

# Continuous Conversion of MSW-derived Waste Paper to Bio-Ethanol Using a 100L 6-stage Continuous Stirred Reactor Separator

*M. Clark Dale<sup>1</sup> and Daniel Musgrove<sup>2</sup>*

<sup>1</sup> Bio-Process Innovation, Inc., 226 N 500 W, W. Lafayette, IN, 47906

<sup>2</sup> Universal Entech, LLC., 5501 N. 7th Avenue PMB 233, Phoenix, Arizona 85013

## ABSTRACT

Biomass is an abundant renewable energy resource that is not widely used due to inherent high costs of converting the substrate into marketable forms such as liquid fuels. To further the technological advancements of bioconverting cellulosic substrates to liquid fuels, many have looked to low cost feedstocks such as Municipal Solid Waste (MSW). Bio-Process Innovation, Inc., and Universal Entech, LLC collaborated on a pilot scale project to demonstrate bio-ethanol production from MSW-derived waste paper. The project consisted of: 1) separating paper from MSW using a unique pulverizer-air classifier, 2) taking the collected light-fraction waste paper stream and pulping the entire unsorted fraction using a low-shear mechanical pulper to produce a clean paper pulp fiber stream, and 3) enzymatically/fermentatively converting the pulp fiber to bio-ethanol using a 100 L pilot scaled Continuous Multi-stage Stirred Reactor Separator (CMSRS). The CMSRS employs gas stripping of ethanol along with the simultaneous saccharification and fermentation of the cellulose fibers and allows recycle of the fermentation broth/cellulase enzyme. The pilot system included a continuous 'hot saccharification/slurry tank' reactor vessel followed by a multi-stage enzymatic Simultaneous Saccharification and Fermentation coupled with gas phase stripping of the ethanol. Broth recycle from the CMSRS effluent was implemented using a 'sludge separating vessel' to maximize re-use of cellulase enzymes. Descriptions of each of the unit operations and performance of the pilot conversion unit are described.

**KEYWORDS:** Enzymatic hydrolysis, Cellulose conversion, waste paper, MSW, bio-ethanol

## INTRODUCTION

In 2001, the International Energy Administration estimated world energy consumption at approximately 404 quads (a quad of energy, or quadrillion British thermal units, is equivalent to 170 million barrels of oil). Biomass is one of the most abundant natural resources on earth. According to the National Renewable Energy Laboratory, the annual world production of biomass is estimated at 2,740 "quads".<sup>1</sup> Meaning, the world produces eight times as much biomass energy as it consumes from all sources of energy. In fact worldwide, biomass is only the fourth largest energy resource after coal, oil, and natural gas. The conversion of biomass to marketable forms of energy, such as fuel bio-ethanol, holds the promise of helping the world move from a fossil fuel based economy to a renewable carbohydrate economy.

Plant biomass is primarily a mix of cellulose, hemicellulose and lignin. Economically converting biomass to bio-ethanol is difficult due to the tough and resistant nature of its lignocellulosic structure. Historically, there have been five "cost" barriers to enzymatically producing bio-ethanol from biomass:

- 1) collection and storage of low density biomass substrates,
- 2) 'pretreatments' needed to make cellulose polymers susceptible to enzymatic attack,
- 3) cellulase enzymes needed to hydrolyze the substrate,
- 4) distilling low concentrations of ethanol produced, and
- 5) converting hemicellulose sugars (xylose & arabinose) to ethanol or other bio-products.

The use of waste paper and other cellulosic waste materials found in Municipal Solid Waste (MSW) is a potential substrate for bio-ethanol production that resolves or assists in negating the first two cost barriers. For the most part, infrastructure and networks are already in place to collect and process MSW, which itself is a 'negative value' commodity or resource. And pretreatment costs as the papermaking process acts as an effective pre-treatment process in itself, allowing good enzymatic attack of paper pulp fibers. In addition, MSW-derived waste paper has little xylose, hence negating the 5<sup>th</sup> cost barrier. In preliminary trials, separated waste paper converted quickly and nearly completely to bio-ethanol and less than 15% of the biomass solids converted to xylose, which alleviates a barrier to commercialization.

