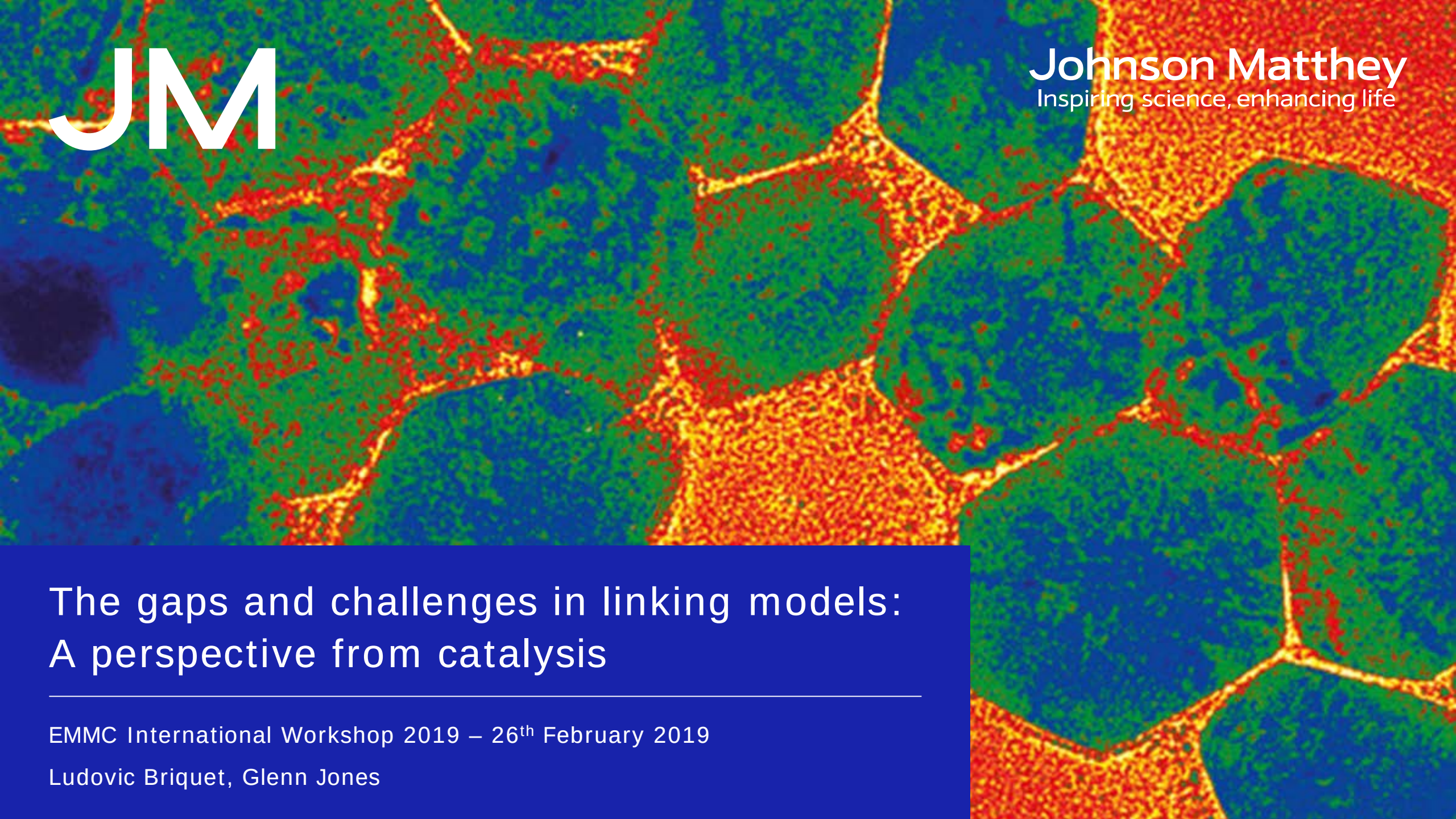

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Johnson Matthey
Inspiring science, enhancing life

The gaps and challenges in linking models: A perspective from catalysis

EMMC International Workshop 2019 – 26th February 2019

Ludovic Briquet, Glenn Jones

Outline

- Introduction
  - Who is Johnson Matthey?
- Catalysis is truly a multi-scale problem
- Non scientific challenges
- Scientific case studies
  - Technical gaps

Johnson Matthey

A speciality chemical company and a world leader in sustainable technologies

Our vision is for a world that's cleaner and healthier; today and for future generations

Clean Air

- Light duty catalysts
- Heavy duty catalysts
- Stationary Emissions control

Efficient Natural Resources

- Chemical catalysts for petrochemical and oleochemical industries
- Precious Metal Refining
- Advanced Glass Technologies
- Syngas
- Chemical Products
- Diagnostic services for oil and gas industry

Health

- Active Pharmaceutical Ingredients
- Controlled Substances – Opiates
- Custom Pharma solutions

New Markets

- Battery Technologies
- Fuel Cells
- Atmosphere Control Technologies
- Medical Devices
- Catalysts
- New Business Development

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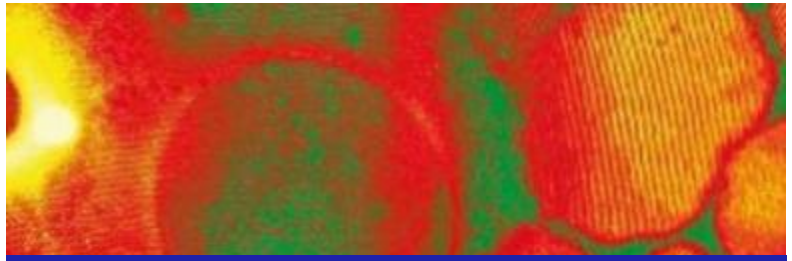
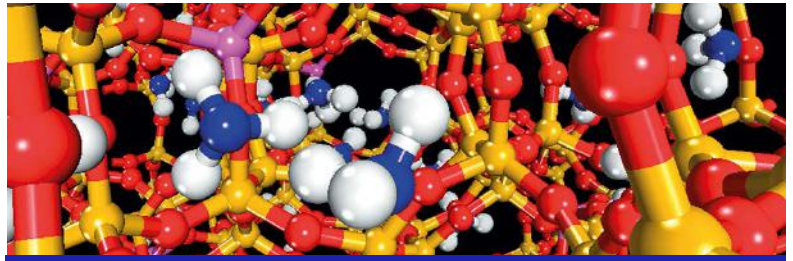
World class science and technology expertise

