18th ICC - INTERNATIONAL CONGRESS ON CATALYSIS

Lyon, France 19.07.24

ACCELERATING MULTI-SCALE MODELING OF CATALYTIC DEVICES THROUGH MACHINE LEARNING SURROGATES

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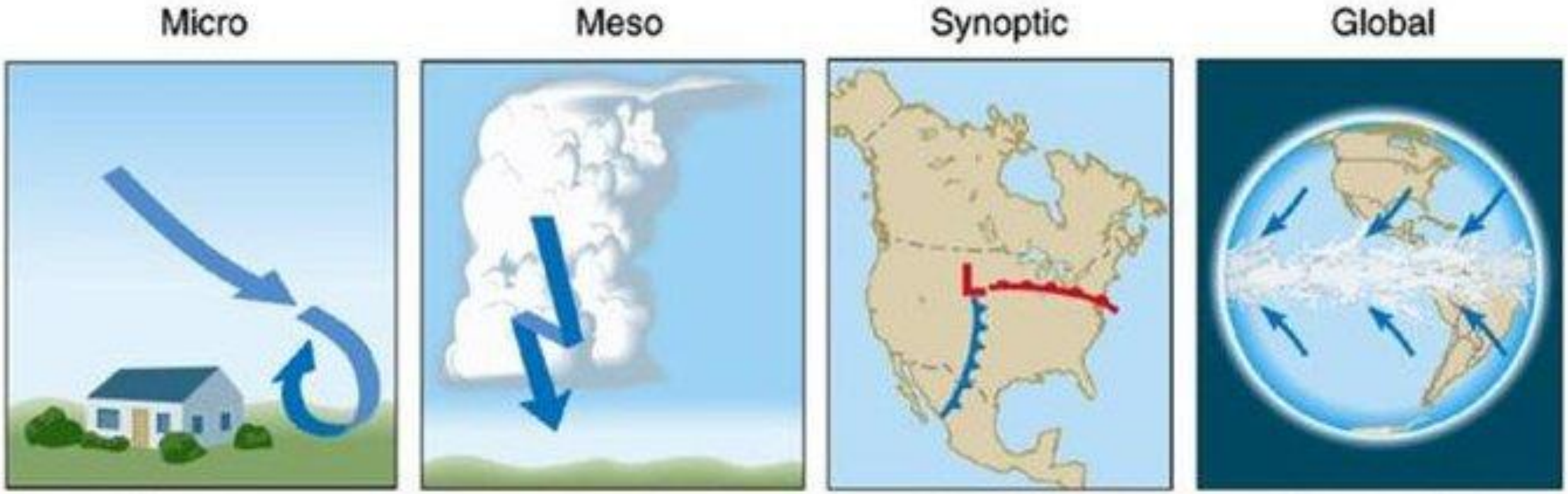
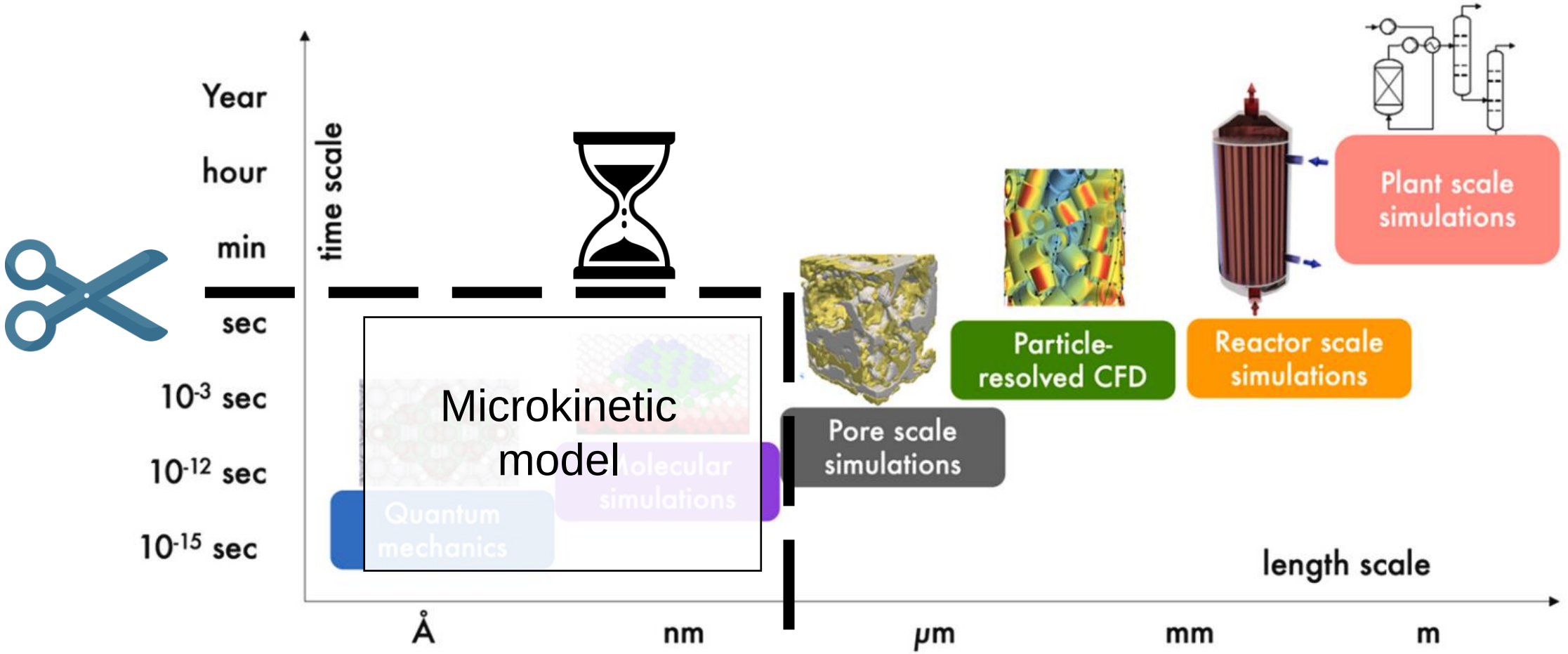
Real life problems are multi scale



