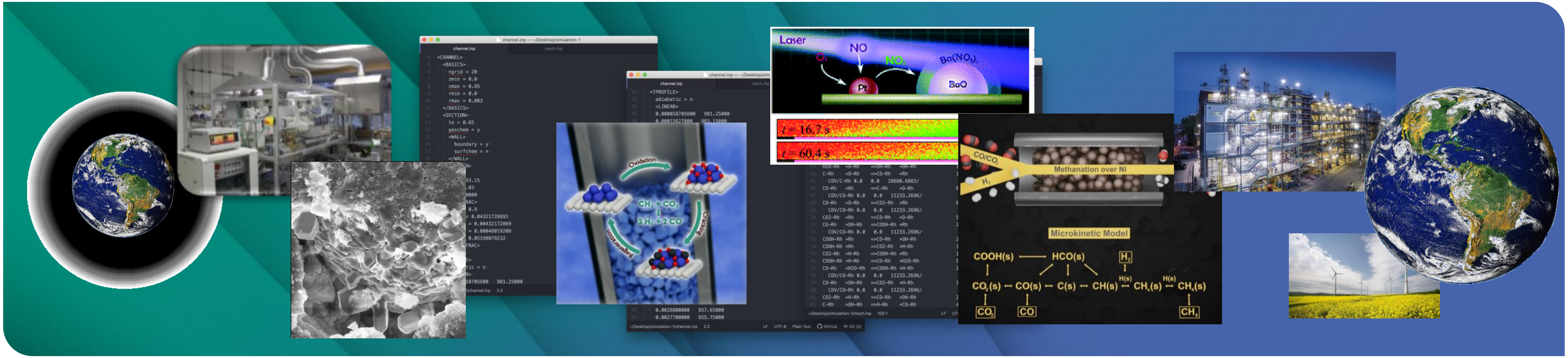
Annual Meeting on Reaction Engineering 2023
15 - 17 May 2023
DECHEMA-Haus, Frankfurt am Main

Digitalization in Catalysis and Reaction Engineering: More than just a Buzzword?

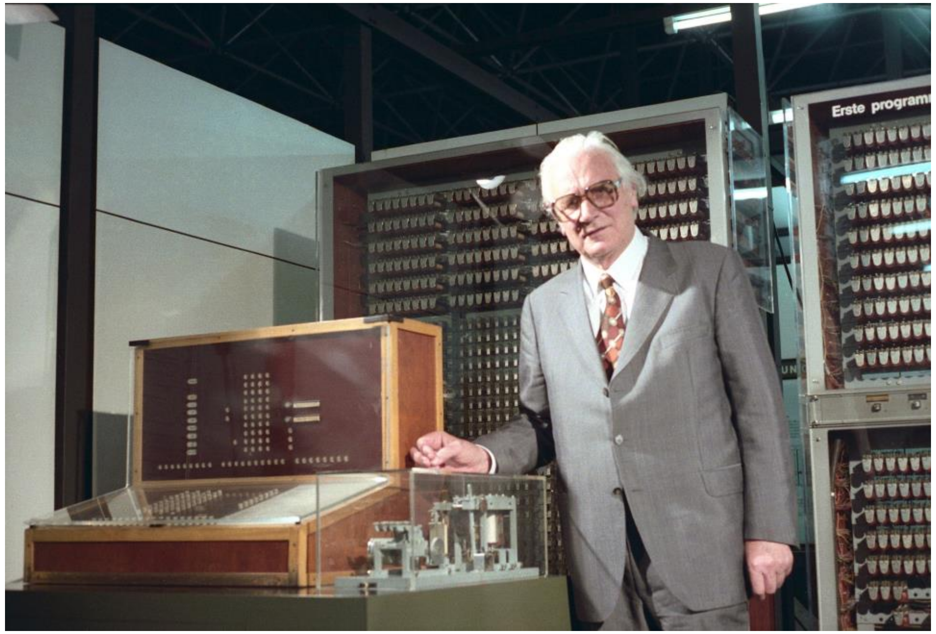
Olaf Deutschmann, KIT, Germany

Stephan A. Schunk, hte GmbH, BASF SE, Germany



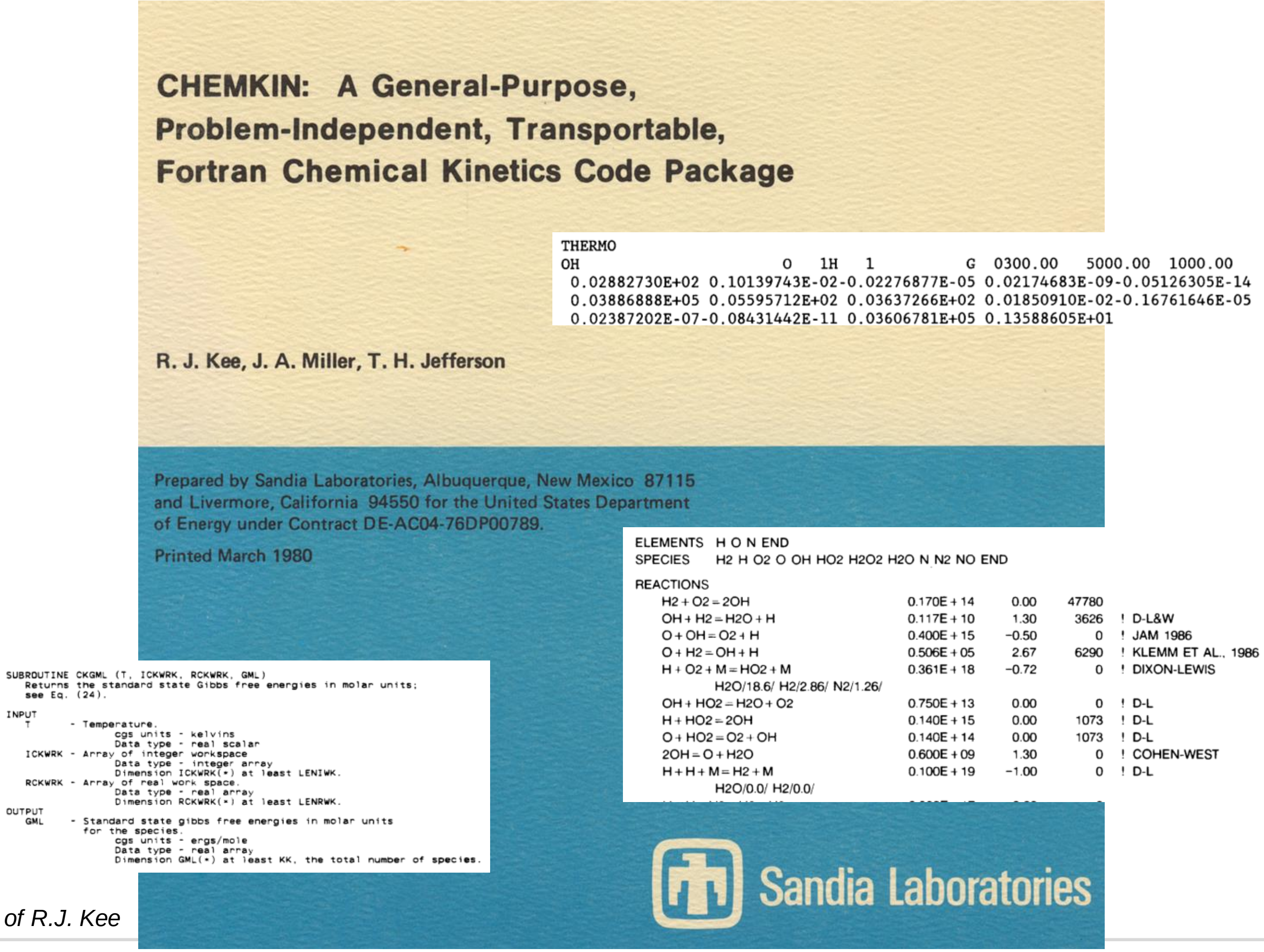
Digitalization in science and engineering
Anything else than new

Konrad Zuse and his first computer,
the Z3, from 1941



Source: Deutsches Museum
https://www.ingenieur.de/technik/produkte/konrad-zuses-z3-computer-welt-80
accessed 3.5.2023

First computer codes modeling reactive flows, 1979



CHEMKIN: A General-Purpose, Problem-Independent, Transportable, Fortran Chemical Kinetics Code Package

```
THERMO
OH                       O   1H   1          G  0300.00   5000.00  1000.00
 0.02882730E+02 0.10139743E-02-0.02276877E-05 0.02174683E-09-0.05126305E-14
 0.03886888E+05 0.05595712E+02 0.03637266E+02 0.01850910E-02-0.16761646E-05
 0.02387202E-07-0.08431442E-11 0.03606781E+05 0.13588605E+01
```

R. J. Kee, J. A. Miller, T. H. Jefferson

Prepared by Sandia Laboratories, Albuquerque, New Mexico 87115 and Livermore, California 94550 for the United States Department of Energy under Contract DE-AC04-76DP00789.

Printed March 1980

```
ELEMENTS   H O N END
SPECIES    H2 H O2 O OH HO2 H2O2 H2O N N2 NO END

REACTIONS
   H2+O2=2OH                 0.170E+14   0.00   47780
   OH+H2=H2O+H               0.117E+10   1.30    3626  ! D-L&W
   O+OH=O2+H                 0.400E+15  -0.50       0  ! JAM 1986
   O+H2=OH+H                 0.506E+05   2.67    6290  ! KLEMM ET AL., 1986
   H+O2+M=HO2+M              0.361E+18  -0.72       0  ! DIXON-LEWIS
        H2O/18.6/ H2/2.86/ N2/1.26/
   OH+HO2=H2O+O2             0.750E+13   0.00       0  ! D-L
   H+HO2=2OH                 0.140E+15   0.00    1073  ! D-L
   O+HO2=O2+OH               0.140E+14   0.00    1073  ! D-L
   2OH=O+H2O                 0.600E+09   1.30       0  ! COHEN-WEST
   H+H+M=H2+M                0.100E+19  -1.00       0  ! D-L
        H2O/0.0/ H2/0.0/
```

```
SUBROUTINE CKGML (T, ICKWRK, RCKWRK, GML)
   Returns the standard state Gibbs free energies in molar units;
   see Eq. (24).

INPUT
   T      - Temperature.
                 cgs units - kelvins
                 Data type - real scalar
   ICKWRK - Array of integer workspace
                 Data type - integer array
                 Dimension ICKWRK(*) at least LENIWK.
   RCKWRK - Array of real work space.
                 Data type - real array
                 Dimension RCKWRK(*) at least LENRWK.
OUTPUT
   GML    - Standard state gibbs free energies in molar units
            for the species.
                 cgs units - ergs/mole
                 Data type - real array
                 Dimension GML(*) at least KK, the total number of species.
```

Sandia Laboratories

Courtesy of R.J. Kee

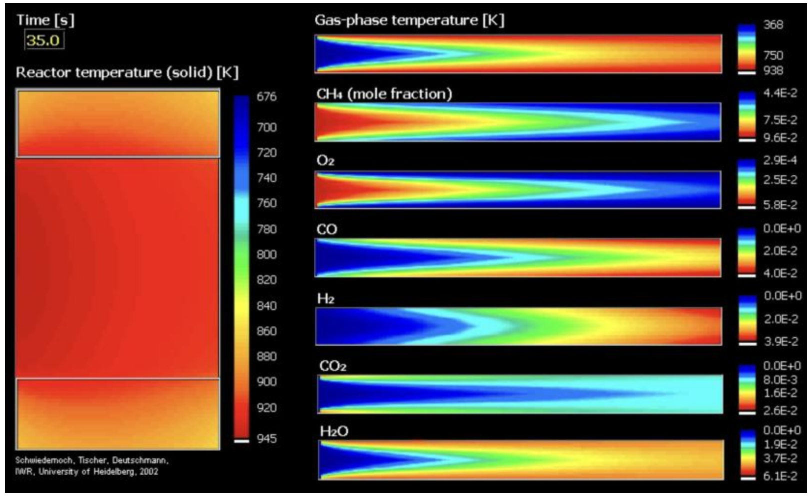
Digitalization in Catalysis and Reaction Engineering
Numerical simulation of the behavior of chemical reactors

Hydrogen from methane by catalytic partial oxidation


ConocoPhillips GTL demonstration plant, Ponca City, OK, USA, 2003

$CH_4 + ½\ O_2 \rightarrow 2\ H_2 + CO$

$CH_4 + 2\ O_2 \rightarrow 2\ H_2O + CO_2$



R. Schwiedernoch, S. Tischer, C. Correa, O. Deutschmann, Chem. Eng. Sci. 58 (2003) 633.

Digitalization in Catalysis and Reaction Engineering
Numerical simulation of the behavior of chemical reactors

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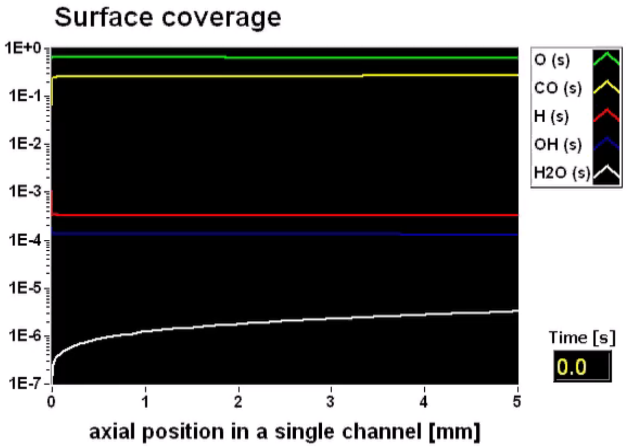
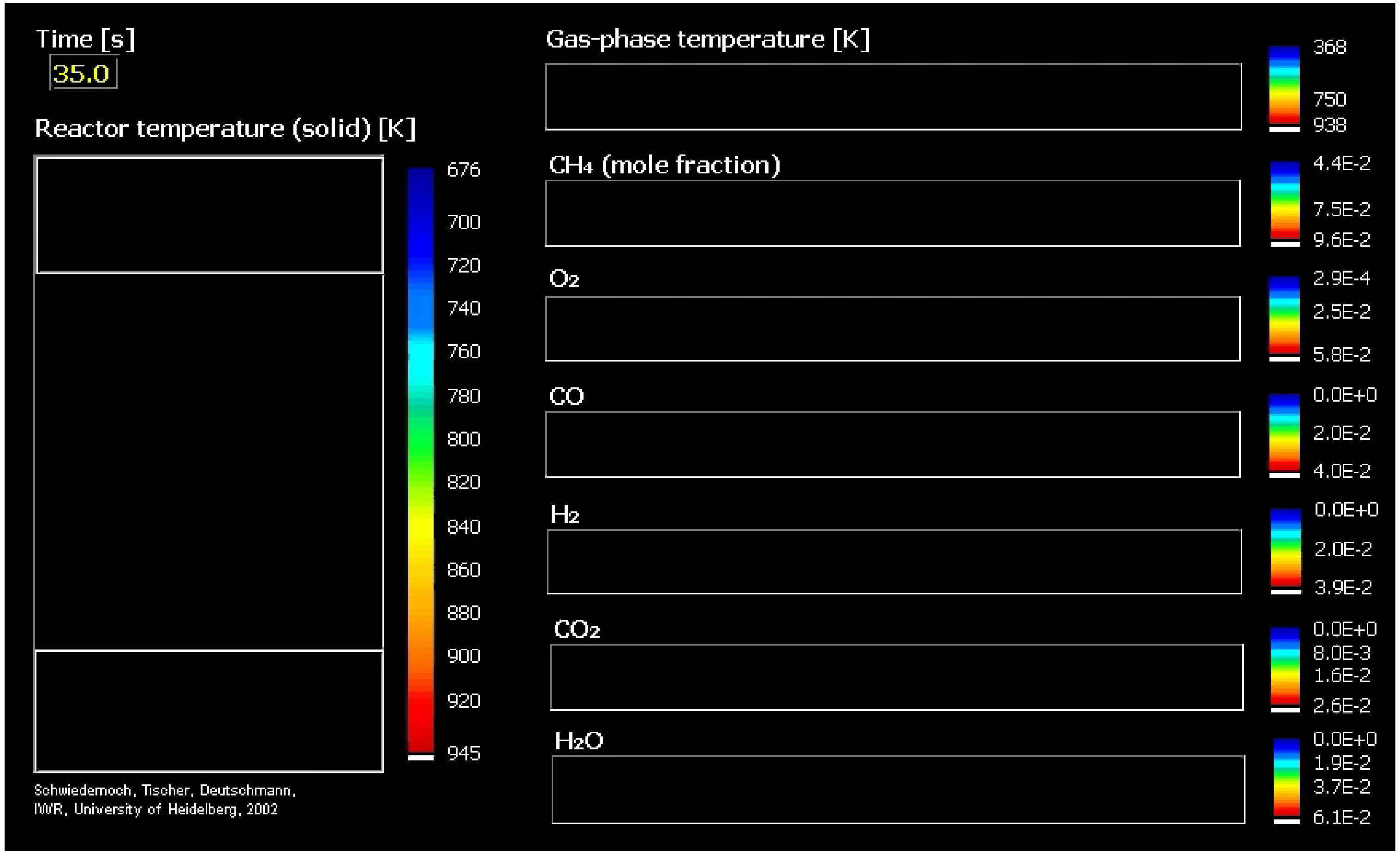


Schwiedernoch, Tischer, Deutschmann, IWR, University of Heidelberg, 2002

R. Schwiedernoch, S. Tischer, C. Correa, O. Deutschmann, Chem. Eng. Sci. 58 (2003) 633.



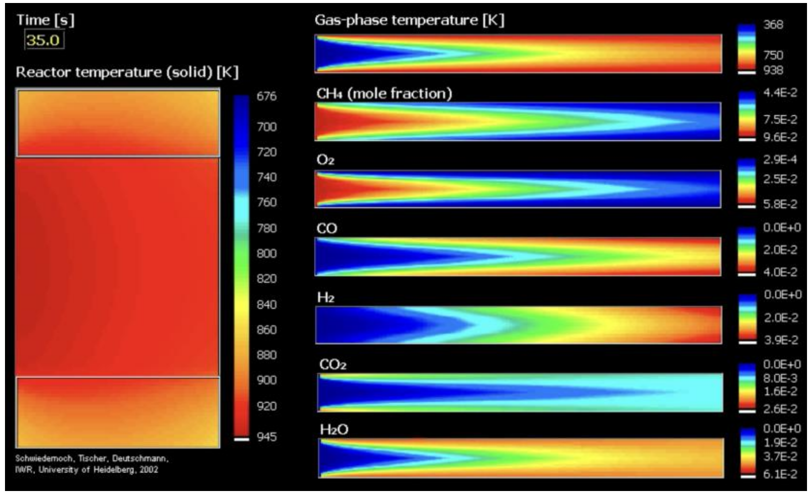
Digitalization in Catalysis and Reaction Engineering
Numerical simulation of the behavior of chemical reactors

Hydrogen from methane by catalytic partial oxidation


ConocoPhillips GTL demonstration plant, Ponca City, OK, USA, 2003

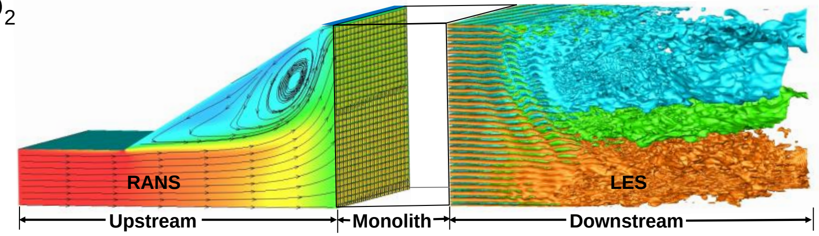
$CH_4 + \frac{1}{2} O_2 \rightarrow 2 H_2 + CO$

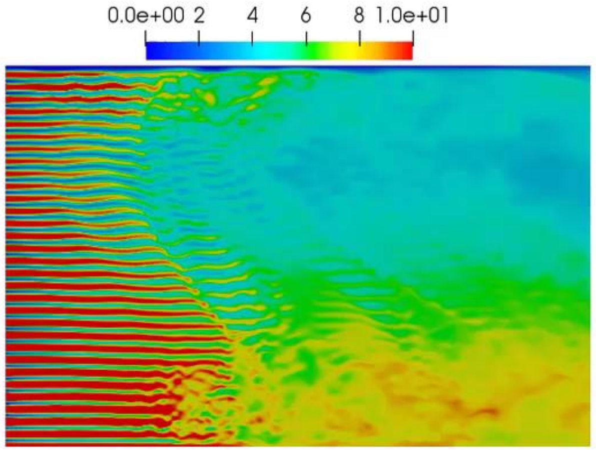
$CH_4 + 2 O_2 \rightarrow 2 H_2O + CO_2$



R. Schwiedernoch, S. Tischer, C. Correa, O. Deutschmann, Chem. Eng. Sci. 58 (2003) 633.

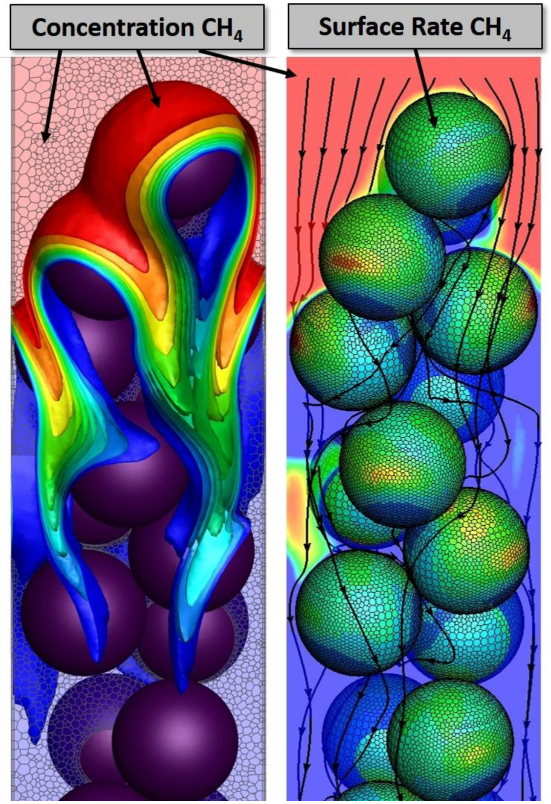
Laminar – turbulence transition behind a monolithic catalyst





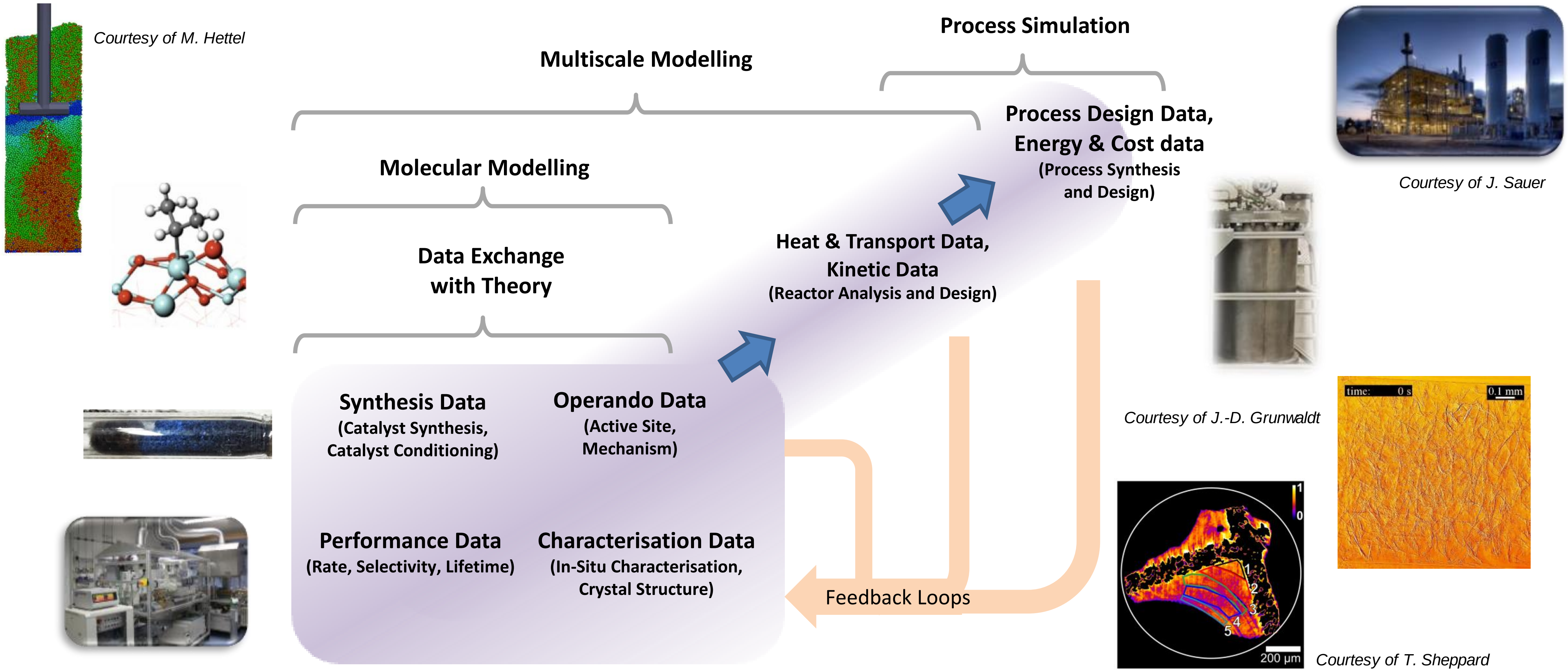
M. Hettel, E. Daymo, T. Schmidt, O. Deutschmann. Chem. Eng. & Processing: Process Intensification 147 (2020) 107728.

