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Optimizing Molecular Simulation Approaches for Predicting Physicochemical Behaviors in Underground Hydrogen Storage

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Hydrogen has attracted significant attention as a large-scale and long-term energy storage medium for renewable energy. For the realization of underground hydrogen storage (UHS), accurate predictions of physicochemical properties such as interfacial tension and mutual solubility with other substances are essential. However, understanding hydrogen behavior under subsurface conditions remains challenging because reliable high-pressure experimental data are limited and uncertain.

In this study, we propose optimal approaches for predicting hydrogen properties, specifically H_2–hydrocarbon mutual solubility and H_2–water interfacial tension, via molecular simulations.

The reproducibility of mutual solubility in H_2–hydrocarbon systems was evaluated using the Gibbs Ensemble Monte Carlo method, and approaches to improve model accuracy were proposed. The TraPPE-UA Force Field [1,2] was used for hydrocarbons. The intermolecular potential for hydrogen was based on the literature [3] reproducing pure H_2 properties. A modified mixing rule was newly applied to describe the interactions between H_2 and hydrocarbons. Simulations were performed for linear hydrocarbons (C5, C6, C10, and C16) at 323–503 K and 1.7–55 MPa, and agreement with experimental data was assessed using the average relative deviation (ARD). By applying the modified mixing rule, the ARD was reduced to below 2%, demonstrating excellent agreement with experimental values over a wide range of conditions.

For the H_2–water interfacial tension, we demonstrated that the low density of hydrogen leads to significant variations in calculated results depending on the treatment of cutoffs and long-range dispersion interactions. Notably, although long-range dispersion corrections (assuming a homogeneous bulk phase) had a minor effect on interfacial tension, they had a substantial impact on bulk density. Based on these findings, applying the Ewald method proved critical for treating long-range dispersion interactions for H_2-water interfacial systems. We further examined the trade-off between accuracy and computational cost to establish robust guidelines for simulation settings in molecular dynamics simulations of interfacial systems involving hydrogen.

[1]Martin, M. G.; Siepmann, J. I. *J. Phys. Chem. B* **1998**, *102*, 2569−2577.

[2]Martin, M. G.; Siepmann, J. I. *J. Phys. Chem. B* **1999**, *103*, 4508−4517.

[3]J.O. Hirschfelder, C.F. Curtiss, R.B. Bird, *Molecular Theory of Gases and Liquids*, Wiley, New York, 1954.