Title: Linking Microbial Funneling to Hydrogenolysis-Based Lignin Depolymerization to Produce 2-Pyrone-4,6-Dicarboxylic Acid from Phenolic Monomers

Authors: Canan Sener[1,2*] (csener@wisc.edu), Jose M. Perez,[1,2] German E. Umana,[1,2] Shamik Misra,[1,2] Yaoping Zang,[1,2] Christos Maravelias,[1,2,3] Steven D. Karlen,[1,2] John Ralph,[1,2] Timothy J. Donohue,[1,2] and **Daniel R. Noguera**[1,2]

Institutions: [1]DOE Great Lakes Bioenergy Research Center, Madison, WI; [2]University of Wisconsin-Madison, Madison, WI; [3]Princeton University, Princeton, NJ

Project Goals: To develop a biomass-to-bioproduct processing chain to produce 2-pyrone-4,6-dicarboxylic acid (PDC) through microbial funneling of the phenolic monomers derived from catalytic depolymerization of lignin.

Abstract Text: When designing viable biorefineries, it is imperative to maximize the value obtained from all the components of lignocellulosic biomass. Lignocellulosic biomass is composed of 70-85 wt% polysaccharides (cellulose and hemicellulose) and 15-30 wt% lignin, a heteropolymer of aromatics. As a bountiful source of renewable carbon for the sustainable production of fuels and chemicals, the polysaccharide fractions have been extensively studied and various processes have been developed to produce valuable liquid fuels and commodity chemicals. The lignin fraction, being difficult to fractionate and process, often is burned for its energetic value to generate process heat and electricity. However, lignin is the largest source of renewable aromatics and, as such, strategies to valorize lignin are required for a bio-refinery to produce aromatic-derived chemicals currently derived from fossil fuels.

“Lignin-first” strategies have been developed to extract value from lignocellulosic biomass in the form of commodity chemicals and liquid fuels from both the lignin and polysaccharide fractions.[1] These strategies liberate the lignin in “native-like” form from the plant cell wall and convert it through catalytic processing[2] or by protection-group chemistry[3] before it can be degraded under the deconstruction processing conditions.

In our previous work, we developed a lignin-to-bioproduct processing chain to produce 2-pyrone-4,6-dicarboxylic acid (PDC) through microbial funneling of the phenolic monomers obtained by catalytic depolymerization of isolated lignin.[4] We used chemical and biological upgrading in tandem to extract greater value from the lignin fraction by converting lignin-derived aromatics to PDC. In this previous work, we first isolated lignin from lignocellulosic biomass under mild reaction conditions using gamma-valerolactone (GVL) and water as the solvent system and dilute sulfuric acid as a catalyst. In this scheme, lignin degradation was minimized during the biomass fractionation by the solvent system, as well as low process temperatures (<120 °C). The technoeconomic analysis of the processing chain showed the cost of lignin isolation to have a large impact on the minimum-selling-price (MSP) of PDC.

In this work, we modified the "lignin-to-bioproduct" processing chain by eliminating the costly biomass fractionation step. In the new scheme, a "biomass-to-bioproduct" processing chain, we combine biomass fractionation and catalytic depolymerization of lignin into a single step, while simultaneously preserving the structure of the polysaccharide fraction. We demonstrate that the lignin fraction of poplar wood can be successfully depolymerized by hydrogenolysis over a Pd/C catalyst into a mixture of monomeric and oligomeric phenolic compounds. Using this strategy, we were able to increase the monomer/oligomer product yield on a per kg of biomass basis. We further show that, using an engineered strain of *Novosphingobium aromaticivorans* DSM12444, this complex mixture of aromatic compounds containing syringyl, guaiacyl, and *p*-hydroxyphenyl aromatics can be upgraded to PDC. Moreover, we show that PDC can be extracted from the culture broth with a simple separation and purification step (*e.g.*, precipitation with sodium chloride). Furthermore, the sugar stream was subjected to enzymatic and microbial digestion to liquid fuels to produce value-added products from both the lignin (phenolics) and polysaccharide fractions.

Combining these improvements over the previous process we demonstrate that the most expensive parts of the lignin-to-bioproduct (PDC) processing chain can be improved by using a lignin-first biomass-to-bioproduct processing chain. Most importantly, we show that tandem processes utilizing both chemical and biological upgrading can significantly improve the upgrading of a complex feedstock such as lignocellulosic biomass.

References/Publications

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