Reaction rate in fixed bed reactor using micro kinetics



E. A. Daymo, M. Hettel, O. Deutschmann, G. D. Wehinger. Chem. Eng. Sci. 250 (2022) 117408.

Digitalization in Catalysis and Reaction Engineering
National research data infrastructure



D. Linke, *C. Wulf, M. Beller, T. Boenisch, O. Deutschmann, S. Hanf, N. Kockmann, R. Kraehnert, M. Oezaslan, S. Palkovits, S. Schimmler, S.A. Schunk, K. Wagemann. ChemCatChem 13 (2021) 3223.*

→ A. Fedorov Tue 10:00

Digitalization in Catalysis and Reaction Engineering
Outline

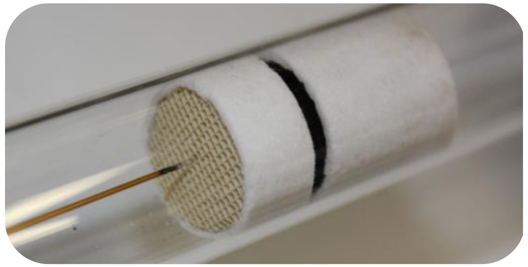
- Introduction –long history of digitalization in science and engineering (OD)
- **Automated workflows on the lab scale – models for heterogeneous catalysis (OD)**
- Feedback loops –from lab to pilot plant (SAS)
- Application of digital tools for scale-up on industrial scale (OD)
- Prediction of materials properties on atom scale using AI for catalyst discovery (SAS)
- Democratization of tools and services: RDM @ NFDI4Cat (SAS)

Variety of experimental methods for a better understanding of chemical kinetics and interaction with mass and heat transport

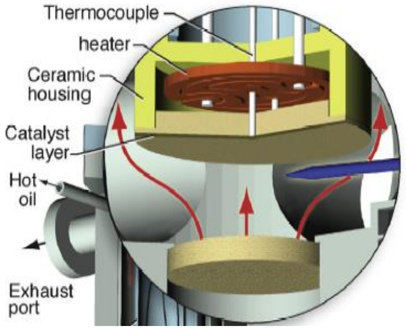
End-of-pipe

Fixed bed reactor

Capillary techniques

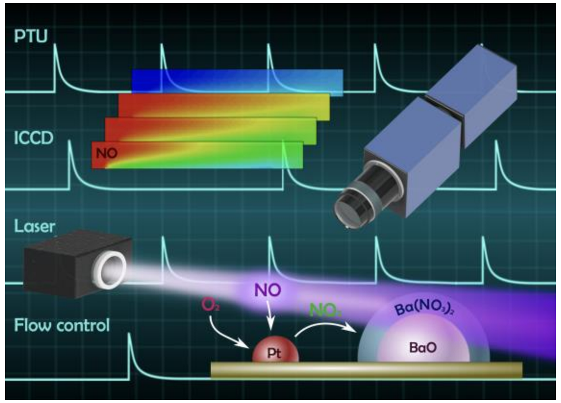


Monolithic reactors



Stagnation flow reactor

Laser diagnostics

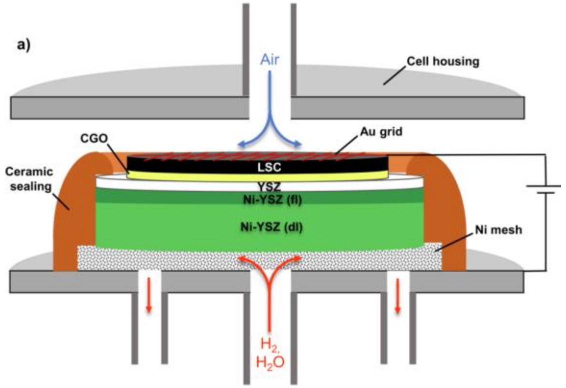


Catalytic wall reactor

Specific flow reactors

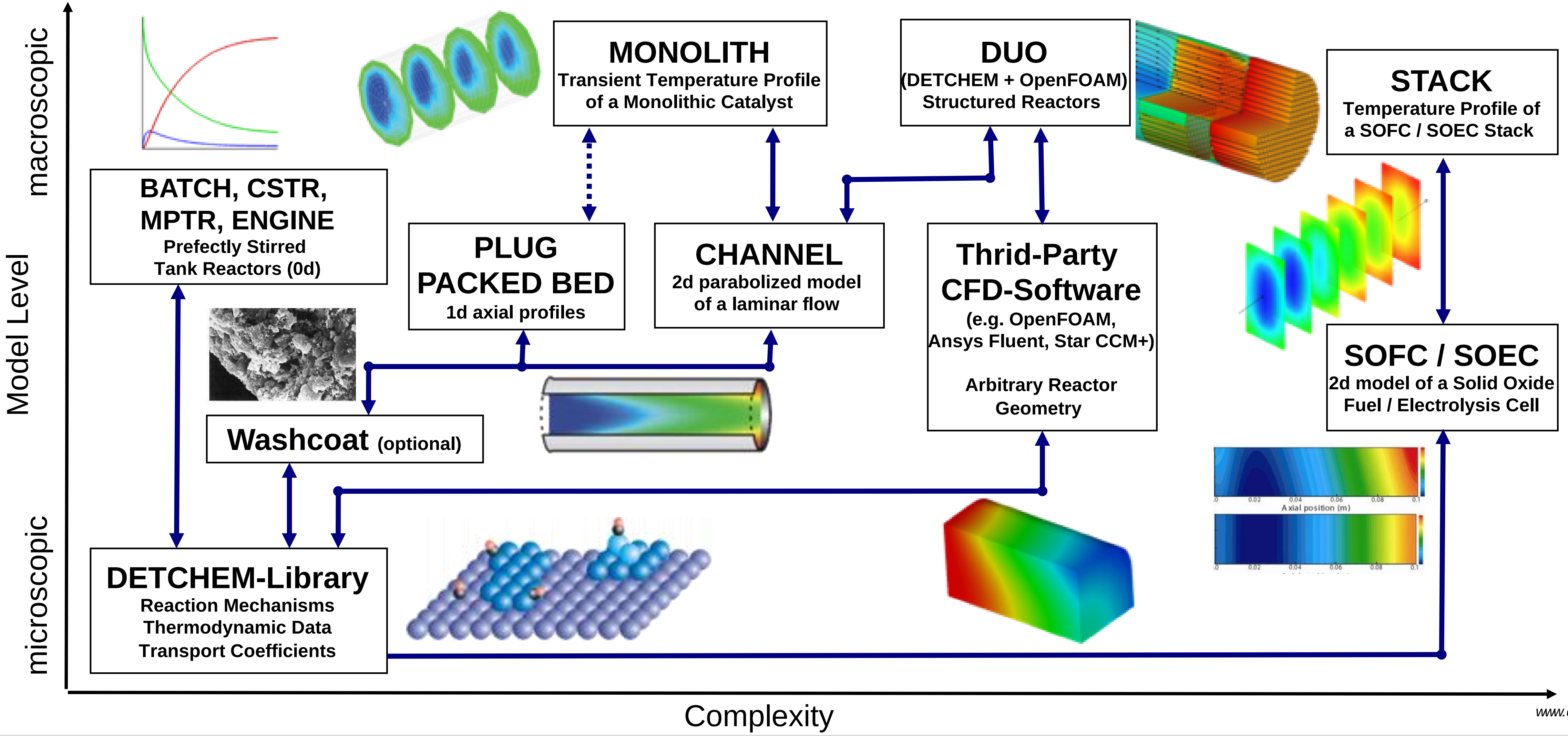
High-temperature pressure reactor

Electrochemical characterization



Button cell

Software package DETCHEM for numerical simulation of a wide variety of chemical reactors



www.detchem.de

Modeling of heterogeneous chemical reactions: Development of reaction mechanisms with associated kinetic data

Karlsruhe Institute of Technology

Experimental Surface science
(XPS, XRD, IR, TEM, STM, AFM…)

Initial reaction mechanism
(idea)

Theoretical Surface science
(DFT, UBI-QEP, MC…)

Modeling of lab reactors
(including gas phase kinetics and transport models)

Lab experiments

(conversion, selectivity, spatial/temporal profiles, coverage...)

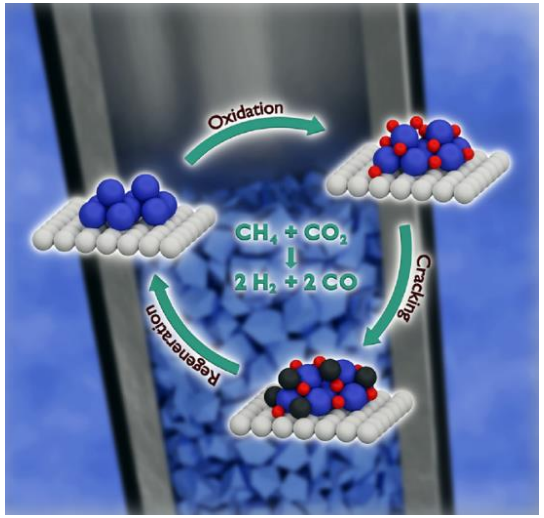
Comparison of experiments and simulation

Technical reactors

Sensitivity & reaction flow analysis

Thermodynamic consistency

Reliable mechanism



BASF The Chemical Company

hte the high throughput experimentation company

THE LINDE GROUP Linde

Bundesministerium für Wirtschaft und Technologie

A.Giehr, L. Maier, S. Schunk, O. Deutschmann. ChemCatChem 10 (2018) 751.
PhD theses of K. Herrea-Delgado, T. Roussiere, L. Kahle, A. Giehr

Automated workflows in catalysis and reaction engineering: Speed-up of model development by CaRMeN

Karlsruhe Institute of Technology

meta data of experiments

Simulation

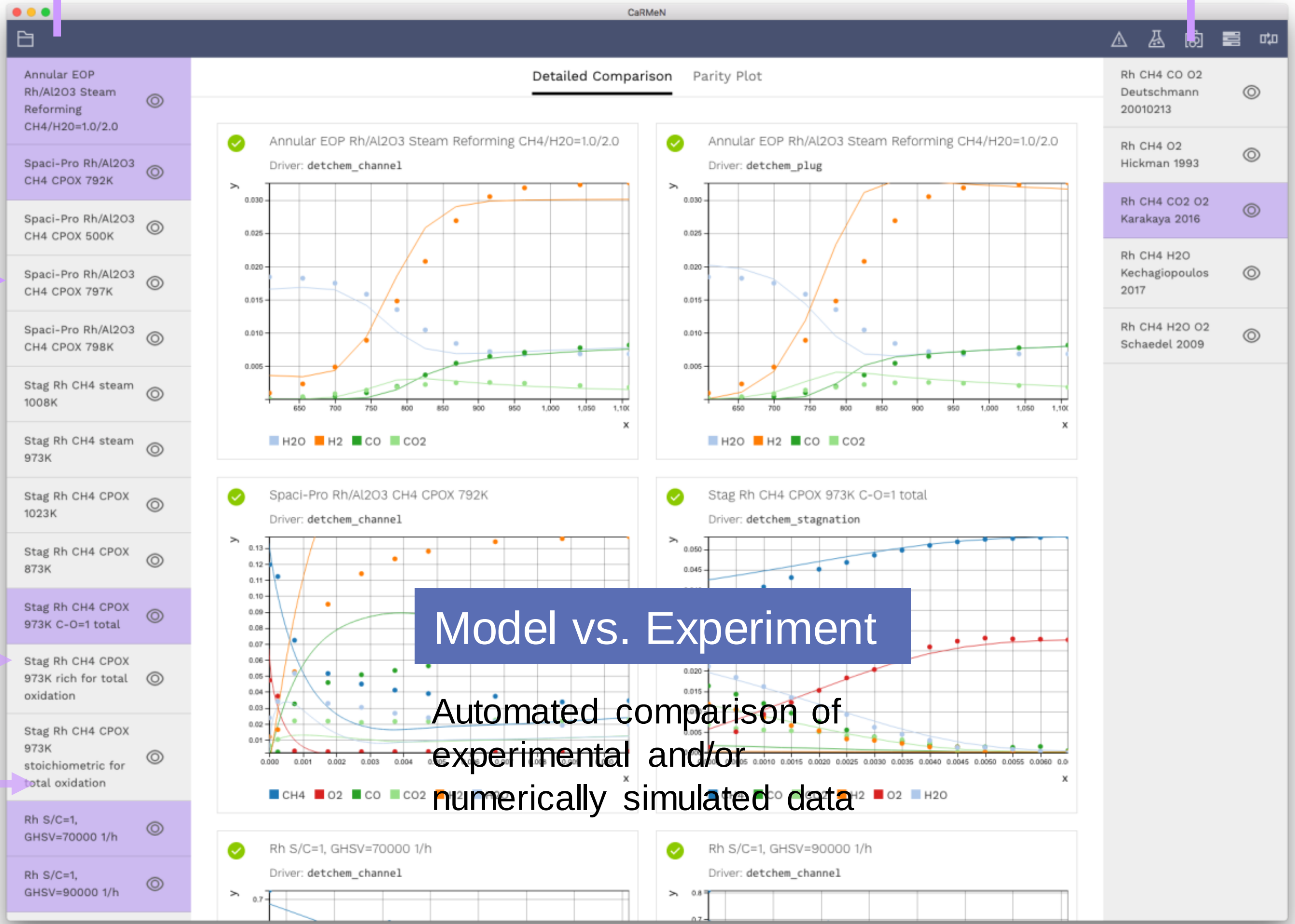
data of physical & chemical models

Performance

Experimental data including all metadata for different reactors, conditions, sources for given chemical system

Synthesis

Characterization



Model vs. Experiment

Automated comparison of experimental and/or numerically simulated data

Combines chemical and physical models

Models

and can be used for easy comparisons of different

- reaction mechanisms,
- transport models,
- computer codes …

H. Gossler, L. Maier, S. Angeli, S. Tischer, O. Deutschmann. PhysChemChemPhys 20 (2018) 10857; Catalysts 9 (2019) 227; www.detchem.de/software/carmen

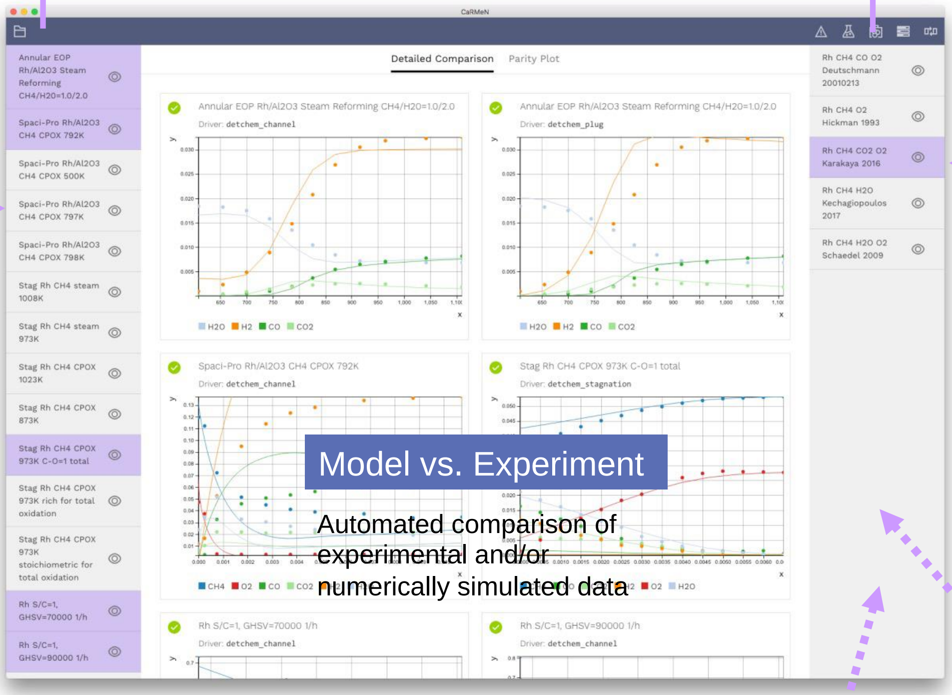
Automated workflows in catalysis and reaction engineering: CaRMeN as data basis for model parametrization and optimization

Karlsruhe Institute of Technology

meta data of experiments — **Simulation** — data of physical & chemical models

Performance

Experimental data including all metadata for different reactors, conditions, sources for given chemical system



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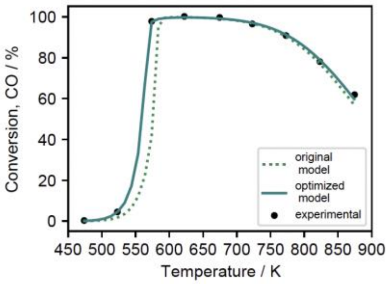
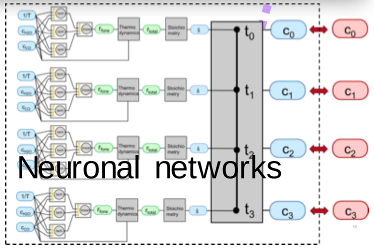
→ Poster 6.01 Chacko et al.

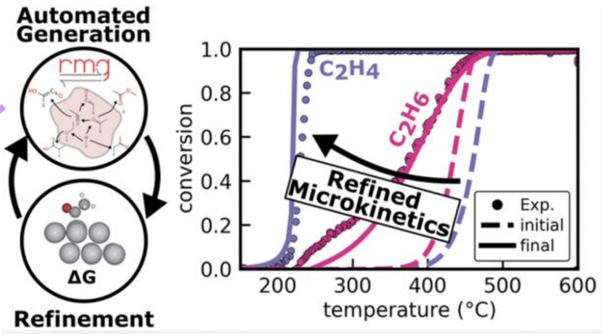
Optimization



R. Chacko, K. Keller, S. Tischer, A.B. Shirsath, P. Lott, S. Angeli, O. Deutschmann. J. Phys. Chem. C (2023)



→ Martin Votsmeier Tue 10:25

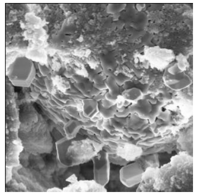


__B. Kreitz__, P. Lott, J. Bae, K. Blöndal, S. Angeli, Z.W. Ulissi, F. Studt, __C.F. Goldsmith__, O. Deutschmann. ACS Catal. 12 (2022) 11137.

H. Gossler, L. Maier, S. Angeli, S. Tischer, O. Deutschmann. PhysChemChemPhys 20 (2018) 10857; Catalysts 9 (2019) 227; www.detchem.de/software/carmen

Digital twin of reactor and direct transfer of performance and meta data: Software tool Adacta

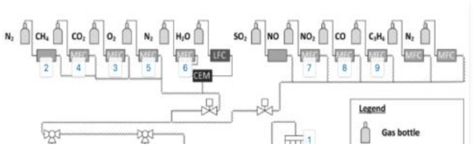
- Focus on **traceability** of data
- Captures exact details to an experimental setup
- Setups composed of **devices and samples**
- **Time line** as independent variable to visualize relationships
- Data exactly associated with recording device
- **Catalyst/materials history** monitored including efficient **search tools**



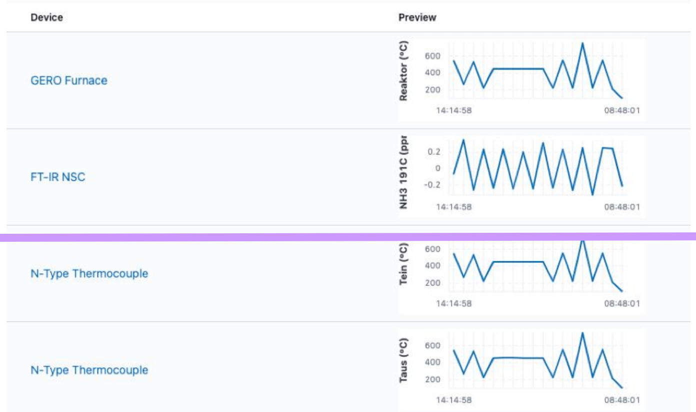
Catalyst data

ADACTA

Y1(X0)	Y2(X0)	Y3(X0)	Y4(X0)	Y5(X0)	Y6(X0)	Y7(X0)
Nitrogen diatomi	Carbon monoxid	Methane (CH4)	Air	Nitrogen diatomi	Sulfur dioxide (S	Water (
ml/min	ml/min	ml/min	l/min	ml/min	ml/min	ml/min
N2 Traeger	CO 500ml/min	CH4	5l/min Air	N2	SO2 50 mL	H2O 75
0	0	3,889379	0,450598	0,479986	0	0
0	0	3,889308	0,449088	0,479947	0	0
0	0	3,888306	0,451563	0,479987	0	0



Reactor data



Analytics data

Data archive and extraction of metadata for setting up input files for models and simulation

H. Gossler, J. Riedel, E. Daymo, R. Chacko, S. Angeli, , O. Deutschmann. Chemie Ingenieur Technik 94 (2022) 1798; www.omegadot.software/adacta

Digitalization in catalysis and reaction engineering: Value-added coupling of digitalization tools

Karlsruhe Institute of Technology

Chemotion, SciCat

- Catalyst synthesis information
- Basic and detailed characterization data

Adacta

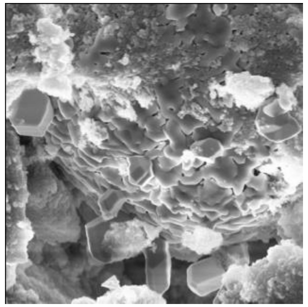
- Digital twin of experimental setups
- Catalyst performance (meta)data

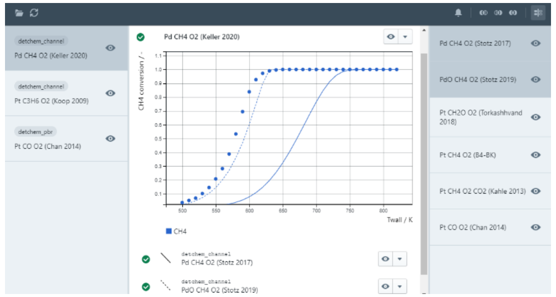
CaRMeN

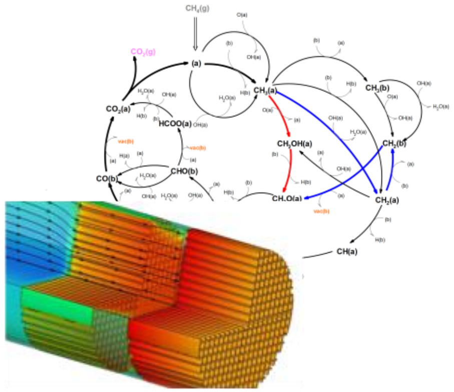
- Automated simulation workflows
- Visualization model vs. experiment data
- Optimization of model parameters

DETCHEM

- Chemical/Physical Models
- Numerical simulation of catalytic reactors







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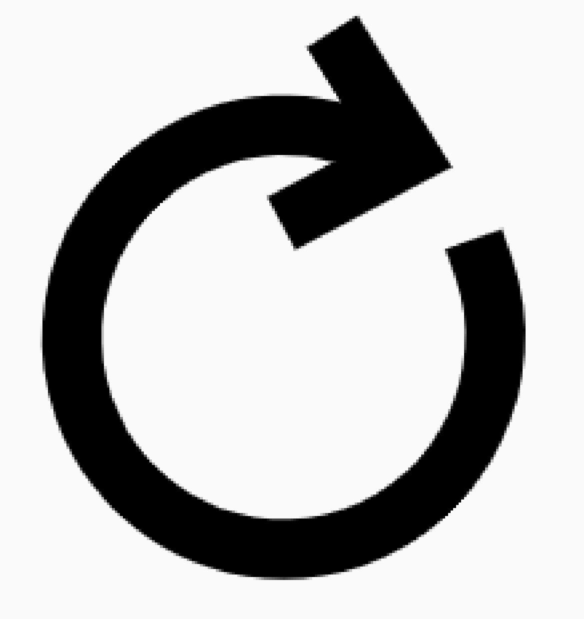
- Democratization of tools and services: RDM @ NFDI4Cat (SAS)

Feedback Loops: Cause and Effect made simple?

