ADVANCES IN CATALYSIS FOR HYDROCARBONS

RESULTS FROM ZEOCAT-3D, C123 & BIZEOLCAT EU RESEARCH PROJECTS

ZEOCAT-3D

C123
VALORISING METHANE RESOURCES

BiZeolCat

Funded by
the European Union

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Catalyst 3D printing for intensified chemical reactors

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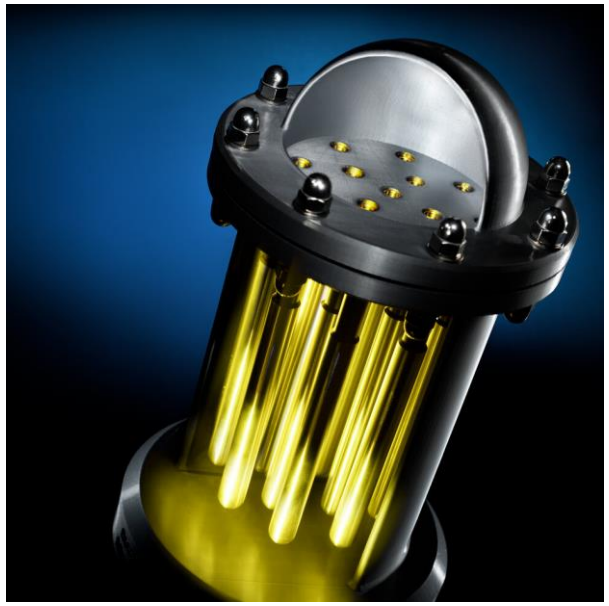
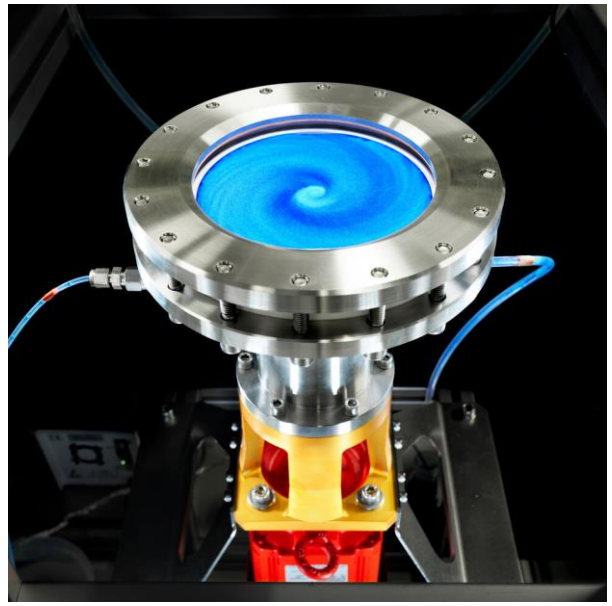
Chemical process intensification

- Intensified processes are characterized by:
  - Lower energy/feedstock consumption
  - Higher efficiency
  - Less byproducts
  - Lower physical footprint

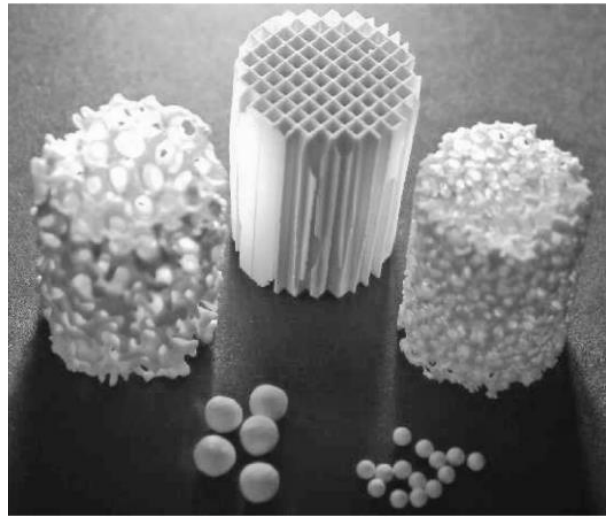
- Common intensification options:
  - **Increased heat/mass transfer**
  - Integrated reaction and separation
  - Forced periodic operation





