One-Pot Catalytic Conversion of Organosolv Lignin, Cellulose and of Woody Biomass to Liquid Fuels

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Efficient methodologies for converting biomass solids to liquid fuels would have the potential to reduce dependence on non-renewable petroleum while easing the atmospheric CO2 burden. However, it is a real challenge for chemical catalysis to accomplish facile and quantitative conversion of lignocelluloses to clean liquid fuels, mainly due to the inherent complexity of biomass, resulting in highly complicated product mixtures. Additionally, uncontrolled competing pathways lead to the formation of undesired side products, mainly biochar. Here we report on quantitative catalytic conversions of organosolv lignin, cellulose solids and wood to liquid and gaseous products in a single stage reactor operating at 300-320 °C and 160-220 bar. (UCSB Process) The reaction medium is super-critical methanol (sc-MeOH) and the catalyst, a copper-doped porous metal oxide, is derived from an easily prepared hydrotalcite precursor. The Cu20-PMO catalyzes methanol reforming (CH3OH \( \rightarrow \) 2 H2 + CO) thus the reducing equivalents for all processes originate from the solvent itself. We applied various reaction parameters for organosolv lignin1, cellulose and sawdust substrates2 and the extent of solubilization and product profiles were determined. For example, the reaction time required for solubilization of sawdust was probed in a series of experiments where mixtures of pine sawdust (100 mg), Cu20-PMO (100 mg) and MeOH (3.0 mL) were heated at 320 °C for various time intervals. The extent of wood conversion was determined by rigorous analysis of the solid residues and was found to be 75% within 30 min and essentially quantitative within an hour. We have also observed that the product spectrum, albeit still complex, displays unprecedented selectivity towards lower boiling alcohols. While the gas phase consists of predominantly hydrogen, the major liquid product is a mixture of C2-C6 aliphatic alcohols and methylated derivatives thereof. We postulated that two classes of alcohols can be obtained: HAE (higher alcohols and ethers) and CAE (cyclohexyl alcohols and ethers), the former group being of cellulosic and the later of ligninic origin. We will also discuss holistic approaches that help evaluate complex biomass related transformations, specifically product mixtures originating from reactions with organosolv lignin and sawdust.