"…Simple causal reasoning about a feedback system is difficult because the first system influences the second and second system influences the first, leading to a circular argument. This makes reasoning based upon cause and effect tricky, and it is necessary to analyze the system as a whole. …"

— Karl Johan Åström and Richard M.Murray, Feedback Systems: An Introduction for Scientists and Engineers

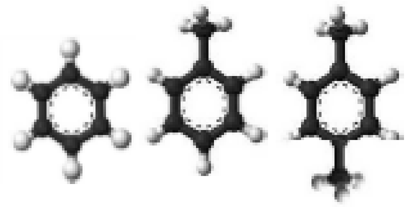
Feedback Loops in Action



From Naphta to Reformate

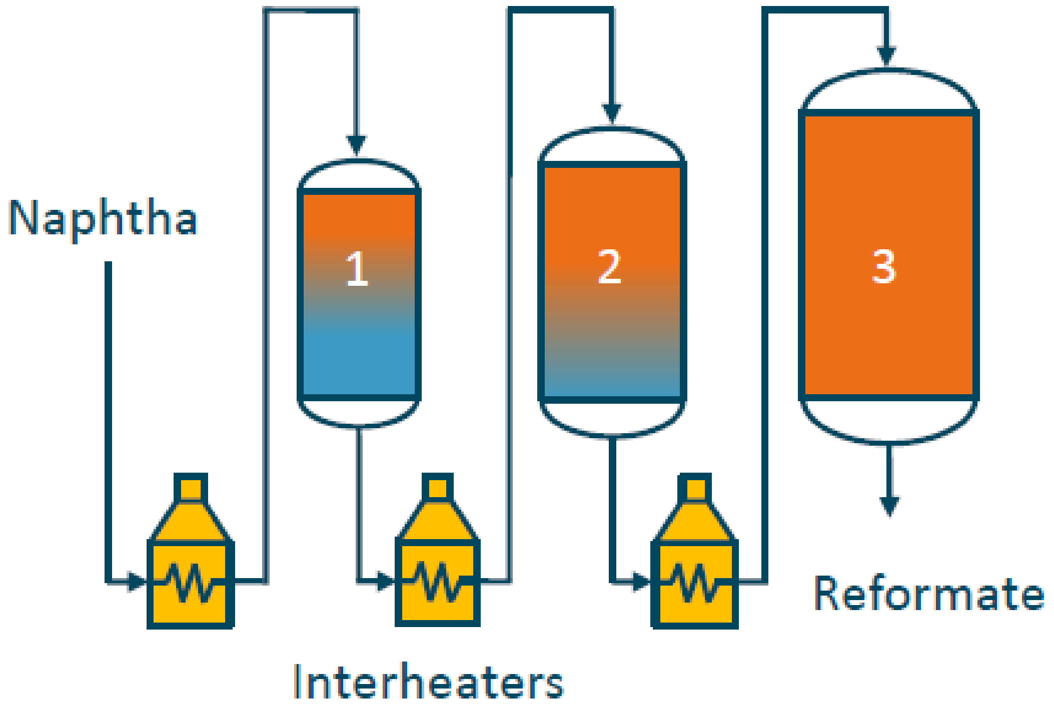
Naphtha *(low octane)* → **H_2** + **Lights C_1-C_4** + **Reformate** *(high octane)*

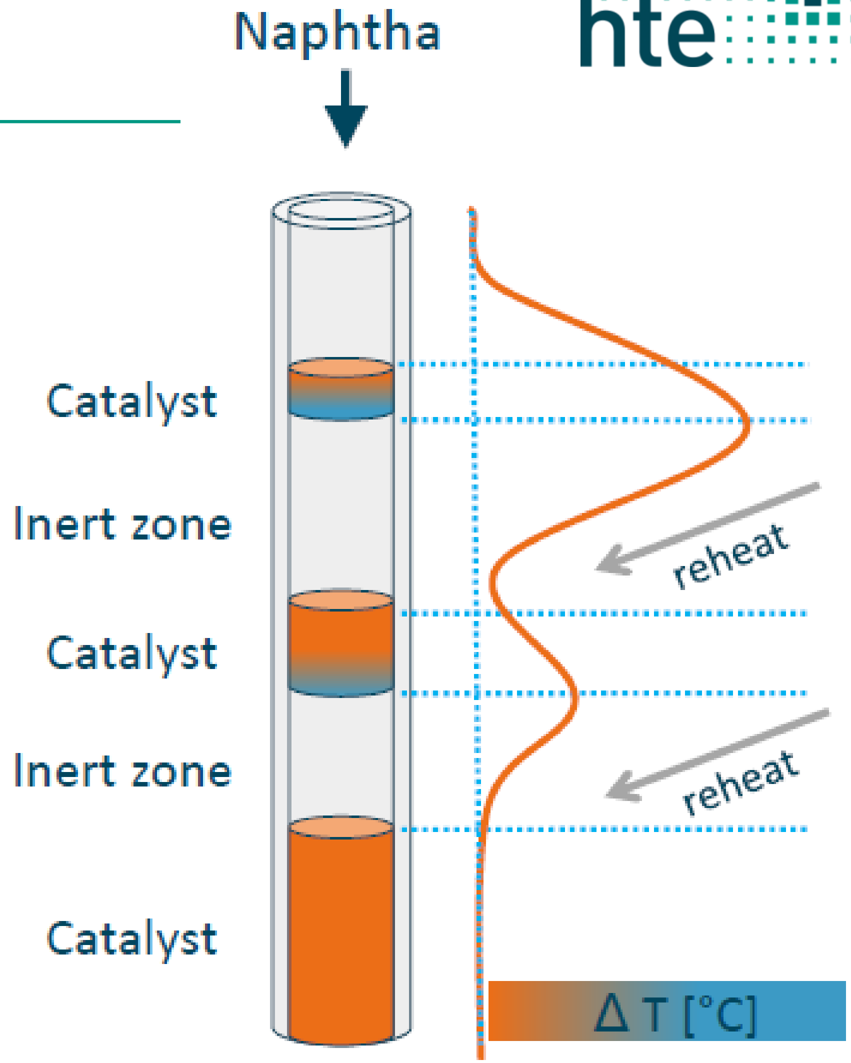
Reformate ↓ **Gasoline** *(high octane)* and ↓ **Aromatics** *(BTX)*



- Challenges
  - Analysis of multi-component feeds (>300 compounds) and products
  - Detection of very small differences in product selectivity (C_5+)
  - Variation of feed & product properties
  - Accelerated decay conditions for fixed-bed catalysts
  - Collection of sufficient data for fast deactivating CCR catalysts

Naphta Reforming on a Technical Scale

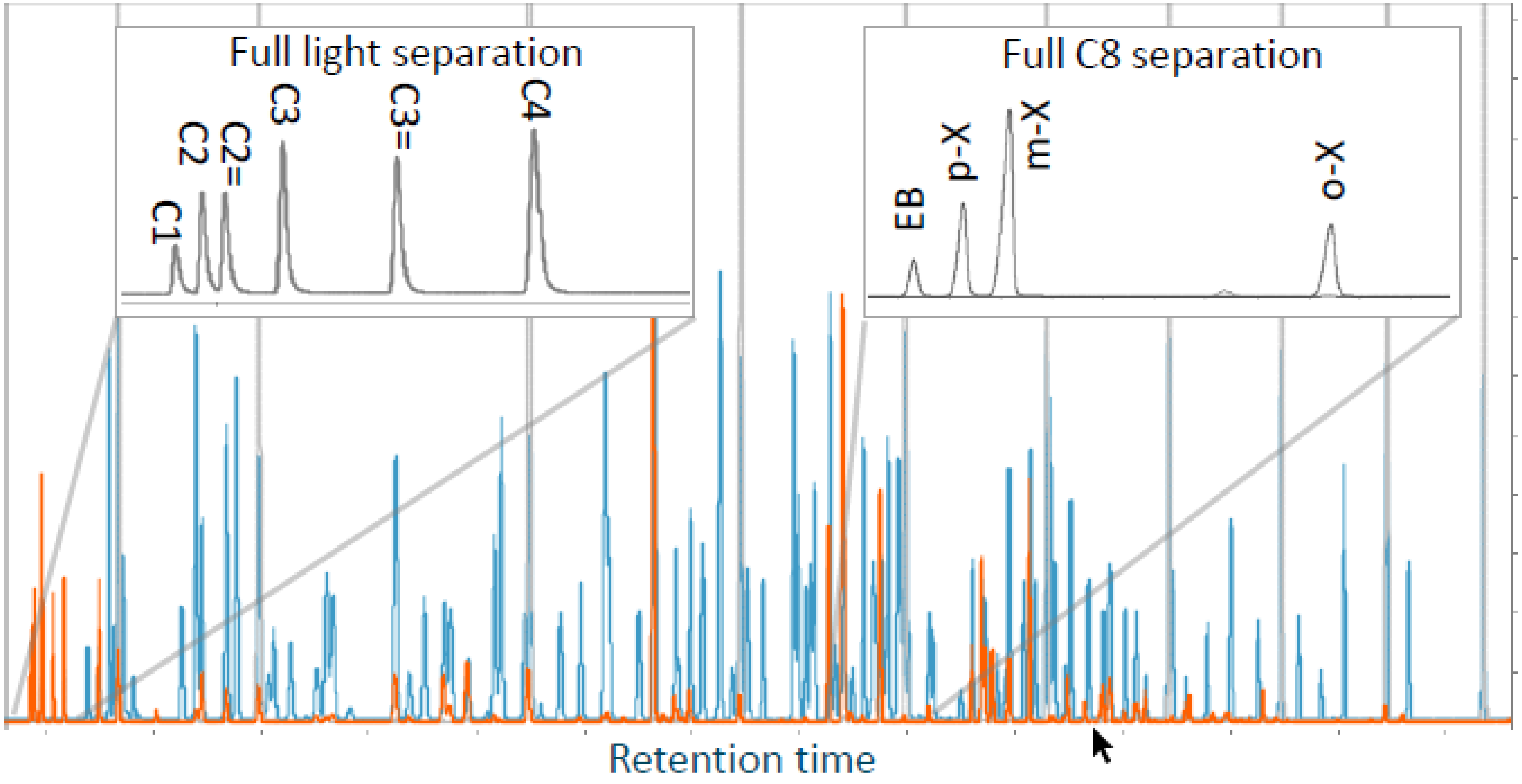




Why Testing under „Iso“-Conditions?

- Comparability on a relative scale
- Comparability on an absolute scale
- Retrieval of kinetic data-sets that link into scale-up criteria

Complexity of the Analysis



C1-C14 Paraffins

Naphtha feed

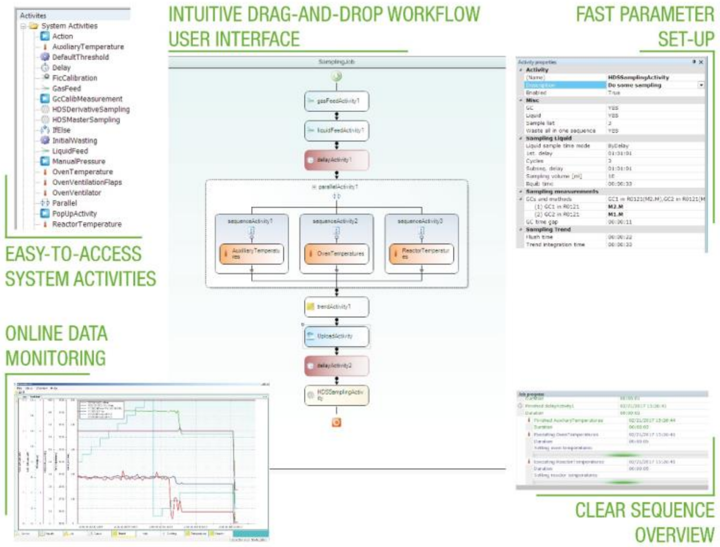
Reformer products

~130 assigned compounds

hte Software Solutions

Combining workflow key steps with one tool

GENERATING DATA	GENERATING KNOWLEDGE

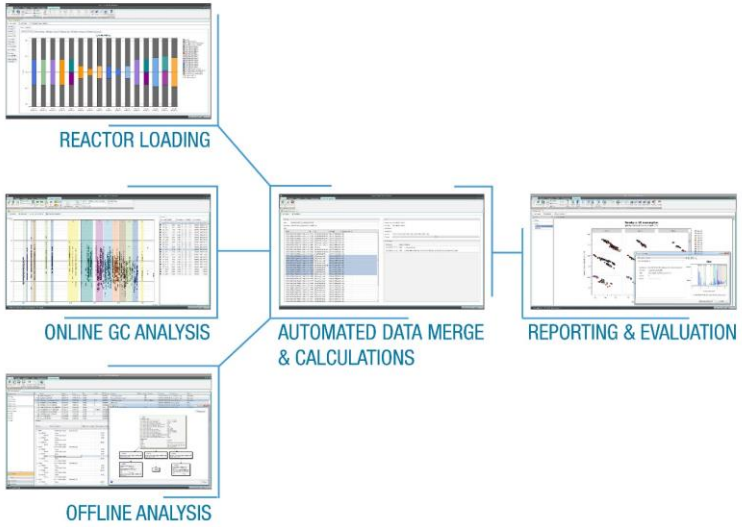


hteControl
Experiment Automation

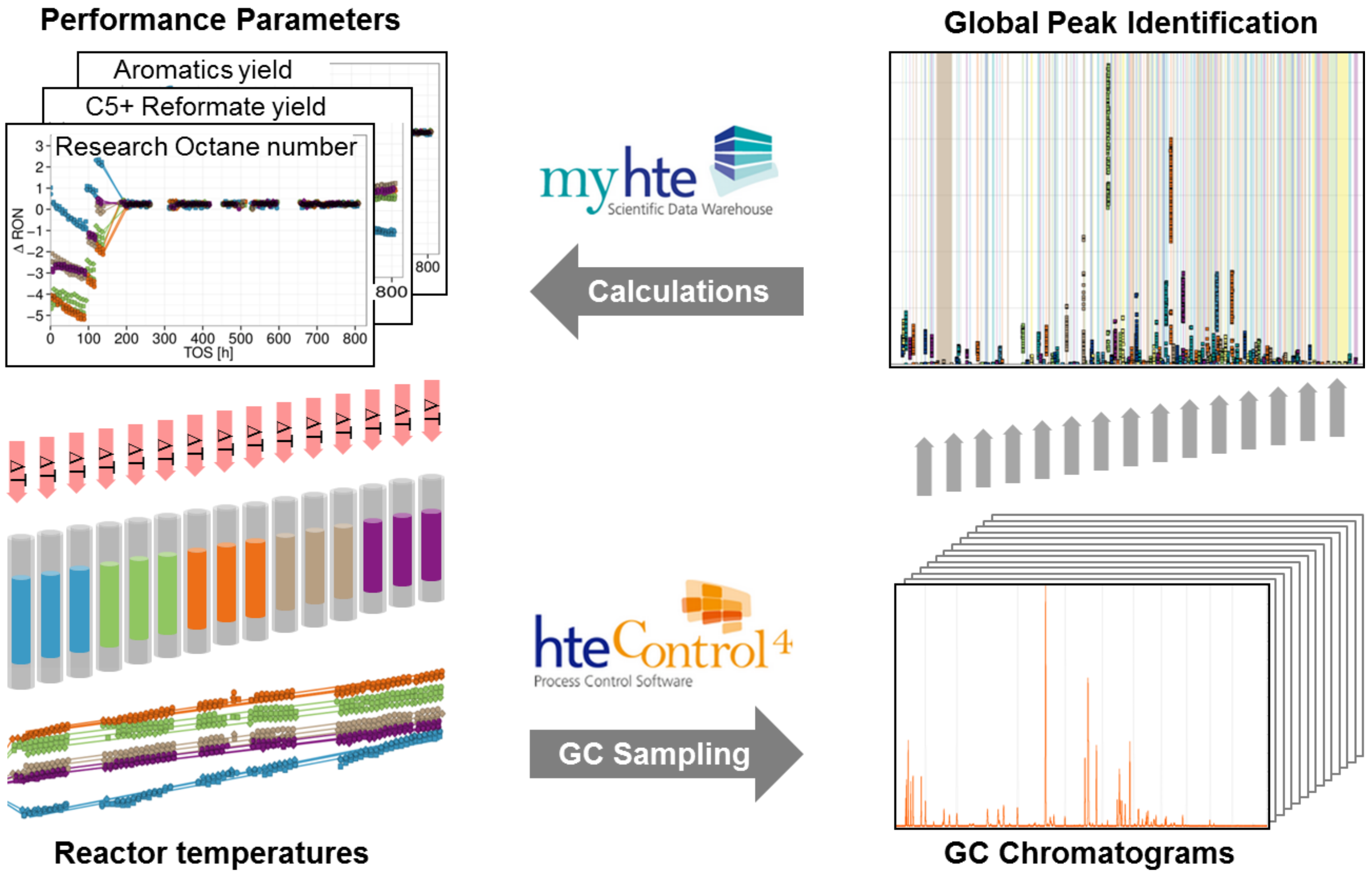
- Recipe editor
- Process control
- Industry standard process visualization
- Intuitive test rig operation
- Fast and efficient failure diagnosis

myhte
Scientific Data Warehouse

- Access data from your desktop
- All data in one database
- Evaluate and create reports
- Integrate your R&D environment

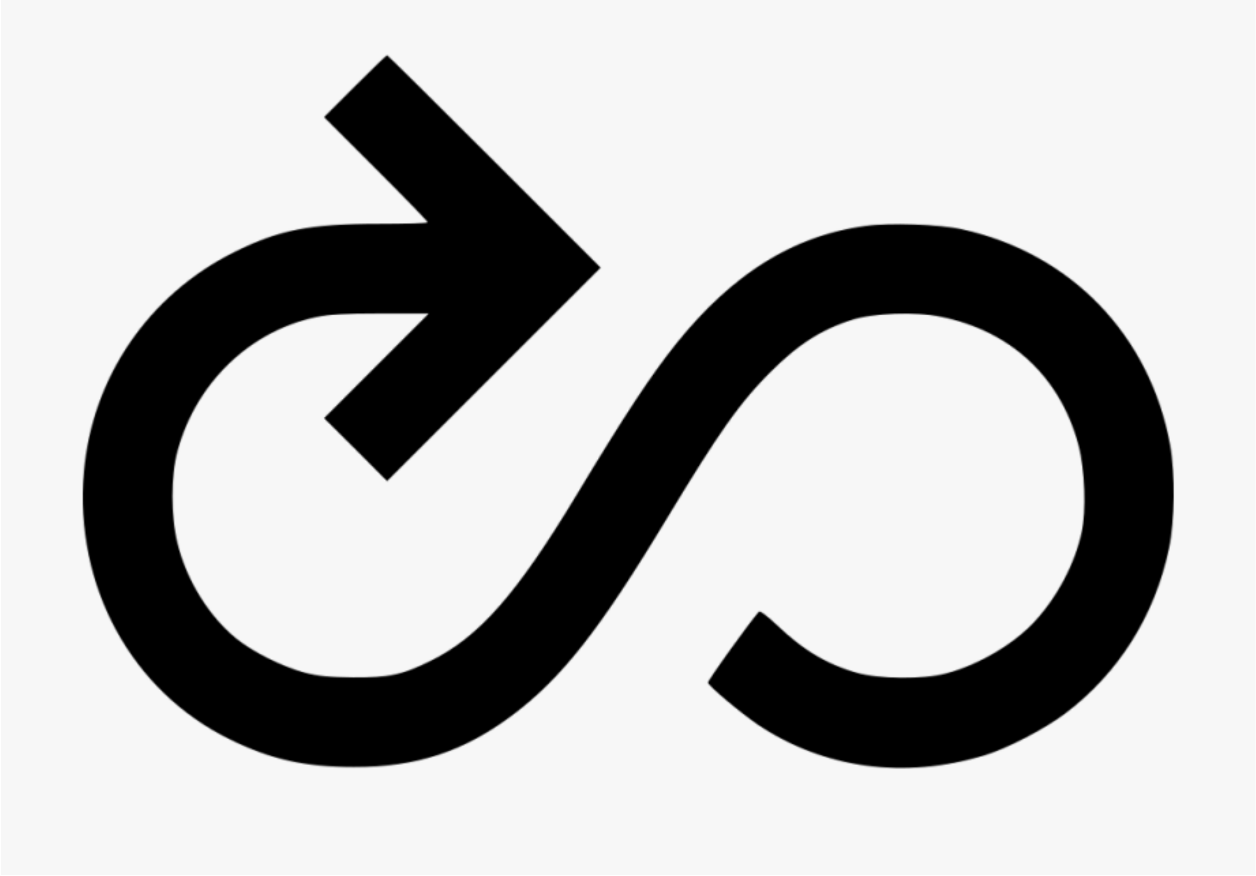


Iso-Operation
Integrated Cyber-Physical Interaction

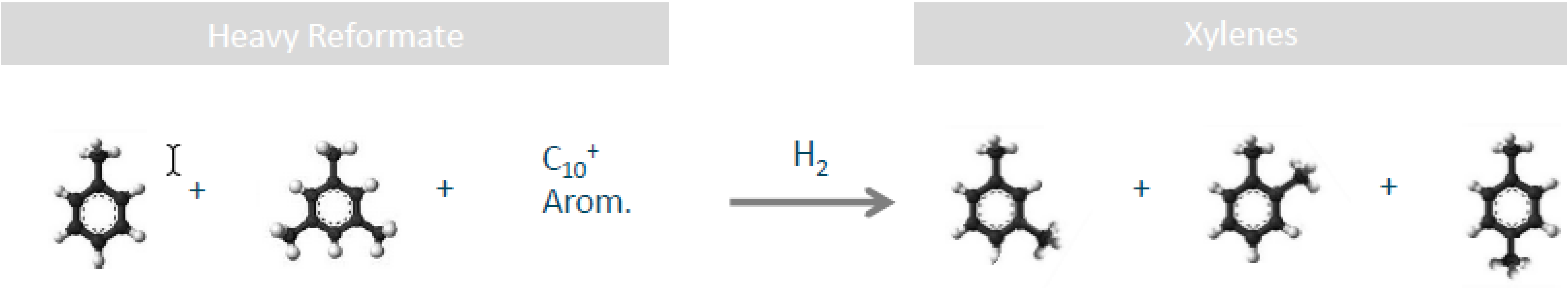


- myhte allows interactive self-adaptation to prechosen values for distinct variables
- This is an excellent example for a self-adaptive cyber-physical interaction

