COMPUTATIONAL AND EXPERIMENTAL STUDIES OF METAL-DOPED ZEOLITES IN CATALYTIC FAST PYROLYSIS

Seonah Kim[‡], Carrie A. Farberow[‡], Calvin Mukarakate[‡], Mark R. Nimlos[‡], David J. Robichaud[‡], Marc Garcia Borràs[§], Daniel Ruddy[‡], Singfoong Cheah[‡], Jesse E. Hensley[‡], K. N. Houk[§] and Joshua Schaidle[‡]

[‡]National Bioenergy Center, National Renewable Energy Laboratory, Golden, CO, National Renewable Energy Laboratory, Golden, CO

[§]Department of Chemistry and Biochemistry, University of California, Los Angeles, Los Angeles, CA

Email address: Seonah.kim@nrel.gov

Fast pyrolysis of lignocellulosic biomass has been considered a promising thermochemical technology to convert biomass to biofuels and/or biomaterials, because this technique is compatible with current petroleum refinery infrastructure. The yield of pyrolysis oil can easily achieve up to 75 wt% but quality of biomass pyrolysis oils is poor including high viscosity and oxygen contents (35-40 wt%). Our primary goal of catalytic fast pyrolysis will be reducing organic oxygen content and increasing carbon number to a range suitable for gasoline, diesel or jet fuels at the same time through various reactions. Dehydration and dehydrogenation over acidic zeolites are important reaction classes in deoxygenation and coupling reactions during the upgrading of biomass pyrolysis oils. Zeolites are chosen for catalytic fast pyrolysis upgrading due to the effectiveness and economical advantages. We will show improvement of catalyst and process performance by: optimizing transition metal and incorporated zeolites (selectivity) to understand the reasons for improved catalyst reactivity and selectivity and leverage findings in this area. The resulting Brønsted and Lewis acidity upon zeolite modification is assessed and the consequence for the activation barriers are evaluated. Hybrid QM/MM ONIOM model calculations were performed and our computational results are compared against experimental GC-MS characterization of the intermediates formed for both target reactions.