

Understanding the Interactions of Cellulose with Ionic Liquids and Ionic liquid/water binary mixture: A Molecular Dynamics Study

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Project Goals:

Understanding the mechanism of cellulose dissolution and regeneration in ionic liquid.

Biomass pretreatment using ionic liquids has received significant attention over the past 5 years. It has been reported that certain ionic liquids can dissolve cellulose at relatively high loadings, and that the cellulose can be precipitated through the addition of an anti-solvent. In this work, we perform molecular dynamics simulations to study cellulose dissolved in imidazolium-based ionic liquids at high biomass loading (20 wt%). The interactions of the [C2mim][OAc] with the I β cellulose structure at room temperature and the interactions within the cellulose structure at 120 °C were studied. The results show that both cation and anion of [C2mim][OAc] can easily penetrate into the cellulose crystal structure, but that the anion in particular forms strong hydrogen bonds with cellulose. Our results also show that the preferential conformation of the methylhydroxyl group of cellulose solvated in [C2mim][OAc] are in the gauche-trans (gt) conformation, in contrast to the dominant trans-gauche (tg) conformation of the cellulose I β found in water or after pretreatment with ammonia. Because of the gauche-trans (gt) conformation found mainly in the cellulose II crystal structure, we hypothesize that the regenerated cellulose from the similar pretreatment conditions are composed of the cellulose II structure. This hypothesis was verified by XRD experiments. MD simulations were also carried out to study fundamental intermolecular interactions that drive the subsequent regeneration of cellulose in complex mixtures of ionic liquids, water and cellulose. The structural analysis of cellulose with different concentration of ionic/water binary mixtures provides new insight into the molecular driving forces present in this ternary system.

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