Feedback Loops in Action

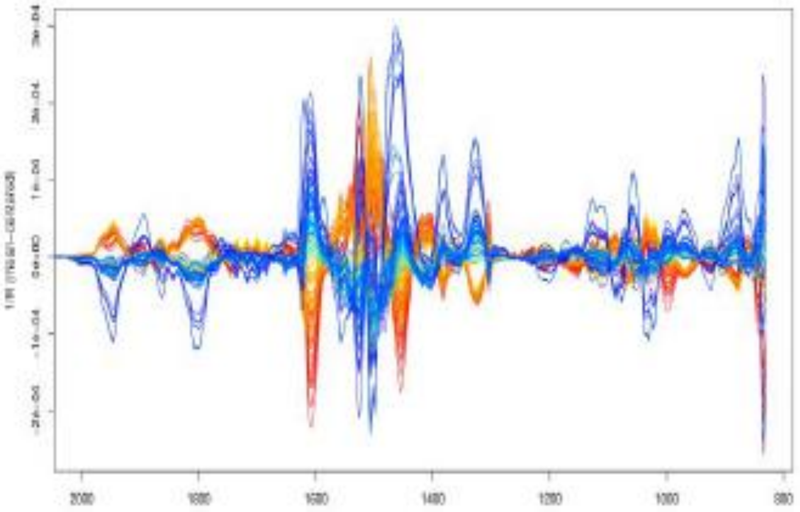


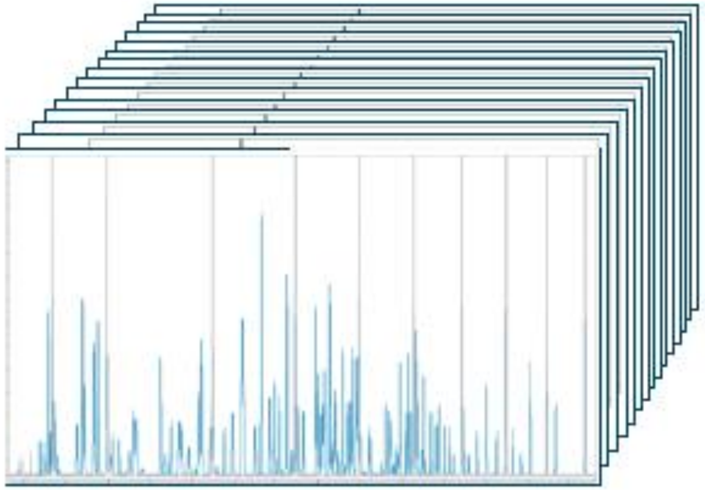
Transalkylation of Aromatics



- Conversion of C7 and C9 to maximize xylenes (p-xylene)
- Scopes
  - Identification of best performing catalyst
  - Deactivation at higher severities
  - Quality control

Multicomponent Mixtures Overlay of wavelengths: chemometrics

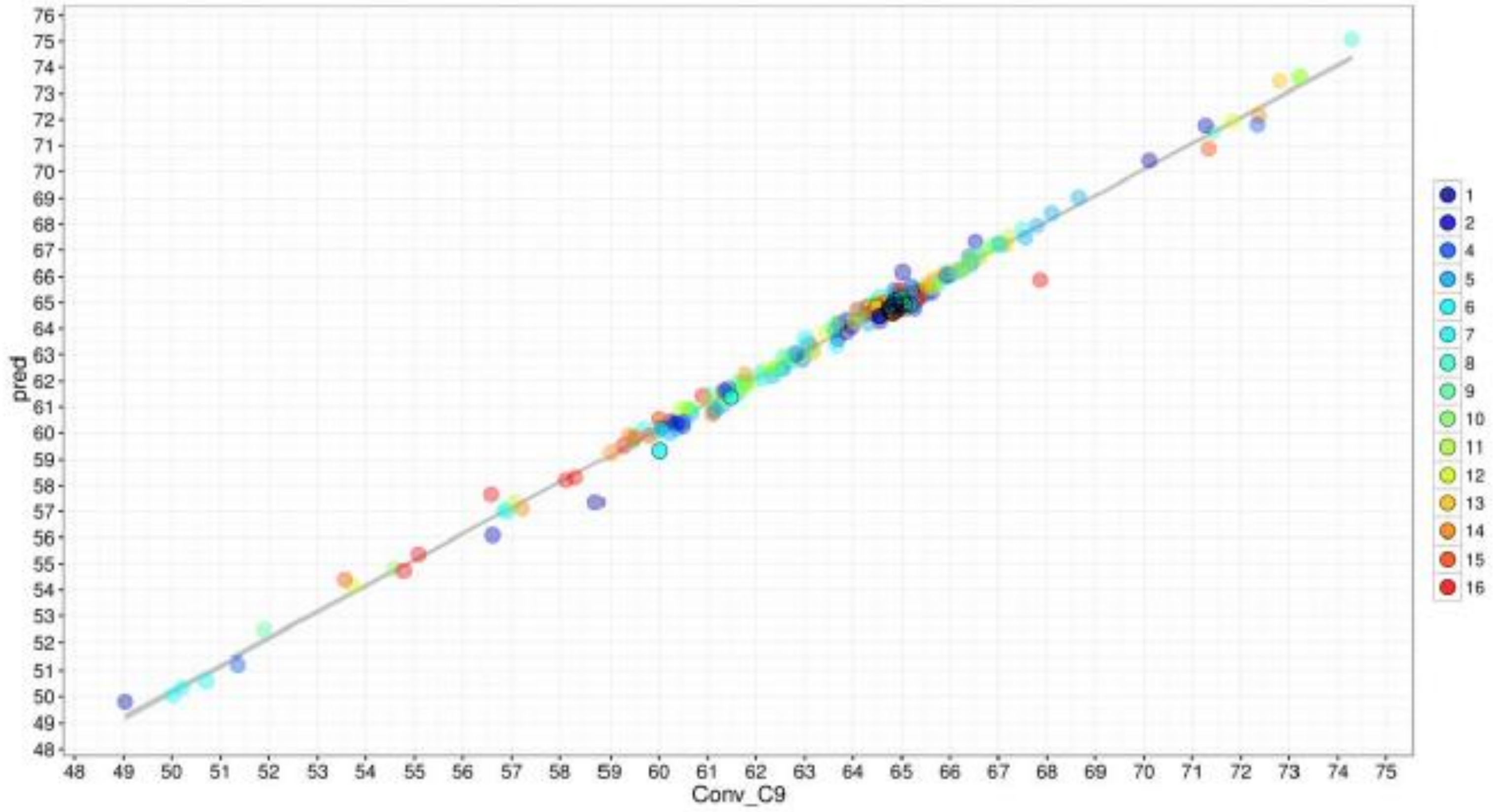




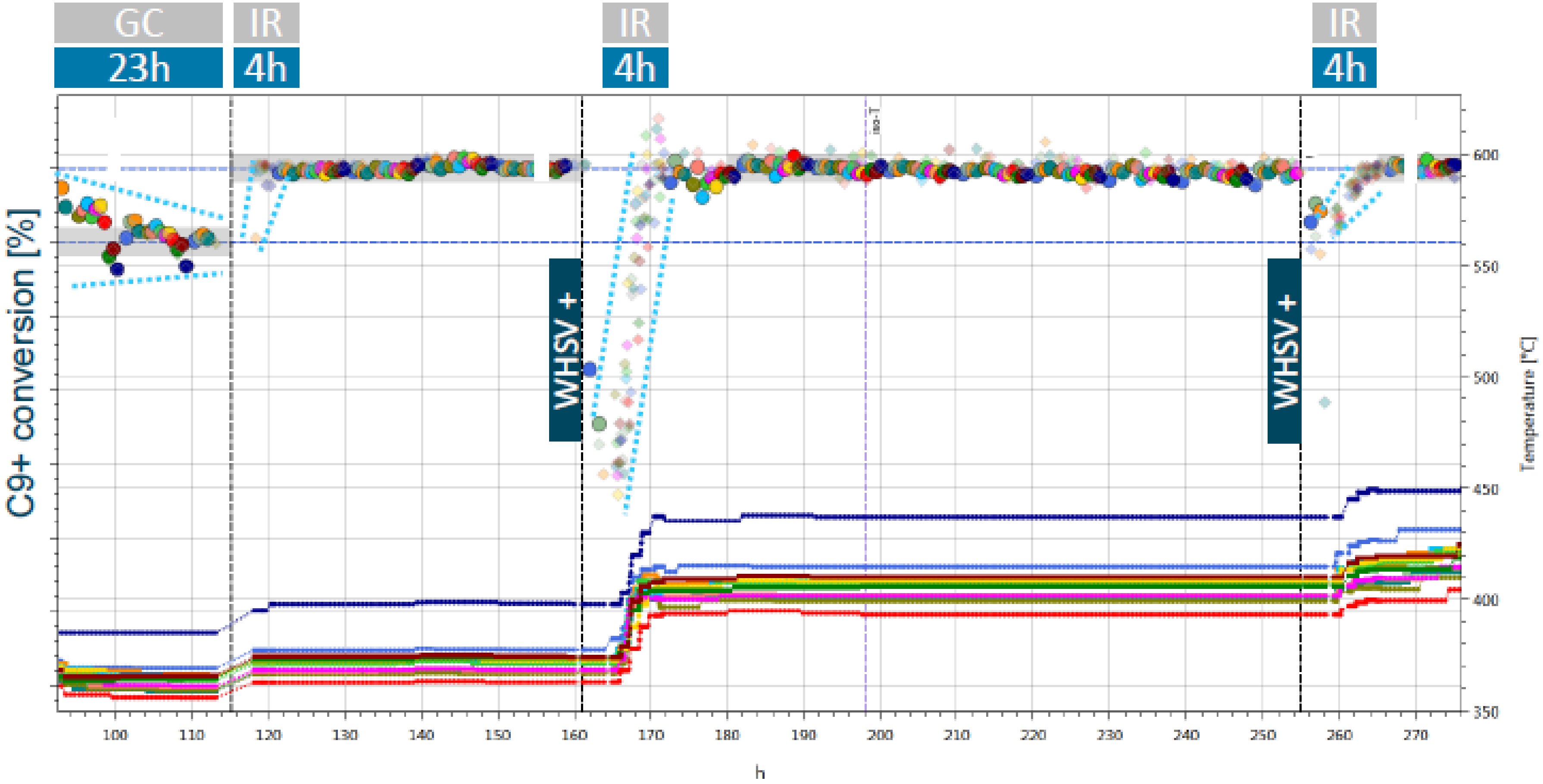
- IR spectra

- GC chromatograms
  - Selectivities
  - Conversions
  - RON, MON

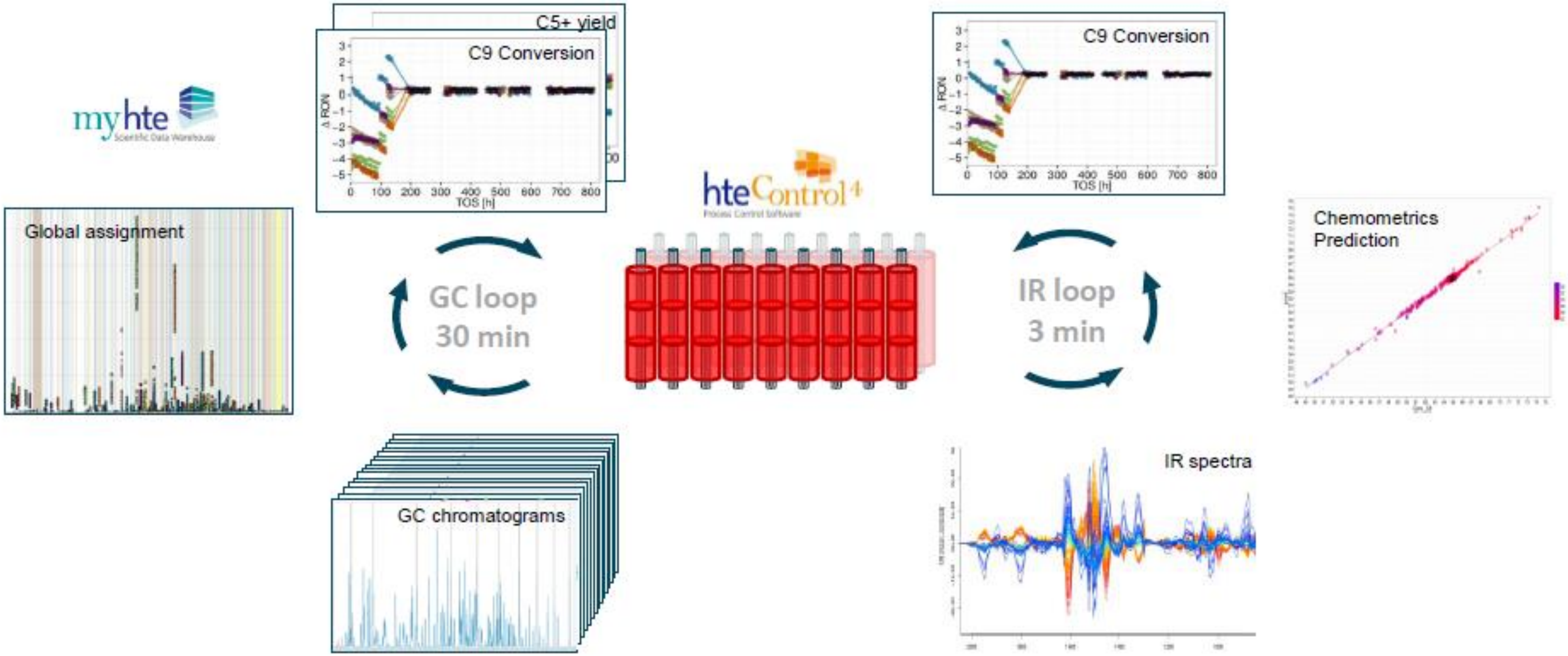
Transalkylation Prediction of C9+ conversion



Feedback Loop
Achieving Constant Conversion



Fast Feedback Loop based on Chemometrics the Power of FTIR Spectroscopy



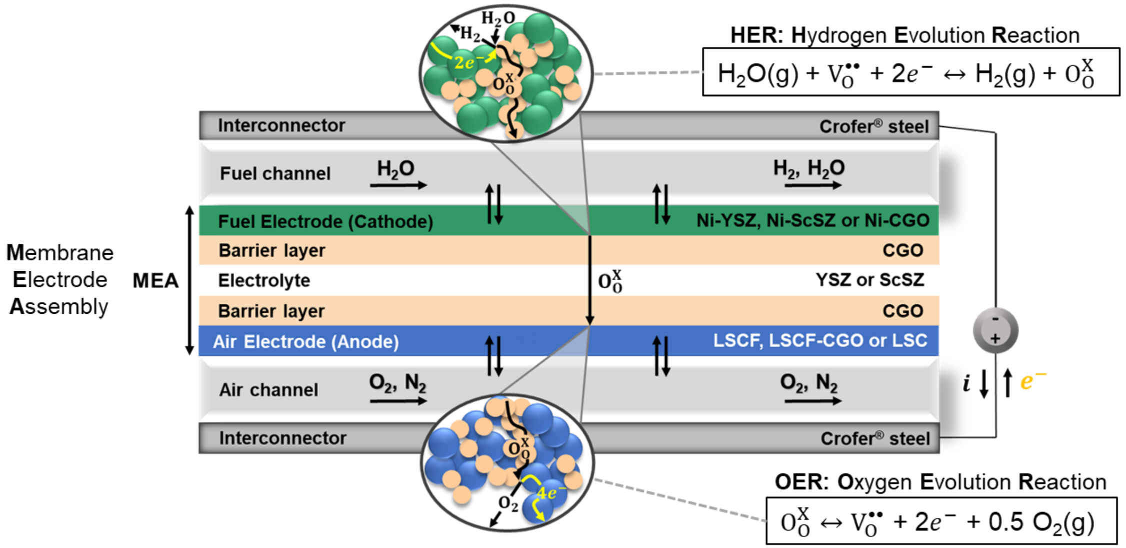
Digitalization in Catalysis and Reaction Engineering
Outline

- Introduction –long history of digitalization in science and engineering (OD)
- Automated workflows on the lab scale – models for heterogeneous catalysis (OD)
- Feedback loops –from lab to pilot plant (SAS)
- **Application of digital tools for scale-up on industrial scale (OD)**
    - High-temperature electrolysis
    - Carbon-free chemical energy carriers
    - CO_2 emission reduction in steel production
- Prediction of materials properties on atom scale using AI for catalyst discovery (SAS)
- Democratization of tools and services: RDM @ NFDI4Cat (SAS)

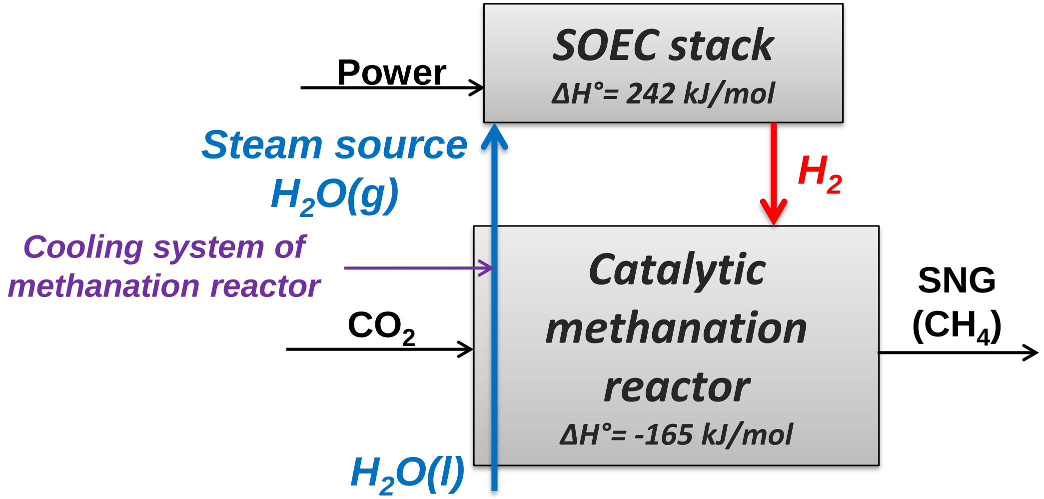
Hydrogen from high-temperature electrolysis: Process intensification trough coupling of thermal and chemical processes

Karlsruhe Institute of Technology

Solid-Oxide Electrolysis Cell (SOEC) operated >700°C



Power-to-Methane: Thermal integration of SOEC stack & methanation reactor for



METHQUEST

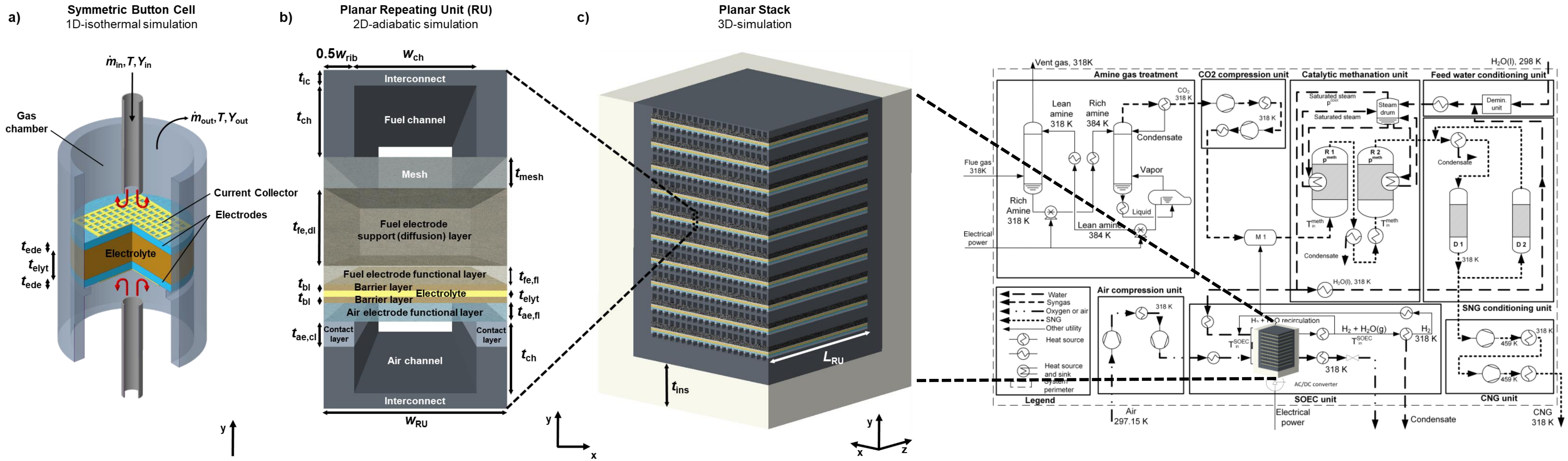
Bundesministerium für Wirtschaft und Technologie

- What is the best SOEC configuration for thermal integration?
- Stack simulation based on physical and chemical characterization of the SOE Cell
- Optimization of design parameters and operating conditions

L. Wehrle, D. Schmider, J. Dailly, A. Banerjee, O. Deutschmann. Appl. Energy 317 (2022) 119143.

Hydrogen from high-temperature electrolysis: Development chain from button cell to stack to system

Electrochemical characterization | Coupling with channel flow and 2D heat transport | Coupling with 3D heat transport models | Coupling with upstream and downstream devices and processes



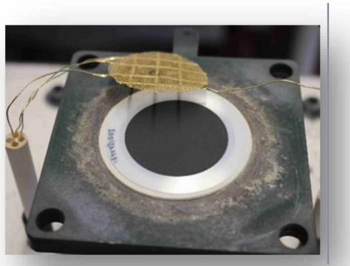
L. Wehrle, D. Schmider, J. Dailly, A. Banerjee, O. Deutschmann. Appl. Energy 317 (2022) 119143.