Shaped catalyst materials

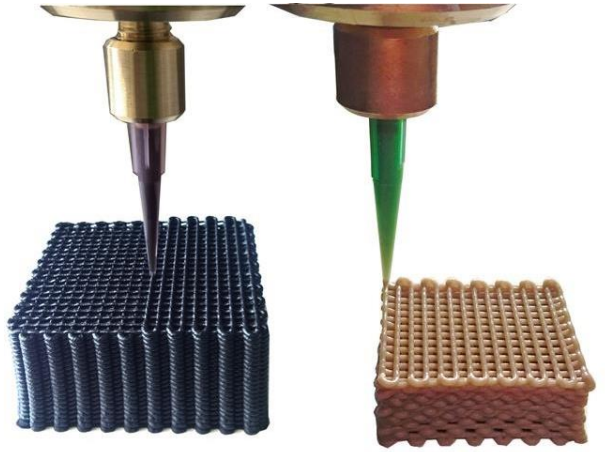
- Catalyst geometry and packing influences
  - Pressure drop
  - Heat/mass transfer
  - Residence time distribution
  - Internal diffusion limitations
- Randomly packed beds
  - High pressure drop and moderate heat transfer
  - High catalyst holdup
- Cellular structures
  - Low pressure drop and excellent heat transfer
  - Low catalyst holdup

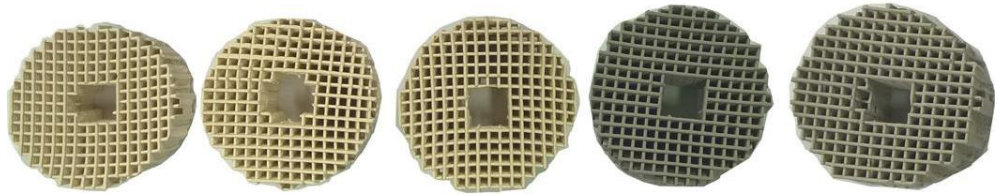


Kraushaar-Czarnetzki, B. and Peter Müller S. (2009). Shaping of Solid Catalysts in *Synthesis of Solid Catalysts*, K.P. de Jong (Ed.).

3D printing of catalysts

- Currently still fairly complex and relatively expensive
- Printed catalyst needs to possess
  - Catalytic properties
  - Porosity
  - Intended dimensions
  - Mechanical stability

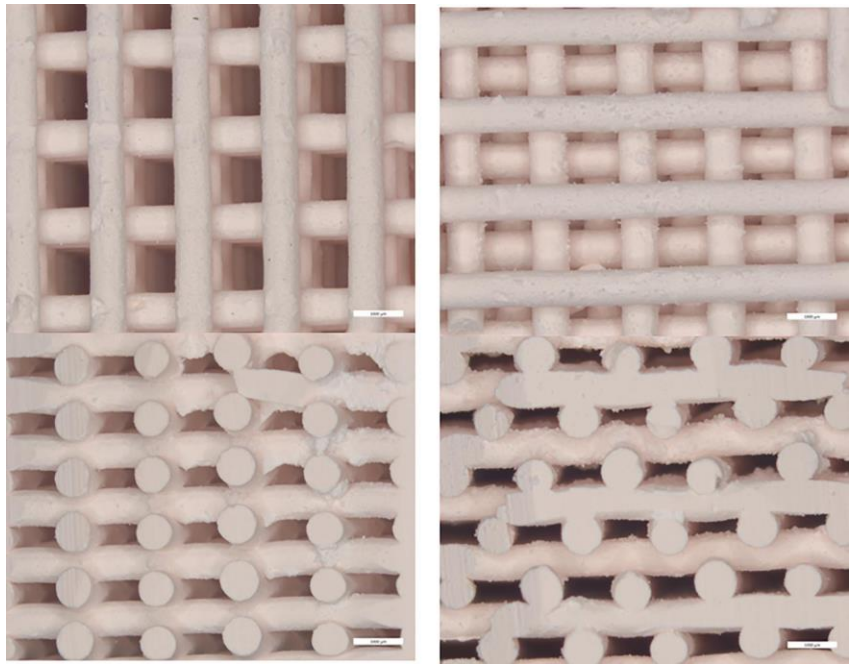




Middelkoop, V. *et al. Chem. Eng. J.*, 2019, 357, pp 309-319.

