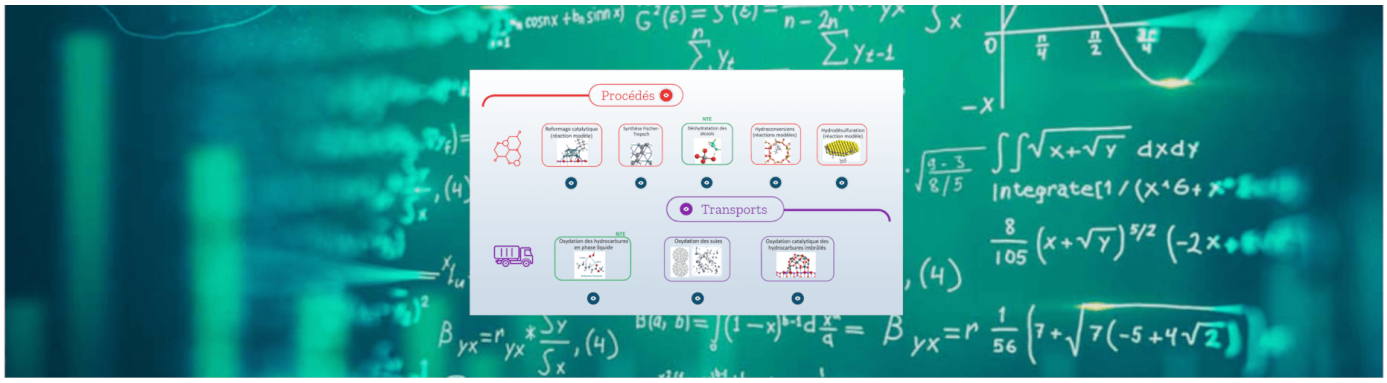


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News

Fundamental Research

Chemical sciences | Catalysis and reaction kinetics | Biosciences and biotechnologies | Biocatalysis

Using chemistry and quantum calculation techniques, researchers at IFPEN have developed predictive kinetic models directly from atomic scale through to reactor scale without the need to parametrize reaction rate constants k. Applied in the field of processes and transport, these models have proved extremely promising for performance prediction.

Multi-scale modeling for chemical processes

In 2014, researchers at IFP Energies nouvelles began working on the development of **a multi-scale modeling methodology for chemical processes** (Figure 1), using *ab initio* quantum calculation techniques. Having used *ab initio* quantum calculation as a tool for determining reaction mechanisms and kinetics in order to construct kinetic models to predict process performance (reaction rates, selectivity), they compared this specific simulation approach, linking the atomic scale and reactor scales, to experimental results contained in the literature or obtained in the context of the research project itself.

In addition to the application objectives, the significant advances achieved by the project relate to the acquisition of knowledge about the physicochemical systems studied, as well as the consolidation of the multi-scale simulation methodology.

