State-of-the-art computer simulations to explore the Fischer-Tropsch mechanism on Cobalt.

The limited availability of easily accessible petroleum has stimulated the search for alternative energy resources. Fischer-Tropsch synthesis plays a critical role in the production of clean fuels from syngas (CO + H₂). Syngas can be obtained through conversion of coal, natural gas or biomass.

Many experimental and theoretical studies have been conducted to elucidate the Fischer-Tropsch mechanism. Despite these studies, there is still a considerable debate in the literature regarding the underlying mechanistic pathways. This debate exists because the intrinsic kinetics of FT catalysts are difficult to probe under typical FT conditions. Experimental techniques often probe the ensemble of elementary reaction steps, rather than each step individually. Theoretical studies like density functional theory (DFT) calculations can be used to investigate the key reaction pathways underlying the FT mechanism. The challenge for a theoretician lies in bridging the gap from an atomic simulation to a full reactor model.

The first step towards a reactor model for FT is constructing a microkinetic model (MKM). Within such a model, the transient behavior of all intermediate species is modeled by solving sets of ordinary differential equations. These ODEs are defined by the large set of elementary reaction steps obtained from DFT. The state-of-the-art is improving the microkinetic model to include various aspects of influence on the overall activity and selectivity of the FT process. The cobalt catalyst is present in the form of nanoparticles. Hence, the morphology of these nanoparticles is important. A good model should include the complex distribution of exposed surfaces on the nanoparticle. Furthermore, lateral interactions like repulsion between adsorbed species may inhibit or promote certain pathways. Transport phenomena may strongly dictate conversion and selectivity in an industrial application and should be included in a reactor model.

Possible projects

1. Construct MKM simulations to investigate the interplay between terrace and step sites on a typical FT catalyst nanoparticle. Where on the nanoparticles is the undesired product methane formed? Does CO dissociate on the steps to provide a monomer species that migrates to the terrace?
2. Investigate the effect of lateral interactions on the surface composition of the catalyst and the elementary reaction steps occurring on this surface. Do lateral interactions increase the speed of monomer formation? Does the inclusion of lateral interactions into the microkinetic model help explain the results observed in experimental studies?
3. Develop a reactor model that bridges the gap between the intrinsic kinetics of MKM and the transport phenomena that occur in a typical reactor. What happens when readsoption of products is taken into account for a plug flow reactor?

Task

During the project you will be made familiar with scientific computing techniques. These include density functional theory simulations and microkinetic simulations. There will be room to learn typical programming languages like C++ and Python. The simulations and model development are aimed towards a full reactor model for FT catalysis.

For further information:

Emiel Hensen (Helix, STW 3.35, Tel 5178, e.j.m.hensen@tue.nl)
Bart Zijlstra (Helix, STW 4.42, Tel 8063, b.zijlstra@tue.nl)
Ivo Filot (Helix, STW 4.28, Tel 4952, j.a.w.filot@tue.nl)