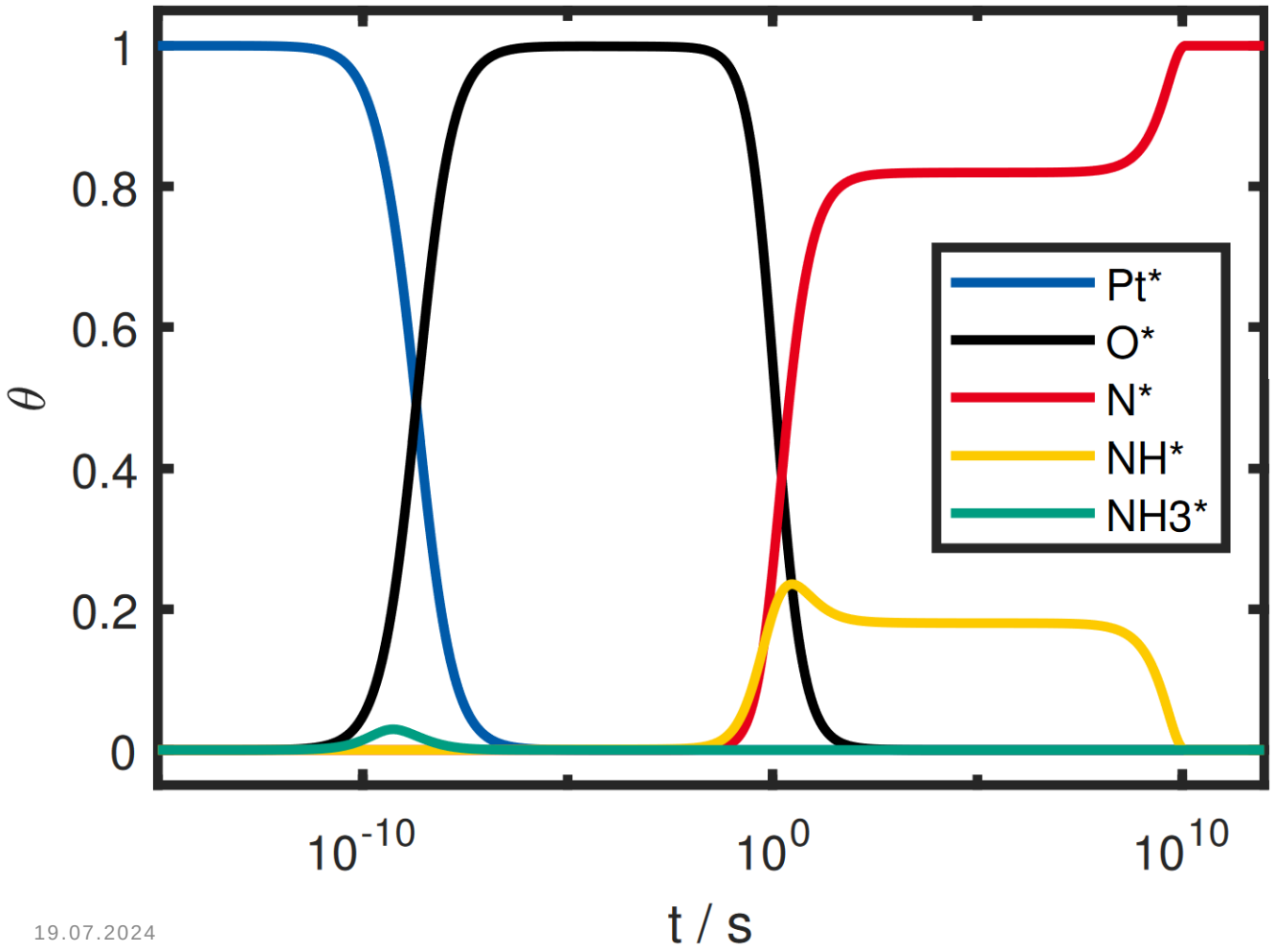
„Scales of atmospheric motion“
Florida State University - Neil O’Brien’

