A Supercritical Water Approach to Cellulosic Sugars:
Lifecycle Energy, Greenhouse Gas and Water Implications

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Abstract
The lifecycle fossil energy, greenhouse gas emissions, and water use are evaluated for a supercritical water approach to sugar production from cellulosic feedstocks. Bark and lignin are used for process heat. If grid electricity is used, the lifecycle fossil energy input is 9.0 MJ/kg of fermentable sugar. If the bark and lignin are used for combined heat and power production, supplemented by natural gas, the lifecycle fossil energy input is 3.9 MJ/kg of fermentable sugar. Lifecycle water consumption is 7 liters per kg of fermentable sugar. If grid electricity is used for process electricity, lifecycle water withdrawal is 30 liters per kg of fermentable sugar, 90% of which is from off-site grid electricity production. If on-site combined heat and power is used for electricity and process heat, the lifecycle water withdrawal is 3 kg per kg of fermentable sugar. Lifecycle greenhouse gas emissions are 522 g CO$_2$e and 320 g CO$_2$e per kilogram of fermentable sugar for the grid electricity and CHP scenarios, respectively.

1. Goal and Scope

There are currently three main processes for converting biomass for biofuel or biochemical applications: enzymatic hydrolysis, acid hydrolysis, and gasification. In enzymatic hydrolysis and acid hydrolysis respectively, enzymes and acid are used to hydrolyze the cellulose. In gasification, high temperature and pressure are used to produce a syngas of primarily CO and H$_2$.

Here we address a different, fourth approach to cellulosic biomass conversion in which supercritical water is used to break down the cellulose into usable sugars. This process may have the advantages of faster processing than with enzymatic or acid hydrolysis, as well as elimination of the need for hydrolysis enzymes, gasification enzymes, or hydrolysis acid, and higher yield than gasification.

A key motivation for use of biomass to produce chemicals and fuels is the potential for lower consumption of fossil fuels and lower greenhouse gas emissions, without excessive water consumption. Here we evaluate the lifecycle fossil energy use, greenhouse gas emissions, and water use of fermentable sugar production using the supercritical water approach to cellulosic processing. These sugars can be used for production of biofuels and biochemicals.

Figure 1 shows the system boundary of the lifecycle assessment. The front-end includes collection of hardwood, described in the next section, which is chipped either off-site or on-site. The on-site operations include some chipping, the full sugar production process,
and may include production of ethanol or other bioproduct as well. This analysis stops at sugar production; analysis of the ethanol production step is also underway.

Figure 1. Scope of Life Cycle Assessment for the Supercritical Water Approach to Biomass Conversion. This analysis includes wood collection through sugar production.

2. Inventory Analysis

2.1 Forestry, Wood Collection, and Delivery
Two types of biomass resources are anticipated as process feedstocks: hardwood mill residuals and low value hardwood. Hardwood primary mill residuals are the non-bark outer wood pieces that are left from cutting roundwood into lumber at sawmills. These residuals will be chipped at the sawmill or will be brought to the processing facility and chipped on-site. In addition, low value hardwood may be chipped in the forest or will be brought to the facility and chipped on site. In this analysis we assume half is chipped on-site and half is chipped off-site. The biomass feedstock is anticipated to be hardwood from eastern US hardwood forests, with low-intensity logging and natural regeneration.

The processes included in this part of the analysis include chainsaw handfelling, chainsaw delimbing, loading, skidding, chipping in the forest, and transportation. We draw on the US Life-Cycle Inventory Database (NREL 2011), which includes data on processes related to forestry, wood, and biomass, and has been used in a number of lifecycle assessments of forest biomass (Hsu et al. 2010). Although the database does not have information specifically on hardwood forestry practices in the southeast, it does have data on energy consumption of hardwood forestry activities that we expect to be largely independent of location.

Specifically, we use 0.19 standard machines hours per green cubic meter for chainsaw hand-felling, 0.08 standard machine hours for large loader operation, 0.10 standard machine hours for chainsaw delimbing, and 0.18 standard machine hours for skidding with a wheeled cable skimmer. Chainsawing is reported to consume
0.76 kg of diesel fuel per hour. We use a hardwood oven dry density of 580 kg/m$^3$ (NREL 2011).

For off-site chipping, not included in the NREL database, we refer to Nati et al. (2010), who report in-woods chipping diesel energy use as 0.72 l/t for large screen (60 x 240 mm) chipping of hybrid poplar, 1.09 l/t for large screen chipping of white pine, and 1.22 l/t for medium screen (60 x 40 mm) chipping of white pine. In our analysis we use 1.0 l/t.

