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Hot Paper

Interconnected Digital Solutions to Accelerate Modeling of the Reaction Kinetics in Catalysis

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Microkinetic modeling is a widely used tool in the domain of heterogeneous catalysis to gain valuable insights about the fundamental surface kinetics, crucial to designing improved catalysts. The development of a microkinetic model is a multi-step process that demands expertise, a wide variety of experimental techniques, substantial computational resources, and extensive time and effort. In light of these challenges, automation within catalysis research is becoming increasingly important to allow exploration of a broader range of catalytic systems in a shorter timeframe. To this extent, a variety of digital tools and software have been developed to accelerate the development of microkinetic models. This work aims to highlight a selection of these tools that address the various challenges confronting the researchers in this field. These tools address diverse aspects, from the efficient storage of research data that allows easy retrieval and reuse, to the establishment of automated workflows that harness state-of-the-art numerical solvers and algorithms that reduce manual effort. Through the use of automation, these aim to expedite and streamline the development and validation of models for catalytic systems, thereby reducing errors and increasing efficiency.

1. Introduction

In the era of data and information revolution, automation has emerged as the key driving force for technological progress. Advancement in the field of catalysis is especially important considering its multifaceted contributions to the modern world (Figure 1). The wide range of length and time scales involved from catalyst synthesis to the technical reactor and commercial processes, and the increasingly complex data generated due to a variety of interdisciplinary phenomena involved, underscore the need for digital transformation in the catalysis research to expedite the integration of knowledge generated from laboratory-scale experiments to industrial applications.[1] Automation and data-driven learning have already begun to revolutionize the various individual aspects of catalyst research, ranging from establishment of databases that enable efficient data reuse to modeling workflows leveraging advanced machine learning algorithms. These solutions enable researchers to rapidly explore a diverse range of catalysts and their compositions, structures, and operating conditions.[2–9] This, in turn, facilitates the identification of novel catalytic systems and reaction pathways.[10,11] However, the integration of these digital solutions into a more synergistic approach remains unrealized.

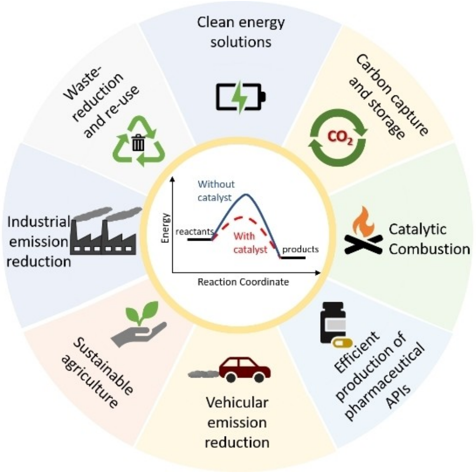

Figure 1. Various applications of catalysis in the modern world.

In general, an essential aspect of catalyst development involves kinetic modeling, because it plays a crucial role in achieving scale-up. It also provides a fundamental understanding of the underlying physical and chemical phenomena such as interactions of the elementary reaction steps with transport processes. This deep understanding serves as the foundation for designing the next generation of catalysts and reactors. In the following sections, we will provide a concise overview of modeling techniques with a focus on microkinetic modeling and discuss the distinct challenges encountered in developing microkinetic models (MKMs). We have identified the following key steps as vital components of MKM development: Efficient archival and easy reuse of experimental data, reaction pathway generation, numerical simulation and validation, and parameter optimization and, we have structured the remainder of this

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article accordingly. This concept article showcases various computational tools and workflows which can help in the acceleration of the development of MKMs and overcome some of its current challenges.

2. Kinetic Modeling in Catalysis

Multiscale modeling approaches are used to bridge atomic-scale models to industrial catalysts leading to knowledge-driven catalyst design. A bottom-up approach to multiscale modeling typically commences with the construction of atomistic models to probe the energetics of elementary reaction steps. These elementary steps are then used in the development of kinetic models, combined with macroscopic heat and mass transfer models.

