Mechanistic Insights and In Silico Identification of Novel Cationic Cores in Ionic Liquids for Cellulose Dissolution

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Cellulose, a highly versatile material, faces challenges in processing due to its limited solubility in common solvents. Ionic liquids (ILs) have been demonstrated high solvating capacities for cellulose.[1] However, the experimental development of ILs with optimal cellulose solubilities remains a time-consuming trial-and-error process. Based on an extensive literature survey, approximately 7000 ILs have been synthesized to date, of which around 330 have been experimentally evaluated for cellulose dissolution. These include 108 unique cations and 83 unique anions. Notably, the majority (~80%) of the evaluated cations are based on imidazolium or ammonium core structures. Given this limited structural diversity, there is a need to explore and identify novel cationic cores using in silico approaches to accelerate the discovery of more effective cellulose-dissolving ILs.

In this study, an electric double layer (EDL) formed by ILs at the cellulose interface was investigated using molecular dynamics (MD) simulation, providing new insights into the underlying mechanism of cellulose dissolution in ILs. This mechanism elucidates the influence of cationic core structures on cellulose dissolution. To identify novel cationic cores in ILs with high cellulose solubility, an automated MD simulation workflow was established to evaluate cellulose solubility in ILs.[2] The accuracy of the MD simulation workflow was widely validated across virous ILs. Employing this automated computational framework, we have screened hundreds of computationally generated novel cationic cores in ILs, produced by a machine learning-based ion generator.[3] Numerous structures exhibiting high cellulose-solvating capabilities were identified, and the screening results corroborate the structure-activity relationship revealed by the proposed dissolution mechanism.

References

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Mengyang Qu received his Ph.D. from the Chinese Academy of Sciences in 2023. After that, he joined the research group of Prof. Kenji Takahashi at Kanazawa University as a Postdoctoral Researcher. He has over seven years of experience in computational chemistry. Currently, his research interest focused on the development of novel ionic liquids for applications in biomass processing and batteries using both computational chemistry and machine learning.