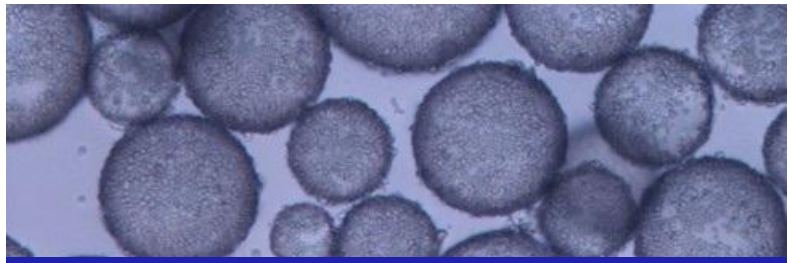


Characterisation and modelling

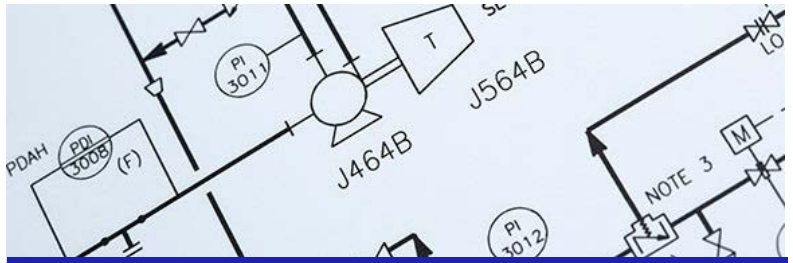
Chemical synthesis



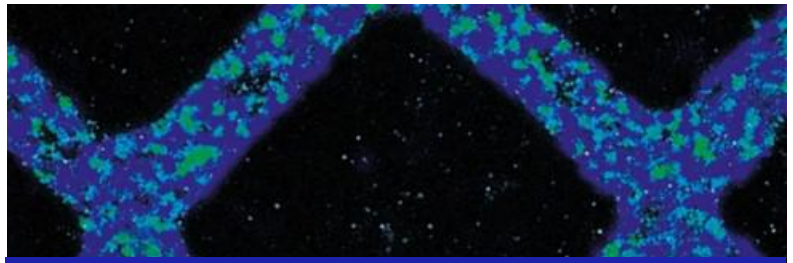
Material design and engineering



Product formulation



Process optimisation



Surface chemistry and coatings

Pgm chemistry and metallurgy

Catalysis and advanced materials

Electrochemistry

We serve global markets

Automotive

Pharmaceutical and medical

Chemicals

Oil and gas

