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### Countercurrent Flowthrough Pretreatment of Cellulosic Biomass

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# Link Energy Fellowship Report by Véronique Archambault-Léger

## 1. Narrative summary

### *Introduction*

Producing fuel from lignocellulosic biomass is of interest in the context of developing a sustainable global energy system<sup>1</sup>. Cellulosic biomass, such as agricultural residues, wood and grasses, is the most promising source of biomass for very large scale energy production. Sugar cane residues such as bagasse, precollected at sites with substantial existing infrastructure, are widely seen as a particularly promising low hanging fruit, large scale and renewable lignocellulosic biofuel resource.<sup>2,3</sup> In the case of biological conversion of cellulosic biomass to sugars, recalcitrance results from incomplete accessibility of attack by microbes and their saccharolytic enzymes. As employed in the biological conversion field, pretreatment refers to the key limiting process of preparing the biomass to overcome biomass recalcitrance. There are a wide variety of pretreatment processes, which decisively improve the subsequent yield of sugar hydrolysis<sup>4</sup>, but account for a substantial fraction of the cost of overall conversion<sup>5</sup>.

The conundrum of conventional pretreatment is that conditions severe enough to yield reactive fiber are accompanied by sugar degradation. Solubilized sugars have less time to degrade when operating in a flow through (FT) configuration as the liquid is removed from the reactor. In addition, recondensation of solubilized lignin and hemicellulose on cellulose fibers upon cooling occurs to a lesser extent in a FT configuration as compared to configurations without FT.<sup>6,7</sup> Consistent with this understanding, a FT configuration has clear potential to produce highly reactive solids while minimizing sugar degradation and removing significantly more lignin and hemicellulose compared to PT in non FT configurations at the same temperature and residence time<sup>4,6,8</sup>.

The aim of my work as a Link Fellow was to develop an analytical framework incorporating fluid mechanical, thermodynamics and kinetic factors to determine the feasible domains of hot water FT

pretreatment. I addressed the mechanical complexities of arranging a bed of biomass for FT configuration and analyzed the energy requirements, capital costs and sugar dilution in order to realize the advantages of FT pretreatment at scale.

### ***Results***

Although continuous counter-current flow operation is common in the wood pulp and paper industry and has been reported for wheat straw pretreatment at a pilot scale, operating continuous FT pretreatment at scale is challenging and few studies have reported related fluid mechanics. I investigated fluid mechanical properties of cellulosic feedstocks to inform identifying conditions for which FT pretreatment can be implemented in a practical context. Measurements of pressure drop across packed beds, viscous compaction and water absorption were reported for milled and not milled sugarcane bagasse, switchgrass and poplar, and important factors impacting viscous flow were deduced. Using biomass knife-milled to pass through a 2 mm sieve, the observed pressure drop was highest for bagasse, intermediate for switchgrass and lowest for poplar. The highest pressure drop was associated with the presence of more fine particles, greater viscous compaction and the degree of water absorption. Using bagasse without particle size reduction, the instability of the reactor during pretreatment above 140 kg/m<sup>3</sup> sets an upper bound on the allowable concentration for continuous stable flow.

Many kinetic models have been reported describing the kinetics of pretreatment using hot water. My experimental results with sugarcane bagasse show that the tradeoff between increasing fiber reactivity and sugar degradation can be substantially mitigated by carrying out pretreatment in a FT mode. As a Link Fellow, I developed a model incorporating both kinetics and mass transfer and evaluated a variety of reactor configurations. Simulated results compared well to the literature for bagasse pretreated in both batch and FT configurations. A variety of FT configurations, including counter-current flow, cross flow, discrete counter-current and partial FT, resulted in sugar degradation that was very low (1 to 5%) and 5 to

20 fold less than a conventional plug flow configuration. Performance exhibits strong sensitivity to the extent of hemicellulose solubilization, particularly for conventional plug flow configuration.

