HPC Simulation of Co-Solvent Pretreatment of Biomass Confirmed by Experiment

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Background

- Previous BESC work has shown that reaction of biomass in aqueous tetrahydrofuran (THF) in CELF (Co-solvent Enhanced Lignocellulosic Fractionation) pretreatment is highly effective at delignification and deconstruction of biomass for biofuels an bioproducts. However, the molecular mechanism of these effects is not known.

Approach

- Molecular dynamics simulations were performed of whole cellulose fibers and single cellulose chains in a THF-water co-solvent at various temperatures on the ORNL TITAN supercomputer.
- The simulations were complemented by THF-water experiments on cellulose, measuring its solubilization and imaging the product residue.

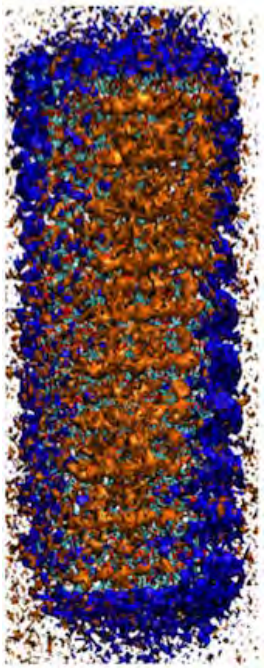
Outcome

- THF and water phase separate on the surface of cellulose fibers, hydrating its glycoside linkages, and a single cellulose chain is preferentially bound by water, possibly enhancing the hydrolysis of cellulose.
- Consistent with the simulation predictions, the experiments show greater cellulose solubilization in the co-solvent than in water-only solutions.

Significance

- The simulations provide a molecular-level mechanism of the enhanced biomass delignification and cellulose hydrolysis due to the equivolume addition of THF in water.
- The use of THF-water in CELF pretreatment has many advantages over other co-solvents, such as improving the solubilization of cellulose and the ability to independently tune the extent of cellulose and lignin solubilization based on reaction conditions.

Cellulose in THF-Water



Supercomputer-derived spatial distributions of THF (orange contours) and water molecules (blue contours) around the cellulose fiber. THF binds to the hydrophobic and water binds to the hydrophilic regions on the fiber surface.

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Mostofian B, Cai CM, Smith MD, Petridis L, Cheng X, Wyman CE, Smith JC, "Local Phase Separation of Co-Solvents Enhances Pretreatment of Biomass for Bioenergy Applications," *JACS* in press (2016).