Logpile structures

- Usually produced via Direct Ink Writing
- Honeycomb monolith-like
  - Open structure
  - Intermediate porosity
  - Operating sweet spot?



Lefevere, J. *et al. Chem. Eng. J.*, 2018, 349, pp 260-268.

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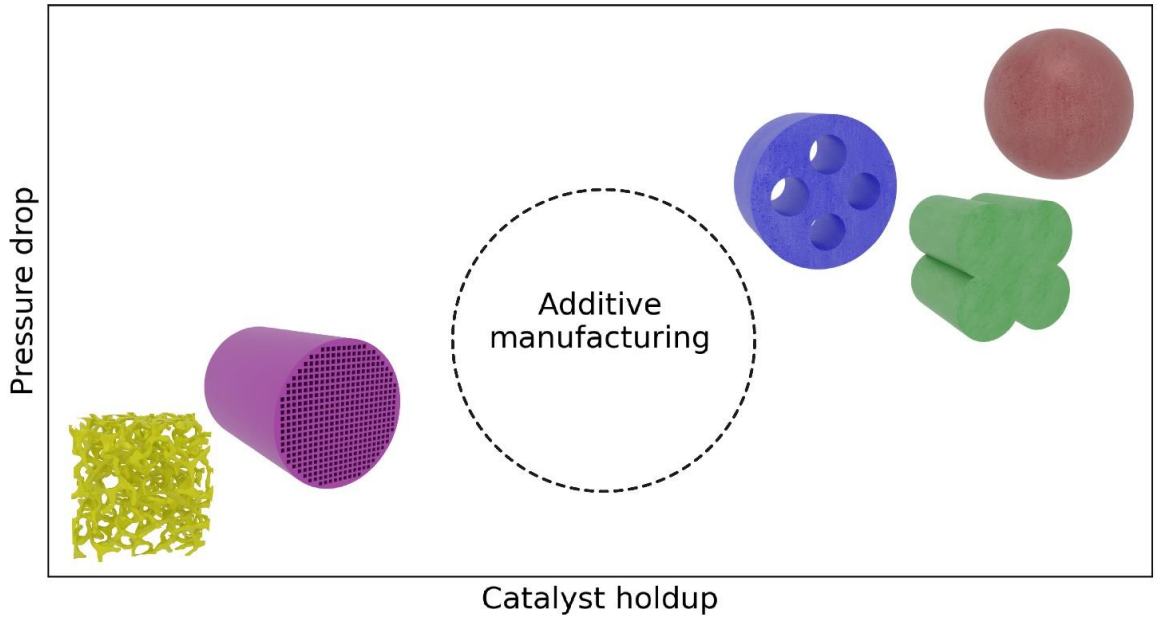
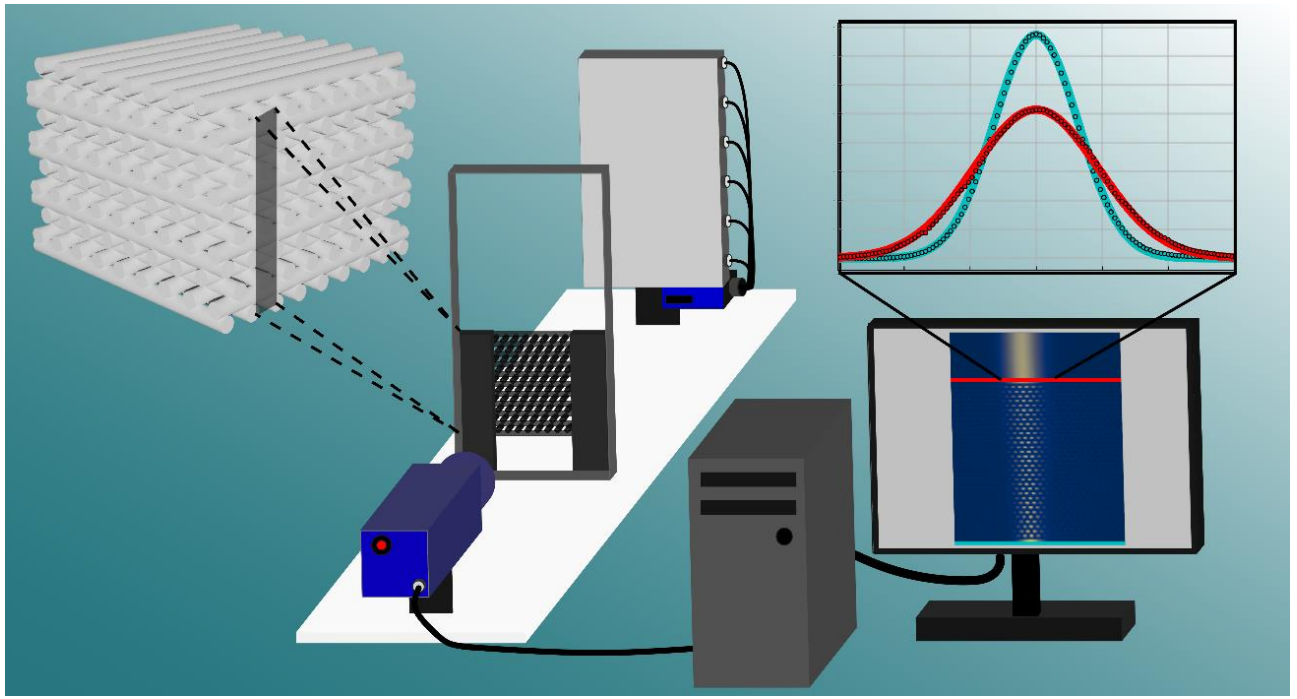