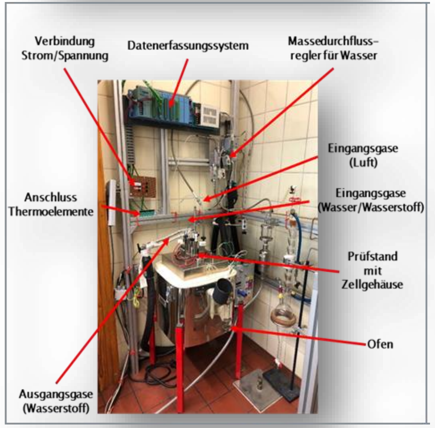
→ Poster 4.02 Furst et al.

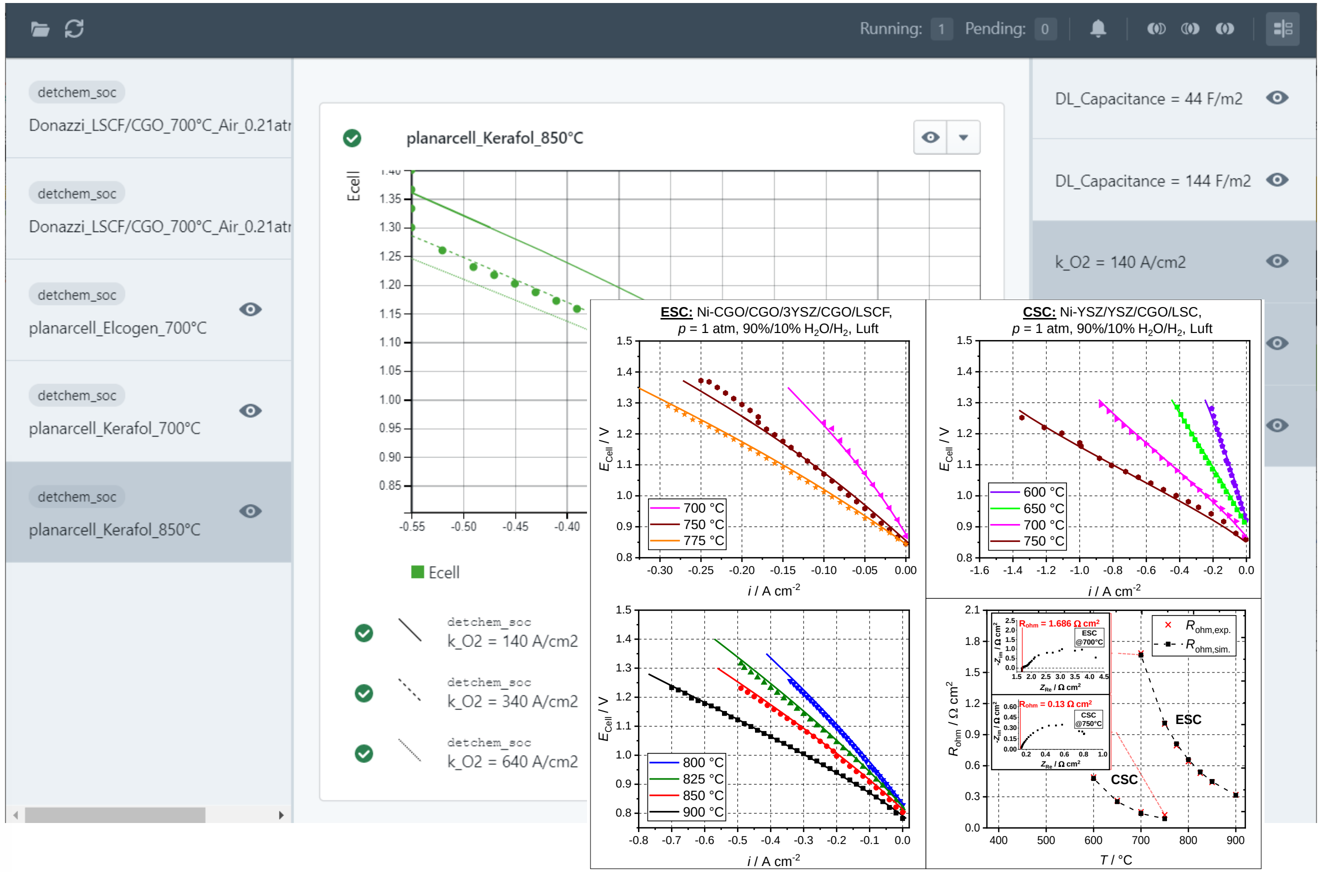
Hydrogen from high-temperature electrolysis: Digitalization tool CaRMeN accelerates parametrization

Electrochemical characterization

i-V EIS



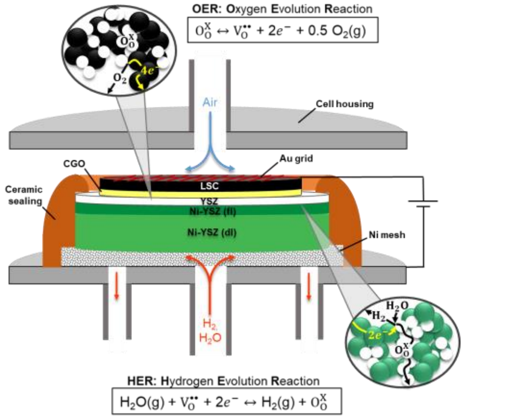




Electrochemical model parameters

Parameter	Value	
Cell type	ESC	CSC
Cathode material	Ni-CGO	Ni-YSZ
Exchange current density / (A cm^{-1} or A cm^{-2})	$5.9\times10^{3}\times e^{-111900/RT}\times p_{H2}^{0.01}\times p_{H2O}^{0.08}$	$0.68\times e^{-105000/RT}\times p_{H2}^{-0.1}\times p_{H2O}^{0.33}$
Anodic CT-coefficient	0.75	0.6
Cathodic CT-coefficient	0.25	0.4
Anode material	LSCF	LSC
Exchange current density / A cm^{-2}	$8.5\times10^{2}\times e^{-138000/RT}\times p_{O2}^{0.11}$	$7.6\times10^{5}\times e^{-150000/RT}\times p_{O2}^{0.22}$
Anodic CT-coefficient	0.5	0.65
Cathodic CT-coefficient	0.5	0.35

$$i_{0,H_2/H_2O} = A_{H_2/H_2O}\exp\left(-\frac{E_{H_2/H_2O}}{RT}\right) f(p_{H_2}, p_{H_2O})$$



L. Wehrle, D. Schmider, J. Dailly, A. Banerjee, O. Deutschmann. Appl. Energy 317 (2022) 119143.

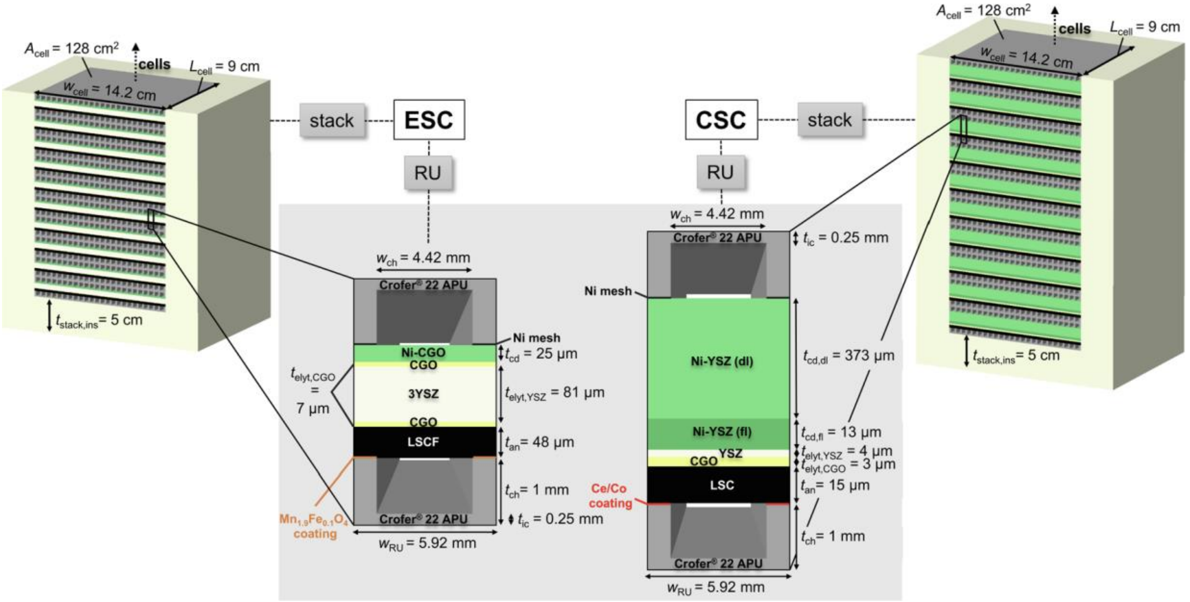
- Next step: Automatic parameter adaptation → Poster 1.01 Jägerfeld et al.

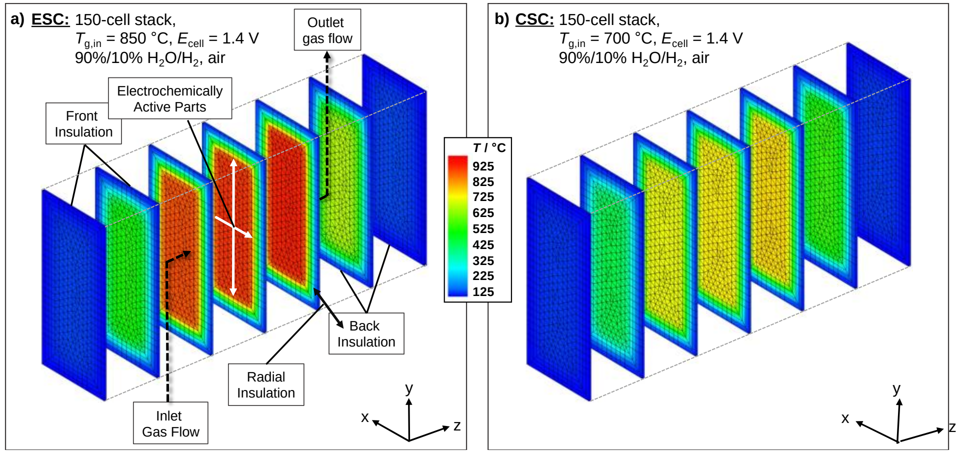
Hydrogen from high-temperature electrolysis: Digitalization and multi-scale modeling supports scale-up

Benchmarking SOEC-stack designs for industrial Power-to-Methane systems

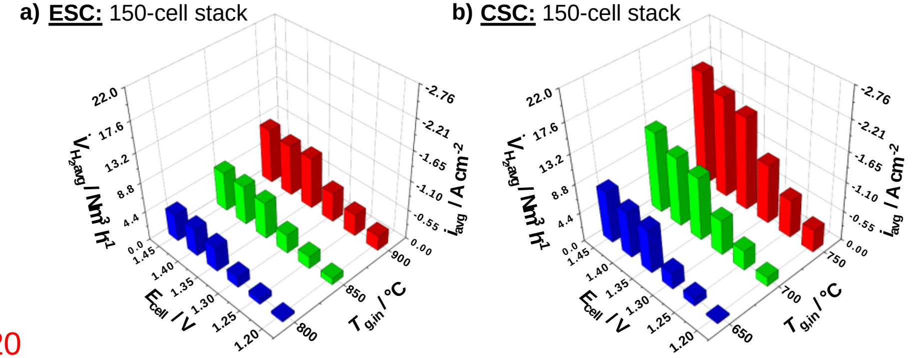
Electrolyte-supported cell (ESC)

Cathode-supported cell (CSC)





90% H_2O, 10% H_2, air, 80% steam conv.

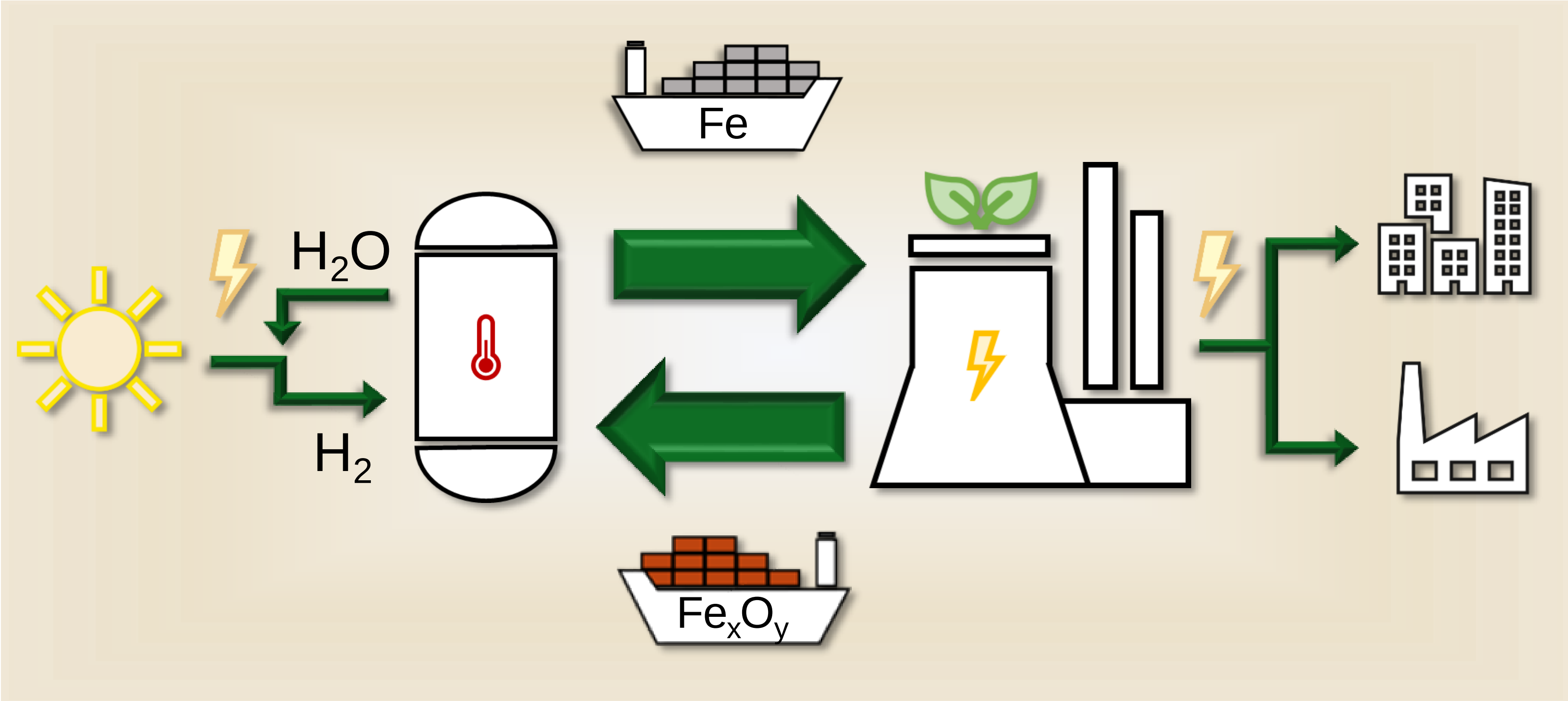


L. Wehrle, D. Schmider, J. Dailly, A. Banerjee, O. Deutschmann. *Appl. Energy 317 (2022) 119143.*

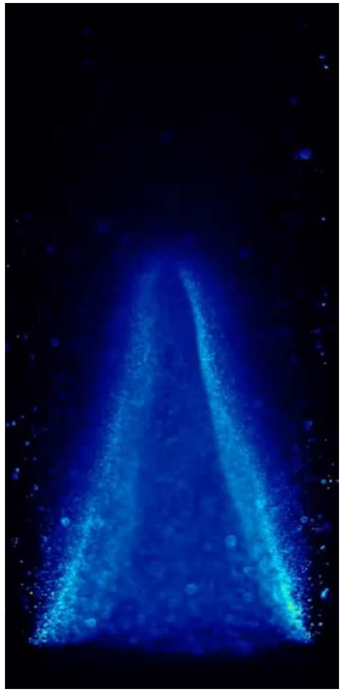
→ Lukas Wehrle, Tue 11:20

Reactive metals as carbon-free energy carriers for a circular energy economy: Iron cycle

- Metals: Promising solution for efficient storage of renewable energy in amounts of 100 TWh due to high volumetric energy density, easy handling, use of existing infrastructures



Iron powder flame

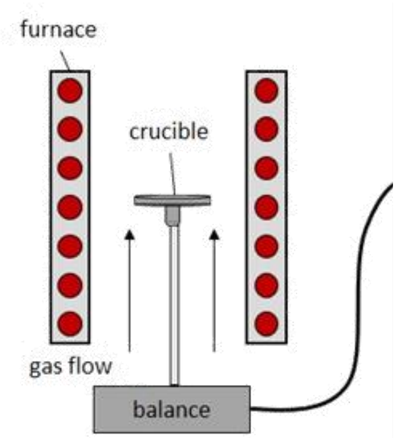


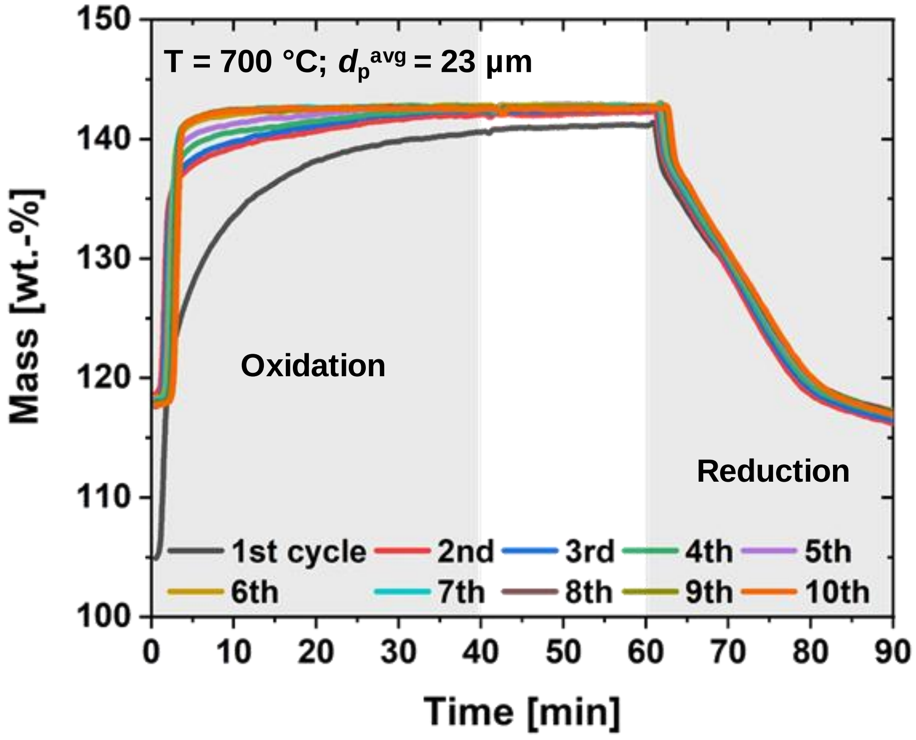
J.M. Bergthorson. *Prog. Energy Combust. Sci.* 68 (2018) 169.
P. Debiagi, J. Janicka, C. Hasse et al. *Renewable Sustainable Energy Rev.* 165 (2022) 112579.
C. Kuhn, A. Düll, P. Rohlfs, S. Tischer, M. Börnhorst, O. Deutschmann. *Appl. Energy and Combust. Sci.* 12 (2022) 100096.

M. Fedoryk, B. Stelzner, S. Harth, S., D. Trimis. Appl. Energy and Combust. Sci. 13 (2023)

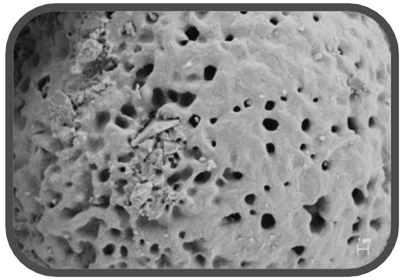
Iron as carbon-free energy carrier: Cycling demands a detailed understanding of kinetics and morphology

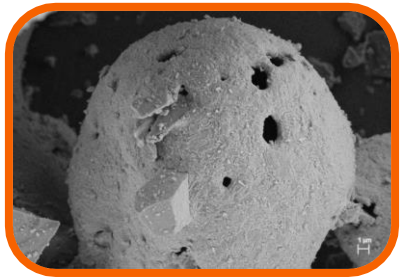
- TGA analysis: Cycling by oxidation with air and reduction with H_2 works
- Significant morphological and reaction rate variations