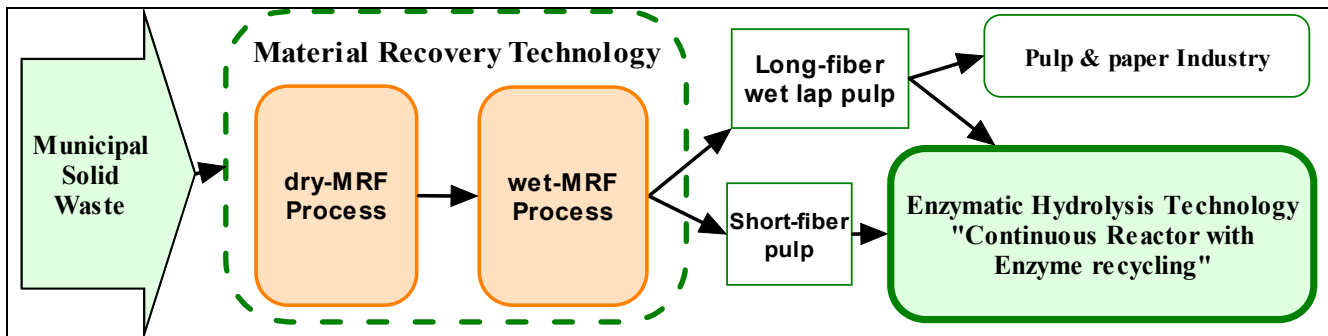
### ***Paper Fraction in MSW***

In 2001, US residents, businesses and institutions produced more than 229 million tons of MSW. Currently 30% is recovered, recycled or composted, 15% is burned, and the remaining 56% is disposed of in a dwindling number of landfills. Of the remaining portion that is landfilled, studies indicate anywhere from 30% to 60% is composed of paper and paperboard (Scleser 1993, 1994, US EPA 2002). Goodman and Walters (1991) estimated that about 66 million tons/yr of paper and paperboard were discarded as MSW in 2000, along with 19 million tons of yard waste. Office paper and magazine stock have a lower lignin and hemi-cellulose make-up than newsprint. Separating waste paper from MSW; 1) reduces the volume of MSW landfilled by 30 to 50%, and 2) can serve as a low to negative cost cellulosic substrate for bio-ethanol production. If 60% of the waste paper stream in MSW in the US were converted to bio-ethanol at a conversion rate of 0.4# ethanol/1 pound paper, MSW-derived waste paper could produce as much as 20 billion gallons/yr of bio-ethanol.

### ***Process Scheme Tested***

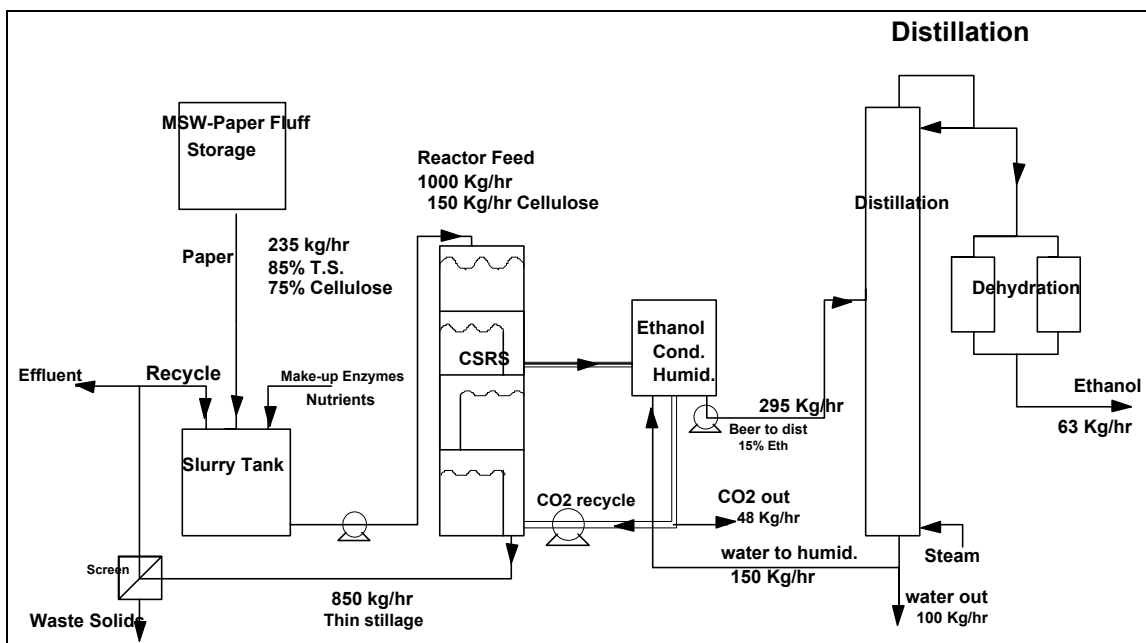
This research involves the evaluation of three process technologies integrated into a process scheme designed to maximize the recovery of MSW-derived waste paper and its subsequent conversion to bio-ethanol (see figure 1). Universal Entech, LLC (UE) and Bio-Process Innovation, Inc. (BPI) are collaborating to develop an innovative waste biomass to ethanol conversion process. The companies are working to integrate their respective processing technologies into a cost effective system for recovering low-grade MSW-derived waste paper and converting the cellulosic material into pulp and/or bio-ethanol. The two Material Recovery Facility (MRF) technologies, while commercially available, require additional pilot testing in order to measure their effective "integration" into one waste processing system. At this 'concept' stage of testing, the results and findings for process efficiencies are preliminary. Subsequent testing and studies will include economic assessments.