Rosseau, L.R.S. *et al. Front. Chem. Eng.*, 2022, 4:834547.

Logpile structures: quantifying the potential

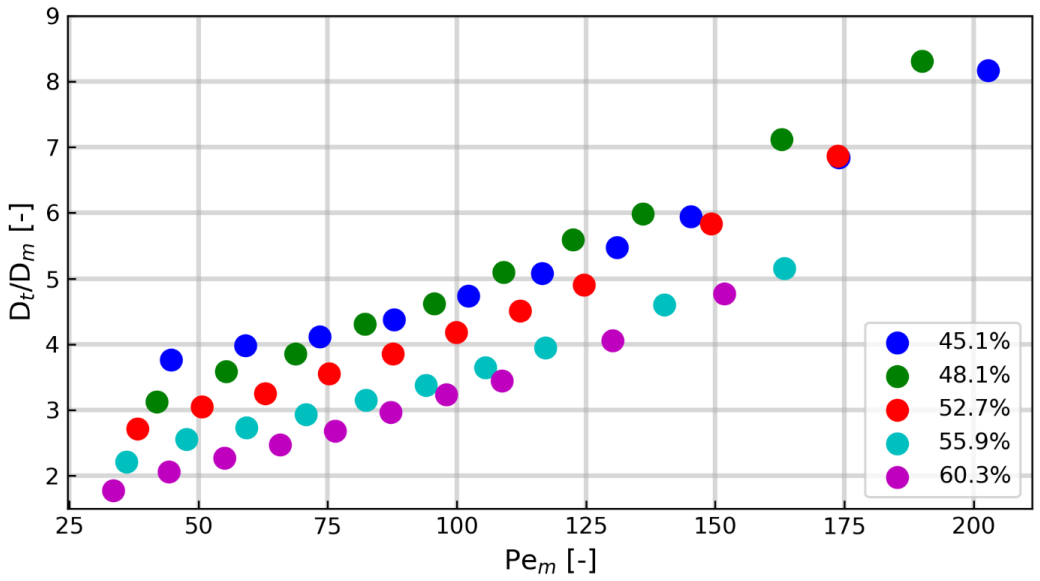
- Inject IR-absorbing tracer gas
- Use IR camera to visualize gas
- Determine mixing phenomena