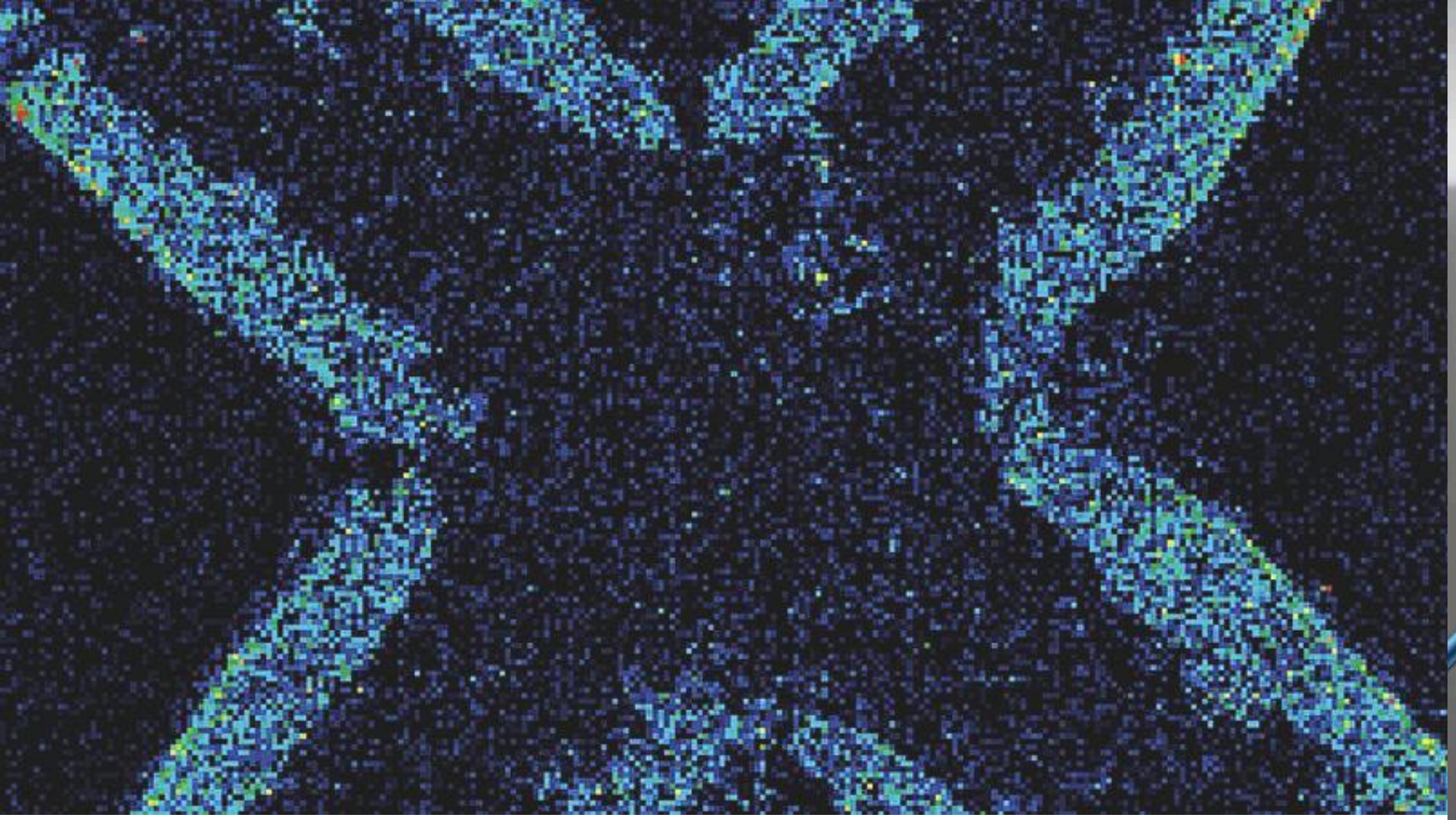
Agrochemicals and fertilisers

Food and beverage

Energy generation and storage

Glass

Other industrial



Catalysis: A truly multi-scale problem

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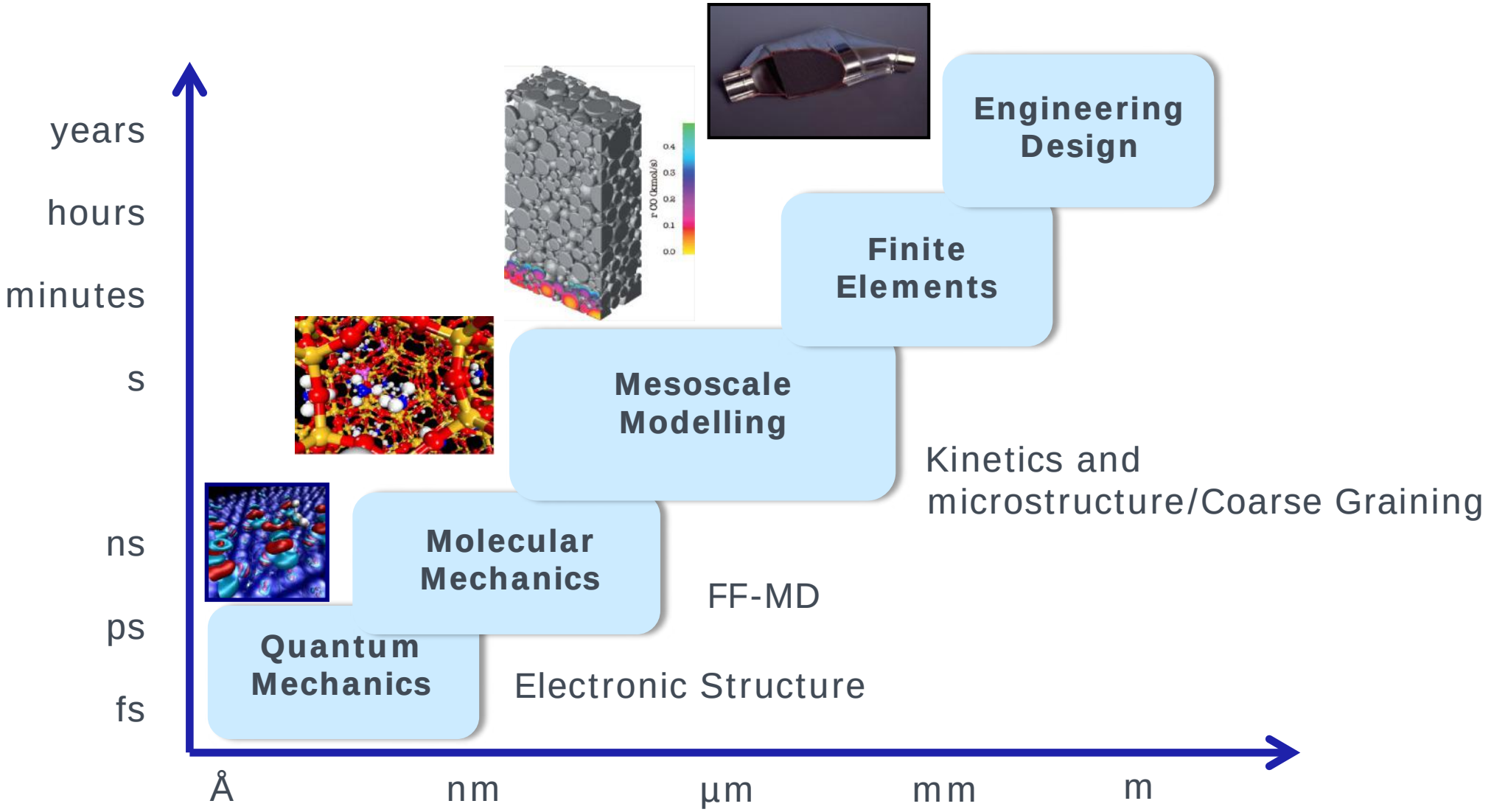
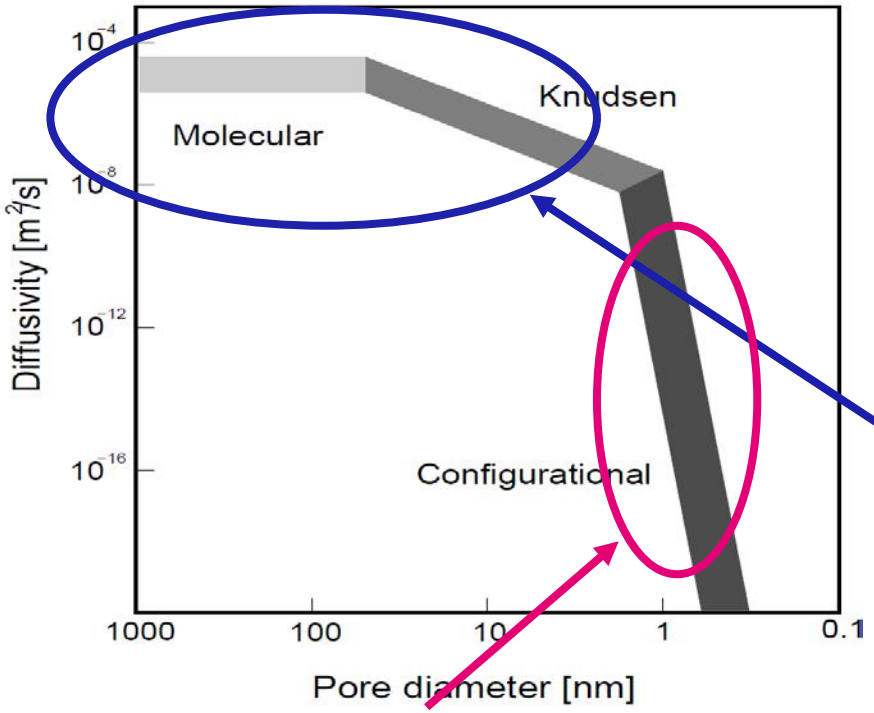
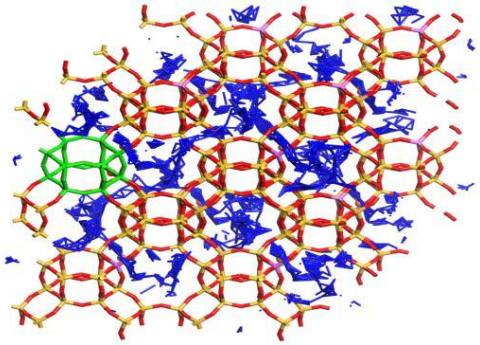
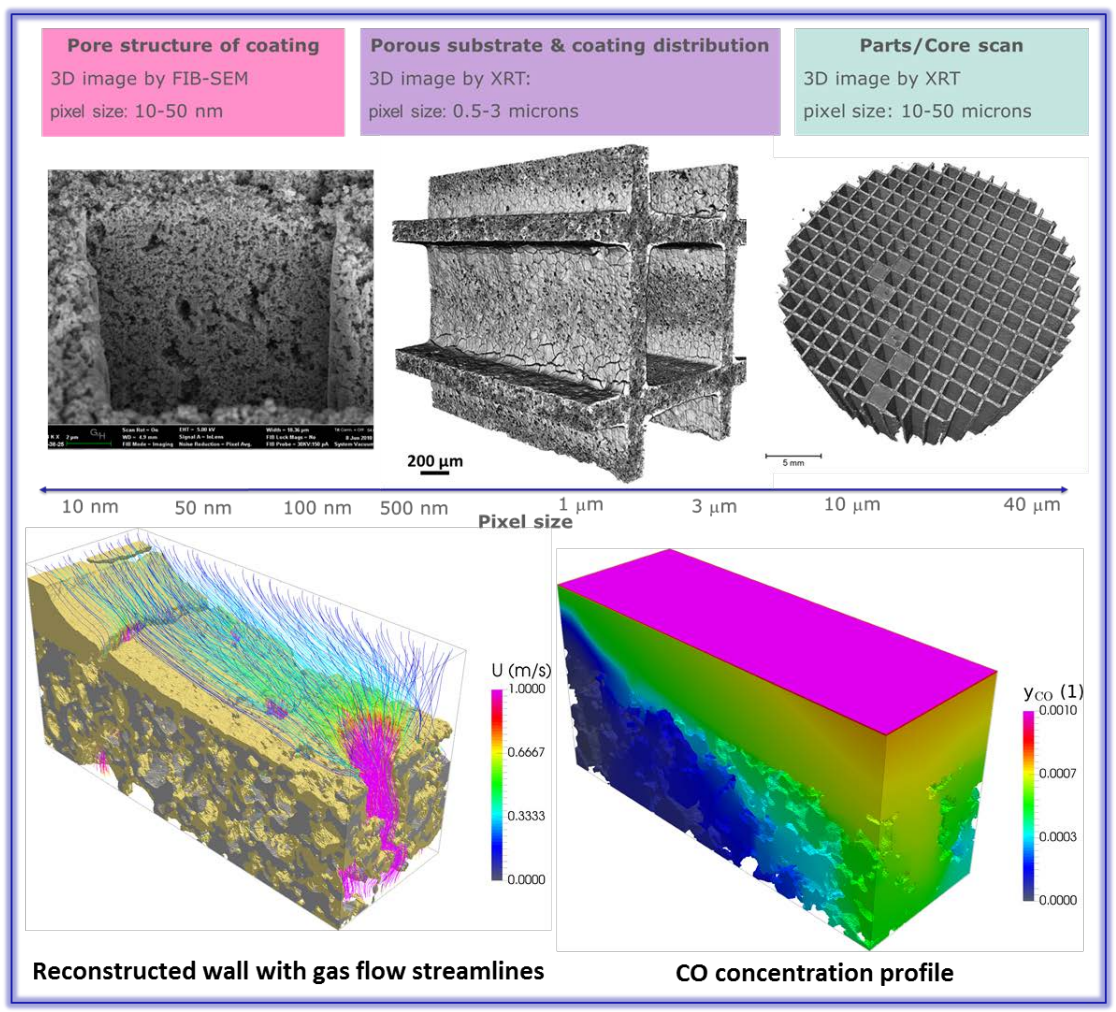
Computational material modelling is multiscale by nature