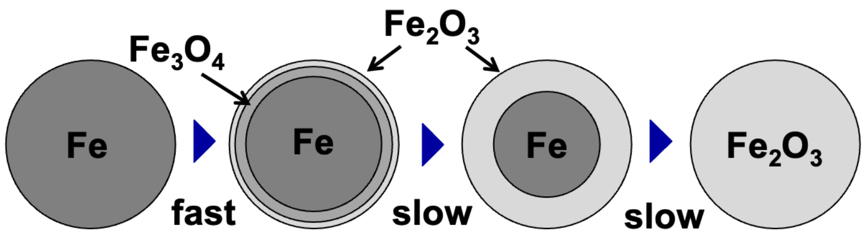


SEM images of iron particles after one and ten cycles



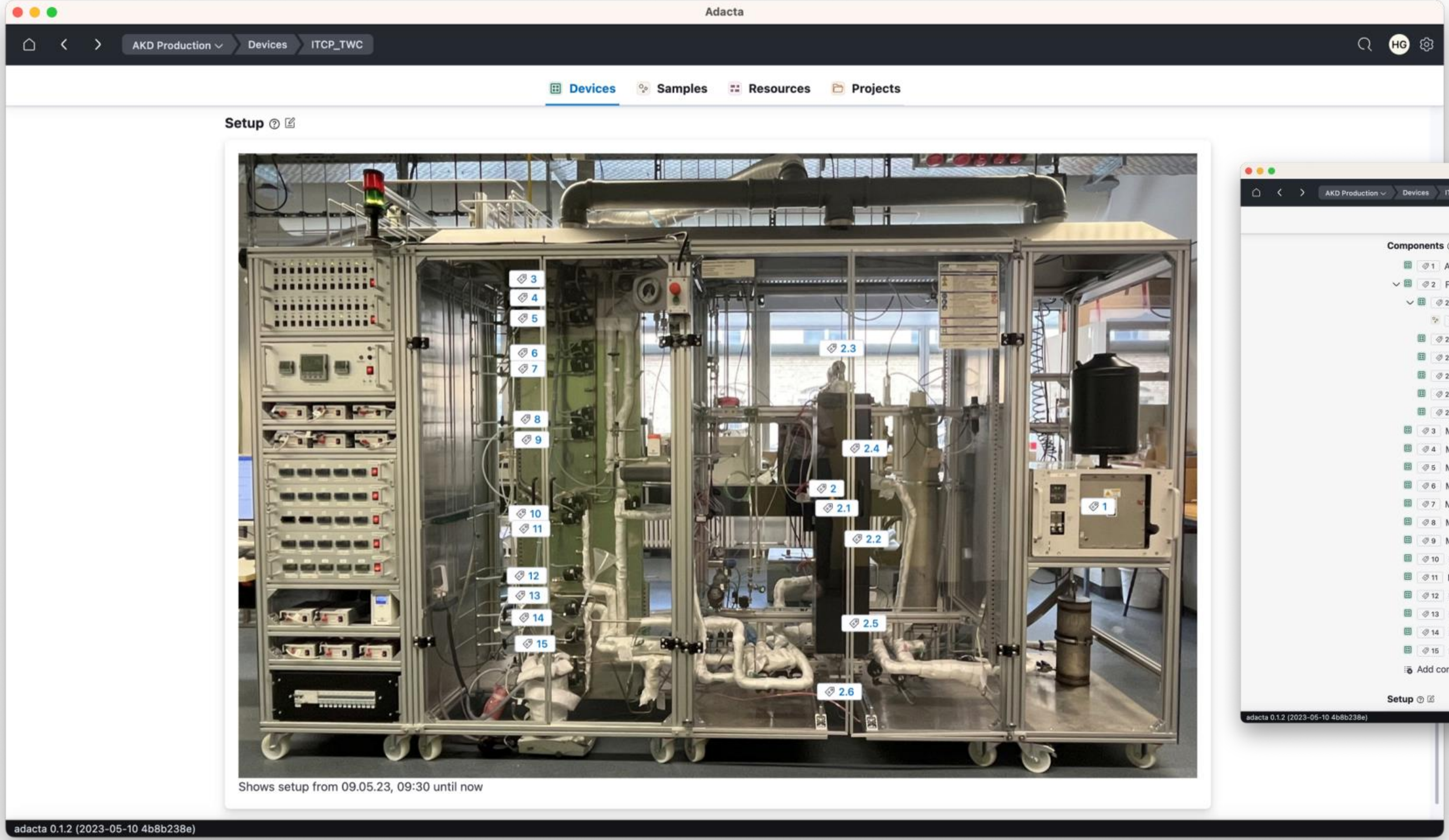


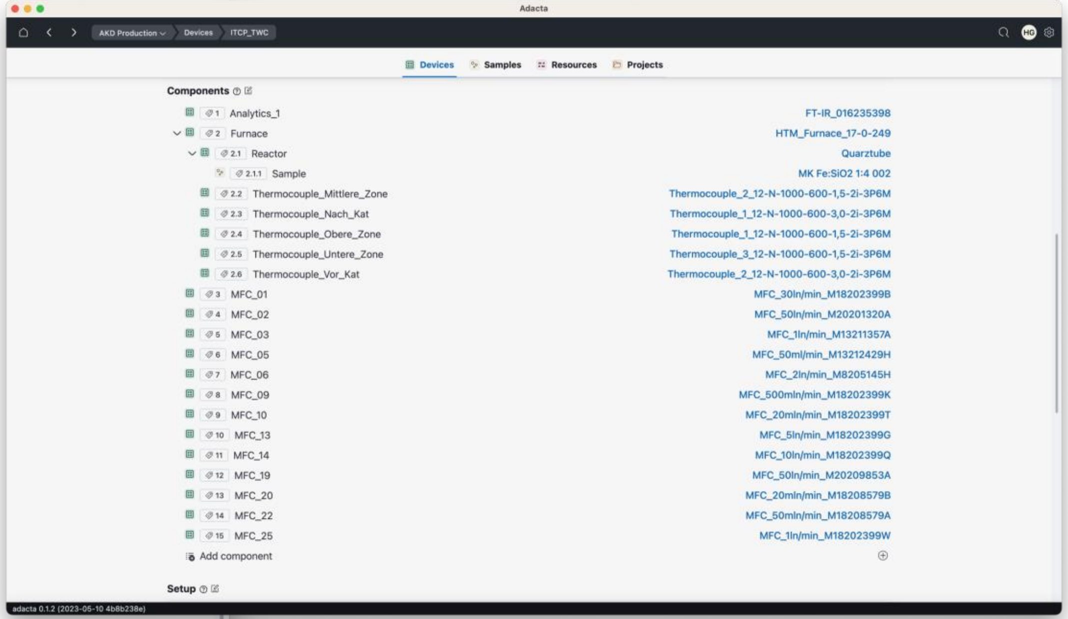
Particles morphology alteration



→ Poster 9.08 Kuhn, Knapp et al.

Iron as carbon-free energy carrier: Monitoring and archiving all experimental details by digital twin using Adacta



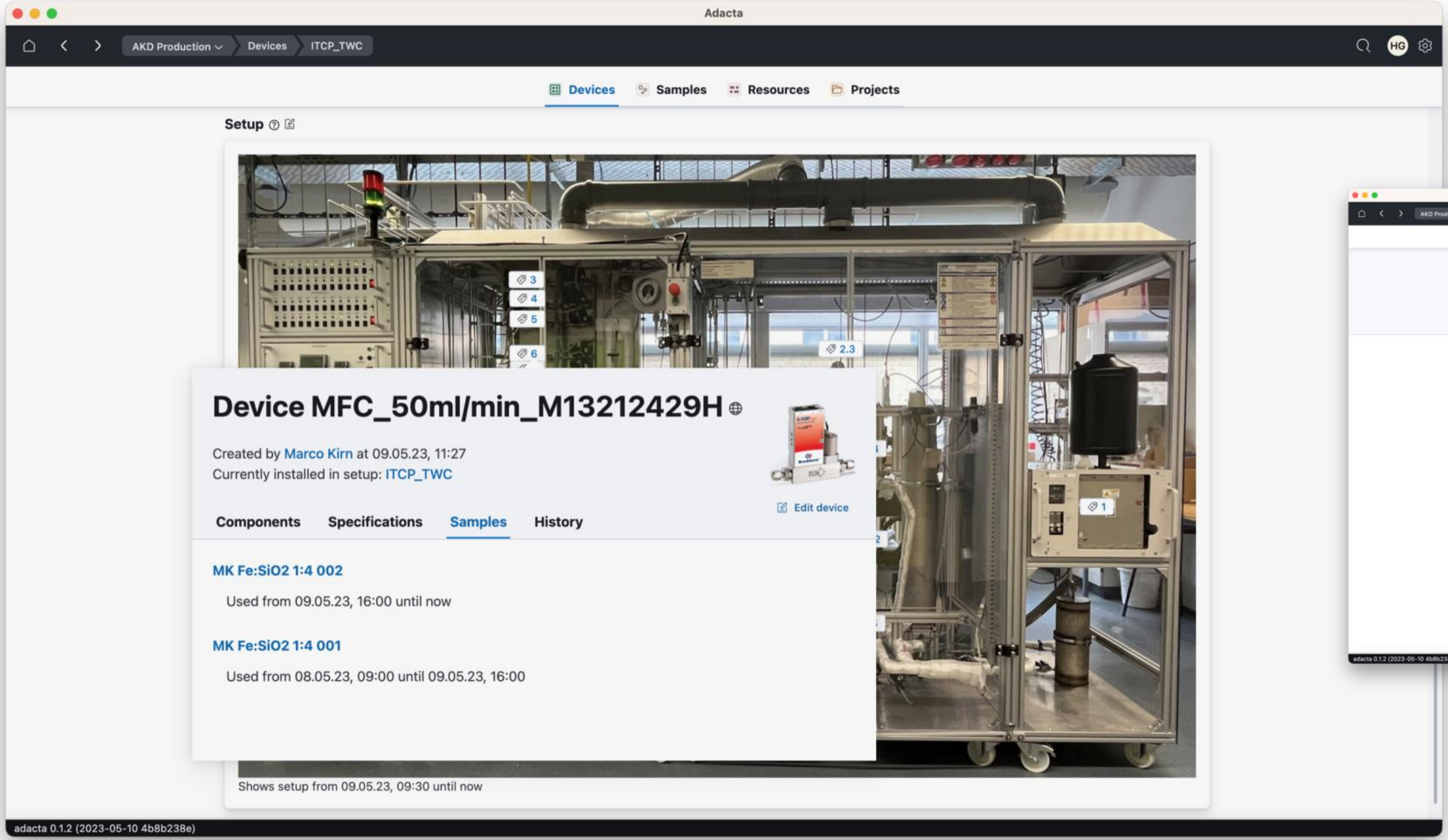


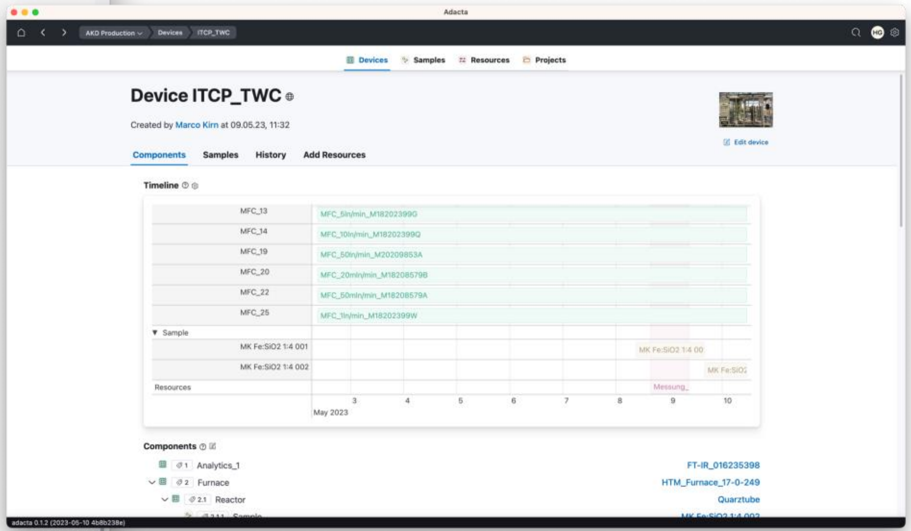
Configuration of experimental setup

H. Gossler, J. Riedel, E. Daymo, R. Chacko, S. Angeli, , O. Deutschmann. Chemie Ingenieur Technik 94 (2022) 1798.
www.omegadot.software/adacta

Iron as carbon-free energy carrier: Monitoring and archiving all experimental details by digital twin using Adacta

Karlsruhe Institute of Technology





All information on setup, devices, samples, operating conditions, performance data et al. at any time accessible from web

H. Gossler, J. Riedel, E. Daymo, R. Chacko, S. Angeli, , O. Deutschmann. Chemie Ingenieur Technik 94 (2022) 1798.
www.omegadot.software/adacta

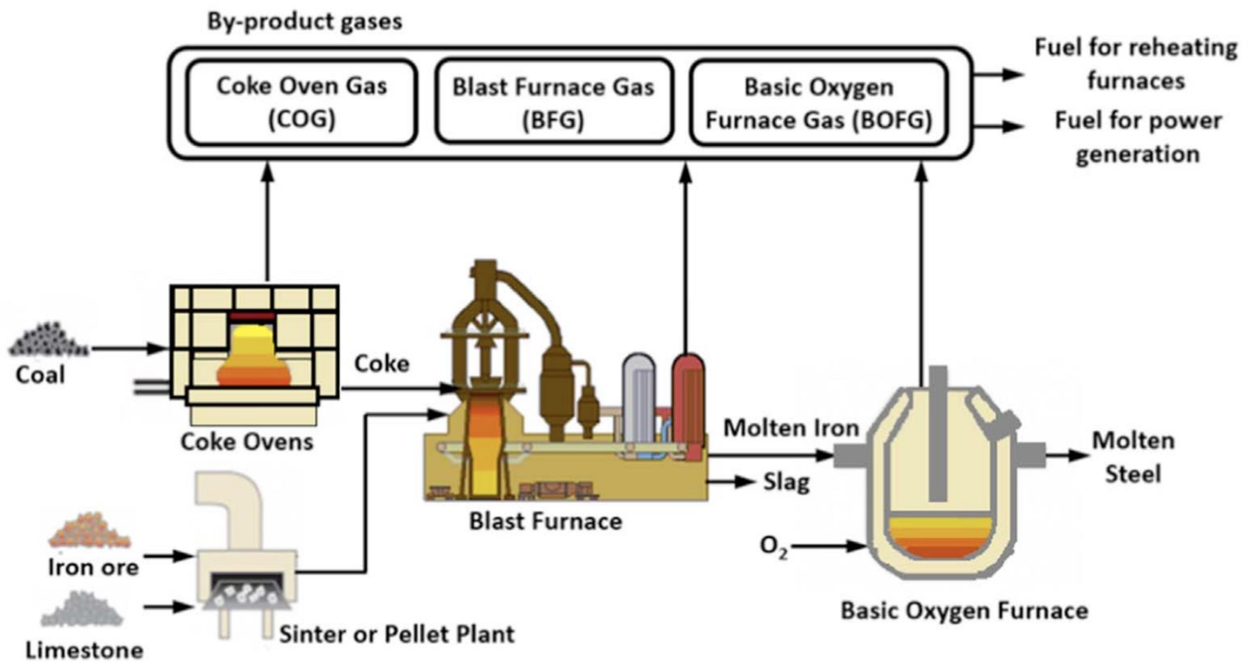
Reduction of CO_2 emission from flue gases of steel industry by dry reforming of methane and re-use in the process

Karlsruhe Institute of Technology

- Steel industry is responsible for 7–9% of the global direct CO_2 emissions
- 70% of steel production by coke-dependent blast furnace - basic oxygen furnace route

$$Fe_2O_3 \xrightarrow{C,\ H_2} Fe$$

- Steel plant off-gases contain value components



W. Uribe-Soto, J.-F. Portha et al., Ren. Sust. Energy Rev. 74 (2017) 809.

Feed name	COG	BFG
CH_4 (vol%)	→ 22.0	0.0
C_xH_y (vol%)	2.0	0.0
CO_2 (vol%)	1.2	→ 21.6
H_2O (vol%)	4.0	4.0
H_2 (vol%)	60.7	3.7
CO (vol%)	4.1	23.5
N_2 (vol%)	5.8	46.6
Ar + O_2 (vol%)	0.2	0.6

S. Angeli, S. Gossler, S. Lichtenberg, G. Kass, A. Agrawal, M. Valerius, K. P. Kinzel, O. Deutschmann. Angew. Chemie Intl. Ed. 60 (2021) 11852.

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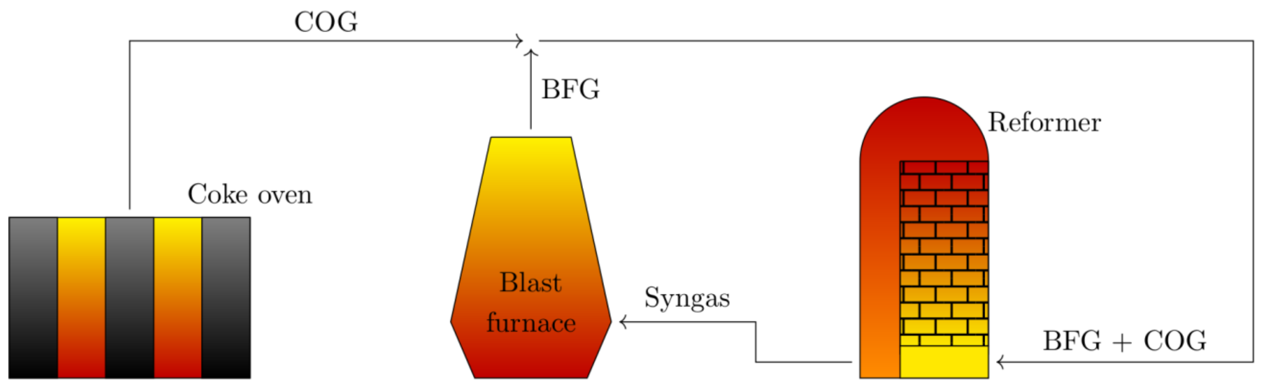
$$Fe_2O_3 \xrightarrow{C,\ H_2} Fe$$

- Use of high-temperature Coke Oven Gas (COG) and Blast Furnace Gas (BFG)
- "Dirty" components of COG and BFG would poison the catalyst
- Reforming of COG/BFG mixtures in the regenerative heat exchanger (cowper) at 800°C-1400°C
- Syngas use for iron ore reduction in blast furnace

$$CH_4 + CO_2 \rightarrow 2\ H_2 + 2\ CO$$



P. Blanck, G. Kass, B. Kanz, K.P. Kinzel, O. Deutschmann. Energy Adv., subm .

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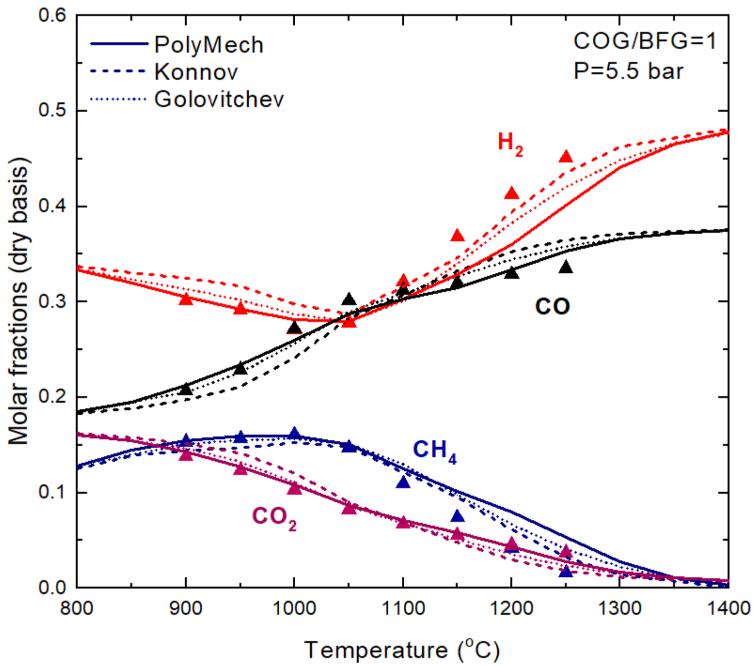
Reduction of CO_2 emission from flue gases of steel industry
Digitalization used for understanding and process development

Karlsruhe Institute of Technology

- Understanding reaction kinetics in the gas-phase and model development for scale-up
- Four models available in literature but none of them ever tested for these conditions
  - PolyMech [1]: 558 reactions, 83 species
  - Golovichev [2]: 690 reactions, 130 species
  - Konnov [3]: 1231 reactions, 129 species
  - AramcoMech [4]: 3037 reactions, 581 species

$$CH_4 + CO_2 \rightarrow 2\,H_2 + 2\,CO$$

[1] S. Porras, C. Schulz, U. Maas et al., Combust. Flame 212 (2020) 107.
[2] V. I. Golovitchev, F. Tao, J. Chomiak, SAE Tech. Pap. 1999-01-3552.
[3] A. A. Konnov, Combust. Flame 156 (2009) 2093.
[4] C.-W. Zhou, H.J. Curran et al., Combust. Flame 197 (2018) 423.

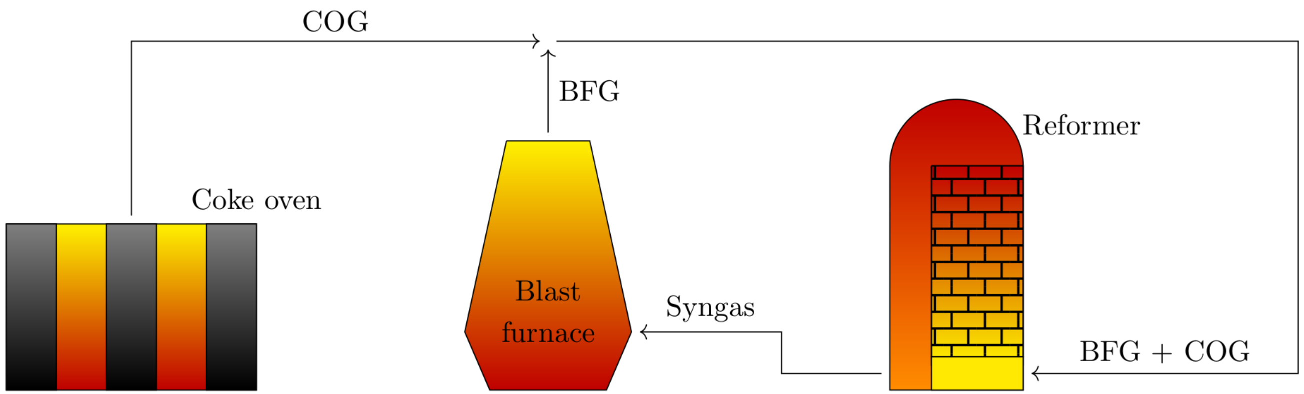


S. Angeli, S. Gossler, S. Lichtenberg, G. Kass, A. Agrawal, M. Valerius, K. P. Kinzel, O. Deutschmann. Angew. Chemie Intl. Ed. 60 (2021) 11852.

Reduction of CO_2 emission from flue gases of steel industry
Digitalization tools support scale-up and operation strategies

Karlsruhe Institute of Technology

- Maximum reduction of 78% CO_2 can be achieved
- Potential of reducing 12 % of steel plant CO_2 emissions
- World-wide implementation → 0.5% less global CO_2 emissions
- DETCHEM and CaRMeN now used for scale-up by industry



S. Angeli, S. Gossler, S. Lichtenberg, G. Kass, A. Agrawal, M. Valerius, K. P. Kinzel, O. Deutschmann. Angew. Chemie Intl. Ed. 60 (2021) 11852.

→ Poster 9.06 Blanck et al.

