

先読みシミュレーションセミナー

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大阪大学 附属図書館 理工学図書館 図書館ホール

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**Free energy computations by machine learning-aided molecular dynamics
simulations**

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Abstract:

First principles (FP)-based simulations have become an indispensable method for predicting the thermodynamics and kinetics of homogeneous and interfacial electrochemical reactions. Various methods have been proposed and applied to compute the free energies of molecules and adsorbates, predicting their potential windows and reaction rates. However, due to the significant difficulty in conducting statistical samplings over the entire phase space—which often requires computationally expensive multiple nanosecond-scale molecular dynamics (MD) simulations—most simulations still heavily rely on simple statistical models (e.g., harmonic oscillators), statically optimized quasi-minimum structures, or approximate implicit solvation models. These approximations often make it difficult to judge whether the results are exact theoretical predictions or artificial results specific to the approximated structures intentionally chosen to reproduce experimental observations, especially when examining the effects of electrolytes that fluctuate anharmonically due to thermal motion. Here, I show that machine learning surrogate models can solve this problem. Machine-learned force fields (MLFFs) can accelerate the required nanosecond MD simulations by orders of magnitude. Additionally, subsequent thermodynamic integration from the MLFF to the FP potential energy surface can accurately correct errors of MLFFs, yielding first principles results. Validation calculations on electrochemical reactions in aqueous electrolytes demonstrated that this method can accurately predict the redox potentials of atoms and molecules. Applications to electrolyte-Pt interfacial systems revealed that hydrogen-bond defects play an essential role in the activation of the oxygen reduction reaction on Pt catalysts.