Operando modeling of zeolite catalyzed processes using first principle molecular dynamics approaches

Veronique Van Speybroeck[1]

[1] *Center for Molecular Modeling, Ghent University, Technologiepark 46, 9052 Zwijnaarde, Belgium.*
** Email : Veronique.vanspeybroeck@ugent.be*

Zeolites belong to the most widely investigated inorganic materials and have found widespread use in many industrial catalytic processes. An in depth understanding of zeolite catalysed processes is mandatory for optimizing product selectivities, conversion efficiencies and the lifetime of the catalyst. However catalytic solids used in industrial applications show a high degree of complexity with spatial and temporal heterogeneities at various length and time scales. Modelling can contribute greatly towards understanding the function of the catalyst, provided the catalyst is modelled in a realistic way taking into account true active sites, defects and provided modelling accounts for realistic working conditions, such as the true nature of the feedstock, temperature and pressure effects, moisture, … Operando modelling can certainly not be achieved using one single technique, instead a combination of molecular dynamics techniques, enhanced sampling methods, microkinetic models, machine learning approaches are currently being explored.

Within this talk, I highlight how first principle ab initio molecular dynamics simulations may contribute towards understanding **the catalytic function in zeolites at operating conditions**. Many concepts are illustrated for the methanol-to-hydrocarbons (MTH) process taking place over acidic zeolites, which is one of the prototypical examples in zeolite catalysis where the catalyst selectivity and lifetime are the result of a complex interplay between various factors such as topology, acidity, operating conditions, transport phenomena, etc. The crux in further catalyst optimization is the ability to follow the formation of carbonaceous species during operation, track their reactivity and transport through the pores of the zeolite. Herein we show how advanced operando spectroscopic measurements, first principle modelling and kinetic measurements may yield crucial information on the reactivity and transport within confined environments. The approach followed in this talk, shows how a synergistic modelling experimental approach may unravel essential nanoscale insight into the nature and reactivity of complexes within confined environments at operating conditions which may lead to further optimization of important industrial catalysts.

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