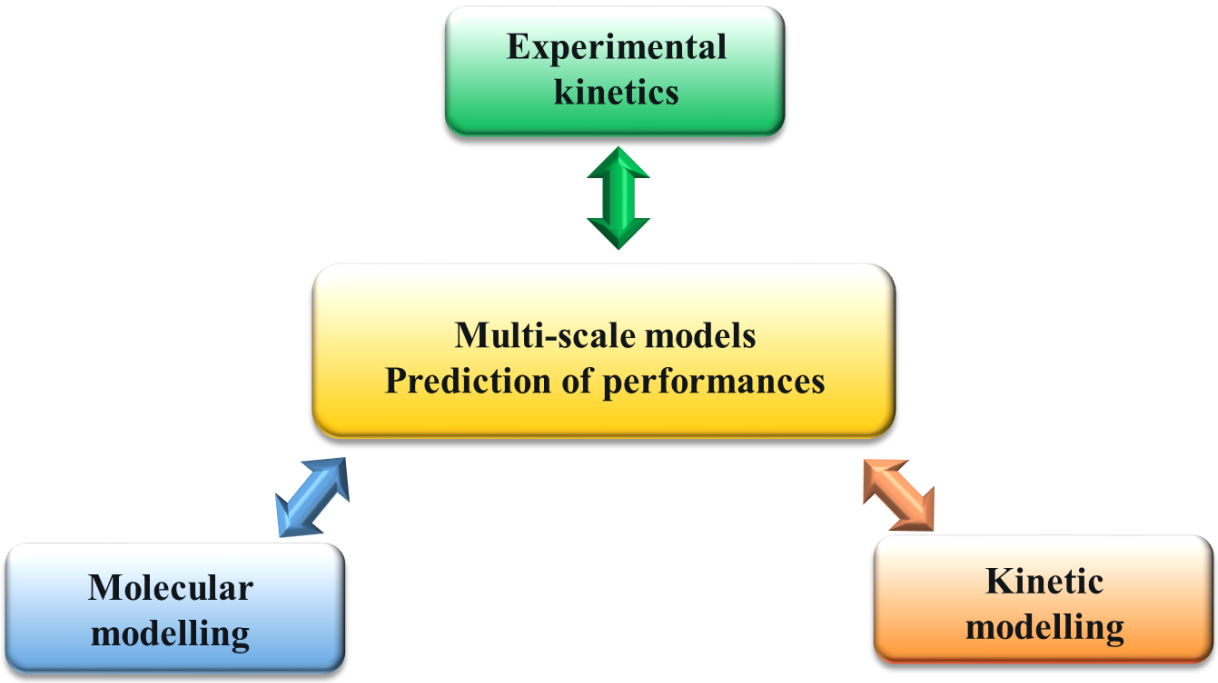


Figure 1-Objective and methodology implemented within the framework of the EYRING project.

What has been the problem with kinetic modeling in industrial processes so far?

The development of predictive kinetic models **is fundamentally important for any industrial process** involving matter conversion reactions. Such models make it possible **to anticipate process performance**, particularly under the effect of a change in operating conditions (temperature, pressures or concentrations, or even a change of catalyst, etc.).

It is common practice for such models to be parametrized using reaction rate constants k that are unknown in advance, the value of which is determined by adjusting rates and selectivities **predicted on the basis of reference experimental data**. When several models are being considered, according to the mechanistic hypotheses advanced, the one giving the best mathematic agreement with the reference data is generally retained. However, this approach makes developing models that are valid for ranges of conditions outside those sampled in advance an arduous task.

Chemistry and quantum calculation at the heart of the multi-scale approach

The strategy adopted to overcome this obstacle was hinged around the construction of kinetic models based **on mechanisms identified by atomic scale modeling** (using quantum chemistry techniques, consisting in solving the Schrödinger equation, and not only on the basis of hypotheses).

Taking things further still, research teams **also identified reaction rate constants k directly using quantum calculation techniques**, without the need for a parametrization step.

As a result, this multi-scale approach provides **a platform for moving directly to the kinetic model on the reactor scale from atomic scale calculations**. Moreover, given their more robust

foundations, the kinetic models thus obtained should theoretically prove better in terms of their predictive capacity.

A few examples of applications in the field of processes and transport

The choice of applications tackled in the project (Figure 2) was dictated by several criteria:
(i) upstream availability of robust ab initio models for the systems studied;
(ii) feasibility of experiments in conditions not limited by diffusion phenomena;
(iii) interest in applied projects. Cases of interest were thus selected in the fields of both processes and transport.

Figure 2. Case studies retained for the EYRING project (NET: New Energy Technologies).

A promising multi-scale modeling approach

IFPEN's researchers tested the multi-scale modeling approach on numerous, chemically varied cases: it has been shown **to be extremely promising for performance prediction**, in the case of model reactions representative of complex environments.

In consolidated cases, the kinetic models developed are likely to be incorporated into simulators integrating the process as a whole, in order to improve their predictive character and robustness with respect to changes in operating conditions. Given the fields concerned, this improved predictability may lead **to an increase in the share of less-polluting fuels and chemical intermediates** - particularly bio-based - and improved energy and environmental performances during their production.

The project did nevertheless **help identify limitations inherent to the current implementation of *ab initio* calculations** and chart the course for future research. These limitations will be the subject of fundamental research efforts, particularly within the context of the FERMI project launched in 2020, which will focus on essential methodological aspects (Machine Learning, reaction rate constants with enhanced accuracy).

Moreover, the project's level of scientific dissemination is extremely high (22 publications, 27 guest lectures, and 22 oral papers submitted, 6 scientific awards, including the 2016 and 2020 Yves Chauvin prizes won by Kim Larmier and Jérôme Rey).

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