**Extracting Value for Biomass**

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The biorefining of bioresources has evolved over the past two decades from primarily focused on cellulosic ethanol to addressing the challenge of atomic economy whereby the biopolymers of the plant cell wall are efficiently fractionated for fuels, chemicals, materials and power. From this perspective, the challenges of lignin valorization have rapidly evolved with lignin first pretreatments taking a dominant research priority. An overarching hypothesis to lignin first pretreatments is that the matching of amphiphilic characteristics of solvents with those of biomass drives the dissociation of their component molecules resulting in structural disruption of these complex biomaterials. Based on this hypothesis, the fundamental physical properties that underly these effects are under active investigation. We hypothesize that efficient solvent pretreatment is dictated by the preferential solvation of lignin and influenced by (i) dipole:dipole forces, (ii) hydrogen bonding, (iii) hydrophobic solute-solvent interactions, (iv) LCC hydrolysis, and (v) polymer depolymerization, which is investigated utilizing a broad swath of solvent systems, structural determinations before and after pretreatment and finally supported by advanced molecular modelling efforts. Our investigations include, to-date, +80 solvent systems and although, no one single parameter has been successfully in predicting cosolvent based delignification our current data, especially for THF and Cyrene, is clearly supportive of hydrophobic-hydrophilic interactions between lignin surfaces and the solvent. The presentation will summarize our current research studies in lignin first pretreatments.