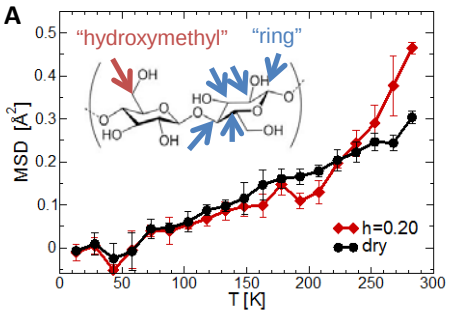
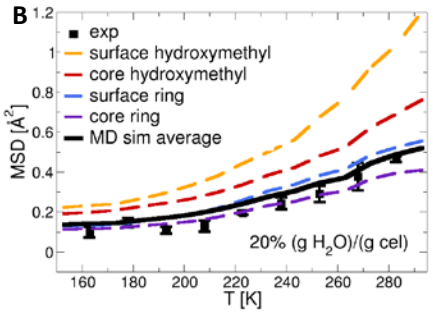
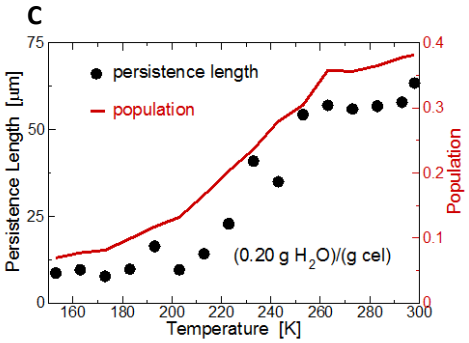
A Counterintuitive Result: Hydrated Cellulose is more Rigid than Dry Cellulose due to Increased Surface Fluctuations

Scientific Achievement

Cellulose hydrated at 20% w/w has increased hydrogen atom fluctuations and surface conformational disorder than drier cellulose hydrated to 5% w/w, yet is more rigid (>240 K).







(A): Experimental mean square displacements (MSD) as a function of temperature at two hydration levels. (**B)**: MSD obtained from simulation, decomposed into contributions from surface and core hydroxymethyl and chain atoms, see inset in A. (**C)**: Temperature dependence of the cellulose rigidity (persistence length, left) and surface disorder (the fraction of glucose monomers that are not in the TG conformation, right).

Significance and Impact

The structural role of cellulose in plant cell walls is directly linked to its rigidity. A detailed description is provided of how hydration-dependent fluctuations (structure) and disorder (dynamics) at the cellulose surface lead to enhancement of cellulose microfibril rigidity (mechanics). This result adds novel insights into the complex action of moisture on plant cell wall structure and strength.

Research Details

- *Neutron scattering experiments, performed at the SNS, probe nanosecond dynamics of cellulose hydrogen atoms;*
- *Molecular dynamics simulations, performed at NERSC, are first compared to experiments by calculating experimental observables (mean square displacements) and then employed to interpret experiments;*

Petridis, L.; O'Neill, H. M.; Johnsen, M.; Fan, B.; Schulz, R.; Mamontov, E.; Maranas, J.; Langan, P.; Smith, J. C. (2014) Hydration Control of the Mechanical and Dynamical Properties of Cellulose. *Biomacromolecules 15*, 4152-4159.

U.S. DEPARTMENT OF ENERGY | Office of Science | OAK RIDGE National Laboratory | NC STATE UNIVERSITY | UNIVERSITY of VIRGINIA | Massachusetts Institute of Technology | UNIVERSITY OF RHODE ISLAND | PENNSTATE | CLSF