Forests can be a sink for carbon, absorbing carbon into above-ground biomass (trees), below ground biomass (roots) and soil. The forests modeled here are assumed to be mature forests; additional carbon storage in mature forests is expected to be low compared to a young forest. Consistent with the low-intensity, natural regeneration hardwood forest system modeled here, we assume zero net change in carbon storage in the forests due to the use of forest materials.

Use of land to produce feedstock for bioenergy production may displace previous land use activities. If these previous activities are moved, in whole or in part, to a different location, either directly by the previous land manager or indirectly through market forces, this indirect land use change may also have energy, greenhouse gas, and other impacts that are considered as part of the lifecycle impact of bioenergy systems (Searchinger et al. 2008). However, the feedstock for this process consists of low-diameter trees and wood residues; use of these materials does not result in a change in land use. Accordingly, no direct or indirect land use change is modeled here.

**Transportation**
The biomass is assumed to be transported 80 km, with an energy use of 3 MJ/ton-km (US DOE 2008).

Table 1 summarizes the energy use for the forestry, wood harvesting, and delivery processes.

<table>
<thead>
<tr>
<th></th>
<th></th>
<th>MJ/green ton</th>
</tr>
</thead>
<tbody>
<tr>
<td>Forestry - chainsawing</td>
<td>6.3</td>
<td></td>
</tr>
<tr>
<td>Forestry - loading</td>
<td>100.8</td>
<td></td>
</tr>
<tr>
<td>Forestry - skidding</td>
<td>165.0</td>
<td></td>
</tr>
<tr>
<td>Biomass transportation</td>
<td>240.0</td>
<td></td>
</tr>
<tr>
<td>Chipping off site</td>
<td>19.3</td>
<td></td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td>531.4</td>
<td></td>
</tr>
</tbody>
</table>

**2.2. Chemical Inputs**

Inputs to the on-site biomass processing include lime, sulfuric acid and ammonia. The energy and greenhouse gas impacts of production of these materials, drawn
from the NREL database, are included in the assessment of off-site impacts (NREL 2011).

Table 2. Chemical Inputs

<table>
<thead>
<tr>
<th>Process Inputs</th>
<th>Electricity Input (kWh/kg)</th>
<th>Natural Gas Input (MJ/kg)</th>
<th>Rate of Use (kg/kg sugar)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sulfuric Acid</td>
<td>0.066</td>
<td>--</td>
<td>0.19</td>
</tr>
<tr>
<td>Lime</td>
<td>0.036</td>
<td>5.495</td>
<td>0.0019</td>
</tr>
<tr>
<td>Ammonia</td>
<td>0.14</td>
<td>5.04</td>
<td>0.003</td>
</tr>
</tbody>
</table>


2.3. On-site operations

2.3.1 On-site auxiliary operations

On-site auxiliary operations include chipping of any incoming unchipped wood, and operations associated with wood handling. Whereas off-site chipping used diesel power, on-site chipping is electrically powered. Discussions with chipping operators indicate electricity requirements for chipping is 9 kWh per green ton (Floyd, 2011).

2.3.2 Sugar Model

Figure 2 illustrates the key sugar production processes in converting biomass into a sugar solution of xylose and glucose. The inputs are biomass, electricity, heat, ammonia, lime, and sulfuric acid. Gypsum is a co-product from of the recovery processes. Although this gypsum may have some economic value, here it is evaluated as a waste that is trucked to a land disposal site. Wastewater pretreatment is also included in the facility operations. The process is modeled using Aspen software.

Figure 2. Diagram of sugar production processes in a supercritical water approach to conversion of biomass to fermentable sugars.
**Fractionation:** The first process step is fractionation, in which the incoming wood chips are reduced to 20 mesh size using a collision mill, corresponding to a diameter of about 850 microns. In the fractionation process, the solids and liquids are separated, with the solids containing largely cellulose, xylan and lignin. The solids go to cellulose hydrolysis, discussed below; the liquids go to C5 recovery, also discussed below.

