Developing Semi-Synthetic Composite Materials for Investigating Cellulose and Matrix Polymer Interactions During Pretreatment

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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.

Plant cell walls comprise of three main components cellulose, hemicellulose, and lignin. The spatial arrangement of these components with respect to each other and changes among them during pretreatment of biomass are not clear. This is due to the complex and polymeric nature of plant cell walls. In order to understand the interactions of the plant cell wall components during pretreatment, we are developing semi-synthetic composite materials comprising of binary combinations of hemicellulose, lignin, and cellulose. In this study, hemicellulose-cellulose composites were prepared by synthesizing bacterial cellulose from *Acetobacter xylinus* in presence of glucomannan or xyloglucan dissolved in the growth media. Quantitative saccharification was carried out and the ratio of glucomannan to cellulose was 1:3 while the xyloglucan to cellulose was 1:5.

X-ray diffraction showed a significant decrease in the crystal size and crystallinity of cellulose synthesized in presence of hemicelluloses. In situ small angle neutron scattering (SANS) was used to study morphological changes during dilute acid pretreatment. The cellulose in the composite material was deuterated to provide contrast between it and the hemicellulose. SANS measurements were performed in 45% D_2O solvent, the contrast match point for protiated hemicellulose, making it possible to extract size and shape information of cellulose. The samples were heated to 170°C at 5°C/min, held at this temperature for 5 mins, before cooling to 25°C. SANS profiles were recorded at 1 min intervals. Structural changes in cellulose in presence of hemicelluloses (either xyloglucan or glucomannan) were observed during the temperature rise from 120 to 170°C. Analysis of the changes in cellulose structure and morphology supported by X-ray diffraction and compositional analysis will be presented.

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