Distribution of hydrolysis energies by molecular simulation and machine learning

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To address the issues of the long-term containment of high-activity nuclear waste, several solutions have been studied, among which vitrification, which has been adopted by many countries. The glass containers are projected to be stored in deep clay wells: this raises the question of how the long-term exposure to underground water will alter the glass by hydrolysis, as it will need to withstand both autoirradiation and external degradation from the environment for geological timescales. Hence, this induces research into special formulations to obtain desirable properties in order to efficiently contain the radionuclides, but also into accurately modelling the interplay of the different mechanisms of glass alteration.

Experiments have shown that the alteration is a complex and multistep process. The rate of corrosion is influenced by many factors (temperature, pH, water composition…) but are mainly driven by glass structure and composition[1]. However, realistic long-term experiments and simulations are not tractable: we then endeavor to predict the distribution of the hydrolysis energies instead.

Using simplified models of nuclear glasses, we calculate dissociation and reformation barriers (see Figure 1) using molecular simulations[2], with both classical and ab initio molecular dynamics (MD) coupled with rare event sampling techniques such as the Blue Moon ensemble method[3] or the potential of mean force (PMF) method. These calculations are also used to evidence descriptors which can account for the variations of energetic barriers. Machine learning[4] (ML) methods allow us to show the best correlations between descriptors and the barriers. The end goal of this project is to develop a ML model that can quickly and accurately predict hydrolysis energies using a given starting configuration. Further down the line, this model could be integrated into Monte-Carlo simulations of glass alteration.

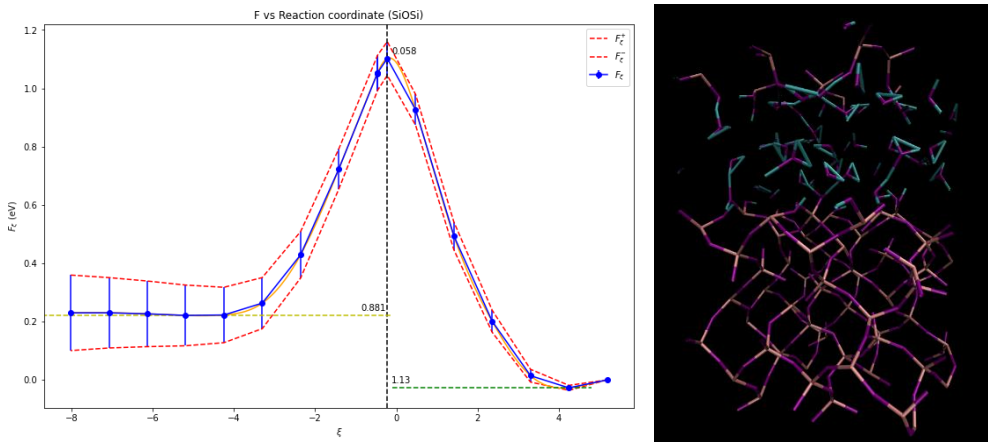

Figure 1. Left: Hydrolysis free energy profile obtained with the Blue Moon ensemble method on a silicate cluster. Right: Water-silicate glass interface.

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