**Figure 1 Process Scheme for Converting MSW-derived Waste paper to Bio-ethanol**



A unique reactor system, called the Continuous Multi-stage Stirred Reactor Separator (CMSRS) has been developed and tested by BPI to address and resolve the additional cost barriers presented above, specifically the issue of enzyme costs and low ethanol concentration. The CMSRS is designed so that a large fraction of the cellulase enzyme can be recycled/re-used after the conversion process. Utilizing the CMSRS should have the effect of reducing the amount of fresh enzyme needed by a factor of 2 to 4 X. The CMSRS reactor technology allows ethanol stripping from the reactor during the multi-stage cascade conversion of substrate to bio-ethanol. This technology is particularly beneficial to a Simultaneous Saccharification and Fermentation (SSF) of biomass as the low temperature stripping of the final inhibitor, ethanol, is accomplished at a low fermentation temperature, allowing the broth to be recycled to the feed make-up. A schematic of the proposed CMSRS pulp to ethanol process is shown in Figure 2 with approximate flows for a system handling 1000 Kg/hr of 15% cellulosics.

**Figure 2 Proposed Process Flow Diagram for MSW-derived Waste paper to Bio-ethanol**



The combination of the BPI bioconversion technology and the two waste paper fiber recovery technologies (dry-MRF and wet-MRF) were tested and are presented in this paper.

## **Pilot Scale Trials**

### **1) Waste Paper Separation from MSW**

MSW-derived waste paper was recovered from full truckload samples (approx. 4,000 kg) of residentially collected MSW. The complete load was processed through an integrated pulverizer-air classifier, with no pre- or post screening for removing dirt, grit, grass and other fine materials. The processed MSW was separated into 2 streams; 1) a paper-rich light fraction, consisting of paper, film plastic, a small percentage of plastic bottles/aluminum cans, and fines (grass, dirt, etc) and 2) a heavy fraction consisting of the balance of the MSW stream. Separation efficiencies (i.e. recovery of light fraction) averaged 25% by weight and 40% by volume. A 200 kg composite sample of the light fraction was sent to a pulper manufacturer for paper/pulp recovery.

### **2) Pulp Fiber Separation**

Three separate 40 kg composite samples were taken from the one 200 kg sample and 3 batch pulping test runs were conducted using a low-shear mechanical pulper. Each test run used 105 deg. F process water (no pulping chemicals or wetting agents) at an approximate 10% consistency. The contents of the pulper were diluted to 2% consistency using ambient tap water and screened through a 9mm slotted rotating screen to remove plastics, overs, non-pulped material and to recover the pulp fiber. The liquid stream containing the accepts was further processed through a secondary disk screen equipped with 20 mesh clothe. Approximately 80 kg of pulped fiber at 25%TS was collected and combined from the 3 batch runs and used to conduct the pilot scale bio-ethanol trials.

### **3) Pulp Fiber to Ethanol Conversion**

Bench-scale trials were conducted at the BPI lab. The composition of the MSW-derived pulp fiber was determined and is represented in Table 1. These compositions were determined using standard NREL procedures (LAP001-LAP 009, NREL, 1999). The pilot plant follows the basic design as per Figure 2, except that ethanol is not condensed, but instead absorbed into a water stream in a countercurrent 6" ID packed column to simplify the pilot operation.

