For each subsequent feed, paper sludge aliquots were autoclaved at 121 °C for 15 min and added to the reactor at every 12 h intervals, until the pre-determined final paper sludge concentration was reached. All the fermentations were allowed to continue for 168 h.

A CCD to optimize fed batch fermentation for the final ethanol concentration and yield as a percentage of the theoretical maximum was designed using Statistica Version 10 (StatSoft Inc., Tulsa, OK, USA). For a design to be rotatable with all the points equidistant from the centre point [18], the axial spacing should be set as $(2^k)^{0.25}$, where "k" represented the number of independent variables investigated. For this study, the axial spacing was calculated as 1.41421. The response desirability profiler function in the Statistica software was used to optimize the SSF process. A desirability input value of 0 was selected for the ethanol concentrations