Integrated chemical and biological catalysis for the valorization of ionic liquid-based biorefinery lignin

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Project Goals: Establish the scientific knowledge and new technologies to transform the maximum amount of carbon available in bioenergy crops into biofuels and bioproducts.

Lignin, a complex and heterogenous polymer composed of phenylpropane moieties, is the most abundant bio-renewable source of aromatics. Due to its heterogeneity and recalcitrance to chemical or enzymatic depolymerization, lignin is currently under-utilized and routinely combusted to generate process heat in the paper and pulp industry. Chemical routes to lignin depolymerization and subsequent valorization are efficient, but can generate toxic chemicals, making the process unsustainable. Enzymatic depolymerization is selective but inefficient at degrading lignin in solid state. We have developed an integrated Chem-Bio route that combines the higher efficiency of chemical lignin depolymerization with the higher selectivity of enzymes to produce targeted value-added chemicals and compounds amenable to biological upgrading. We first studied depolymerization of Pd/ZrP catalyzed hydrodeoxygenation (HDO) of ionic liquid (IL) pretreated lignin from poplar. GC-MS analysis and molecular weight distribution profiling showed the process efficiently cleaved C-O bonds as a function of temperature and time. Based on analysis of lignin content, the process produced high yields (>29.3%) of a lignin oil with minimum char formation (<15%). In general, products obtained after HDO of lignin such as phenols and guaiacols are not biocompatible, so we investigated the potential of highly active and thermostable laccases from Cerrena unicolor to upgrade the HDO products to biocompatible compounds. We observed that laccases could detoxify up to 89.0% non-biocompatible HDO products to such as syringic acid and vanillin with a selectivity of 48.2% and 40.0%, respectively. Thus, our integrated chemical and biological approach enables the conversion of lignin oil into platform molecules.

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