From quantum mechanics to machine learning : Bridging length and time scales in modeling nanoporous materials at operating conditions

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Nanoporous materials used in industrial applications like 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.[1] Within this respect, the terminology spatiotemporal behaviour in realistic nanostructured materials has been coined, referring to the entanglement between the dynamics of the materials and its spatial properties.[2] Modelling realistic materials having defects, active sites at true operating conditions of temperature, pressure, etc poses a tremendous challenge.

Within this lecture I will first set the scene on accessible length and time scales in modelling nanostructured materials.[3] Realistic crystal particles used in applications have sizes varying from the 50 nm to the micrometer scale and dynamical phenomena within for example catalysis span a wide range from the (sub)picosecond time scale for molecular vibrations to seconds or hours for macroscopic observations. Currently there exists still a huge length/time scale gap between experimental and theoretical attainable scales. To understand the main bottlenecks in bridging the length-time scale gap, one needs to carefully inspect all steps of a typical modelling exercise. The sequence of a molecular modelling exercise starts with building a realistic atomistic representation for the material under consideration. In a next step one needs a method to evaluate the forces between the atoms and obtain a reliable description of the Potential Energy Surface (PES) after which the desired thermodynamic and kinetic properties can be derived.

The level of theory used to describe the PES largely determines the accessible length and time scales of current simulations. In principle one needs to use quantum mechanical methods to evaluate the interatomic forces. Currently this is mostly done using Density Functional Theory (DFT) in combination with a proper Exchange Correlation functional. The revolution brought by DFT to the field of computational chemistry was tremendous and led to the Nobel Prize in Chemistry in 1998 to Walter Kohn "for his development of the density-functional theory" and John A. Pople "for his development of computational methods in quantum chemistry.[4] Despite these successes, we are currently facing the limits of DFT to further upscale towards longer length/time scales. With DFT, we can typically simulate length scales in the nanometer range while typical DFT based molecular dynamics simulations extend to hundreds of picoseconds. Classical force fields have been used to extend accessible length and time scales, however here one looses the quantum accuracy as the interactions between the atoms are described by simple analytical potentials. Recently the field of Machine Learning Potentials (MLPs) is steadily making its entrance in the field of nanostructured materials.[5] In this case the interatomic interactions are described by a numerical potential derived from underlying quantum mechanical data. MLPs hold the potential to extend the accessible length and time scales while retaining quantum accuracy. Within this contribution, I will show some of our recent results where we derived MLPs for nanoporous frameworks and applied the methods to describe reactive events and diffusion in zeolites and flexible behaviour within nanoporous materials and defective materials reaching to the mesoscale.[6-7] The rise of Machine Learning methods does not make fundamental research on quantum mechanical methods to describe the electronic structure problem for challenging problems unnecessary. A correctly trained MLP will at best be as accurate as the underlying quantum mechanical data on which it was generated. The advantage of MLPs relies in the fact that when having access to very accurate training data set generated with highly accurate quantum mechanical methods, highly accurate MLPs could be trained having an accuracy comparable to the underlying highly accurate training data

The shown examples in this contributions, originate from timely materials/applications in the field of catalysis, sensing and sorption.

References

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