Transport processes range over several orders of magnitude







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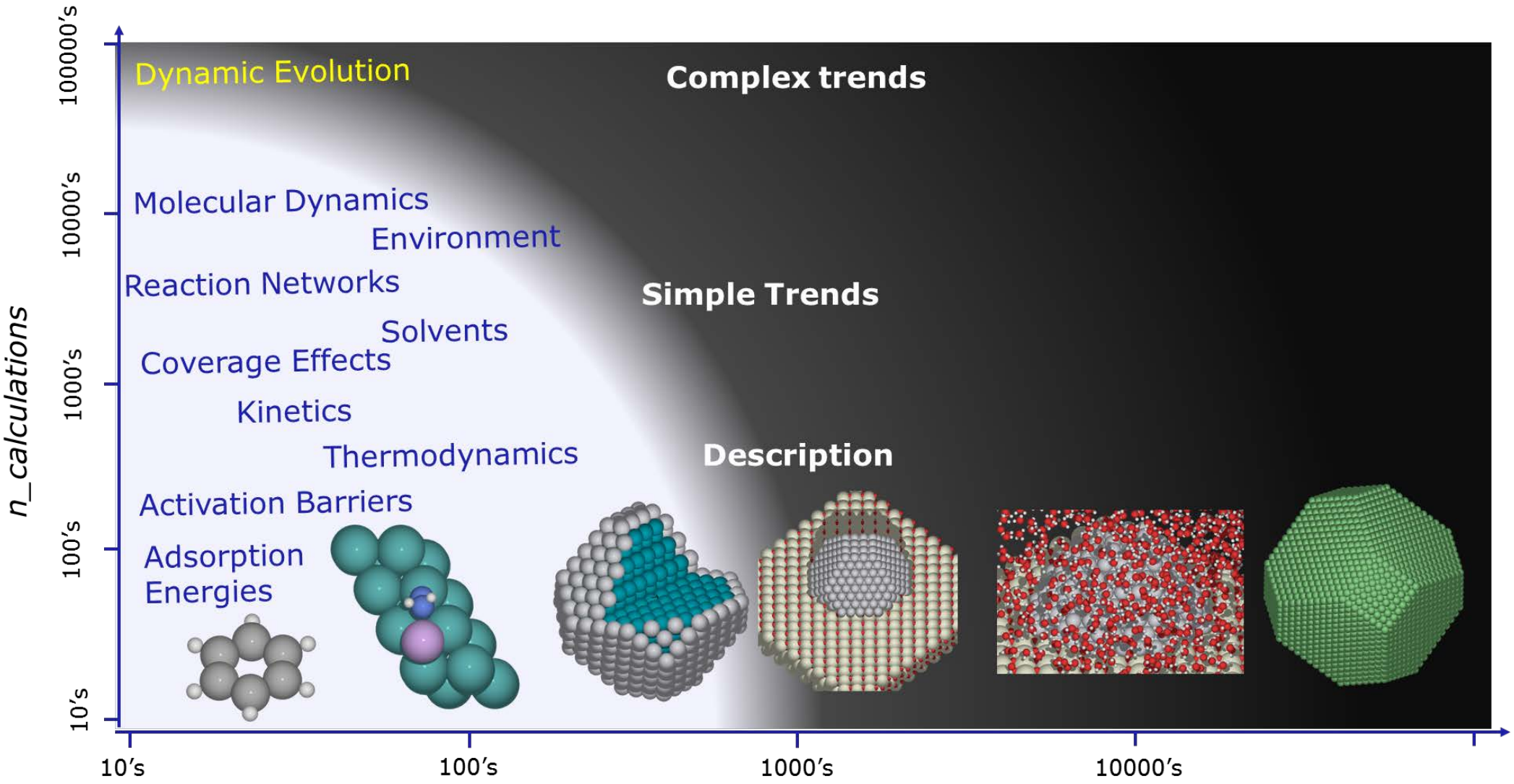
We will always need approximations