The development of kinetic models spans a spectrum of complexity, from empirical rate law expressions to more intricate Langmuir-Hinshelwood models and ultimately to detailed microkinetic models. Along this continuum, models become progressively accurate and gain predictive power. More importantly, the kinetic parameters associated with these models acquire deeper physical significance as one transitions from pure regression-based models to first-principle based microkinetic models (MKMs).[12]

Two types of MKMs are predominant: mean-field microkinetic models (MF-MKMs)[13,14] and kinetic Monte Carlo (KMC) simulations.[15–17] MF-MKMs are based on the mean-field approximation, which assumes a uniform distribution of all surface species and active sites at the nanometer scale. To account for the lateral interactions between adsorbates, which have been recognized as important for correctly predicting the catalytic activity, coverage-dependent activation energies are employed. On the other hand, KMC simulations model the spatial distribution of catalytic sites by tracking the positions of adsorbates on a defined-lattice along with detailed lateral interactions. Although KMC simulations are more accurate than MF-MKMs, they are constrained by their large computational demand. Hence, MF-MKMs are more commonly used as a practical tool to study heterogenous catalysis.

A typical MKM development process involves selecting reaction pathways that are likely to be the most important to describe the catalytic transformation of reactants into products. The equilibrium constants for these elementary reactions are determined from the changes in the Gibbs free energy. The corresponding enthalpy and entropy changes are obtained from experimental data or estimated through ab-initio density-functional theory (DFT) calculations, the unity bond index-quadratic exponential potential (UBI-QEP) method, linear scaling relations, or Brønsted-Evans-Polanyi (BEP) relations.[18–21] The

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Hendrik Goßler obtained his PhD in Chemistry from the Karlsruhe Institute of Technology in 2019 under the supervision of Prof. Olaf Deutschmann. His research focused on numerical optimization of combustion processes coupled with detailed reaction mechanisms. Currently, he is engaged in digitalization efforts, concentrating on developing software for research data management and workflow automation. Serving as the principal architect of the CaRMeN and Adacta software, Hendrik has played a key role in their development. Additionally, he holds the position of CEO and co-founder at omegadot, a company providing specialized software solutions and consulting services in the field of chemical technology.

Olaf Deutschmann holds the Chair in Chemical Technology at Karlsruhe Institute of Technology. He studied physics and chemistry in Magdeburg, Berlin, and Heidelberg, receiving his PhD in 1996 and venia legendi in 2001. Before coming to Karlsruhe in 2003, he also worked at the University of Minnesota and Los Alamos National Laboratory. His work focuses on the development of climate- and environmentally-friendly chemical technologies and novel digitalization tools. Today's research topics include carbon capture, C-free chemical energy carriers, emission control, fuel and electrolysis cells, and circular economy. The software packages DETCHEM, CaRMeN, and Adacta were developed under his guidance.

rate constants are expressed in terms of Arrhenius parameters that is, the pre-exponential factor (or sticking coefficients for adsorption reactions), the activation energy, and the temperature-dependency parameter.

One of the most significant challenges in MKM development lies in accurately estimating these parameters for all the elementary steps within the MKM.[22] DFT-based methods are very successful in providing reaction energetics in heterogenous catalysis,[23] although are limited by their high computational cost for larger systems. Additionally, commonly used DFT functionals such as the generalized gradient approximation (GGA) suffer from issues such as the self-interaction error (SIE) and the inadequate capture of van der Waals (vdW) interactions.[24] Therefore, hybrid functionals and vdW-inclusive functionals are used to mitigate SIE and incorporate long-range interactions in DFT calculations. The emergence of new DFT functionals, accelerated with the use of machine learning and linear scaling DFT methods are gradually enabling their use for larger systems.

In addition to selecting a suitable electronic structure method for energy calculations, it is also important how the catalyst particles and the support are approximated. While the periodic slab model is suitable for larger nanoparticles (>5 nm), more precise models are required for smaller nanoparticles. This is further complicated by the increasing evidence in the literature that the catalytic support can also influence the catalytic activity due to an increased charge transfer between the clusters and the support, and the formation of metal-support interfaces which could act as active sites for certain reactions.[25] Cluster-support interactions in DFT calculations are currently not carried out on a large scale due to their computational expense. It is also well-known that DFT-derived energy barriers have an inherent error between 0.1 to 0.2 eV.[26] Moreover, DFT-calculations are conducted on model surfaces, which do not account for dynamic changes in the catalytic surface during the operating conditions and in the presence of intermediates. Consequently, it is common practice to adjust the parameters from DFT to match the MKM predictions to experimental observations.[27] However, the conventional approach to manually elucidate the reaction pathways and the subsequent kinetic parameter calculation and optimization is laborious and time-consuming, and therefore offers many opportunities for automation.

