Segregation of Organosolv Solvents at the Cellulose Interface Influences Water Dynamics and Cellulose Deconstruction

**Objective:**
- Determine how common water-organosolv cosolvent systems interact with cellulose, which is key to understanding cellulose solubilization in biomass pretreatment for bioenergy and bioproducts

**Approach:**
- The first extensive molecular dynamics simulation study of how four organosolv-water mixtures – acetone, ethanol, tetrahydrofuran (THF) and γ-valerolactone (GVL) – behave on cellulose fiber surfaces.

**Results:**
- We find that qualitative differences exist in the distributions of the cosolvent components at the cellulose surface, with THF-, GVL- (and to a lesser extent) ethanol-water systems phase-separating.
- Median water-cellulose contact lifetimes increase for the cosolvent systems in the order of THF > acetone > ethanol > GVL
- In THF-water, unlike GVL-, acetone- and ethanol-water cosolvents, a significant fraction of surface water is slowed.

**Significance:**
- The surface phase separation and surface-water slowing may be of importance in enhancing reactions breaking down cellulose.

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*(Dynamic Visualization of Lignocellulose Degradation …)*


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*Figure: Two dimensional spatial density profiles of organosolv co-solvents on a cellulose surface. A) acetone, B) ethanol, C) GVL, D) THF. Orange colours indicate the organosolv, cyan/blue water and green cellulose. THF shows the most enhanced local segregation.*