MACHINE LEARNING INTERACTION POTENTIALS FOR TRIBOCHEMISTRY: AN APPLICATION TO ALKYL THIOLATE AT SLIDING COPPER INTERFACES

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KEYWORDS
Modelling in tribology; Machine Learning / AI; Physics of Friction; Transition-State Theory

ABSTRACT

Tribological processes occur by the applied stresses modifying the energy barriers for a transition of the system from one state to another. These mechanisms are at the heart of diverse phenomena such as sliding friction (in the Prandtl-Tomlinson model, fluid viscosity (as in the Eyring model) and tribochemistry [1]. In chemistry, such interconversions are described using transition-state theory so that it is appropriate to analyze the effects of stresses on tribological processes using a perturbation method of transition-state theory proposed by Evans and Polanyi [2]. In particular, this approach can quantitatively predict the rate of a normal-stress-induced surface tribological reaction [3] and develop formulae for pressure-dependent shear strengths [4]. However, being able to utilize such strategies to accurately model the rates of tribochemical reactions will require corresponding accurate atomistic simulations. Empirical or reactive force-fields are generally not precise enough to describe tribochemical reactions with the same quantum accuracy achievable with first-principles modelling of normal stress-induced rates [3,5].

To address this issue, we use machine learning (ML) to create a bespoke potential for tribological systems that permits to extend the time-scale and the system-size of the molecular dynamics (MD) simulations with the accuracy of *ab initio* MD. We use the active learning approach implemented in the Smart Configuration Sampling (SCS) code[6], whose general procedure is outlined in Figure 1. This method trains simultaneously several neural networks and refines the original dataset by selecting relevant candidates through ML MD simulations under tribological conditions. Using the ML generated potential, we calculate the velocity, load and temperature dependence of the shear stress of an alkyl thiolate overlayer on copper, a well-known reference system for tribochemical reactions that is also easy to analyse experimentally [3].

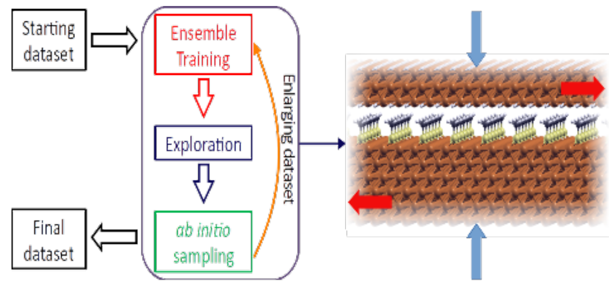

Figure 1 The active learning SCS scheme where the exploration phase is specifically conducted with MLMD simulations of the alkyl thiolate overlayer on copper under tribological conditions

Combining ML MD simulations with the analytical model based on the Evans-Polanyi method [4], we can obtain efficient and precise tribochemical reaction rates for the alkyl thiolate/copper system and better understand the interplay between shear and normal stresses in enhancing tribochemical reactivity.

ACKNOWLEDGMENTS
These results are part of the SLIDE project, which received funding from the European Research Council (ERC) under the European Union's Horizon 2020 research and innovation program. (Grant Agreement No. 865633).

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