PhD project proposals to the Einstein Center of Catalysis (EC^2) for a PhD start in Oct. 2022

Title of Project: **New design-of-experiment strategies for efficient materials discovery in heterogeneous catalysis**

Co-supervisor 1: **Dr. Frank Rosowski**
BasCat, TU Berlin

Co-supervisor 2: **Prof. Dr.-Ing. Jens-Uwe Repke**
Dynamik & Betrieb techn. Anlagen, TU Berlin

Abstract:

Research in heterogeneous catalysis requires a systematic planning to maximize the information derived from a limited number of experiments. Efficient search strategies rely on the so-called Design-of-experiments (DOE). Aim of the proposed work is the development of new methods for the efficient Design-of-Experiment for (A) improved catalyst materials as well as (B) optimal reaction conditions. The proposed DOEs will combine data-driven modelling with micro-kinetic approaches and computational chemistry to incorporate pre-existing chemical knowledge into the planning of experiments. The resulting model-guided search strategies will not only improve the efficiency of catalytic experiments, but provide also data of higher quality and relevance for both research and practical applications.

Extended description version of the project:

Title of Project: **New design-of-experiment strategies for efficient materials discovery in heterogeneous catalysis**

Co-supervisor 1: **Dr. Frank Rosowski**
BasCat, TU Berlin

Co-supervisor 2: **Prof. Dr.-Ing. Jens-Uwe Repke**
Dynamik & Betrieb techn. Anlagen, TU Berlin

1. Overall goal of the project

The proposed work will develop new methods for the efficient Design-of-Experiment (DOE) in catalysis research. Two different types of experimental planning are addressed (A) improved catalyst materials as well as (B) optimal reaction conditions. The proposed DOEs will combine data-driven modelling with micro-kinetic approaches and computational chemistry to incorporate pre-existing chemical knowledge into the planning of experiments. The resulting model-guided search strategies will not only improve the efficiency of catalytic experiments, but provide also data of higher quality and relevance for both research and practical applications.

2. State of the art

Developing new improved catalytic systems for coupled chemo-catalytic reactions faces significant challenges due to inherently high system complexity. Planning experiments that deliver a maximum of information at minimum cost can be guided by appropriate search strategies. So-called design-of-experiments (DOE) are formalized ways to formulate these search strategies.

Common DOEs are based on statistical considerations. However, heterogeneous catalysis and the interaction between different active sites are governed by a multitude of complex and often nonlinear relations between material composition, structure, surface dynamics, chemical kinetics and transport processes. Literature, prior experience and experimental data exist for many reactions and materials of interest. This information can be used to focus the experimental effort. Including domain-specific knowledge is essential in order to the DOE on regions of interest and highest gain in knowledge in the underlying multi-dimensional search spaces.

The Repke group has built up expertise in the generation and optimization of data driven models [1, 2], experimental approaches to study catalytic systems and the analysis of coupled chemical reactions (EC^2 project “Fundamental experimental investigations on mass transfer between different catalytic centers situated in porous matrices”). Further experience includes data driven models for optimization [3] and the online model-based optimal experimental design [1]. Recent projects pioneer the use of machine learning in chemical engineering (DFG SPP 2331, Co-initiated by Prof. Repke).

BasCat develops catalytic systems for the selective conversion of syngas ($CO + H_2$) into valuable chemical feedstocks, e.g. ethanol. Related catalysts represent prime examples of complex systems with coupled active sites [4]. Different data-driven workflows were established [5, 6]. They can connect information from structure databases with DFT and machine learning to guide the identification of improved catalysts. Recently, a large and consistent data set of several thousand

data points measured in syngas conversion was build, including very diverse catalyst compositions and detailed kinetic measurements. This unique data set will form the starting point of the proposed work.

[1] Chemie Ingenieur Technik 92, 2020, 1247-1247. [2] Computer Aided Chemical Engineering 48, 2020, 1189-1194. [3] Computer Aided Chemical Engineering 47, 2019, 311-316. [4] ACS Catalysis 7(11), 2021, 4047-4060. [5] Nature Communications 10, 2019, article # 441. [6] arXiv:2102.08269 [cond-mat.mtrl-sci].

3. Specific aims and workplan

Main objective of the proposed project is the development of new efficient DOE strategies for the exploration of complex systems in heterogeneous catalysis. Using the existing data set on syngas conversion, new search strategies for (A) better catalyst materials as well as (B) optimized reaction parameters, will be aimed for. Targeted experiments will help to further increase the quality of the data, and to validate experimentally proposed correlations and improved catalysts.

To reduce the high dimensionality of the search area for (A) optimized reaction conditions, and thereby reduce the experimental effort, data driven modeling (“black box”) will be employed. It will be used to predict online (during measurements) step-by-step the next best set of parameters in terms of measurement conditions (e.g. GHSV, feed composition, temperature, pressure) that provides the highest possible gain in information. A black box model will be pre-trained with the existing data set in terms of a supervised machine learning. Complementary, microkinetic models (“white box”) of increasing complexity will be developed to describe the kinetic data of different catalysts based on the principles of surface catalysis. Both types of models will be compared in terms of efficiency, performance and prediction quality. Subsequently, hybrid models (“grey box”) will be designed to enable a computationally feasible online prediction of optimal sequential DOEs.

Different challenges are faced by DOEs that help (B) to explore the parameters space in terms of optimal catalyst elemental compositions and phase combinations. Again, data driven models will be developed to identify promising candidates for testing. These models will be then stepwise augmented with existing chemical information, i.e. crystal structures, preferred surface geometries as well as estimated adsorption energies for adsorbed reactive species based on property databases and existing DFT calculations.

The developed DOE approaches will be tested and validated experimentally using existing parallel-reactor-technology for high-pressure syngas-conversion under industrially relevant conditions operated at BasCat.

Collaborations within UniSysCat include the groups of Prof. A. Thomas (catalyst synthesis), Prof. R. Schomäcker (reactor design), Dr. S. Matera (multi scale modeling) and Prof. F. Hess (DFT). On a national level, close interaction with the data science community in catalysis and materials development (NFDI4cat, FAIRMAT) will open up new avenues to efficient re-use of data.