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AB INITIO INFORMED MACHINE LEARNING POTENTIAL FOR TRIBOCHEMISTRY AND MECHANOCHEMISTRY: APPLICATION FOR LUBRICANT ADDITIVES

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ABSTRACT
Mechanochemistry and tribochemistry processes involve multiple physical/chemical interactions induced by extreme conditions including molecular confinement, high temperatures and mechanical stress applied [1]. The simulations of the dynamics of the system when sliding under an imposed external load can reveal the key atomistic mechanisms influencing the tribochemical reactions at the interface, underlying the formation of the friction reducing lubricant tribofilm and the modifications of the activation path for the reactions, due to the concurrent action of shear and load stresses. However, while force fields fall short reproducing the enhanced reactivity arising by quantum effects, simulating these processes by *ab initio* molecular dynamics [2] is severely limited by the size and complexity of the systems of interest and the long-time scale on which relevant events take place. In this work, applying an active learning approach [3] in a careful and innovative way, to a system composed of aromatic molecules on iron, a landmark deep neural network potential has been developed which reproduces the accuracy of *ab initio* interactions at the classical molecular dynamics computational cost and permits to successfully simulate the tribochemical processes occurring at the interface between the lubricant additive molecules and the metal substrate under tribological conditions. The trained neural network potential is shown to accurately reproduce DFT based ab-initio dynamics in the explored tribological conditions, avoiding the shortcoming of reactive force field based approaches. By means of massive molecular dynamics simulations we characterize the tribological properties of self-assembled monolayers (SAM) of gallate molecules sandwiched between iron surfaces. The key mechanisms behind molecular adhesion with the formation of lubricious SAMs were uncovered and correlated to the variation of friction reduction with different molecular geometries. Here we show how machine learning, marrying accuracy with system sizes and long time scales, can pave the route toward a new area in computational tribochemistry.

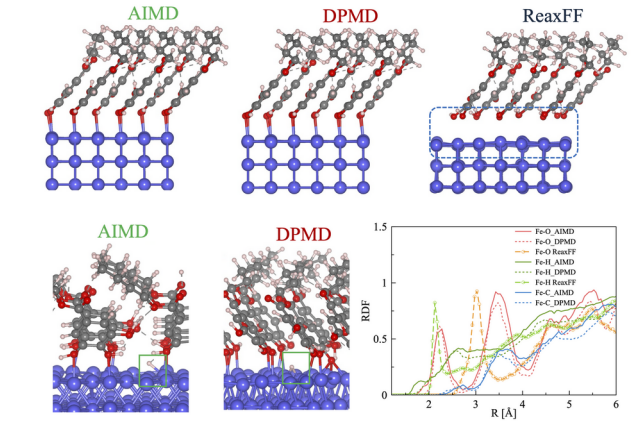

Figure 1 Comparison of relaxed (top) and dynamical (bottom) configurations for butyl gallates between iron surfaces. The radial distribution functions underline the failure of classical force-fields in reproducing the monolayer structure and tribochemistry while sliding under applied normal and shear stresses.

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