3. Data Archival and Reuse

As was emphasized in the GeCats whitepaper,[28] the fundamental requirements for achieving digital catalysis involve the establishment of digital archives that adhere to standardized and FAIR-complaint data storage, along with the development of digital workflows facilitating data exchange and evaluation. To this extent, Electronic Laboratory Notebooks (ELNs) are being developed to meet the specific requirements of the catalysis community to efficiently document, archive and extract information across various stages of catalysis research, especially the data pertaining to catalyst-synthesis and characterization.[29] A recent paper by Marshall et al.[30] introduced the AC/CatLab data archive as a data management strategy that prioritizes automation while integrating ELN functionalities with data archival processes. It is imperative that the data is accompanied by rich metadata that assures data provenance and reproducibility. To address this need, the software Adacta has been developed to enable efficient data archiving by allowing users to create digital twins of their experimental setups, and subsequently tracing the time-stamped measurement data to the specific devices used for measurement.[31] This approach allows to trace the history of the experimental setup in a timeline visualization, enabling even those not directly involved in the data generation to access and extract information, related to the equipment employed or the samples in use during specific time periods. Adacta also provides other visualization features to quickly observe trends in the measured data.

4. Automated Mechanism Generation

The open-source software Reaction Mechanism Generator (RMG)[32,33] can automatically predict reaction pathways and their associated rate constants in MKMs (Figure 2). It relies on a flux-based algorithm to identify important species and reaction pathways combined with thermochemical parameter estimation using thermochemistry databases for Ni and Pt, that are augmented by scaling relations for other metals. Beyond the generation of mechanisms, RMG also provides features for model analysis incorporating local and global uncertainty analysis including the ability to track correlated uncertainties in model input parameters.

Studies by Kreitz et al. showcase the application of RMG in automatic mechanism generation for CO_2 hydrogenation over Ni(111) and for catalytic conversion of exhaust gases from gasoline engines on Pt(111).[34,35] A more recent study demonstrated a universal framework for automated construction of reaction mechanisms, with inherent capability of DFT-con-

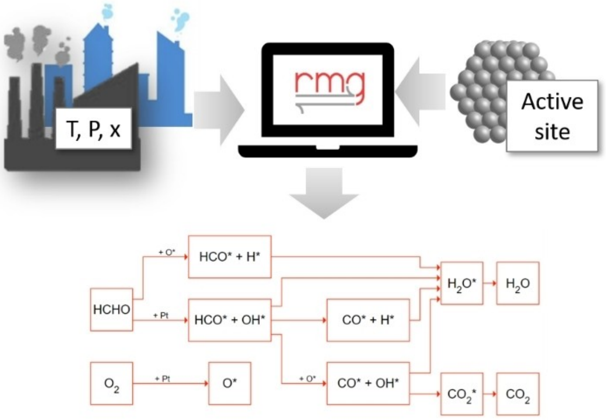

Figure 2. Schematic representation of the Reaction Mechanism Generator for prediction of a reaction scheme.

strained model parameter optimization.[36] This is achieved by sampling an ensemble of microkinetic models within the correlated uncertainty space in the predicted energies of the BEEF-vdW functional and choosing the final model as the one that is able to best reproduce the experimental results.

5. Tools for Numerical Simulation

Simulating reactive flows involves the numerical solution of a set of governing equations for a given reactor configuration to calculate the reaction rates, species concentrations and/or surface coverages. Various MKM solvers are available, each offering its own set of advantages and limitations.[37] Notable options include Chemkin,[38] OpenMKM,[37] DETCHEM,[39] Cantera,[40] CatMAP,[41] CATKINAS,[42] CatalyticFOAM[43] and more.[44–46] Chemkin pioneered the development of simulation codes for gas-phase kinetics which was later extended to support surface kinetics.[14,38] Cantera is a widely-used open-access simulation toolkit for mainly homogenous reactions and can be easily accessed through C++, Python and MATLAB interfaces.[40] OpenMKM toolkit,[37] built on top of Cantera,[40] targets the heterogenous catalysis applications incorporating batch, continuous stirred tank reactor (CSTR), and plug flow reactor (PFR) codes, integrated with pMuTT[47] for thermochemistry input file generation and RenView[48] for reaction path analysis.