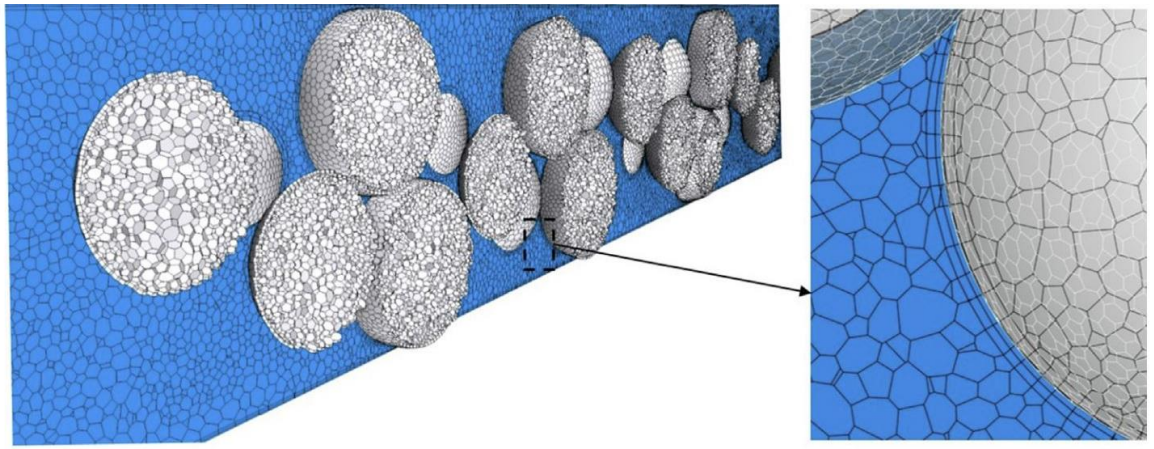
Real life problems are multi scale



Chemical Engineering Research and Design 184 (2022) 39–58

Solving microkinetics is slow



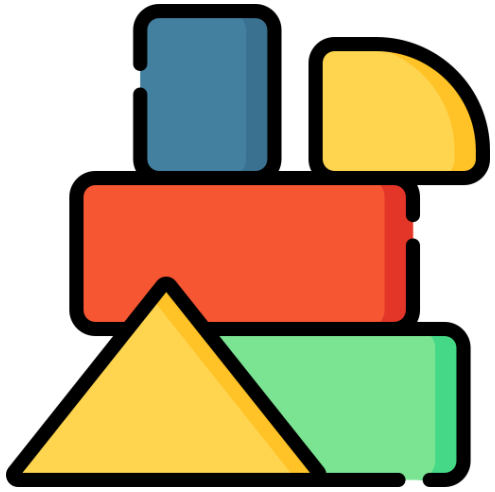


Chemical Engineering Science, 250, 117408

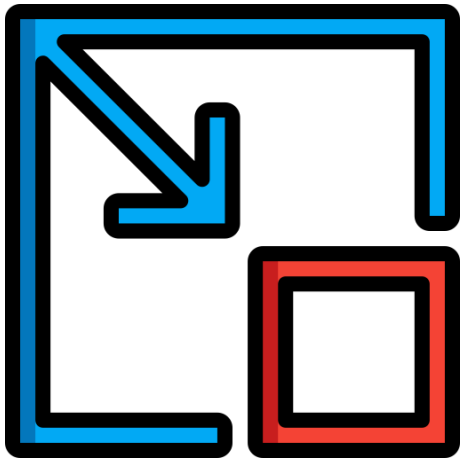
- 70-90%[1]
- ~ 90%[2]
- > 99%[3]

[1] Bracconi, M., et al. (2017), AIChE Journal, 63(1), 95–104.
[2] Nikitin, V. F., et al. (2021), Acta Astronautica, 194, 376–382.
[3] Brown, T. S., et al. (2021), In Lecture Notes in Computer Science: Vol. 12761 LNCS (pp. 23–39)

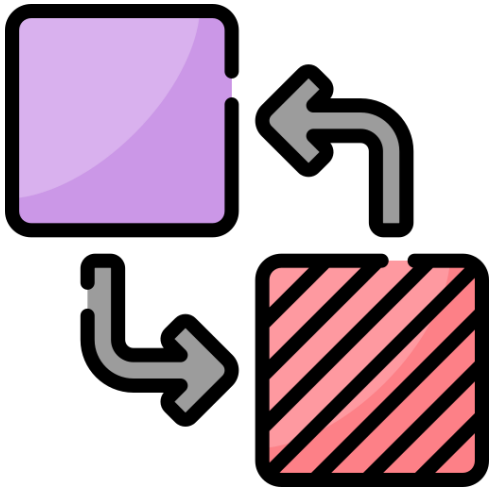
Speed-up strategies



Simplify



Reduce[1,2]



Replace

[1] Computers and Chemical Engineering, 2017, 97, 175–182.
[2] Reaction Chemistry & Engineering,2020, 5, 278.
Made with icons from flaticon.com

Types of surrogates

Splines[1-5]

up to 7 parameters[6]

Random Forests[7,8]

Neural Networks

Physics?

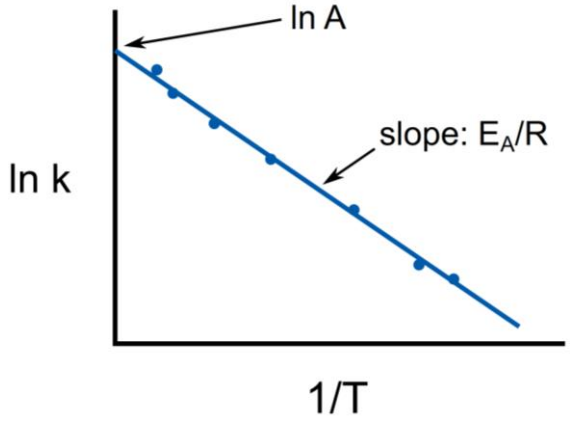
[1] Chemical Engineering Science 2009, 64, 1384–1389.
[2] Catalysis Today 2010, 151, 271–277.
[3] Applied Catalysis B: Environmental 2012, 111-112, 445–455.
[4] Computers & Chemical Engineering 2016, 88, 126–134.
[5] AIChE Journal 2017, 63, 87–94.
[6] Computers & Chemical Engineering 2017, 98, 21–30.
[7] Computers & Chemical Engineering 2018, 115, 286–294.
[8] Chemical Engineering Journal 2020, 400, 125469..