Non-scientific challenges

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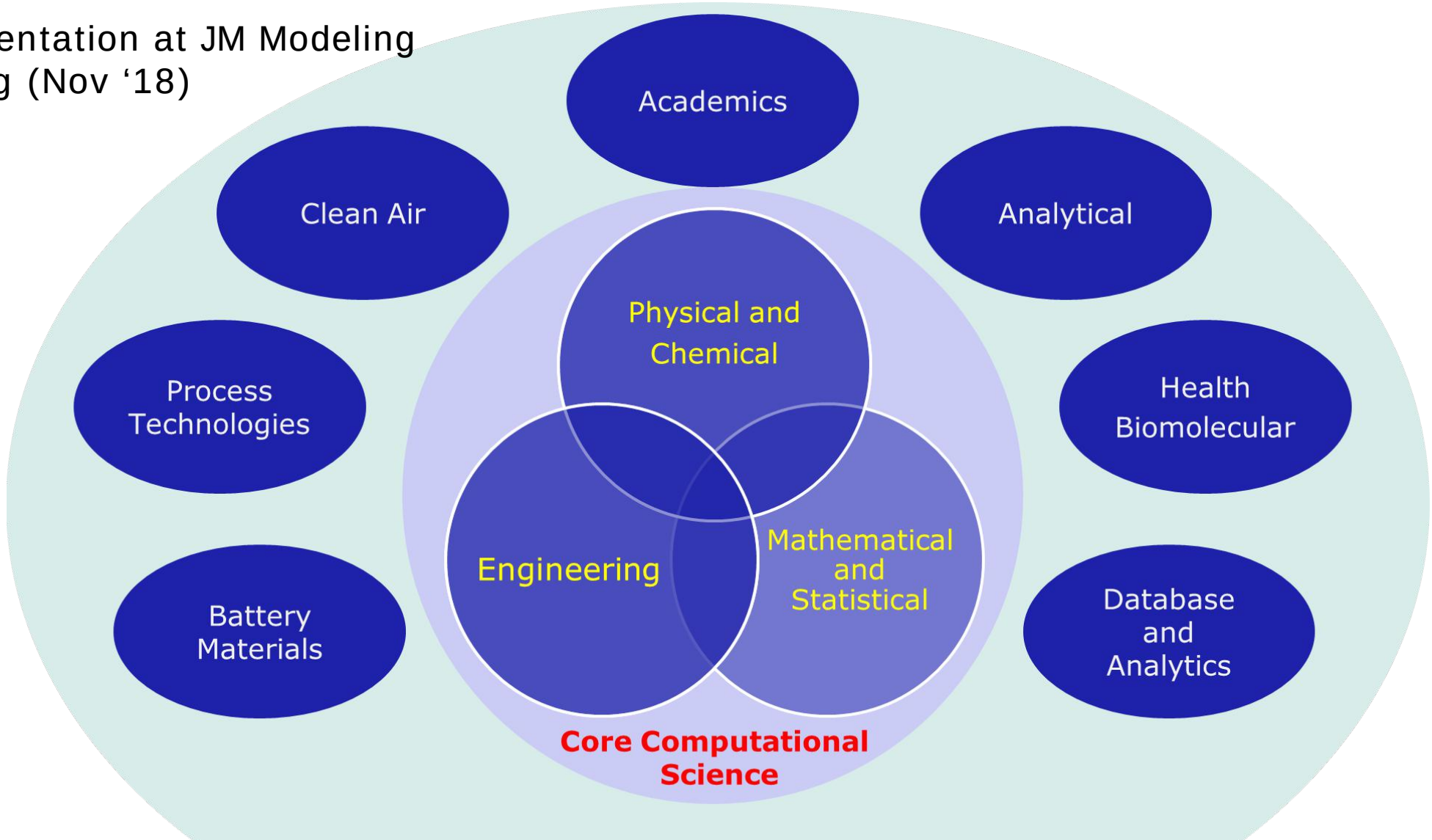
1. Different competencies are required for each level

Representation at JM Modeling Meeting (Nov '18)



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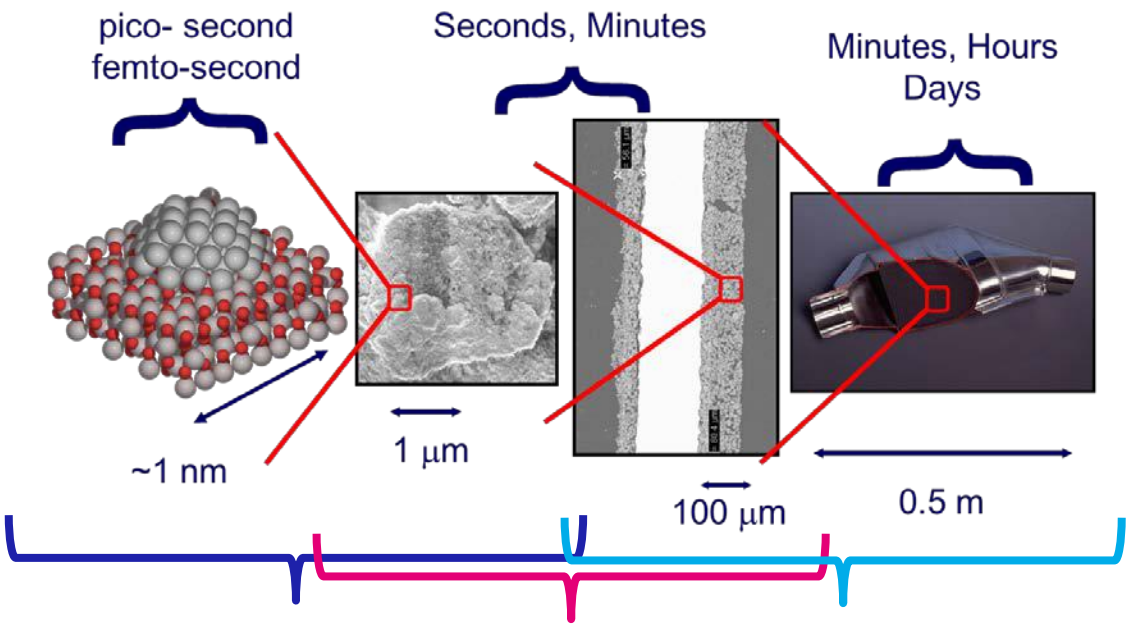
1. Different competencies are required for each level



Science evolves
learn new ways to solve problems

Need to bring in new talent

Improve and influence direction of research

- Atomistic modelling
- Chemistry
- Physics

- Analytical chemistry
- Formulation
- Chemistry
- Physics
- Chemical engineering
- Statistics

