Lignin Valorization by Integrating Chemical Depolymerization and Microbial Funneling

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

Abstract: Lignocellulosic biomass is a bountiful source of renewable carbon for the sustainable production of fuels and chemicals.

Lignocellulosic biomass is composed of 70-85 wt% polysaccharides (cellulose and hemicellulose) and 15-30 wt% lignin, a heteropolymer of aromatic units. Lignin is the largest source of renewable aromatics on the planet; however, it is quite recalcitrant to removal from the polysaccharides and to its own chemical and/or biological upgrading. Maximizing the value obtained from lignocellulosic biomass feedstocks requires production of liquid fuels and commodity chemicals from both the polysaccharide and lignin fractions.

In this work, we have used chemical and biological upgrading in tandem to extract greater value from the lignin fraction by converting it into 2-pyrone-4,6-dicarboxylic acid (PDC). We will show that high-quality lignin can be isolated from lignocellulosic biomass under mild reaction conditions using γ-valerolactone (GVL) and water as the solvent system and dilute sulfuric acid as a catalyst for polysaccharide depolymerization. We demonstrate that the resulting GVL-lignin can be successfully depolymerized by hydrogenolysis over a Pd/C catalyst into a mixture of monomeric and oligomeric phenolic compounds. The product mixture contains compounds with similar chemical structures that are difficult to separate. Indeed, this has been a major bottleneck in obtaining value-added products from lignin. A compelling solution to this problem is the biological funneling of the mixture of aromatic compounds to a single compound. For biological funneling, we use an engineered strain of *Novosphingobium aromaticivorans* DSM12444 to transform the mixture of aromatic compounds containing syringyl, guaiacyl, and *p*-hydroxyphenyl substructures to a single product, PDC. Thus, we show that a complex mixture of lignin hydrogenolysis products can be reduced to a single product that can be extracted from the culture broth with a simple separation and purification step (*e.g.*, precipitation with sodium chloride). Additionally, we show that this strategy is agnostic to the biomass type by successfully applying this strategy to GVL-lignin extracted from hardwoods (poplar and maple) and grasses (switchgrass and sorghum).

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