Arrhenius & mass action law

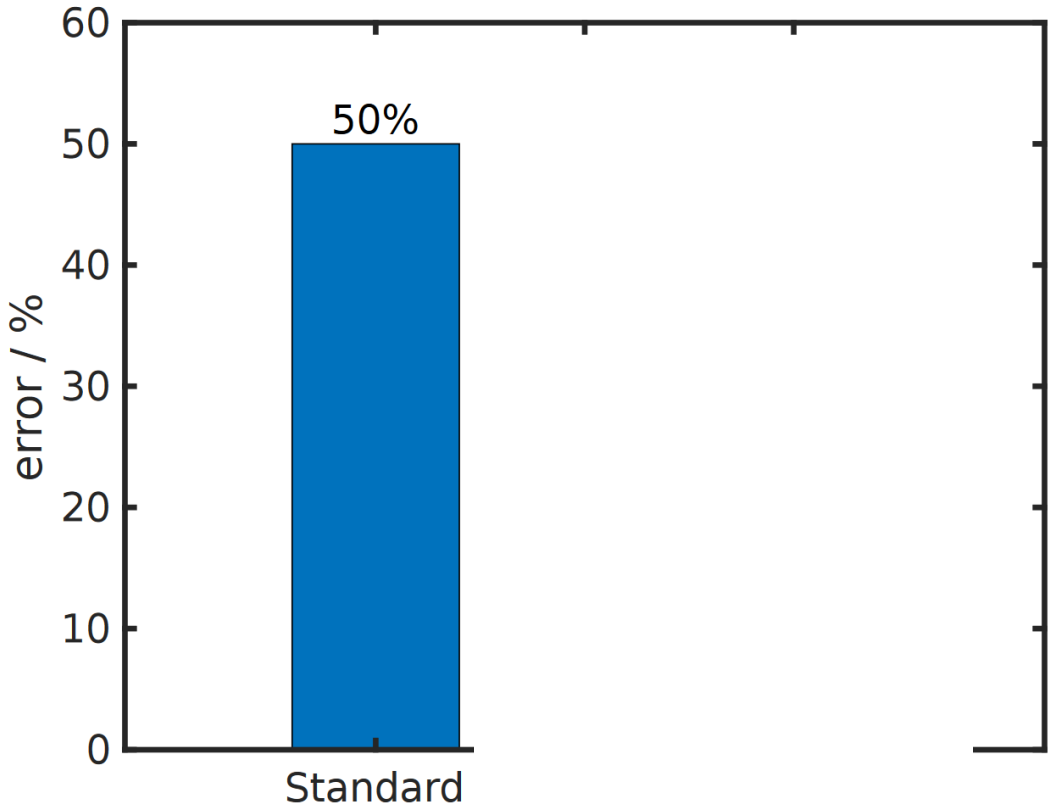
$$r = k_0 \cdot \exp\left(-\frac{E_A}{R\,T}\right) \cdot \prod_{i \in \text{reac.}} a_i^{-\nu_{i,j}}$$

$$\ln r = \ln k_0 - \frac{E_A}{R} \cdot \frac{1}{T} - \sum_{i \in \text{reac.}} \ln a_i$$

$$\dot{s} = r_{\text{ads}} - r_{\text{des}} = r_{RDS}^{+} - r_{RDS}^{-}$$



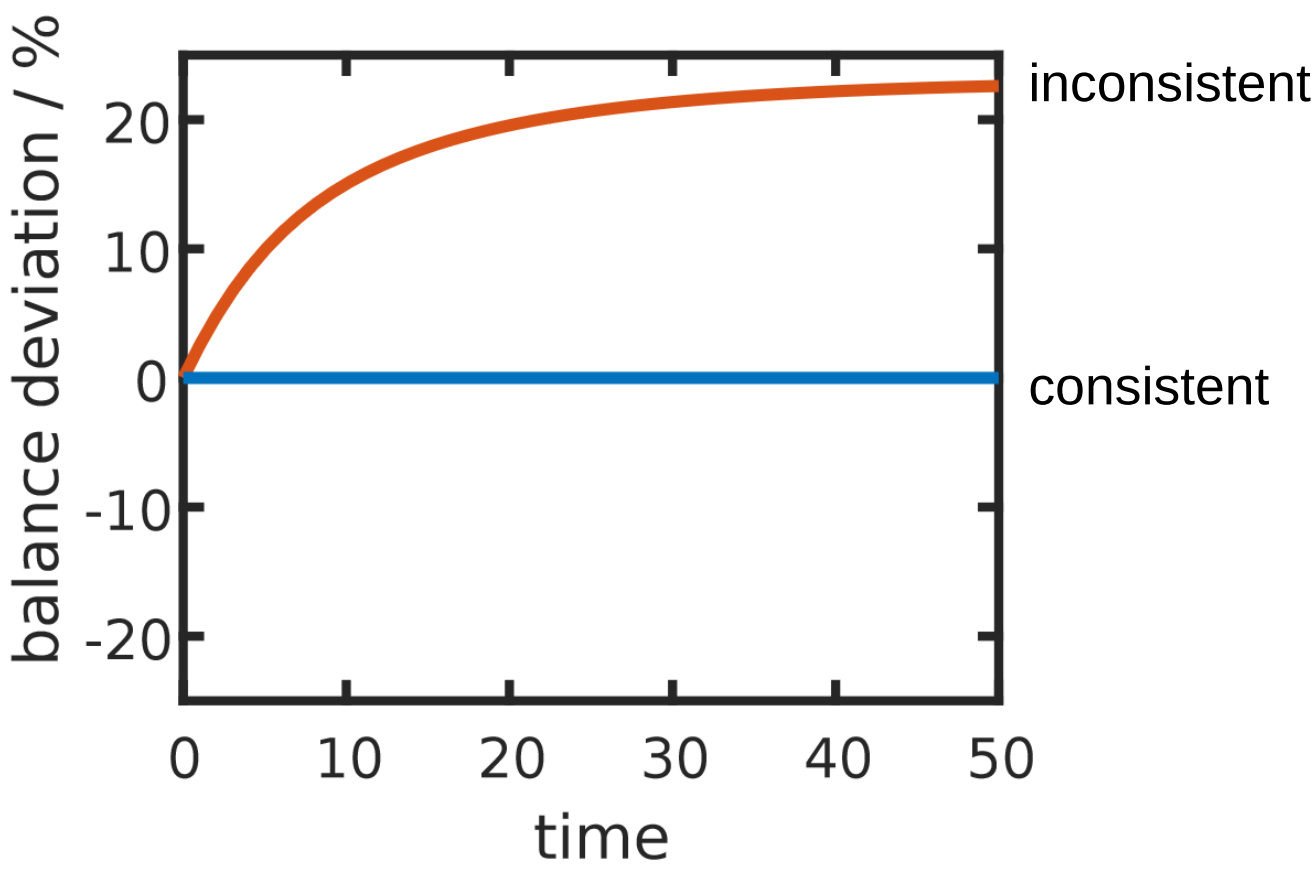
Preferential oxidation of CO on Pt[1,2]