- Chemical engineering
- Chemistry
- Physics
- Statistics

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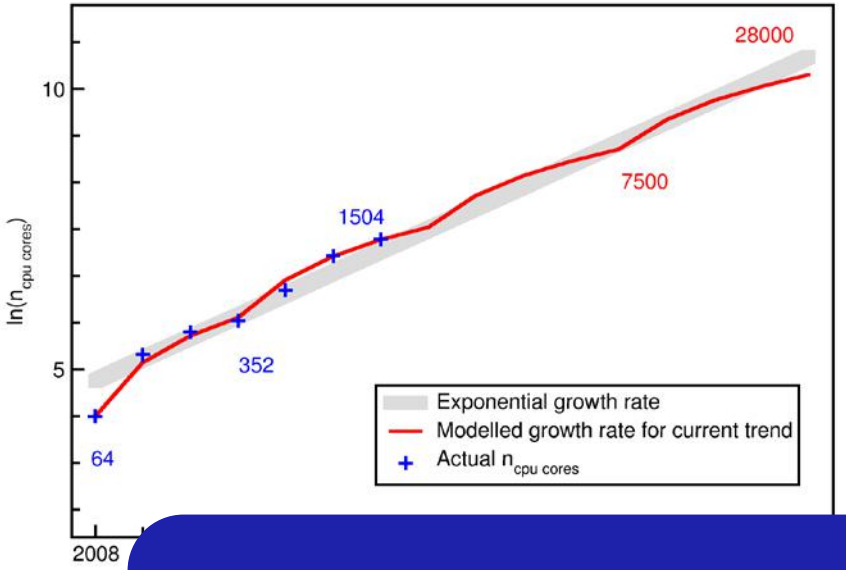
2.Computing capabilities

JM's HPC capabilities vs time:



+ Licensing issues

Current:

- Number of small to medium size clusters disseminated across 3 sites
- Unsupported by corporate IT
  (Users are sys admin)
- Impeding the growth of the department

Mid- to long-term:

- Getting support from corporate IT
- Single company-wide HPC capability vs Cloud?
  - Users located all over the world (latency)
  - Computer requirement are different for each users (CFD vs. DFT)
  - Computing performances
  - Cost

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• 3. Is it worth it? (from an industrial perspective)

Even if all the L&C problem were solved, multi-scale modelling can be expensive
(Manpower / hardware / software)

For each potential project

- What do we wish to learn?
- What would be the return on investment?
- Can we use a simpler way to get the answer?

We need to:

➔ Ensure that we have a well defined problem

Each length/time scale presents a risk to be side-tracked from the original problem

➔ Manage expectations

"Just because the results are in colour, it doesn't mean they are right"*

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* H. Stitt, M. Marigo, S. Wilkinson, T Dixon, Johnson Matthey Technol. Rev. 59 (2015) 75; M.P. Dudukovic, F. Larachi, P.L. Mills, *Chem. Eng. Sci.* 59 (1999) 13

Case study examples

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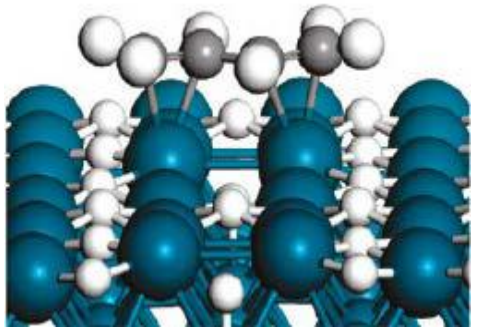
Butadiene hydrogenation

Atomic scale $\xrightarrow{E^{\dagger},\ \Delta H_{ads}}$ / $\xleftarrow{\theta_H}$ **Kinetic model** $\xrightarrow{r_i}$ / $\xleftarrow{T,\ P,\ C_i}$ **Macroscopic scale**

DFT

VASP

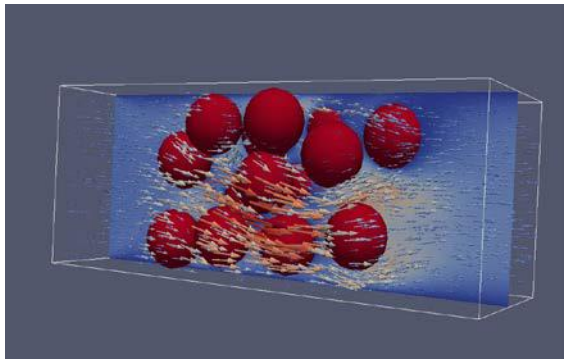


Mean Field Kinetics

CFD

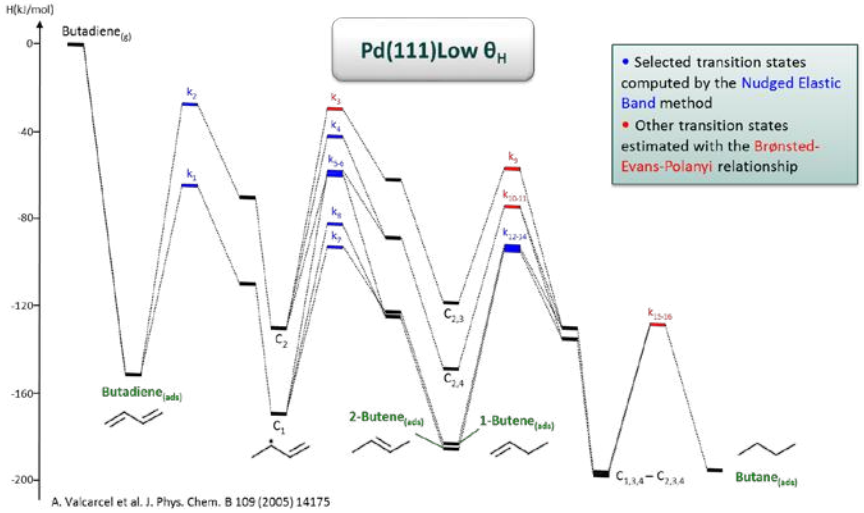
PeliGRIFF

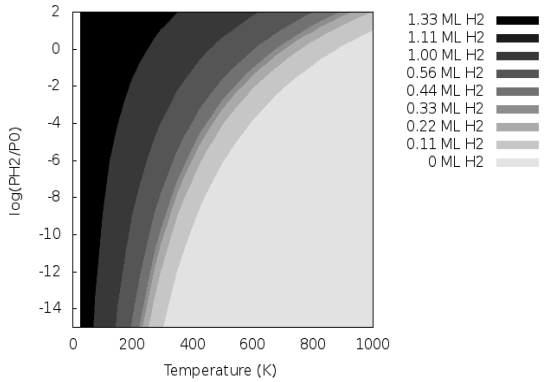
Parallel Efficient LIbrary for GRains In Fluid Flows