DETCHEM (DETailed CHEMistry),[39] a program package written in FORTRAN is specifically developed for the simulation of reacting flows, in particular heterogenous systems. It offers a diverse array of modules tailored for computing the transport and kinetic properties of species participating in both gas-phase and surface reaction mechanisms. It supports several reactor configurations, including batch reactors, CSTRs, PFRs, stagnation flow reactors, packed bed reactors, catalytic monoliths, and models designed for simulating high-temperature fuel and electrolysis cells. DETCHEM also accommodates user-defined expressions for calculating reaction rates, in addition to microkinetic models defined in terms of Arrhenius equations. Its versatility enables its usage in setting up workflows designed to standardize and accelerate the development of microkinetic models.

6. Automated Simulation and Visualization

CaRMeN (Catalytic Reaction Mechanism Network) is a software that can take advantage of metadata-enriched archived experimental data in conjunction with an MKM solver in order to automate the simulation workflows.[49,50] The architecture of CaRMeN is adaptable to any modeling software by configuring an appropriate simulation driver, making it a versatile tool for kinetics studies. These simulation drivers function as interfaces that translate the data formats between CaRMeN and the chosen modeling software. CaRMeN consists of several simulation drivers for a variety of reactor configurations. Many research projects have utilized its capabilities to execute and compare multiple simulation models against the experimental data measured at matching operating conditions.[51–54] It markedly expedites workflows by automating the labor-intensive process of setting up input files required for multiple simulations and postprocessing outputs. At its core, CaRMeN operates by deconstructing a simulation input into manageable subsets or fragments, each of which can be independently accessed and manipulated. These fragments can be thought of as small building blocks that can be reused and combined with other fragments. This modular approach allows users to dynamically assemble these individual subsets within the user interface to initialize multiple complete simulation configurations (Figure 3).

A complete simulation configuration would mean that all information about the reactor type and dimensions, operating conditions, feed compositions, thermodynamic data, transport data, appropriate chemical model, and corresponding experimental data is provided. Consequently, when the need arises to set up multiple simulations with several parameters remaining constant, and the focus is on studying the impact of changes in one or more specific parameters, CaRMeN enables the reuse of the subset containing the constant parameters. This eliminates the redundancy of rewriting the same parameters for each individual simulation, streamlining the setup process. The multiple simulations are then executed in parallel and the outputs are postprocessed and subsequently visualized in the user interface in an automated manner. To summarize, CaRMeN can facilitate in reducing effort and errors in the iterative steps involved in developing microkinetic models.

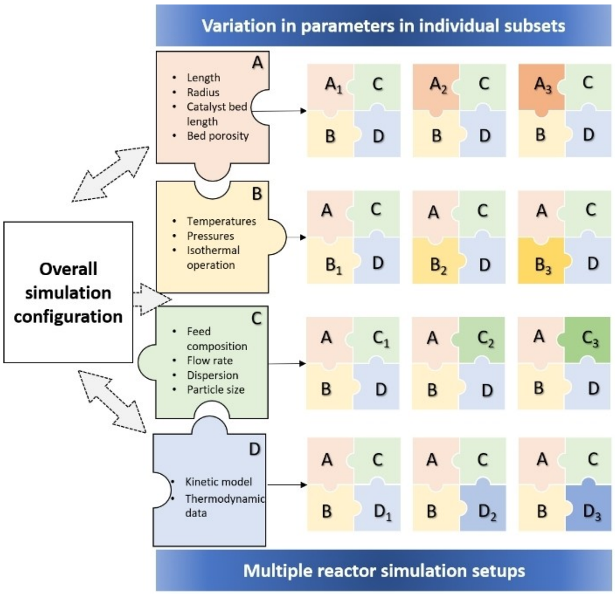

Figure 3. Modular approach followed in the CaRMeN tool to generate multiple simulations.