**Table 3. On-site Process Energy Requirements for Sugar Production Process**

<table>
<thead>
<tr>
<th>Process Stage</th>
<th>Energy Source</th>
<th>Power</th>
<th>Units</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chipping on site</td>
<td>Electricity</td>
<td>6</td>
<td>kWh/green ton</td>
<td>293 kWh/hr</td>
</tr>
<tr>
<td></td>
<td>Diesel</td>
<td>0.15</td>
<td>gal/green ton</td>
<td>990 MJ/hr</td>
</tr>
<tr>
<td>Sugar Production Process + WWTP</td>
<td>Feed</td>
<td>Electricity</td>
<td>1839 kW</td>
<td>1,839 kWh/hr</td>
</tr>
<tr>
<td></td>
<td>Fractionation</td>
<td>Electricity</td>
<td>607 kW</td>
<td>607 kWh/hr</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Heat Requirement</td>
<td>24.73 MMBtu/hr</td>
<td>26,089 MJ/hr</td>
</tr>
<tr>
<td></td>
<td>Cellulose Hydrolysis</td>
<td>Electricity</td>
<td>1599 kW</td>
<td>1,599 kWh/hr</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Heat Requirement (Supercritical Water)</td>
<td>78.74 MMBtu/hr</td>
<td>83,068 MJ/hr</td>
</tr>
<tr>
<td></td>
<td>C5-Recovery</td>
<td>Electricity</td>
<td>38 kW</td>
<td>38 kWh/hr</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Heat Requirement</td>
<td>0.71 MMBtu/hr</td>
<td>747 MJ/hr</td>
</tr>
<tr>
<td></td>
<td>C6-Recovery</td>
<td>Electricity</td>
<td>72 kW</td>
<td>72 kWh/hr</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Heat Requirement</td>
<td>2.51 MMBtu/hr</td>
<td>2,651 MJ/hr</td>
</tr>
<tr>
<td></td>
<td>Utilities</td>
<td>Electricity</td>
<td>864 kW</td>
<td>864 kWh/hr</td>
</tr>
<tr>
<td></td>
<td>Others</td>
<td>Electricity</td>
<td>824 kW</td>
<td>824 kWh/hr</td>
</tr>
<tr>
<td></td>
<td>Subtotal</td>
<td>Electricity</td>
<td>5843 kW</td>
<td>5,843 kWh/hr</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Heat Requirement</td>
<td>106.69 MMBtu/hr</td>
<td>112,555 MJ/hr</td>
</tr>
<tr>
<td></td>
<td>Gypsum disposition</td>
<td>Diesel</td>
<td>180</td>
<td>MJ/ton of gypsum</td>
</tr>
<tr>
<td>Total</td>
<td>Electricity</td>
<td>5,989</td>
<td>kWh/hr</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Heat</td>
<td>112,555</td>
<td>MJ/hr</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Diesel</td>
<td>1,022</td>
<td>MJ/hr</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Heat Credit from Lignin &amp; Bark</td>
<td></td>
<td></td>
<td>133,556 MJ/hr</td>
</tr>
</tbody>
</table>

Data are based on a sugar production rate of 10,323 kg/hr.

**Cellulose hydrolysis:** In the cellulose hydrolysis process, the solids are mixed with water, heated and then treated with supercritical water. The output is a cellulose liquor and lignin. The supercritical water boiler has a substantial heat requirement, about 74% of the entire sugar production process heat requirement.

**Recovery:** Solids are filtered and sugars are produced from the cellulose liquor. Ammonia is added to help detoxification. Calcium hydroxide (Ca(OH)2) is added to
remove sulfuric acid (H₂SO₄), producing solid gypsum. C5-recovery and C6-recovery produce xylose and glucose, with five and six carbon atoms, respectively.

Table 3 summarizes the on-site electricity and process heat requirements.

**2.4 Lifecycle Energy, Water and Greenhouse Gas Inventory for Sugar Production**

**2.4.1. Energy source scenarios**
Lignin and bark will be used to provide process heat for on-site operation, with additional natural gas heat as needed for sugar conversion to ethanol or other products. To explore the implications of the energy alternatives, we consider several scenarios for energy sourcing: sourcing electricity from the grid, and using either (1) natural gas or (2) lignin and bark for the process heat; this comparison highlights the benefits of the use of biomass for heat. In addition we consider the development of an on-site combined heat and power system to produce both electricity and heat using (3) natural gas or (4) lignin and bark supplemented by natural gas as necessary. Note that scenarios (1) and (3) are for comparison purposes only; lignin and bark will be used for heat.

For the grid-sourced electricity scenario, the electricity is assumed to be sourced from Dominion Virginia Power (DVP), which has a fuel mix of 31% coal, 28% nuclear, 10% natural gas, 2% other, and with purchases of 29% (Dominion 2011a). Typical coal-fired generation has an efficiency of about 33%, nuclear efficiency is about 30%, and natural gas generation can be about 50% efficient with use of combined cycle technology. Given the substantial DVP purchase of electricity from unspecified sources, we estimate 33% efficiency of the conversion of primary fuel energy to electricity.

