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09:45-10:15 Modelling realistic nanoporous materials at operating conditions

Abstract

Nanoporous materials used in catalysis, sorption, separations are far from perfect, they possess a broad range of heterogeneities in space and time extending over several orders of magnitude. Furthermore, their functional behaviour is largely determined by the conditions in which they do the work. Modelling realistic materials having defects, active sites at true operating conditions of temperature, pressure, etc poses a tremendous challenge. A typical modelling endeavour consists of various steps, with first construction of an atomistic model which is representative for the material. Within this respect, it needs to be emphasised that true crystals exhibit a broad range of spatial heterogeneities, ranging from the nanometer to the micrometer scale. Furthermore, realistic crystals have a finite size, with a certain morphology, which all affect their properties. Given this complexity construction of realistic molecular models for the materials is a challenge on its own. Second, one needs to select a method to determine the forces between the atoms and construct the potential energy surface. This level of theory largely determines the accessible length and time scales of current simulations. In principle one needs to use quantum mechanical methods however in this case length and time scales are restricted to the nanometer scale and molecular dynamics runs extend to a few hundreds of picoseconds, which are much too small to compare with experimental spatiotemporal windows. With classical force fields one can extend the accessible length and time scales, however one loses accuracy compared to the quantum description and the simple analytical potentials are not straightforward transferable to a broader range of thermodynamic conditions. With the fast evolution of machine learning potentials, a window of opportunity is created to simulate more realistic materials at longer length and time scales than currently accessible with accuracy comparable to the underlying quantum mechanical data from which the MLP is derived. Within this contribution, we highlight some of our recent results where we derived MLPs for nanoporous frameworks and applied the methods to describe reactive events in zeolites and to describe flexible behaviour within nanoporous materials. Finally, when having selected an appropriate level of theory to determine the PES, advanced sampling methods need to be used to efficiently explore all relevant regions of configuration space. Within Professor Van Speybroeck's group they have developed a series of methods to describe so-called rare events like reactive events, transport properties, phase transformation of nanoporous materials. Using the plethora of methods sketched above they will give some examples on how to model spatiotemporal processes in realistic nanostructured materials. Spatiotemporal processes refer to processes where the observed dynamics is entangled with the spatial heterogeneities within the material.[1] Examples are taken from catalysis and diffusion within zeolites, phase transformations in metal-organic frameworks.

[1] V Van Speybroeck, S Vandenhaute, AEJ Hoffman, SMJ Rogge, *Trends in Chemistry* 2021, 3 (8), 605-619.

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