Döppel and Votsmeier, Chem. Eng. Sci. 2022, 262, 117964

[1] Journal of Physical Chemistry B, (2004), 15246-15258, 108(39)
[2] Applied Catalysis A: General, (2011), 174-182, 397(1-2)

Atom balance



Via key species:

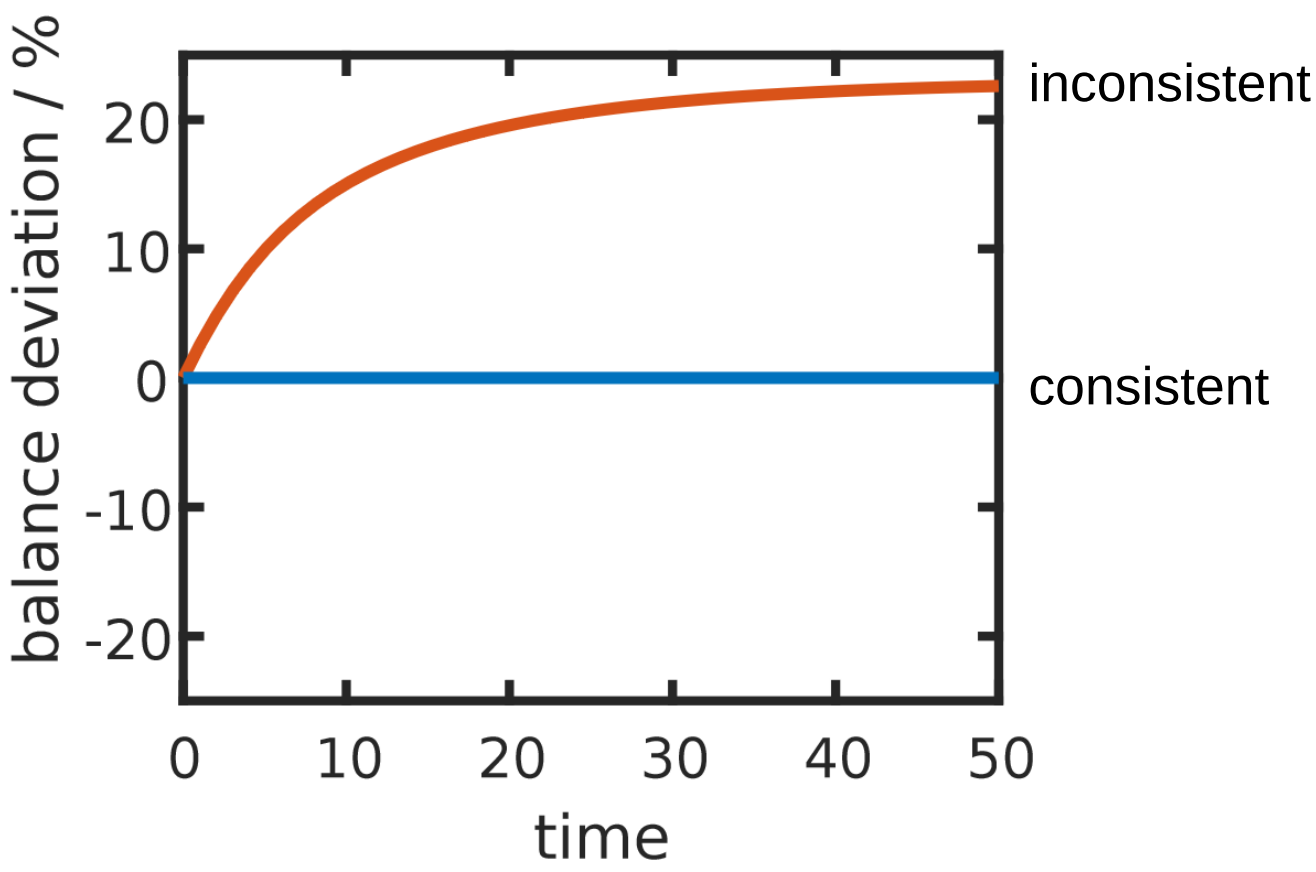
5 species

– 3 elements

= 2 unknowns

Döppel and Votsmeier, Proceedings of the Combustion institute, 2024, in production

Atom balance



Via global reactions:

$$R_{\mathrm{CO-ox}} = \ldots$$
$$R_{\mathrm{H_2-ox}} = \ldots$$
$$R_{\mathrm{WGS}} = \ldots$$

$$\dot{s} = \sum_j \nu_{i,j} R_j$$

Döppel and Votsmeier, Proceedings of the Combustion institute, 2024, in production

Thermodynamics

De Donder relation[1]

