

Fine-Tuning MLIPs for Reactive MD in Catalytic Surface Chemistry

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Machine Learning Interatomic Potentials (MLIPs) combine the computational efficiency of Molecular Dynamics (MD) with the accuracy of density functional theory (DFT). Universal MLIPs, such as M3GNet,^[1] are promising, but they can fail to describe catalytic surface reactivity, particularly bond-breaking and bond-forming events at metal surfaces. Fine-tuning such universal models for specific surface reactivity can lead to more accurate mechanisms. However, conventional MD-based training often fails to sample high-energy transition states, limiting the ability of the model to properly describe the reactive regions of the potential energy surface (PES). We recently introduced our ALPES workflow, combining Active Learning with automated PES exploration in the Amsterdam Modeling Suite (AMS).^[2, 3] ALPES fine-tuning of M3GNet for methanol (CH_3OH) decomposition on Rh(111) shows high-fidelity correlation with DFT energies and forces, with improved description of the PES, including the energy barriers, stable intermediates and transition states. Here, we stress-test ALPES improved models in reactive MD simulations for surface reactions on a range of metal surfaces and reactive molecular systems. Our reactive MD simulations exert external forces in ways inspired by the work of Wang et al.,^[4] pushing the systems beyond equilibrium. Simulations with the fine-tuned M3GNet correctly describe the sequential dehydrogenation upon adsorption.^[5] In contrast, simulations using the universal M3GNet led to C-O bond cleavage upon adsorption, mechanism with a higher energy barrier. These results show that ALPES fine-tuning not only improves the description of PES critical points, but also enables reliable mechanism prediction with reactive MD for catalytic surface chemistry.

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Figures

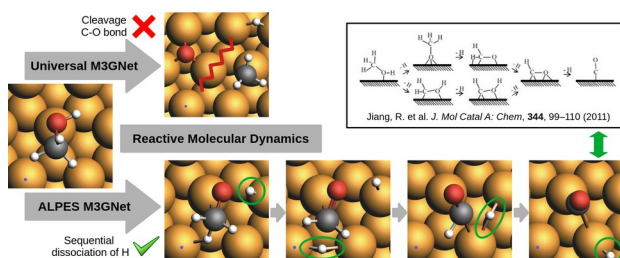


Figure 1. Reactive MD snapshots of methanol decomposition on Rh(111) with universal M3GNet vs fine-tuned ALPES M3GNet models.

References

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