Butadiene hydrogenation

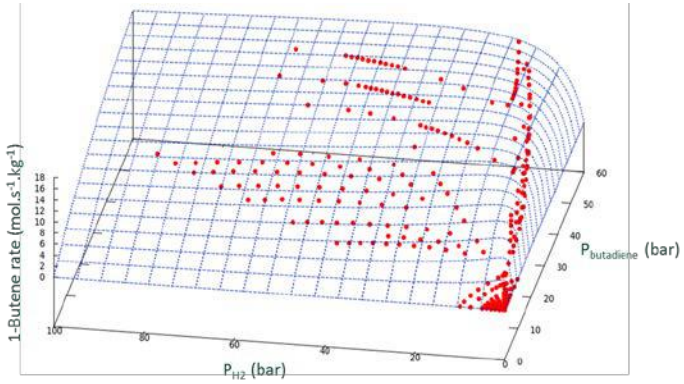
Reaction profile at ≠ θ_H



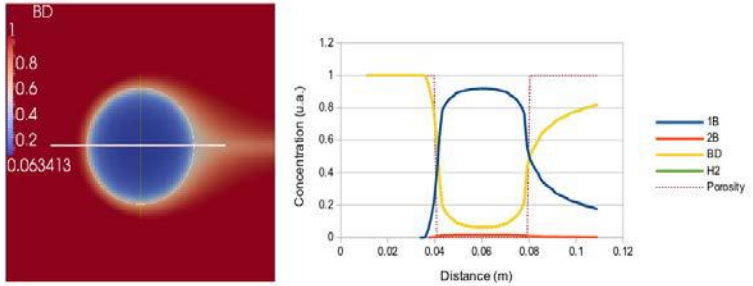


Kinetic law via a μ-kinetic model

$$r = \frac{K_1 P_{BD} P_{H_2}}{\left(1 + K_2 P_{BD} + K_3\sqrt{P_{H_2}}\right)\left(P_{H_2} + K_4 P_{BD}\left(1 + K_2 P_{BD} + K_3\sqrt{P_{H_2}}\right)\right)}$$



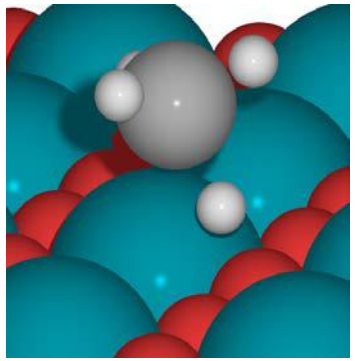
1st tentative 2D CFD modelling



Methane Oxidation

Catalyst reactivity

A) $CH_{4(g)} + O^* + 2O_{2(g)}$
B) $CH_3^* + HO^* + 2O_{2(g)}$
C) $CH_3O^* + HO^* + 1.5O_{2(g)}$
D) $CO_2^* + 2H_2O_{(g)} + 0.5\ O_{2(g)}$
E) $CO_{2(g)} + 2H_2O_{(g)} + * + 0.5\ O_{2(g)}$
F) $CO_{2(g)} + 2H_2O_{(g)} + O$

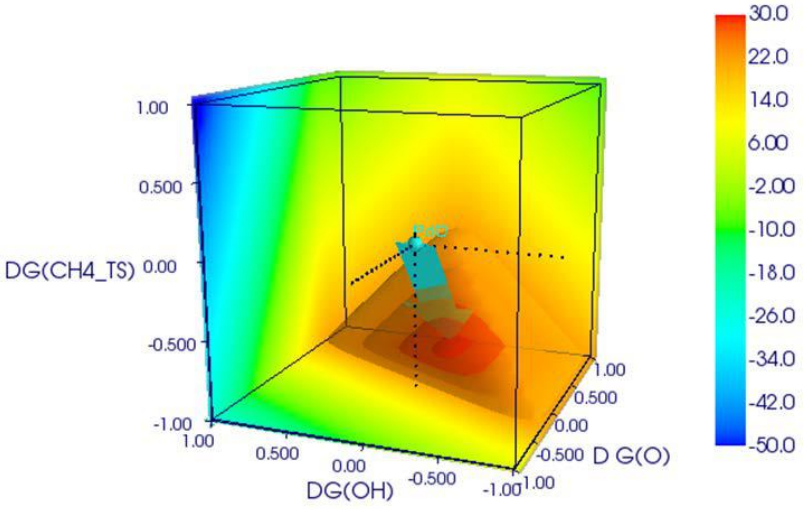


Detailed analysis of the reaction kinetics

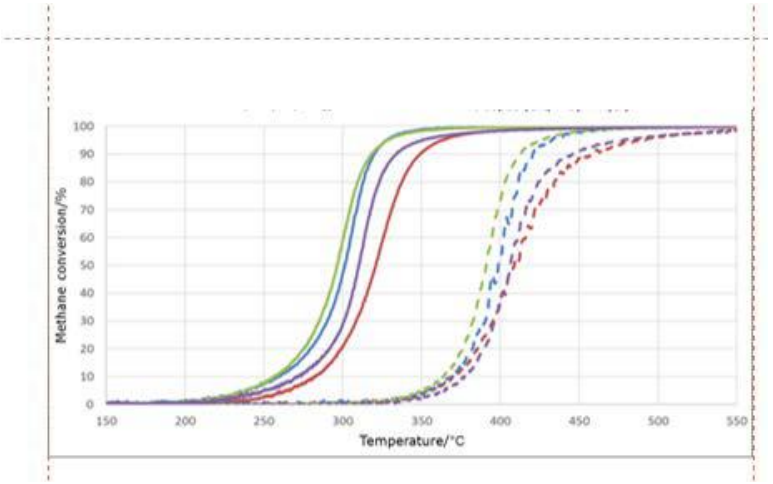
$$X_{TRC,i} = \left(\frac{\partial \ln r}{\partial\left(\frac{-G^o{}_i}{RT}\right)}\right)_{G^o{}_{i\neq j},\, G^o{}_i{}^{TS}}$$

Species	TRC (300K)	TRC (550K)	TRC (800K)
$O_{2(g)}$	-0.12	-0.949	-0.49
$CH_{4(g)}$	-1.00	-1.00	-1.00
O*	-1.00	0.90	0.96
CH_3O^*	0.00	0.00	0.00
CH_2O^*	0.00	0.00	0.00
CHO_2^*	0.00	0.00	0.00
OH*	0.00	-1.90	-1.96
H_2O^*	0.00	0.00	0.00
$CO_{2(g)}$	0.00	0.00	0.00
$H_2O_{(g)}$	0.00	1.90	0.98
CH_4O_TS*	1.00	1.00	1.00