$$r_{\text{eff}} = r^{+} - r^{-}$$

$$= r^{+} \cdot \left(1 - \frac{\prod_i a_i^{\nu_{i,j}}}{K_{\text{eq}}}\right)$$

- Fewer rates to model
- Progress towards correct equilibrium

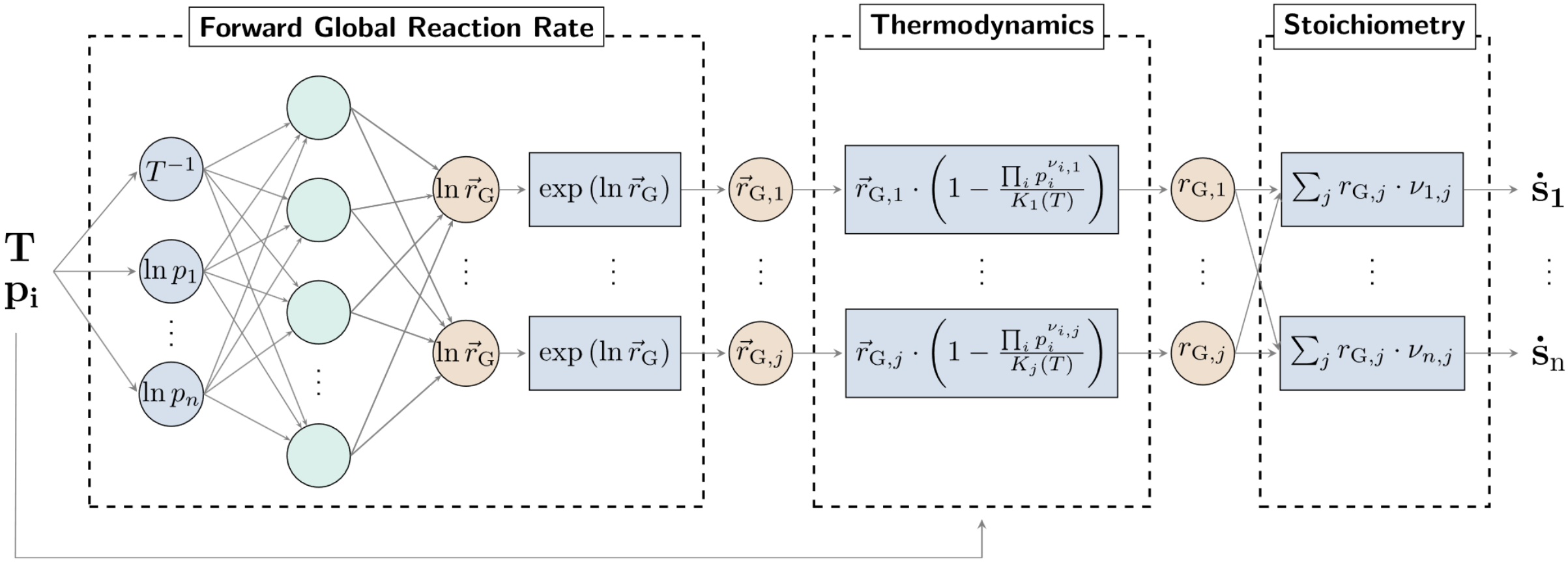
[1] Journal of Catalysis, (1999), 496-505, 185(2)

Physics checklist

- ✓Arrhenius law
- ✓Law of mass action
- ✓Atom balance
- ✓Thermodynamics

- ○ Butler-Volmer expression?
- ○ Charge balance?
- ○ Symmetry?

Global Reaction Neural Network



Kircher, Döppel, and Votsmeier, Chem. Eng. J., 2024, 485, 149863

Applications

- Preferential oxidation of CO on Pt[1]
- Methane steam reforming on Rh[1]
- Methanol synthesis on Cu/Zn[2]
- Ammonia synthesis on Ru[3]
- Methane non-oxidative coupling over single atom Fe/SiO2[3]

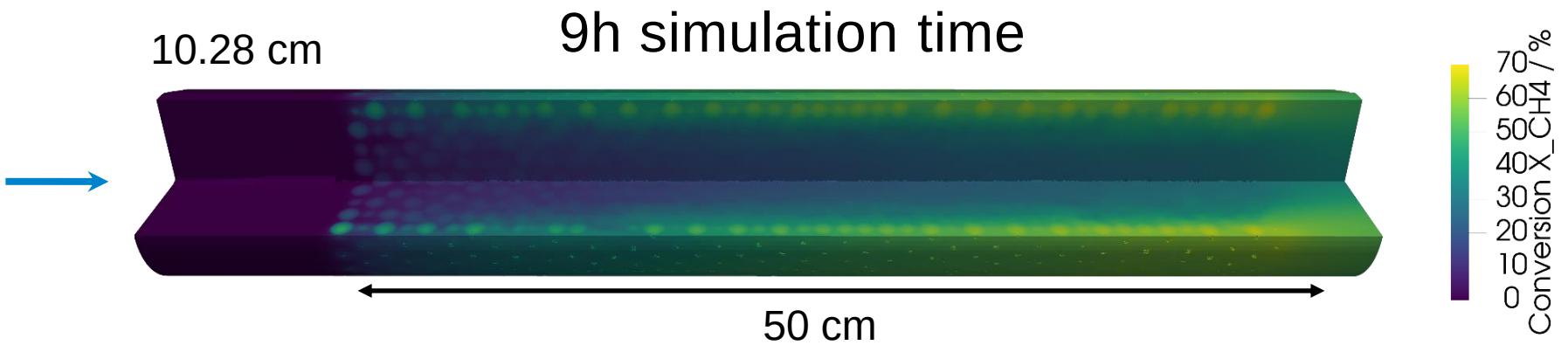
[1] Chemical Engineering Journal, (2024), 485, 149863
[2] Reaction Chemistry & Engineering, (2024), 9, 1047-1060
[3] Reaction Chemistry & Engineering, (2024), 9, 119-131

Reactor Simulation
Packed Bed Steam Reforming

Biermann, Uglietti, Döppel, Kircher, Votsmeier, Bracconi, Maestri, Manuscript in preparation