Moisture	75%
Ash (d.b.)	15%
Kl.Lignin (d.b)	11%
Cellulose(d.b.)	62%
Hemicellulose/Xylan	<5%

1) Hot Saccharification/Slurry Reactor. A 40 liter 316 stainless steel vessel was developed to serve as a Hot Saccharification-Feed Slurry (HSFS) reactor. This reactor was fitted with 1) stirring- using a belt driven ½ horsepower 4 speed drive, 2) temperature control- using an external 500W rubber heating pad and an external thermocouple temperature controller, 3) pH control- using a pH probe and a controller adding sulfuric acid as needed to keep the pH at 4.8. The slurry from the HSFS tank is pumped from the bottom of the reactor using a Masterflex peristaltic pump to the first stage of the CMSRS.

2) Continuous Multistage Stirred Reactor Separator (CMSRS). The CMSRS consists of six 25 cm high by 25 cm ID stacked stages, the first 2 stages are termed 'enriching' stages where SSF of cellulose allows ethanol concentration to increase in the broth, and then the next

4 stages include countercurrent gas/liquid stripping trays where the overflow from the stage contacts a recycled CO<sub>2</sub> stream which strips the ethanol from the fermentation broth. The vessels are stirred with a center shaft mounted 3" marine propeller at 900 RPM using a ½ horsepower belt drive. The drive is attached to a single shaft through all six stages connected by socket adapters between the stages. Total liquid hold-up in the 6 stages is approximately 100 L. Gas flow rate (CO<sub>2</sub>) through the separation trays of the stages was set using a stripping gas to liquid ratio mV/L of 2.5; where V is the molar flow of CO<sub>2</sub>, L is the molar flow rate of the water in the fermentation broth, and m is the partition coefficient ( $m=y_e/x_e$ ) of ethanol at the operating temperature of the CMSRS. At a residence time of the broth in the CSMRS system of 36 hours, a feed rate of 2.8 L/hr of 10% pulp solids (100 g/L pulp solids, 900 g/L water), and an operating temperature of 35 C would require a CO<sub>2</sub> flow rate of 600 gmole/hr or 248 Liters/min.

3) Absorber. The rich CO<sub>2</sub> is taken to the bottom of a 6" ID by 6' tall 304 stainless steel absorber column where it is contacted with a counter current water stream which absorbs the ethanol. The absorber is packed with an absorbent proprietary 'structured packing' developed by BPI. The lean CO<sub>2</sub> from the top of the absorber column is recycled to the CMSRS system. Gas/liquid flow rates (CO<sub>2</sub>/water) were set using a L/mV molar flow ratio of 2.5, where L is the molar flow rate of absorbing water and V is the molar flow rate of CO<sub>2</sub>, and m is the equilibrium coefficient ( $m=y_e/x_e$ ) for of ethanol at the operating temperature of the absorber (32 C.) (King, 1988)

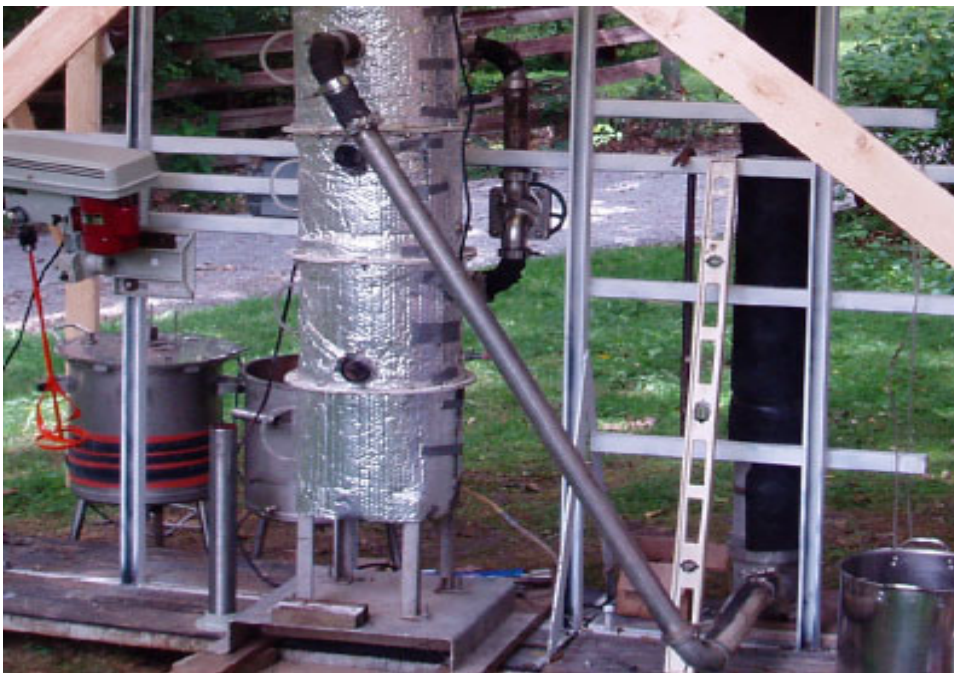


Figure 3. Pilot Pulp to Ethanol Facility during set-up- Hot Saccharification/Slurry Tank on left, bottom 3 stages of CMSRS in middle, Absorber column on Right.

