

Molecular Dynamics Insights into Polypropylene and Polyethylene Aggregation in Ionic Liquids: Towards Rational Solvent Design

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Global plastic waste accumulation necessitates innovative valorization pathways towards a circular economy.^{1,2} The European project WASTE2H2³ aims to address this by developing novel ionic liquid (IL)-based catalytic systems for the deconstruction of polyolefinic plastics into clean hydrogen and valuable decarbonized chemicals. Effective polymer dispersion in ILs is critical for efficient catalytic conversion, thus molecular understanding is paramount for rational IL design.

In this work, we investigated the aggregation behavior of polypropylene (PP) and polyethylene (PE) in different ILs using all-atom classical molecular dynamics simulations. To systematically characterize polymer-solvent interactions, we employed radial distribution function analysis to probe local polymer aggregation and solvation environments, and Voronoi domain analysis to quantify instantaneous and average polymer cluster volumes and surfaces. Our simulation approach was validated against two benchmark ILs. In our preliminary experimental tests, one of them exhibited poor PP dispersion, while the other proved to be an effective solvent for PP. The simulations accurately captured these key experimental trends, reproducing the formation of single polymer aggregates and the more dispersed configurations observed using both ILs.

Building on this validated approach, we screened a diverse set of new ILs, systematically varying the alkyl chain length of their cations or anions, to elucidate trends in their ability to promote PP and PE chain separation. The derived molecular insights establish structure-property relationships essential for guiding the rational design and experimental selection of high-performance ILs towards efficient plastic waste valorization.

References

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