Development and Application of an FMO-based Non-Empirical Scheme for χ Parameter Evaluation

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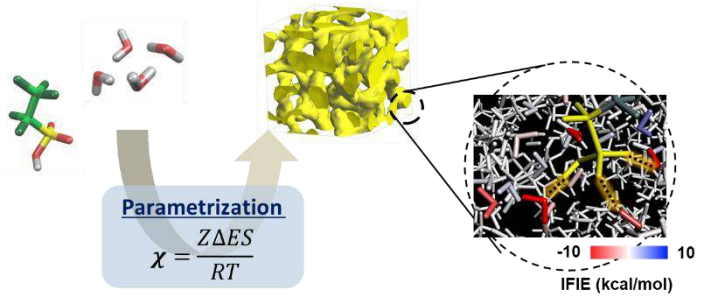
Abstract

Controlling the phase separation structures of molecular assemblies is a crucial for optimizing material properties. Simulation-based predictions have become widely utilized, but all-atom simulations struggle to accurately predict long-term phenomena, and therefore coarse-grained simulations such as Dissipative Particle Dynamics (DPD) are used. Typically, the Flory-Huggins χ parameter is employed to represent interactions between coarse-grained particles, but enhancing the accuracy of χ parameter prediction remains a significant challenge. Approaches for predicting χ parameters from chemical structures include solubility parameters, direct calculations of interaction energies between small molecules, and COSMO methods based on molecular surface charges. Our group has been developing the FMO-based Chi parameter Evaluation Workflow System (FCEWS) [1], which improves calculations based on direct interaction energies by utilizing the Fragment Molecular Orbital (FMO) method [2,3], an efficient quantum chemical calculation approach. This approach specifically targets systems in which microscopic interactions such as polarization and charge transfer dominate phase separation.

In addition to parametrization, we established the FMO-DPD scheme, employing χ parameters computed via the FMO calculation for DPD simulations, and have progressively expanded its application across various systems. Successful correlations with experimental results have been obtained for lipid systems and polymer electrolyte membranes. Ongoing studies include peptides and peptide-like molecules, as well as emulsions containing water-soluble polymers. Furthermore, recent efforts include developing the DPD-based Structure Reverse Mapping System (DSRMS)[4], which assigns atomic details to DPD simulation results for detailed nanoscale analysis using FMO calculations, We are also accelerating parameterization using machine learning[5] and improving parameter evaluation by incorporating many-body configurations. In this presentation, we will summarize our recent progress, applications, and challenges.



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References

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