Catalytic Foam[1] Interface

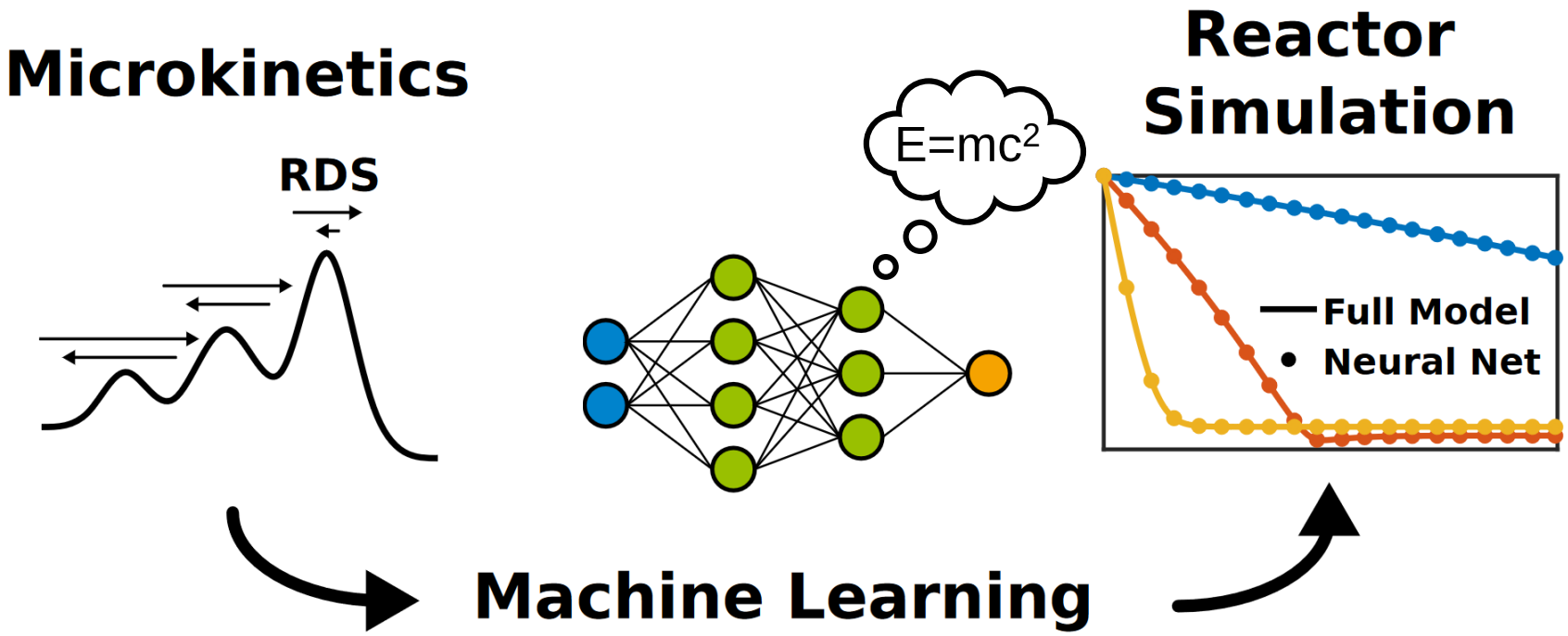
- 2D validation: 120x speed-up
- 3D simulation:



2400 pellets
44.8 mio cells

Summary

- Machine learning surrogates enable detailed reactor simulations
- Embedding physics leads to reliable results



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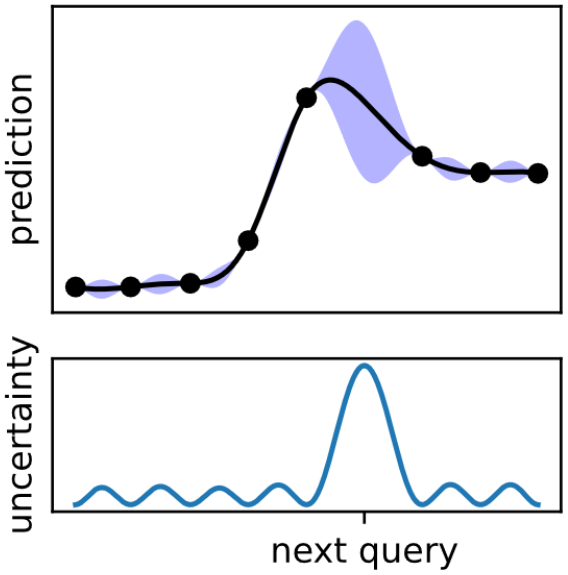
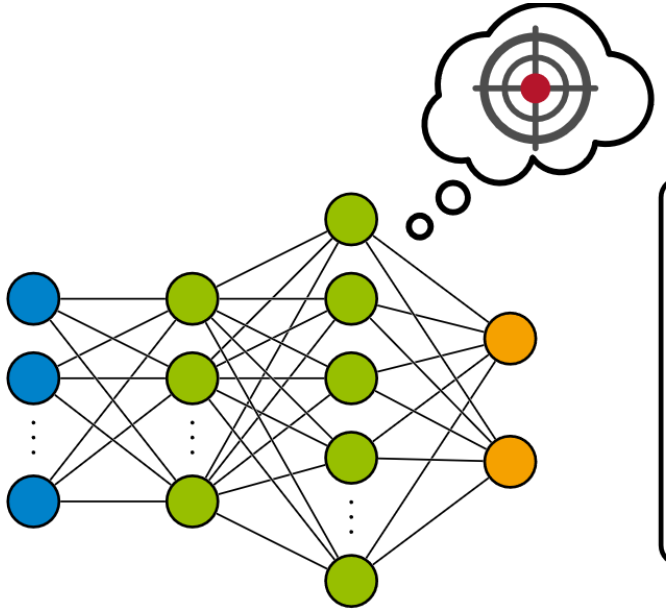
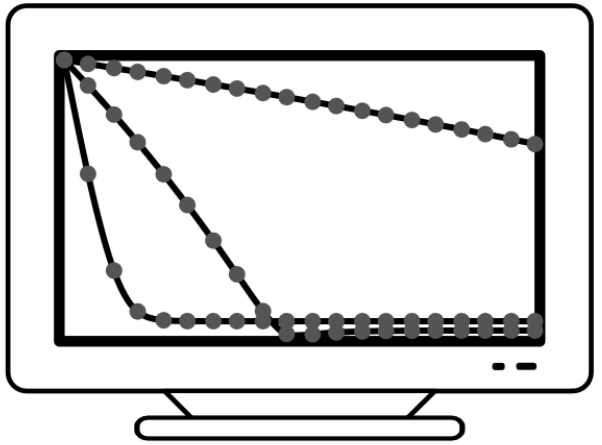
Automated surrogates