Pilot plant at Dillinger Hütte

PAUL WURTH SMS group

P. Blanck, G. Kass, B. Kanz, K.P. Kinzel, O. Deutschmann. Energy Adv., subm .

Digitalization in Catalysis and Reaction Engineering
Outline

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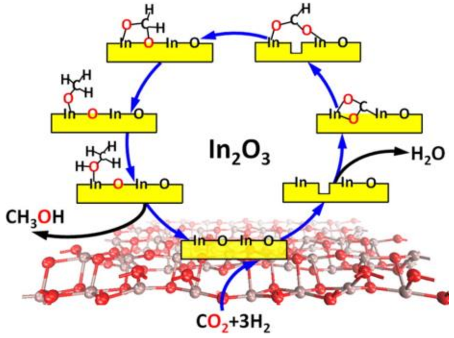
- Democratization of tools and services: RDM @ NFDI4Cat (SAS)

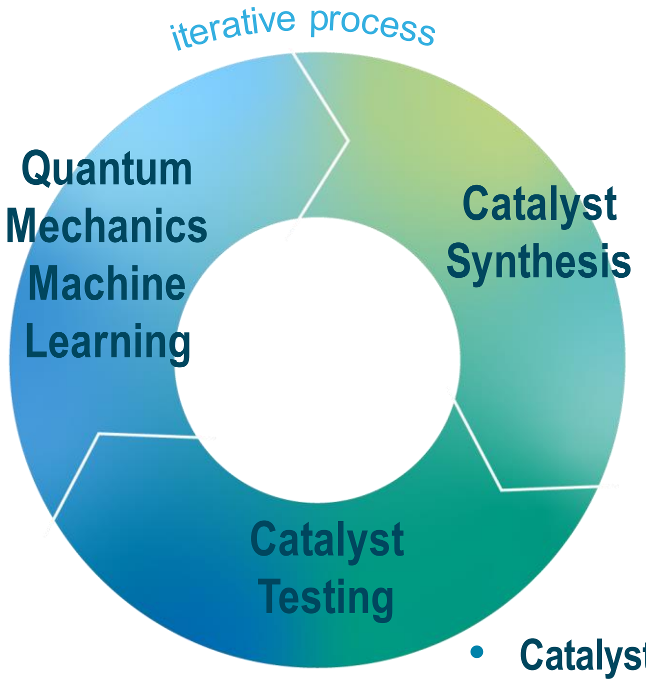
Case Study

Mixed Metal Oxides as Potential Candidates for CO_2 to MeOH

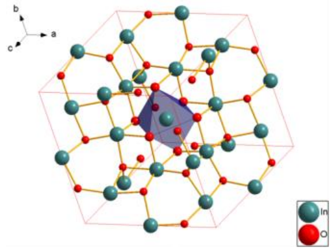
Scope: Development of a predictive tool for catalyst design by correlation of catalyst properties and performance Identification of a competitive catalyst technology for CO_2 to MeOH

- **Model development** (e.g., DFT) and candidate library generation
- **Machine Learning algorithms** to accelerate QM simulations and analyze huge number of structures and compositions
- **Machine learning to find descriptor** to correlate the structure properties with catalytic activity





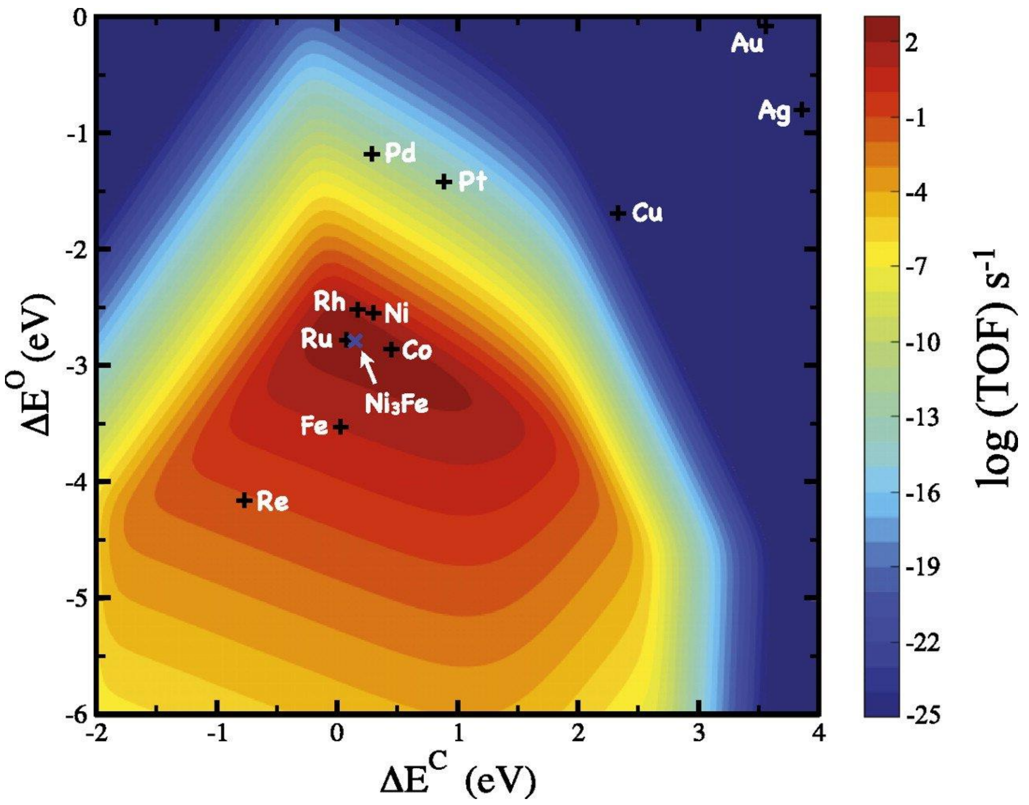
- **Catalyst synthesis** (as bulk material) and characterization
- **Synthesis of supported catalysts** of the most promising materials (including ALD)



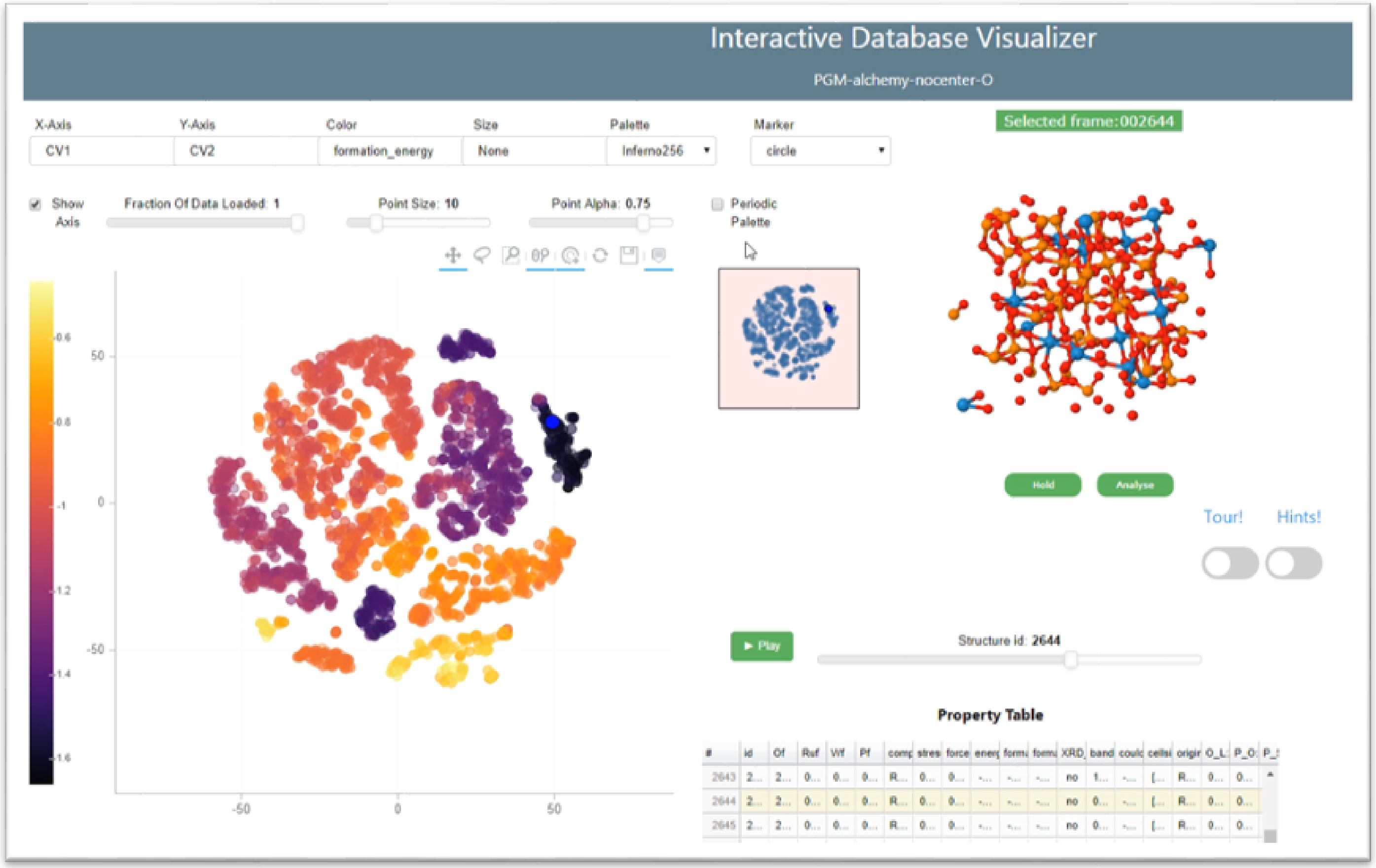
- **Catalyst testing** in high throughput plant: for fast screening and stability tests
- **Kinetic studies** in the profile reactor

Identify Opportunity by Finding Patterns

Theoretical volcano for the production of methane from syngas, CO, and H_2



https://doi.org/10.1073/pnas.1006652108

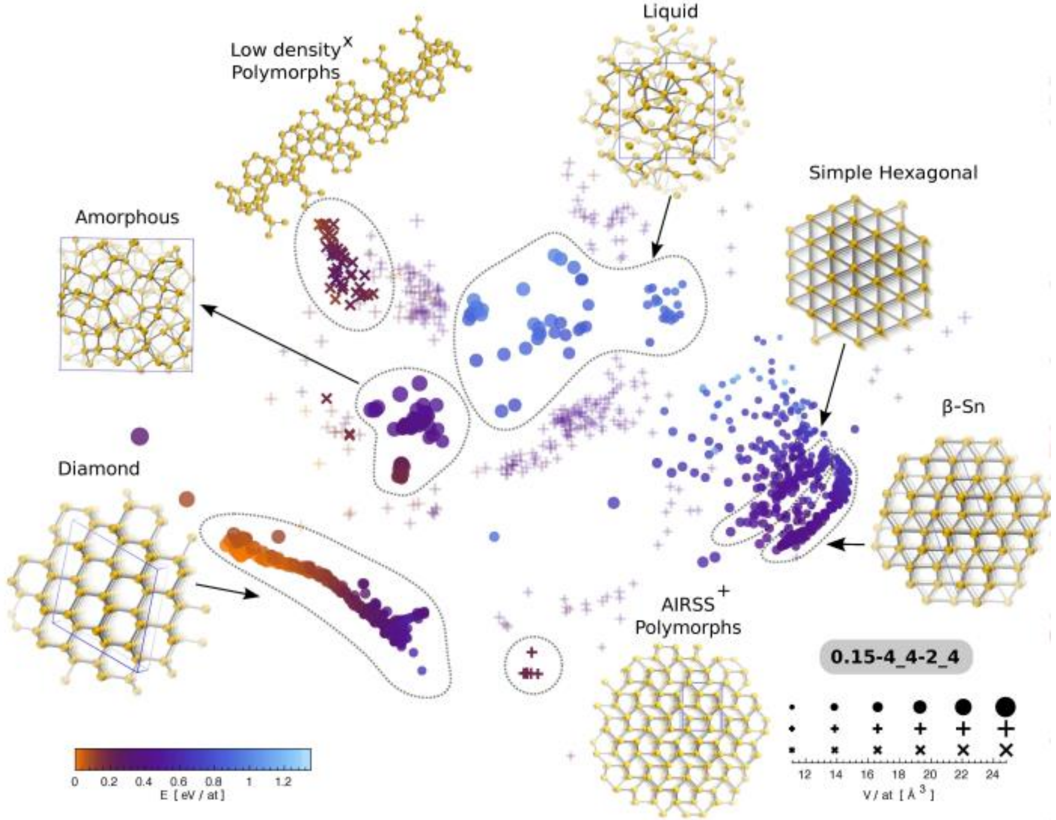


https://github.com/sandipde/Interactive-Sketchmap-Visualizer

Identify Opportunity by Finding Gaps

			Ti = 50	Zr = 90	? = 180
			V = 51	Nb = 94	Ta = 182
			Cr = 52	Mo = 96	W = 186
			Mn = 55	Rh = 104,4	Pt = 197,4
			Fe = 56	Ru = 104,4	Ir = 198
			Ni = Co = 59	Pd = 106,6	Os = 199
H = 1			Cu = 63,4	Ag = 108	Hg = 200
	Be = 9,4	Mg = 24	Zn = 65,2	Cd = 112	
	B = 11	Al = 27,4	? = 68	Ur = 116	Au = 197?
	C = 12	Si = 28	? = 70	Sn = 118	
	N = 14	P = 31	As = 75	Sb = 122	Bi = 210?
	O = 16	S = 32	Se = 79,4	Te = 128?	
	F = 19	Cl = 35,5	Br = 80	J = 127	
Li = 7	Na = 23	K = 39	Rb = 85,4	Cs = 133	Tl = 204
		Ca = 40	Sr = 87,6	Ba = 137	Pb = 207
		? = 45	Ce = 92		
		?Er = 56	La = 94		
		?Yt = 60	Di = 95		
		?In = 75,6	Th = 118?		

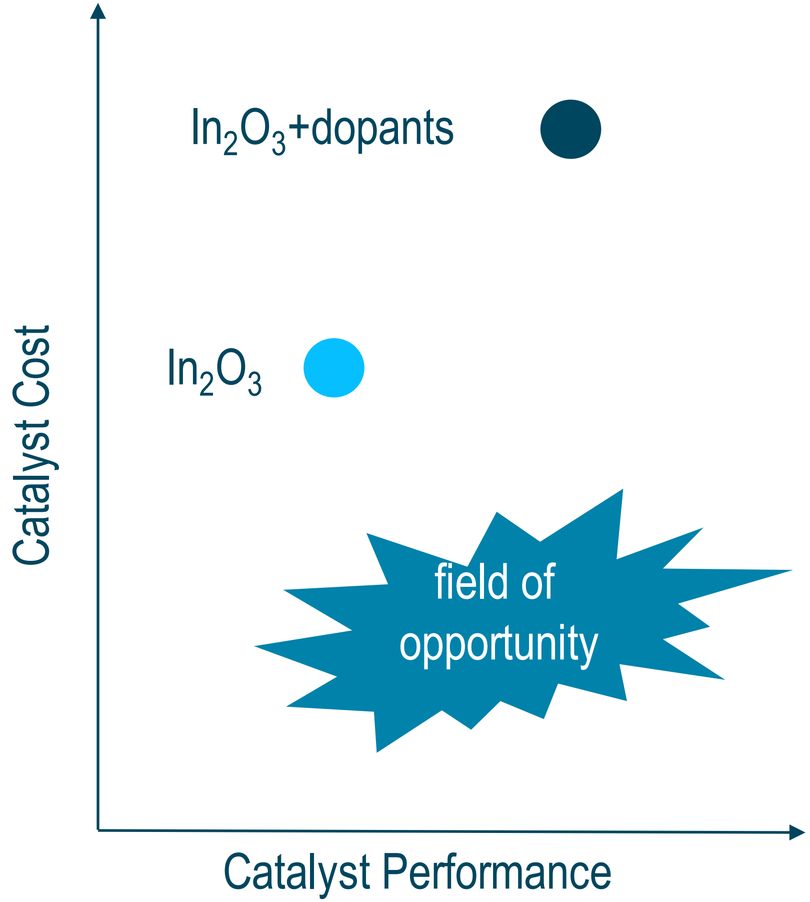
G. Arthur Montreal, CC BY-SA 4.0 via Wikimedia Commons
https://commons.wikimedia.org/wiki/File:First_Version_of_the_Periodic_Table.gif



http://interactive.sketchmap.org/
Comparing molecules and solids across structural and alchemical space, Sandip De et al. *Phys. Chem. Chem. Phys.*, 2016,18, 13754-13769

CO_2 to Methanol

Cost Performance Tradeoff



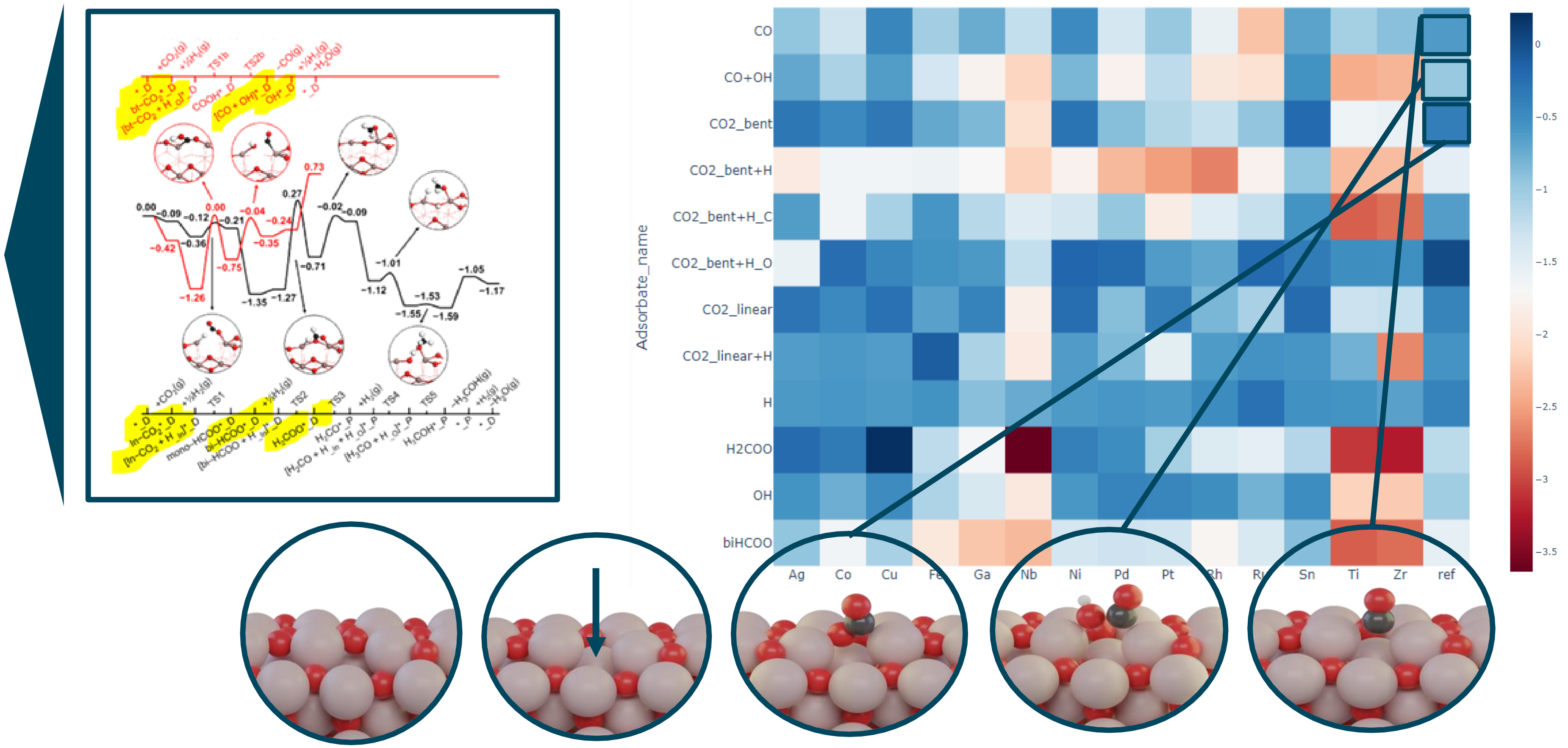
- Costs hard to reduce further
- Patent space is crowded

- Opportunity for IP
- Opportunity for low-cost materials

- Exploit synergy between theory and experiment
- Explore agile ways of combining high throughput simulation and high throughput experiments
- Harvest benefits of AI and Big Data

Probe into In_2O_3+ dopants

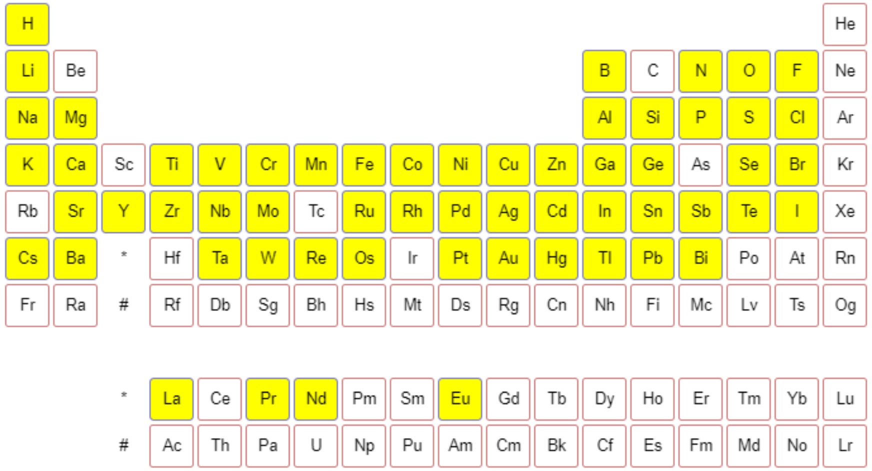
- Surface analysis
- Vacancy formation
- Substitutional doping focus on In_2O_3 (110)
- Reaction intermediates



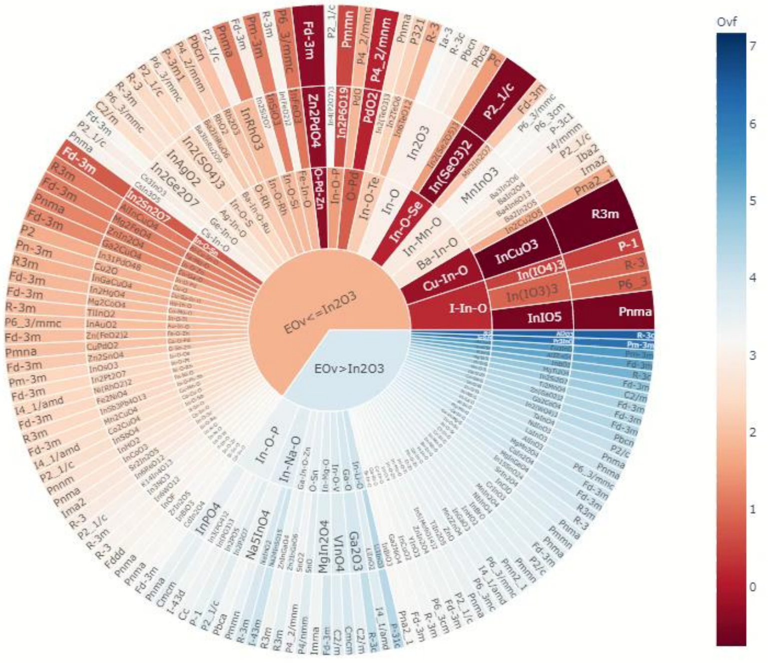
In_2O_3 (1,1,0) O - vacancy

Utilize existing Databases for High Throughput Screening – Compute when necessary!

Calculate key descriptors like vacancy (Ov) formation energies and adsorption energies.

