Combining Deuterium-Labeling and Neutron Scattering to Gain Molecular-Level Insights Relevant to Biomass Deconstruction

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http://cmb.ornl.gov/research/bioenergy/lignocellulose-dynamics

Project Goals: Lignocellulosic biomass comprises the vast majority of biomass on Earth and has the potential to play a major role in generation of renewable biofuels if cost-effective conversion can be achieved. Largely composed of plant cell walls, it is a complex biological composite material that is recalcitrant to the structural deconstruction and enzymatic hydrolysis into sugars that is necessary for fermentation to bioethanol. The Scientific Focus Area in Biofuels is developing "Dynamic Visualization of Lignocellulose Degradation by Integration of Neutron Scattering Imaging and Computer Simulation" for multiple-length scale, real-time imaging of biomass during pretreatment and enzymatic hydrolysis. This is providing fundamental information about the structure and deconstruction of plant cell walls that is needed to drive improvements in the conversion of renewable lignocellulosic biomass to biofuels.

Neutrons have no charge, are highly penetrating, and do not cause radiation damage, allowing measurements of biomass structure under conditions that are relevant to thermochemical and enzymatic deconstruction. Neutrons interact with nuclei making it possible to observe the lighter elements such as hydrogen (H) and deuterium (D) and distinguish these light elements next to heavy ones. Furthermore, the neutron scattering cross-sections of H and D are very different making it possible to selectively highlight different components within a complex system. The scattering length densities of biomolecules such as lipids, proteins and DNA are inherently different which allows structural studies of complex systems or combinations of these molecules by varying the H_2O/D_2O ratio of the solvent. However, the only way to distinguish between components of a system, in which the scattering length densities are all similar, such as with proteins and carbohydrate polymers, is through the use of D-labeling techniques.

We have developed an approach to produce deuterated bacterial cellulose as a model material to investigate different aspects of biomass structure and dynamics that are relevant to biomass deconstruction [1,2]. At the atomic scale, using neutron fiber diffraction it as possible to gain insight in the structural rearrangements that occur in crystalline structure of cellulose microfibrils during conversion from cellulose I to cellulose II. At the mesoscale, small-angle neutron scattering can be used to investigate conformational changes in cellulase enzymes when bound to deuterated bacterial cellulose providing molecular-level details about the inter-domain interactions that cannot be obtained by other means. Model lignocellulose materials composed of individual matrix copolymers (e.g., hemicellulose and lignin) and deuterated bacterial cellulose can be synthesized to obtain new knowledge about matrix copolymer-cellulose interactions and the structural rearrangements that occur during pretreatment. The

aforementioned mesoscale studies take advantage of the contrast between deuterated cellulose and protiated polymers or proteins to distinguish changes in the individual components. The dynamical properties of water when bound to cellulose can be probed using quasi-elastic neutron scattering to further enhance our understanding of water's role and how it partitions in the cellulose supramolecular structure, potentially leading to more efficient pretreatment approaches. The presentation will provide specific details, from the examples above, of how the combination of biodeuteration with neutron scattering can enhance our knowledge of the underlying processes that change biomass morphology during different pretreatment regimes for biofuels production.

References

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