automated training set design

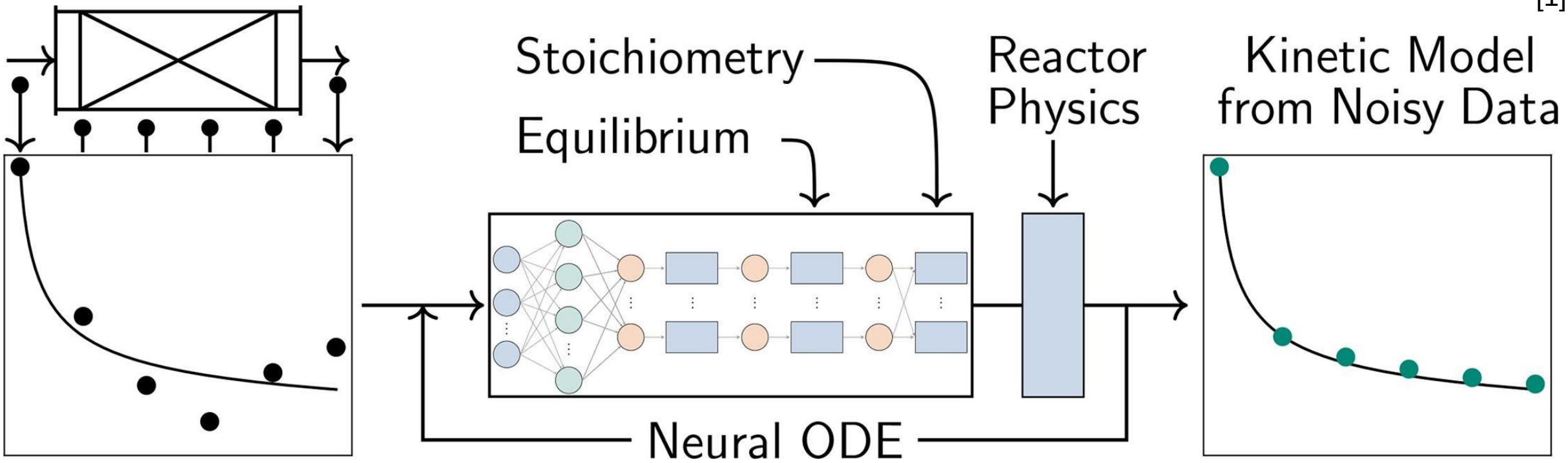
goal-oriented kernel model

fast & accurate reactor simulations

Döppel et al., Chem. Ing. Tech. 2024, 96, 6, 759-768

Model discovery from integral data

[1]



- Methane steam reforming on Rh[1]
- Preferential oxidation of CO on Pt[2]
- CO_2 Fischer Tropsch[3]

[1] Chemical Engineering Journal, (2024), 485, 149863
[2] Computer Aided Chemical Engineering, (2024), 53, 817-822
[3] Chemical Engineering Journal, (2023), 477, 146869

Acknowledgements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T. Kircher


F. Biermann


Prof. M. Maestri


Prof. M. Bracconi

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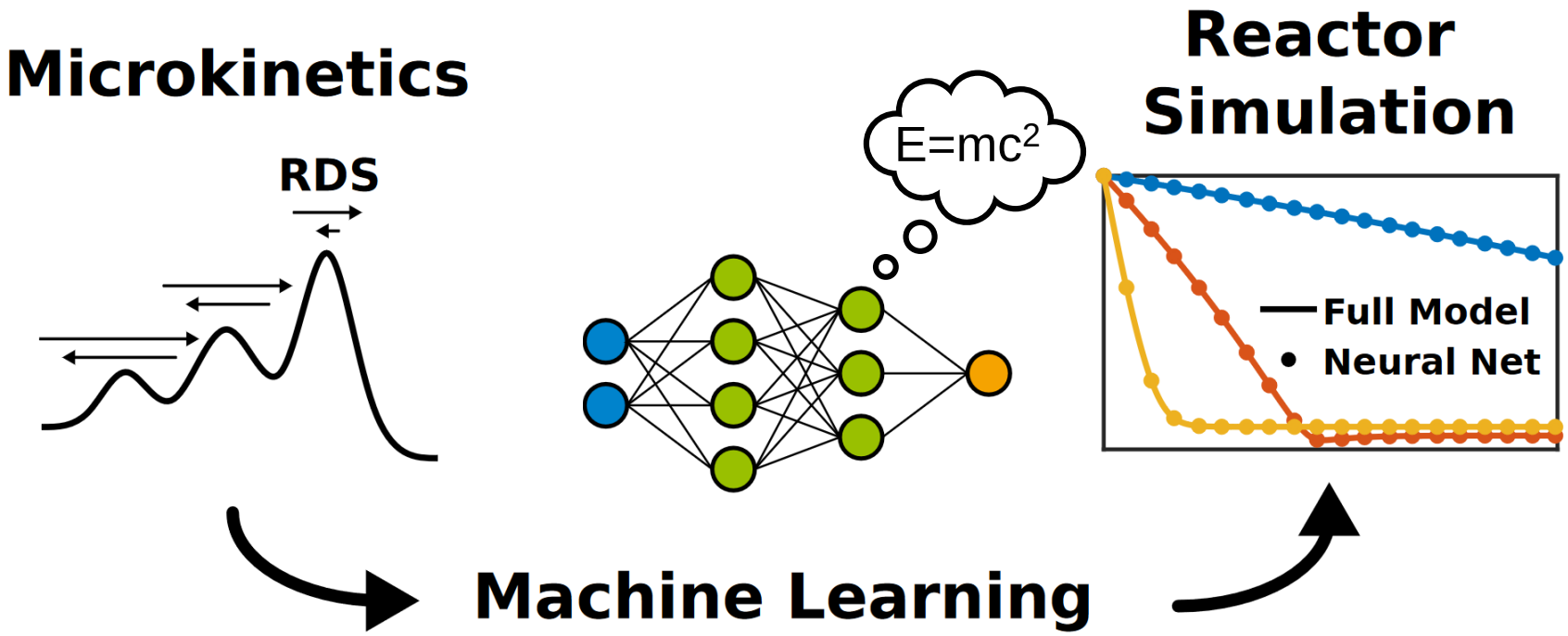

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