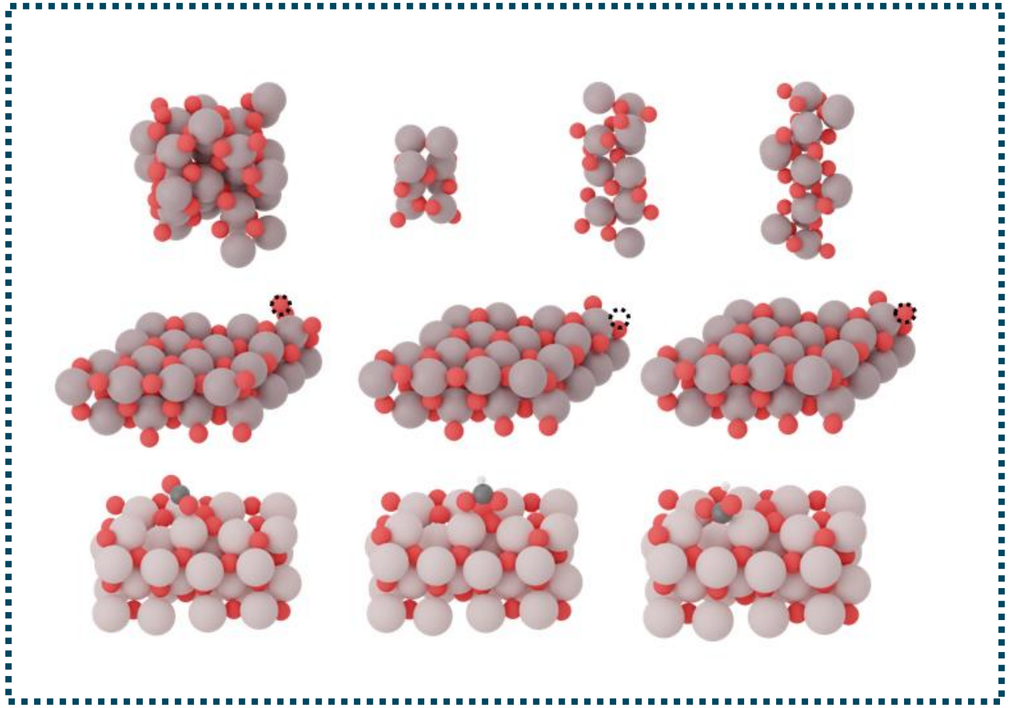


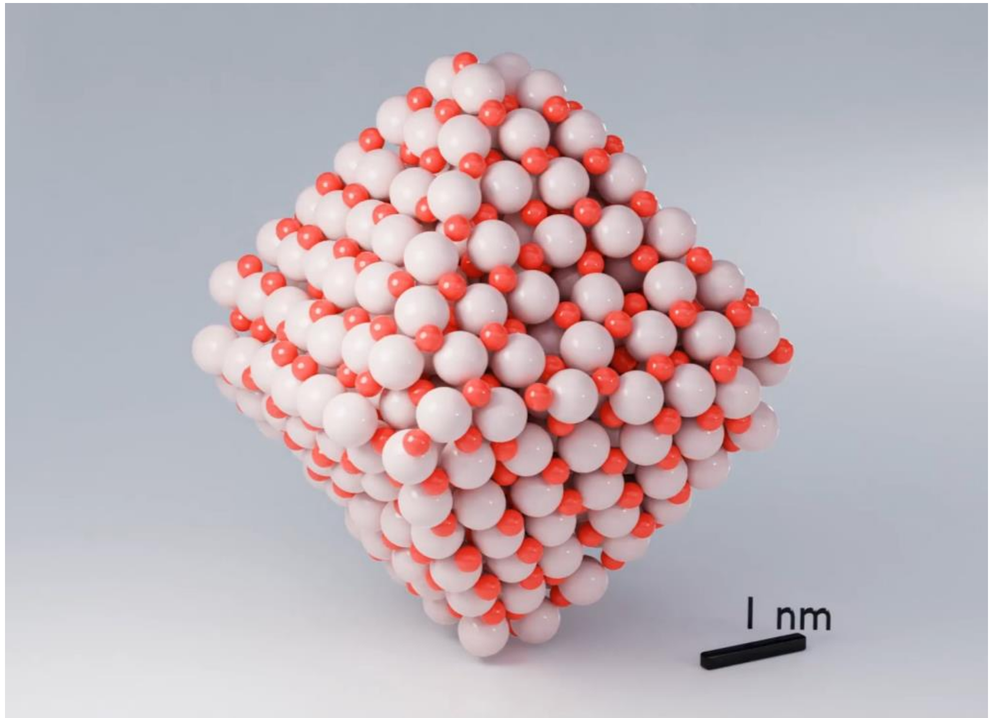
- Not everything is there in databases!
- Compute what we need quickly with high-throughput computation



Glimpse in defect formation energy database

Machine learning at Atomic Scale:
Complexities of Interatomic Interactions

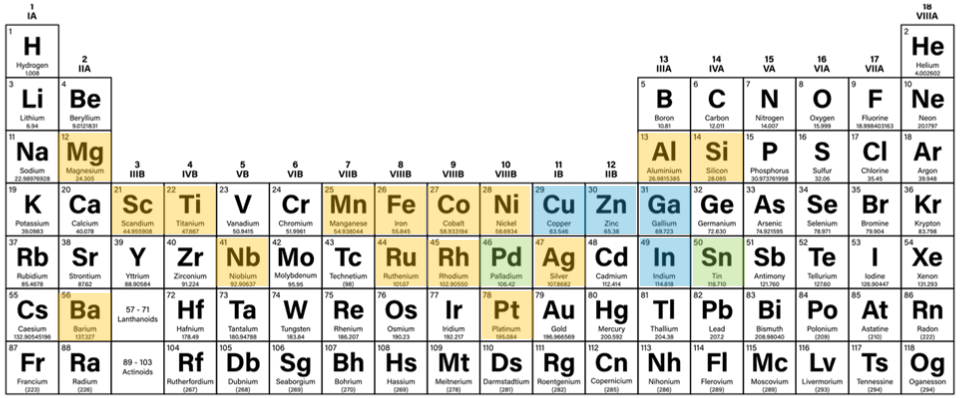


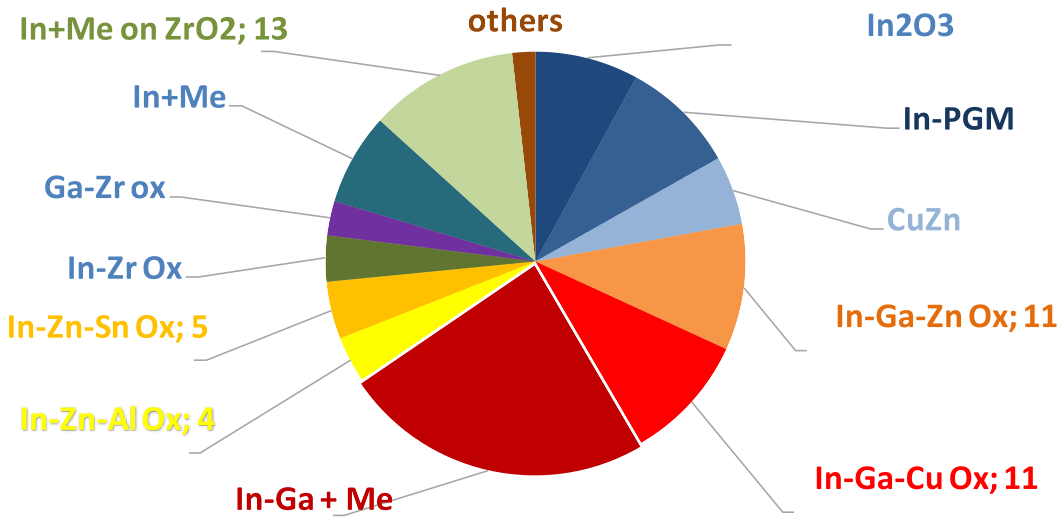


- ML model is 10^3-10^6x faster than reference QM
- Improving at each iteration and converges ~ 5-10 iterations
- ML training takes ~10-24 hours

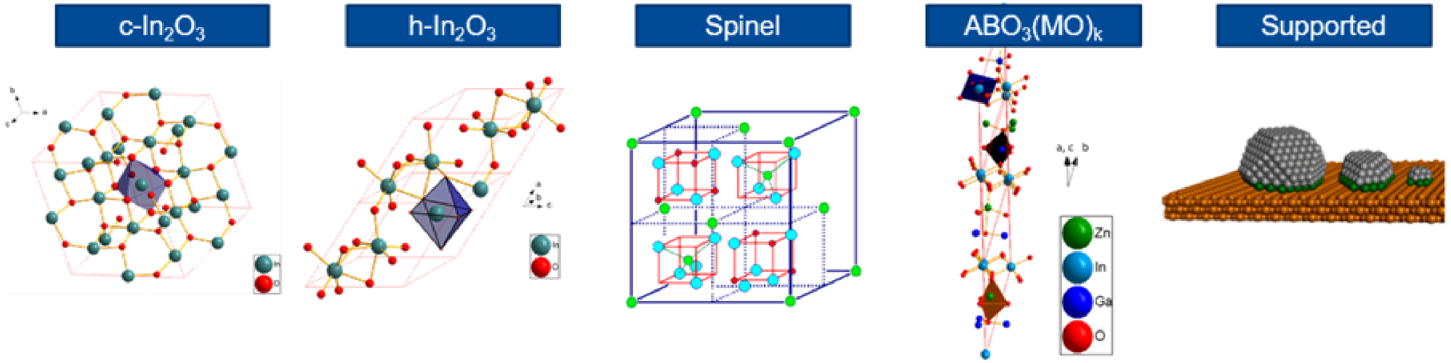
Crystalline Solids predicted by Quantum Chemistry Simulations enabling Understanding of Structure-Performance Relationships

Candidate Elements for Mixed Metal Oxides in the Light of the Target Reaction



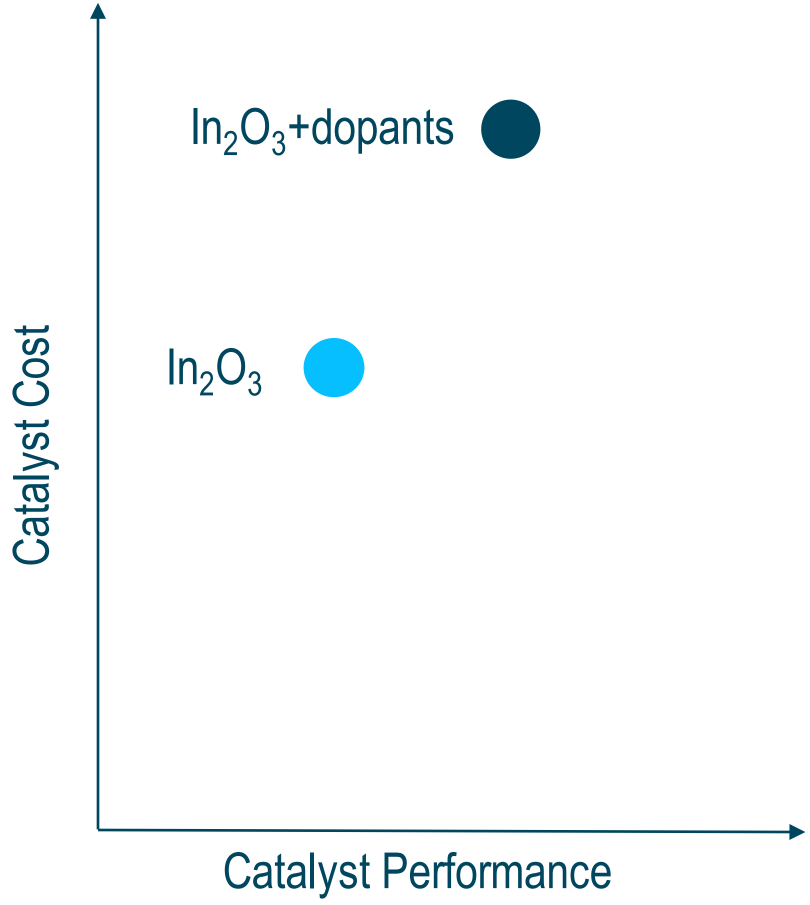


Structure Field Map Proposed for the Target Reaction:



CO_2 to Methanol

Cost Performance Tradeoff



- Costs hard to reduce further
- Patent space is crowded

- ✓ Large IP space available
- ✓ Low-cost materials developed

- Synergy between theory and experiment proven ✓
- Agile ways of combining high throughput simulation and high throughput experiments pay off ✓
- Benefits of AI and Big Data could be harvested ✓

H. Ahi, C. Boscagli, S. De, E. C. Frei, M. Mueller, S. A. Schunk WO002022248460A2

Digitalization in Catalysis and Reaction Engineering
Outline

- Introduction –long history of digitalization in science and engineering (OD)
- Automated workflows on the lab scale – models for heterogeneous catalysis (OD)
- Feedback loops –from lab to pilot plant (SAS)
- Application of digital tools for scale-up on industrial scale (OD)
- Prediction of materials properties on atom scale using AI for catalyst discovery (SAS)
- **Democratization of tools and services: RDM @ NFDI4Cat (SAS)**

NFDI$_4$Cat Vision - Digital Catalysis

A connected community (academia and industry) in catalysis related sciences enabling interlinked knowledge through digital catalysis. Faster innovation through the potential application of artificial intelligence.

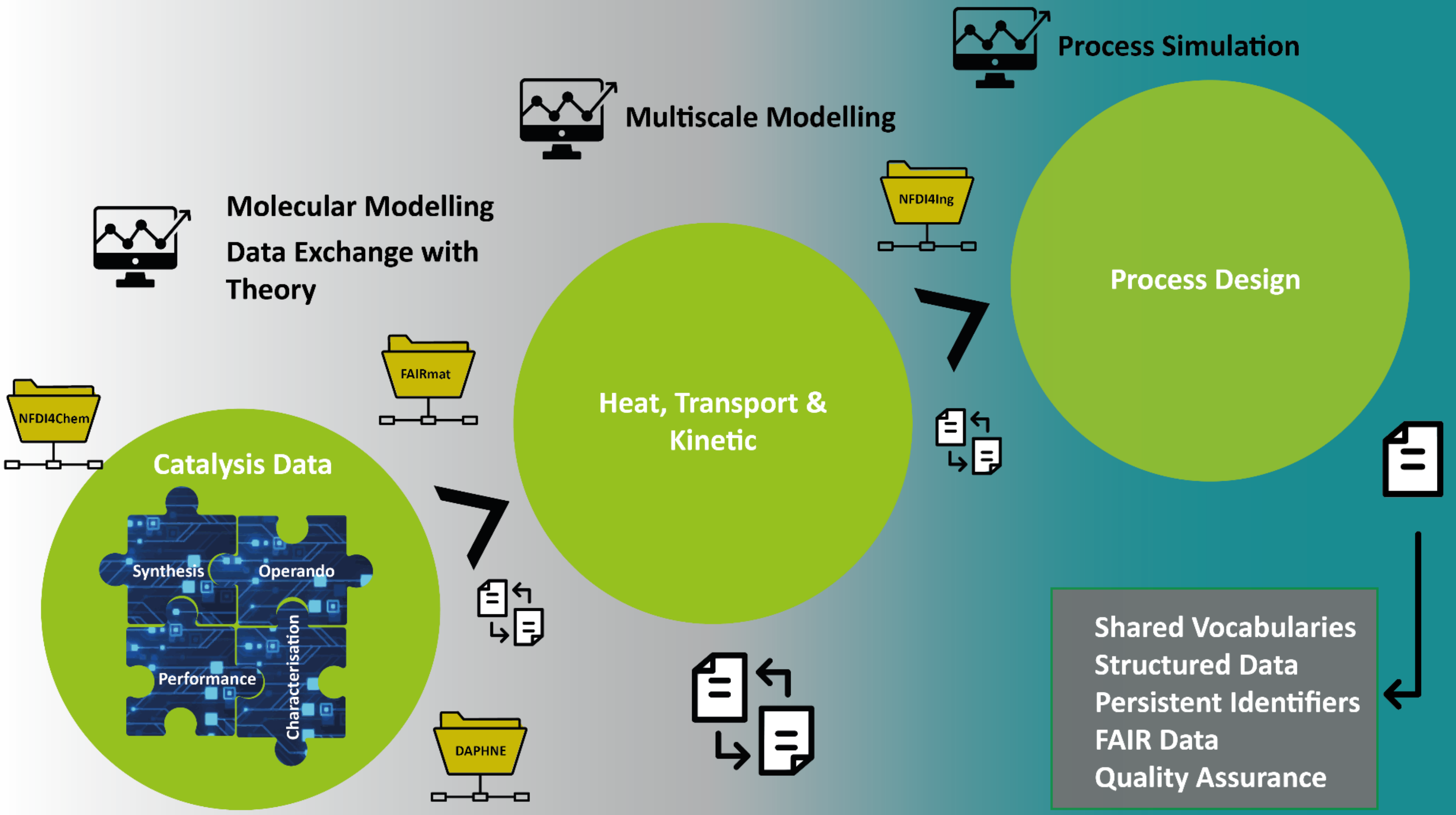
A community with an established research data management: research data planning, collection, archiving are carried out according to the FAIR principles by providing repositories, tools, services, and training.

Easy exchange of homogeneously annotated data and structured knowledge through the creation of shared vocabularies, the use of ontologies and persistent identifiers.

Ensuring the lifelong availability of catalysis research data by establishing a shared and understandable knowledge base.

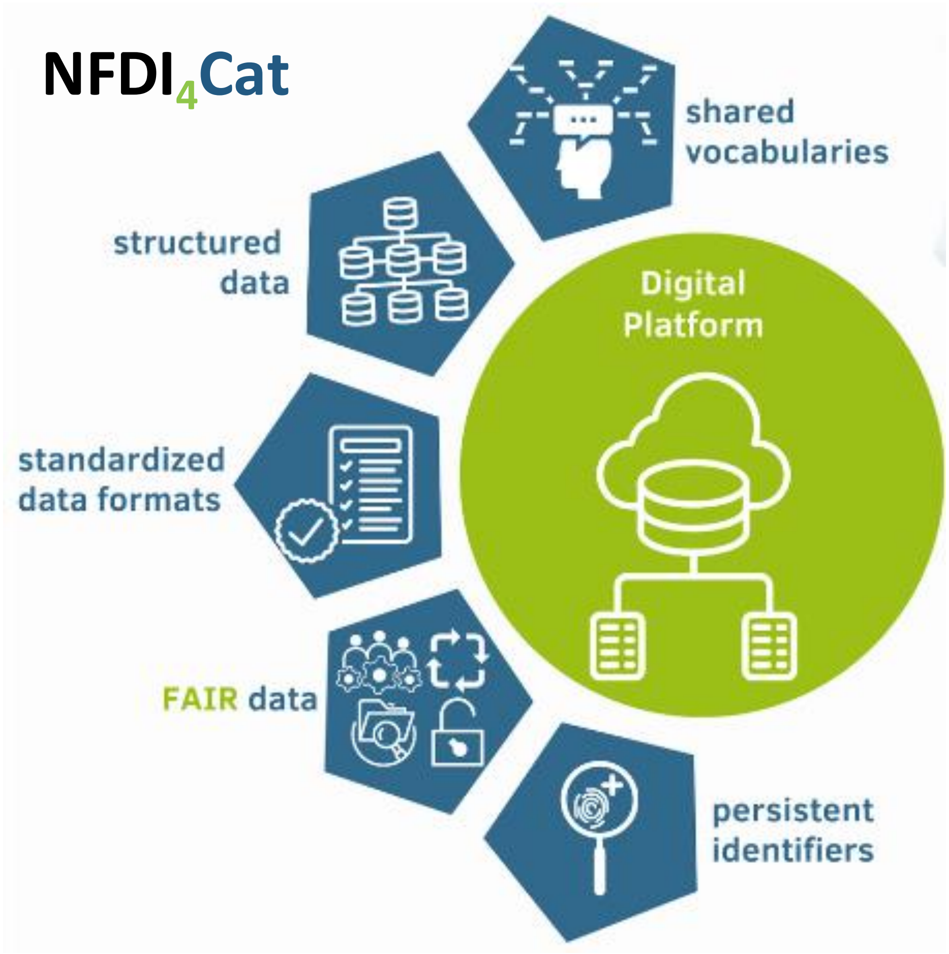
The Value Chain of Digital Catalysis

We are committed to realise the paradigm shift towards digital catalysis in catalysis related sciences along the data value chain and aligned with the real value chain "from molecule to chemical process".



Open Science and Digitalisation

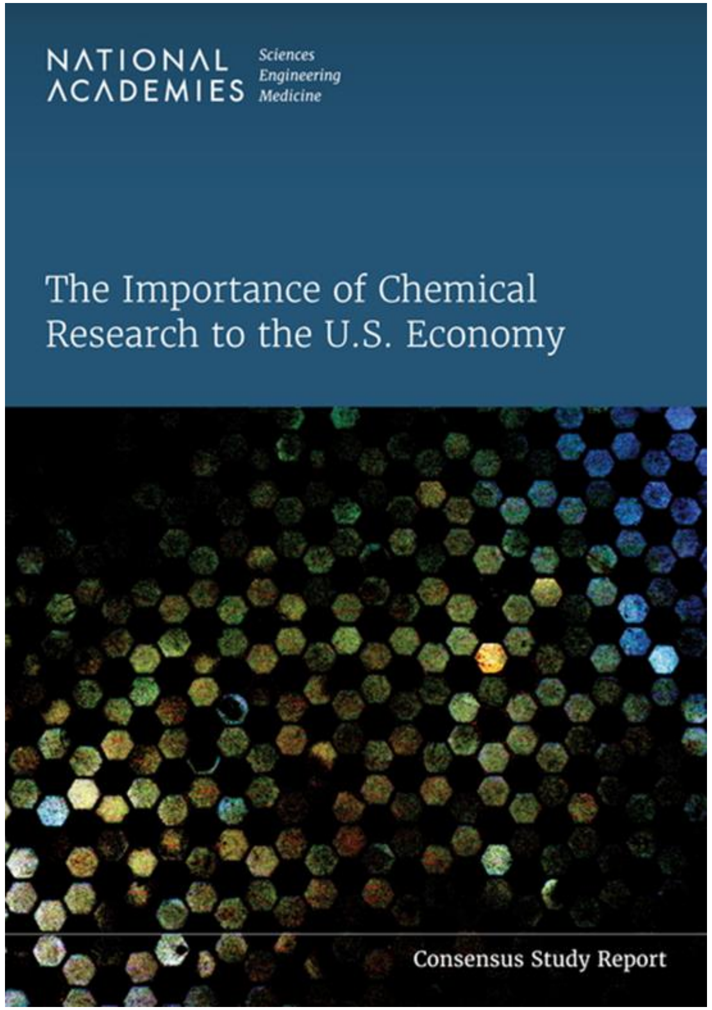
the drivers in *catalysis* and *chemical engineering*



NFDI4Cat – facilitator of cooperation between academia and industry in the field of digital catalysis

NFDI4Cat – the digital enabler for sustainable production of chemicals and energy carriers

NFDI4Cat – Enabler for Enhanced Catalysis Development



The urgent need to tackle sustainability-related challenges is driving the **need to improve the pace and efficiency of discovering new catalysts and developing** new catalytic processes. Toward this end, available **materials, adsorption, and reaction data** are being harnessed using high-throughput computation and ML to gather further insights and predict new materials (Bo et al., 2018). However, this knowledge extraction approach is challenged by a data deficit, or non-uniform reporting of materials-related data that are not computer readable (Himanen et al., 2019). Wulf et al. (2019) note that “in order to make data widely useful, rather advanced and well-coordinated approaches are needed that are beyond what a single group or institution can develop and sustain.” **Digitalization of the catalysis field is essential** to “enable efficient data-driven interdisciplinary development of catalysts and catalytic processes” (Wulf et al., 2019). The **NFDI4Cat** (National Research Data Infrastructure for Catalysis-Related Sciences), supported by the German government, **is an example of such a large-scale effort**. Establishment of an “internet of catalysis,” will guide research along the development chain from molecules to chemical processes.

The creation of digital workflows that bridge theory and experimental studies in catalyst design, characterization, kinetics, and related engineering aspects will accelerate discovery and innovation in the catalysis sciences.

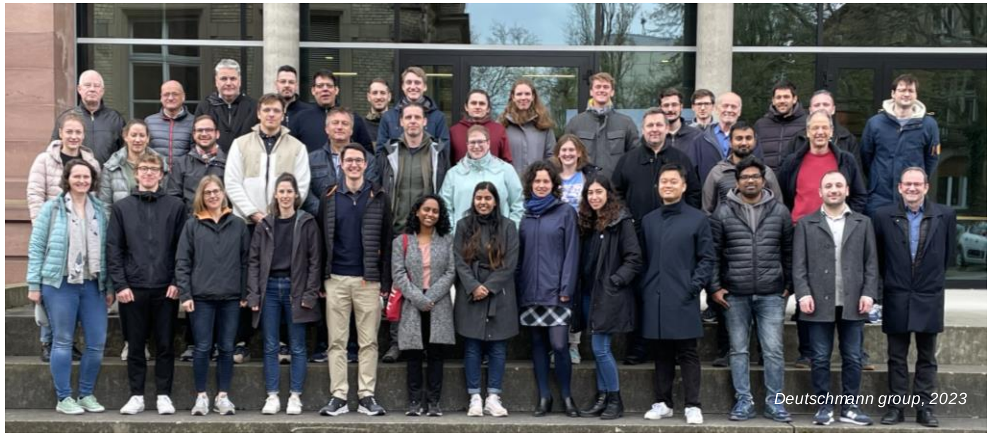
Digitalization in Catalysis and Reaction Engineering: More than just a Buzzword!

Karlsruhe Institute of Technology

hte – the high throughput experimentation company

NFDI4Cat

Acknowledgements


Deutschmann group, 2023

Thank you for your attention

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