FT operation is also challenging because the higher water usage compared to non-flow configuration dilutes the sugar streams and increases energy consumption<sup>8</sup>. I addressed the energy demand, sugar dilution and economic concerns of using FT pretreatment for the bioconversion of sugarcane bagasse and trash to ethanol. FT pretreatment resulted in a lower minimum ethanol selling price (\$0.82/L) than dilute acid (\$1.01-1.19/L), hot water (\$1.13-1.27/L) and steam explosion (\$0.86-1.18/L) (the range represents different studies). Sugar dilution was not a limiting factor provided that extensive heat integration was employed, as is the case in an oil refinery. The ethanol beer to distillation (combined ethanol from the cellulose and hemicelluloses streams) contained 5.0 wt% ethanol. Integrated first generation and second generation plants with no external fuel supplied were examined based on conversion of sucrose, bagasse, and available cane trash. A base case was defined using FT pretreatment which routed all of the bagasse and 31.8% of the available trash to ethanol production. For an alternative “best parameter” case, all of the bagasse and available trash was routed to ethanol production, leaving 1.1% of the feedstock HHV available for electricity exports. Ethanol yields for the base, best parameter and steam explosion cases were 59.9, 81.6 and 50.8 L/wet ton cane respectively, representing increases of 79%, 108% and 67% compared to the first generation plant. My analysis indicated that sugar dilution and energy consumption are not barriers to practical commercial implementation of FT pretreatment, and that FT pretreatment has potential to be economically advantageous compared to hydrothermal and dilute acid pretreatments.

### ***Significance, impact and future directions***

Other scientists looking at FT pretreatment have focused primarily on the important aspect of operative chemistry. I focused primarily on mechanical aspects, thermodynamic considerations, reactor configuration, kinetics and economics, building where possible on the foundation of chemically-oriented

work that others have provided and on insights from the pulping industry. The key challenges for practical implementation of this promising pretreatment method were identified and analyzed to bridge the gap between fundamental understanding and scale up feasibility. Prior to my work, FT pretreatment was mainly regarded as a tool to study the pretreatment reaction, but as an impractical method for commercial implementation. My work contributed to the new perception that hot water FT pretreatment is actually a highly efficient, promising and practical pretreatment method.

Partial FT pretreatment incorporating plug flow and flow regions could provide highly digestible solid fibers and high sugar recovery at one set of reaction conditions, be more mechanically practical than complete counter-current and help address the sugar dilution and energy consumption challenges. The next step toward implementing FT pretreatment of sugarcane bagasse is to test the detailed mechanical design of the reactor at the pilot scale.

### ***References***

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## 2. Scholar contributions

### *Journal publications*

1. V. Archambault-Léger and L. R. Lynd, Fluid mechanics relevant to flow through pretreatment of cellulosic biomass. *Bioresource Technology*, 2014, **157**, 278-283.
2. V. Archambault-Léger, X. Shao and L. R. Lynd, Simulated performance of several reactor configurations for hot water pretreatment of sugarcane bagasse incorporating kinetics and mass transfer. *Chemistry and sustainability*, under review.
3. V. Archambault-Léger, Z. Losordo and L. R. Lynd, Energy, sugar dilution and economic analysis of hot water flow through pretreatment for producing biofuel from sugarcane residues. *Biofuels, Bioproducts and Biorefining*, under review.

### *Conference presentation*

1. Archambault-Leger V., Shao X, Lynd L. Kinetic modeling of sugarcane bagasse hot water flowthrough pretreatment, oral presentation, AIChE Annual Meeting, San Fransisco CA, November 2013.
2. Archambault-Leger V., Shao X, Lynd L. Fluid dynamics relevant to pretreatment of cellulosic biomass, poster presentation, 35<sup>th</sup> Symposium on Biotechnology for Biofuels and Chemicals, Portland OR, May 2013.

## 3. Financial statement

Discretionary funds were not allotted.

## 4. Impact of fellowship

I am truly grateful and honored to have received the Link Energy Fellowship. It allowed me to pursue high quality research freely and without worrying about funding. I was able to determine the key challenges to practical implementation of flow through pretreatment and analyze them to evaluate the practicality of this highly performing pretreatment. The fellowship was a crucial support through the later, most productive years of my doctoral studies. I am also convince that the support will have a lasting impact on my professional career and desire to make a difference in the energy field.