7. Automated Parameter Optimization

Workflows are also needed that can assist with fast optimization of the kinetic parameters within the bounds of their estimated uncertainties. Furthermore, very often researchers need to study the effect of slight modifications in the known catalytic systems such as change in the support material, or addition of promoters. Such changes are first studied experimentally on a laboratory scale and then models are used for scale-up or to gain deeper insights on the underlying processes. In such cases, it is not practical to build a complete microkinetic model from scratch involving expensive DFT computations to ascertain the kinetic parameters. Instead, existing microkinetic models that are applicable for other similar catalytic systems can be fine-tuned, to align them with the experimental data corresponding to the modified system. The high dimensionality of the microkinetic models make it unfeasible to individually set up variations of the initial microkinetic model in order to run simulations. Usage of optimization algorithms are therefore the norm, with a suitable objective function chosen based on the species of interest, range of values, and initial agreement between the experimental and measured quantities. Based on the task at hand and the uncertainties associated with measured data, it may be necessary to use adjustable weights or use logarithmic values of species concentrations in the objective function.

Many literature studies showcase the use of optimization algorithms to optimize global kinetics models wherein the number of kinetic parameters to be tuned are less.[55–57] In the case of microkinetic models, the calculation of gradients of complex governing equations with respect to the kinetic parameters is non-trivial. Gradient-free global optimization algorithms offer a generalized methodology for optimizing these microkinetic parameters irrespective of the reactor configuration.[58,59] Our previous paper showcases a proof-of-concept for the same using the global optimization algorithm Basin-Hopping[60] to optimize high-dimensional microkinetic models for methanation of CO over Ni and methane oxidation over Pd catalysts.[61] Basin Hopping is a stochastic algorithm that searches for a potential global minimum by hopping within the parameter search space as determined by a user-defined step-size value. Once it has made a hop, it attempts to find a local minimum by leveraging a local-search technique such as the Nelder-Mead[62] method. Each new local solution is then compared with the previously best solution, and is either accepted or rejected based on the Metropolis criterion.[63] Since these methods are gradient-free and work well for high dimensional cases, they are quite useful in setting up automated workflows to optimize kinetic models, incorporating any kind of reactor configuration requiring the researcher to only provide appropriate input files for the chosen modeling software as shown in Figure 4.

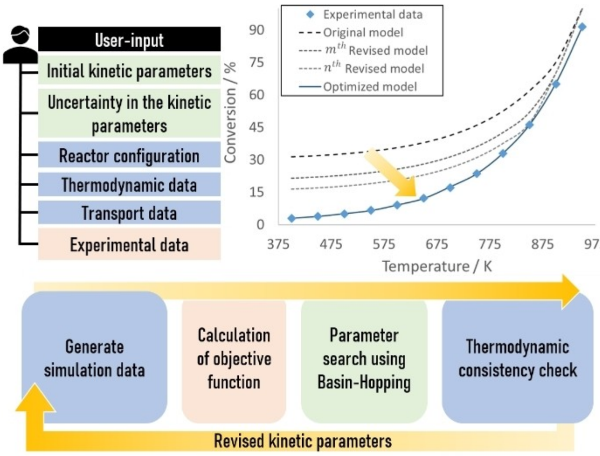

Figure 4. Steps involved in the optimization of kinetic models using the Basin-Hopping algorithm.

It is important to emphasize that the primary objective of kinetic modeling is to obtain meaningful insights about the underlying reaction mechanism. Consequently, techniques such as sensitivity analysis[64,65] and reaction path analysis[66] are employed to identify which parameters and the reaction steps have the most significant impact on the predicted outputs. Campbell's degree of rate control (DRC)[67] method is a highly useful tool to analyze multistep reaction mechanisms to identify the rate controlling transition states and intermediates and can be easily implemented through the open-source code CatMAP.[41] Additionally, both the DETCHEM simulation package[39] and the OpenMKM[37] tool feature modules for sensitivity analysis and reaction path analysis.