Prediction through reaction descriptors



Experimental feedback



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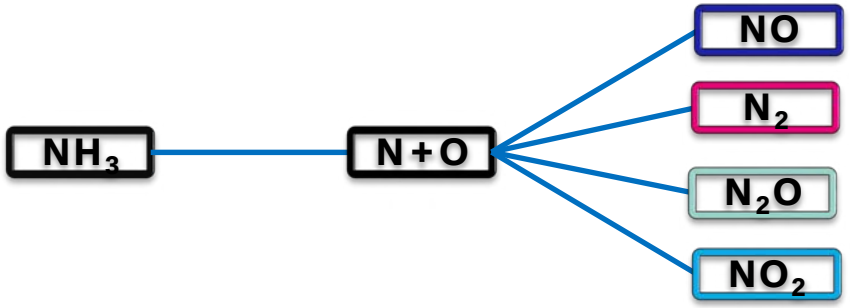
Ammonia oxidation

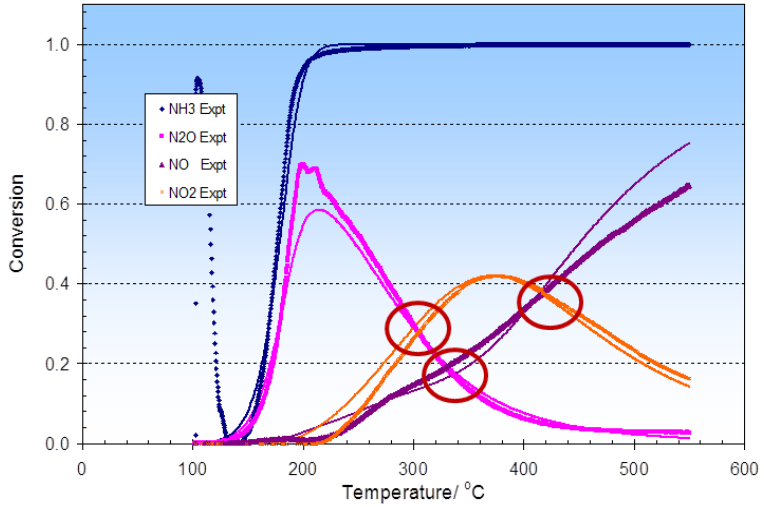
Catalyst selectivity



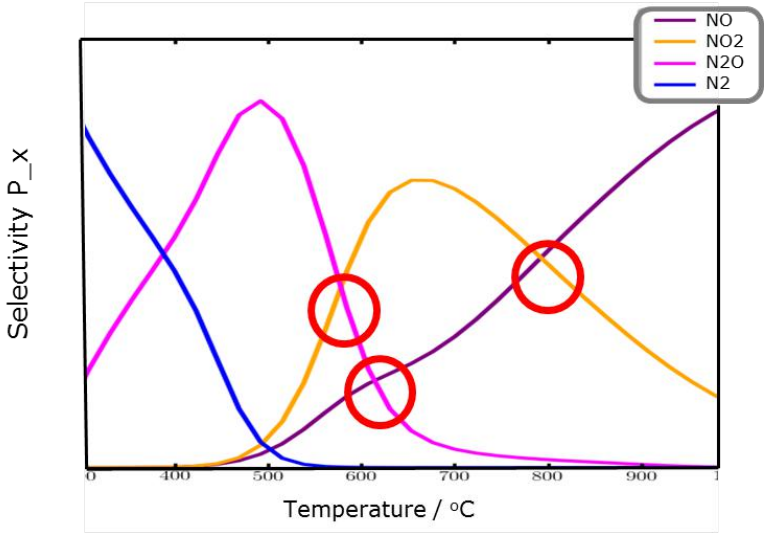
- Selectivity not determined by thermodynamics
- Need for a kinetic model

- Is DFT accurate enough to capture the selectivity of the reaction?
  - 3 regimes correctly predicted
  - Regimes are shifted to higher temperatures
  - High sensitivity to oxygen adsorption energy

Experimental



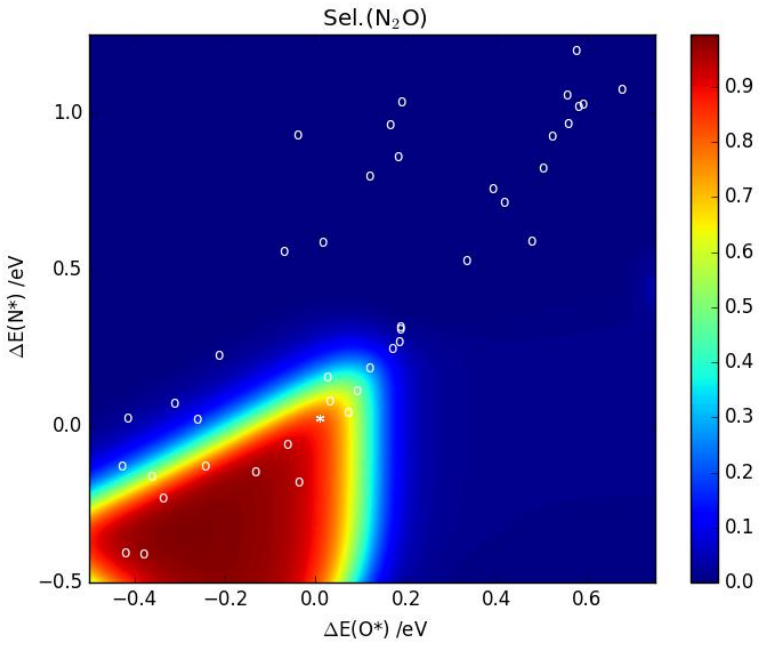
Micro-kinetic Model



Ammonia oxidation

Catalyst selectivity

Selectivity towards N_2O



Descriptors and Approximations allow for fast materials screening

(BEP and linear relationships)

DFT can get trends in selectivity

(we don't expect full quantitative agreement)

The modelling gives leads for new materials

(in addition to insight)

Experimental work is now testing new materials.

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Technical gaps and challenges for multi-scale modelling

- Stiff equations in micro-kinetics models
- Code interoperability ➔ need for a uniformized open input/output format
- Accuracy and error propagation
  - $r(t) = \kappa \frac{k_B T}{h} e^{\frac{\Delta S^{\ddagger}}{R}} e^{\frac{-\Delta H^{\ddagger}}{RT}}$
  - Complex metal oxides
- How to properly describe the meso-scale pore structure?

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Johnson Matthey
Inspiring science, enhancing life

Thanks to:

Glenn Jones
Johanna Espach
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Mehrdad Ahmadi
Christopher Perry

Philippe Sautet
Sarah Gautier
Céline Chizallet
Pascal Raybaud
Jan Verstraete
Jean-Marc Schweitzer

Review of computational modelling work at Johnson Matthey:

M. Sarwar, C. Cooper, L. Briquet, A. Ukpong, C. Perry, G. Jones, Johnson Matthey Technol. Rev. 59 (2015) 257-283