**2.4.2. Lifecycle Energy**

Table 4. Primary Fossil Energy Consumption for Forestry, Wood Collection, Delivery, and Chemical Inputs

<table>
<thead>
<tr>
<th>Off-site</th>
<th>Energy Source</th>
<th>MJ/kg Sugar</th>
</tr>
</thead>
<tbody>
<tr>
<td>Forestry - chainsawing</td>
<td>Diesel</td>
<td>0.03</td>
</tr>
<tr>
<td>Forestry - loading</td>
<td>Diesel</td>
<td>0.45</td>
</tr>
<tr>
<td>Forestry - skidding</td>
<td>Diesel</td>
<td>0.73</td>
</tr>
<tr>
<td>Biomass transportation</td>
<td>Diesel</td>
<td>1.07</td>
</tr>
<tr>
<td>Chipping off site</td>
<td>Diesel</td>
<td>0.09</td>
</tr>
<tr>
<td>Sulphuric acid</td>
<td>Electricity (US Grid)</td>
<td>0.14</td>
</tr>
<tr>
<td>Lime</td>
<td>Electricity (US Grid)</td>
<td>0.0008</td>
</tr>
<tr>
<td></td>
<td>Natural Gas</td>
<td>0.01</td>
</tr>
<tr>
<td>Ammonia</td>
<td>Electricity (US Grid)</td>
<td>0.005</td>
</tr>
<tr>
<td></td>
<td>Natural Gas</td>
<td>0.02</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td></td>
<td><strong>2.53</strong></td>
</tr>
</tbody>
</table>
Table 4 shows the energy balance for front-end processes, and Table 5 summarizes the primary energy requirements of on-site processes for each of the energy scenarios. Adding the off-site energy consumption of 2.53 MJ/kg to the on-site energy consumption from table 5 shows that when grid electricity is used for electricity (and lignin and bark for process heat), lifecycle fossil energy use is 8.96 MJ/kg of fermentable sugar, which is lower by a factor of three compared to what it would be if natural gas were used for heat. With development of an on-site combined heat and power system, in which the lignin and bark are used for electricity and process heat generation, supplemented by natural gas if needed, the on-site fossil energy consumption would be 1.35 MJ/kg fermentable sugar, for a total lifecycle fossil energy consumption of 3.88 MJ/kg of fermentable sugar.

Table 5. Primary Fossil Energy Consumption for On-site Sugar Production

<table>
<thead>
<tr>
<th>On-site Energy Supply Scenario</th>
<th>Energy (MJ/kg Sugar)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Grid Electricity</td>
</tr>
<tr>
<td>(1) Grid Electricity + Natural Gas (NG) Heat</td>
<td>6.33</td>
</tr>
<tr>
<td>(2) Grid Electricity + Lignin &amp; Bark</td>
<td>6.33</td>
</tr>
<tr>
<td>(3) NG CHP for Electricity &amp; Heat</td>
<td>0</td>
</tr>
<tr>
<td>(4) Lignin, Bark &amp; NG CHP for Electricity &amp; Heat</td>
<td>0</td>
</tr>
</tbody>
</table>

2.4.3. Water

We characterize water use in terms of withdrawal – the use and return of water to a local waterbody – and consumption – the permanent or evaporative removal of water. We calculate the off-site water use – that associated with electricity generation and diesel fuel production, and the direct on-site water use.

On-site water use includes process water and water contained in the incoming biomass. Water is recycled within the processes; water leaves the processing facility through the wastewater pretreatment system and in the sugar output stream. On site, a net total of 9.9 kg of water is used per kg of sugar produced, of which 3 kg is returned to the watershed via the wastewater treatment process, and 6.9 of which is consumed.

For off-site water use, based on the grid electricity mix discussed above and the corresponding water withdrawal and consumption associated with coal, natural gas, nuclear and other electricity generation sources (US DOE 2006), we calculate that the water withdrawal and consumption rates corresponding to electricity consumption are 15 gal/kWh_e and 0.157 gal/kWh_e, respectively. For the diesel fuel used in transportation and forestry applications, we use 8 gallons of water per gallon of diesel fuel (Harto et al. 2011).
Table 6 summarizes the lifecycle water withdrawal and consumption. The table shows that the direct water use is 3 liters of water withdrawal and 7 liters of water consumption per kg of fermentable sugar production. While the 7 liters of water consumption comprises essentially the entire lifecycle water consumption, the lifecycle water withdrawal, almost entirely from grid electricity production, is 30 liters per kilogram of fermentable sugar. If on-site combined heat and power is used for electricity production and process heat, and if no additional water is needed for this system, the lifecycle water withdrawal would be 3 liters per kg of fermentable sugar.