8. Summary

The field of microkinetic modeling for heterogenous catalytic systems holds potential for advancement through automation, as it can enhance the knowledge-driven design of catalysts. In this work, we have highlighted a range of tools and software solutions that can contribute to achieving this objective. An overview of how these tools may work together is depicted in Figure 5. The overall workflow of microkinetic modeling involves several key steps, commencing from the generation of an appropriate reaction scheme, followed by the estimation of the kinetic parameters for each reaction, simulation of the reactive system with thermodynamics consideration, validation against experimental data, and optimization of the kinetic parameters within their uncertainties to match the experimental data. The various tools showcased in this work are poised to expedite these steps by automating a variety of tasks that were traditionally performed manually.

The use of data archival tools like Adacta and ELNs ensures data provenance and storage of data in a manner that can be reused long after its generation. The automated mechanism generator RMG not only reduces the effort involved in MKM development but also mitigates errors stemming from personal bias in selecting the reaction steps for consideration. Automated simulation workflows facilitate the execution of multiple models, validation against experimental data and visualization of results. In cases where mismatches with the experimental data occur, minor adjustments to the initial model can be

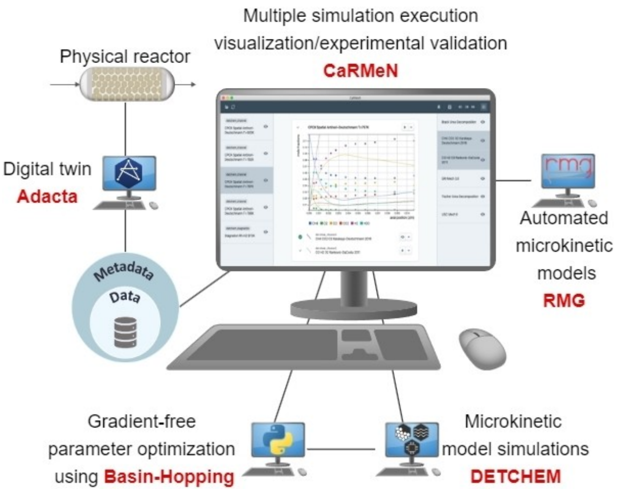

Figure 5. Overview of the various software and tools and their applications during the model development process. (*"Python" and the Python logos are trademarks or registered trademarks of the Python Software Foundation, used by the authors with permission from the Foundation.*)

performed using global optimization algorithms such as Basin-Hopping that are gradient-free and can be configured with appropriate objective function incorporating suitable solvers for running simulations. Consequently, the utilization of automated workflows can accelerate microkinetic modeling and contribute to the development of improved catalysts. However, a more widespread use of automation to enhance catalyst design also entails strengthening the research data infrastructure from the ground up, with a focus on ensuring that the data generated in all aspects of catalysis research are Findable, Accessible, Interoperable and Reusable (FAIR).[68] Collaborative initiatives such as NFDI4Cat[69] in conjunction with other National Research Data Infrastructure (NFDI)[70] consortia like DAPHNE4NFDI[71] and others, are diligently working to establish the necessary infrastructures for the digitalization of catalysis research and related disciplines. These initiatives aim to foster a culture of data sharing and collaboration, further advancing the field of catalysis.

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Conflict of Interests

The authors declare no conflict of interest.

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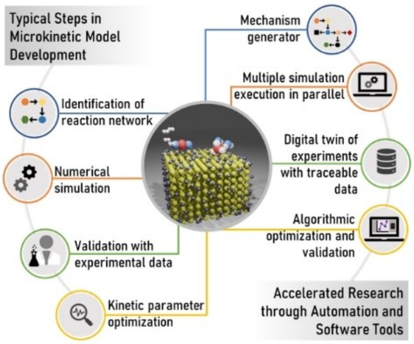
CONCEPT

Microkinetic modeling, typically a time and labor-intensive process, can be significantly expedited through the application of automated software and digital tools. This concept article showcases the utilization of these digital solutions in accelerating the various key steps involved in the development of a kinetic model from identification of reactions and kinetic parameters to subsequent simulation and validation against experimental data.



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Interconnected Digital Solutions to Accelerate Modeling of the Reaction Kinetics in Catalysis