4) Gas recirculation. The CO<sub>2</sub> is re-circulated using a centrifugal 1/2 HP Gast Regen blower. The gas flow is ducted through 2" 304 stainless steel tubing connected by Tri-Clover type dairy fittings and clamps. Gas flow rates are measured using a by-pass orifice rotometer from Cole-Parmer. Gas flow rates are varied using a by-pass loop around the blower fitted

with a sanitary 2" valve. As this valve is closed, more gas is forced through the reactor gas stripper /absorber loop of the pilot plant. Figure 3 shows the pilot plant during set-up.

5) Sludge separation/ broth recycling vessel. A 40 liter 316 stainless steel vessel was fitted to be a 'sludge separator'. The vessel is fitted with a baffle, and the incoming exhausted liquid fermentation broth effluent (ethanol and cellulose depleted) is introduced near to the bottom of one (heavy) side of the baffle, and liquid broth that overflows the baffle (light side) is taken back to the Hot Saccharification/ Feed Surry (HSFS) reactor to make-up the new feed. Sludge is drained from the heavy side of the tank each time feed is introduced to the HSFS.

### **Pilot performance**

The effect of stirrer type and performance on saccharification of a 10% pulp mix was determined using several types of stirrers at various speeds in the HSFS reactor. Lab scale trials indicated that shear/stirring had a very positive effect on liquefaction/saccharification of the pulp fiber. Prior to beginning our continuous pilot trial(s), we tested 1) a 3" 3 bladed marine propeller, 2) a paired set of high speed drum low pitch 4 bladed propellers (Grainger), 3) a high shear dispersion 3" propeller (Design B Blade- Indco, New Albany, IN), and 4) a high pitch 'mixed flow' impeller (MFI31H, Indco). In a set of trials using an 8 to 10% extremely viscous, semi-solid, non-saccharified pulp, the best performance by visual observation was using both the marine and the high shear blades together; with the marine propeller mounted about 6" above the high shear dispersion blade. Using this configuration, the pulp liquefied in approximately 12 hours at 50 C with 50% less enzyme as compared to lab bench-scale trials. Further details on the pilot performance will be presented during the conference.

### **Bibliography**

Dale, M.C. and M.R. Moelhman 2002. Enzymatic SSF of Biomass to Ethanol on Lab and Pilot Scales: A 130 L Multistage Continuous Stirred Reactor-Separator. *Final Report for: Great Lakes Governor's Regional Biomass Program, Indiana Biomass Program, and DOE Inventions and Innovations Program*

Dale, M.C., 1992. A Continuous Multi-Stage Stirred Reactor-Separator for Production of Volatile Fermentation Products. Patent # 5,141,861.

King, J. 1980. *Separation Processes*. McGraw Hill Chem Eng Series. NY. NY.

NREL, ia Western Regional Biomass Energy Program, [www.westbioenergy.org](http://www.westbioenergy.org)

Philippidis, G., D. Spindler, and C. Wyman. 1991. Mathematical Modeling of Cellulose to Ethanol by Simultaneous Saccharification and Fermentation Process. Paper #25. Biotech. Symp. for Fuels and Chemicals. Colorado Springs.

Shleser, Robert. 1994. Ethanol Production in Hawaii. Prepared for the State of Hawaii, Department of Business, Economic Development and Tourism. Honolulu: Energy Division, Dept. of Business, Economic Development and Tourism, State of Hawaii

### **Acknowledgements**

Funding for this project was provided by the Arizona Department of Environmental Quality (ADEQ) Recycling Unit Grant Program and the Indiana Biomass Grant Program. The project was further supported by contributions from BPI and UE. Cellulase enzymes for the pilot scale project were generously provided by Valley Research of South Bend IN (Validase TR cellulase), and a non-commercially available cellulase being developed by Novozyme Biotech

---