Table 6. Lifecycle water withdrawal and consumption for sugar production

<table>
<thead>
<tr>
<th></th>
<th>Withdrawal (L/kg Sugar)</th>
<th>Consumption (L/kg Sugar)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sulphuric acid</td>
<td>0.70</td>
<td>0.0074</td>
</tr>
<tr>
<td>Lime</td>
<td>0.0039</td>
<td>0.016</td>
</tr>
<tr>
<td>Ammonia</td>
<td>0.0040</td>
<td>0.0047</td>
</tr>
<tr>
<td>Forestry</td>
<td>Indirect Water for diesel fuel</td>
<td>0.25</td>
</tr>
<tr>
<td>Biomass transportation</td>
<td>Indirect Water for diesel fuel</td>
<td>0.22</td>
</tr>
<tr>
<td>Chipping off site</td>
<td>Indirect Water for diesel fuel</td>
<td>0.018</td>
</tr>
<tr>
<td>Chipping on site</td>
<td>Indirect Water for electricity</td>
<td>0.034</td>
</tr>
<tr>
<td>Biomass Conversion to Sugar</td>
<td>Direct Water</td>
<td>2.99</td>
</tr>
<tr>
<td></td>
<td>Indirect Water for electricity</td>
<td>26.19</td>
</tr>
<tr>
<td>Total Water Use</td>
<td>30.41</td>
<td>7.22</td>
</tr>
</tbody>
</table>

**2.4.4. Greenhouse gas emissions**
Dominion power reports greenhouse gas emissions of 500 g CO₂/kWh (Dominion 2011b). Dominion’s 28% use of nuclear power results in an average greenhouse gas emission rate that is somewhat lower than the national average of 700 g CO₂e/kWh. Dominion’s reported greenhouse gas emission rate does not include the lifecycle emissions resulting from fuel mining, processing and production.

For diesel fuel, we use a greenhouse gas emissions factor of 90 g CO₂e/MJ (Skone and Gerdes 2005). For natural gas, we use a greenhouse gas emissions factor of 0.075 g CO₂e/Btu (Skone 2011).
Table 7 summarizes the off-site greenhouse gas emissions, which total 223 g CO2e per kg of fermentable sugar.

Table 7. Greenhouse gas emissions for Forestry, Wood Collection, Delivery, and Chemical Inputs

<table>
<thead>
<tr>
<th>Off-site Process</th>
<th>Energy Source</th>
<th>g CO2e/kg Sugar</th>
</tr>
</thead>
<tbody>
<tr>
<td>Forestry - chainsawing</td>
<td>Diesel</td>
<td>2.53</td>
</tr>
<tr>
<td>Forestry - loading</td>
<td>Diesel</td>
<td>40.34</td>
</tr>
<tr>
<td>Forestry - skidding</td>
<td>Diesel</td>
<td>66.07</td>
</tr>
<tr>
<td>Biomass transportation</td>
<td>Diesel</td>
<td>96.09</td>
</tr>
<tr>
<td>Chipping off site</td>
<td>Diesel</td>
<td>7.73</td>
</tr>
<tr>
<td>Sulphuric acid</td>
<td>Electricity (US Grid)</td>
<td>8.77</td>
</tr>
<tr>
<td>Lime</td>
<td>Electricity (US Grid)</td>
<td>0.049</td>
</tr>
<tr>
<td></td>
<td>Natural Gas</td>
<td>0.54</td>
</tr>
<tr>
<td>Ammonia</td>
<td>Electricity (US Grid)</td>
<td>0.30</td>
</tr>
<tr>
<td></td>
<td>Natural Gas</td>
<td>0.78</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td></td>
<td><strong>223</strong></td>
</tr>
</tbody>
</table>

Table 8 summarizes the greenhouse gas emissions from the on-site biomass conversion process. Including the forestry and biomass transportation processes, and the greenhouse gas emissions associated with production of the input chemicals, the lifecycle greenhouse gas emissions are 522 g CO2e and 320 g CO2e per kilogram of fermentable sugar for the grid electricity and CHP scenarios, respectively.

Table 8. Greenhouse gas emissions for on-site biomass conversion to sugar.

<table>
<thead>
<tr>
<th>Energy Supply Scenario</th>
<th>GHG emissions (g CO2e/kg Sugar)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Grid Electricity</td>
</tr>
<tr>
<td>Grid Electricity</td>
<td></td>
</tr>
<tr>
<td>+ Lignin &amp; Bark Heat</td>
<td>290.10</td>
</tr>
<tr>
<td>Lignin, Bark &amp; NG CHP for Electricity &amp; Heat</td>
<td>0</td>
</tr>
</tbody>
</table>
References


Floyd, D. Personal communication, email. April 26, 2011.