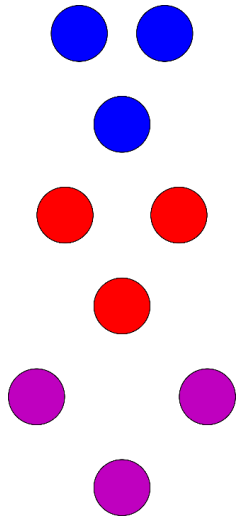


Rosseau, L.R.S. *et al*. *ACS Eng. Au*, 2022, 2:3, pp 236-247.

Logpile structures: quantifying the potential

- Usually produced via Direct Ink Writing
- Honeycomb monolith-like
  - Open structure
  - Intermediate porosity
  - Operating sweet spot?
- Flexibility in operating characteristics

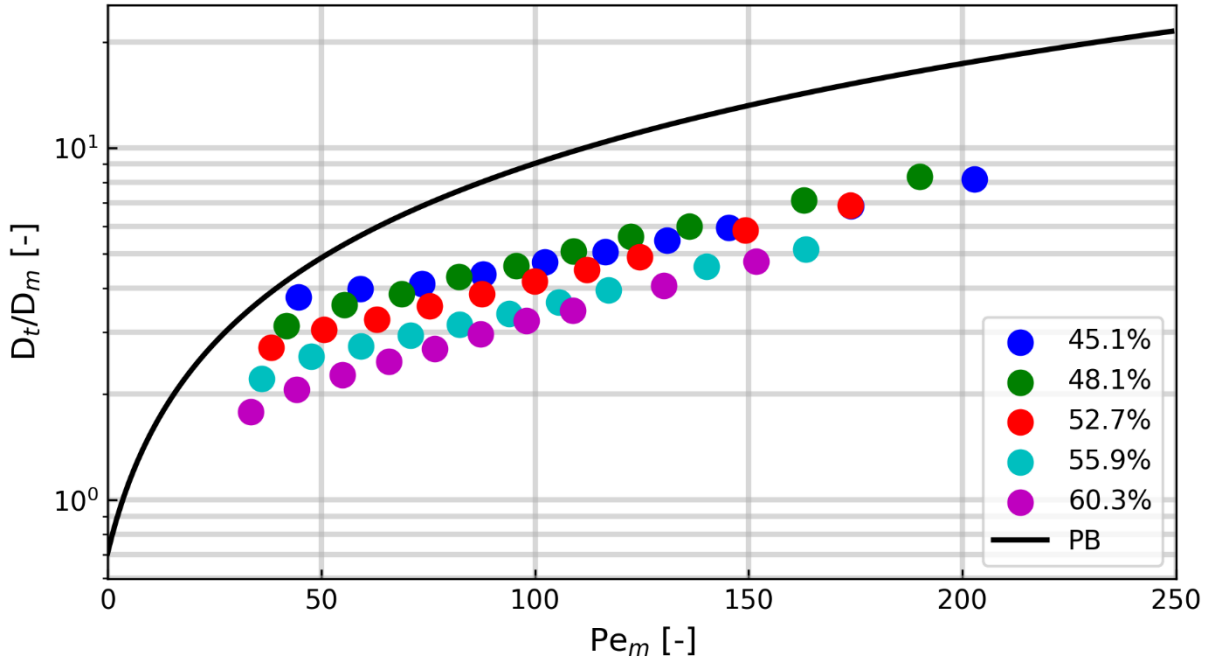




Rosseau, L.R.S. *et al*. *ACS Eng. Au*, 2022, 2:3, pp 236-247.

Logpile structures: quantifying the potential

- Usually produced via Direct Ink Writing
- Honeycomb monolith-like
  - Open structure
  - Intermediate porosity
  - Operating sweet spot?
- Flexibility in operating characteristics
  - But values still relatively low compared to conventional packed bed of spheres



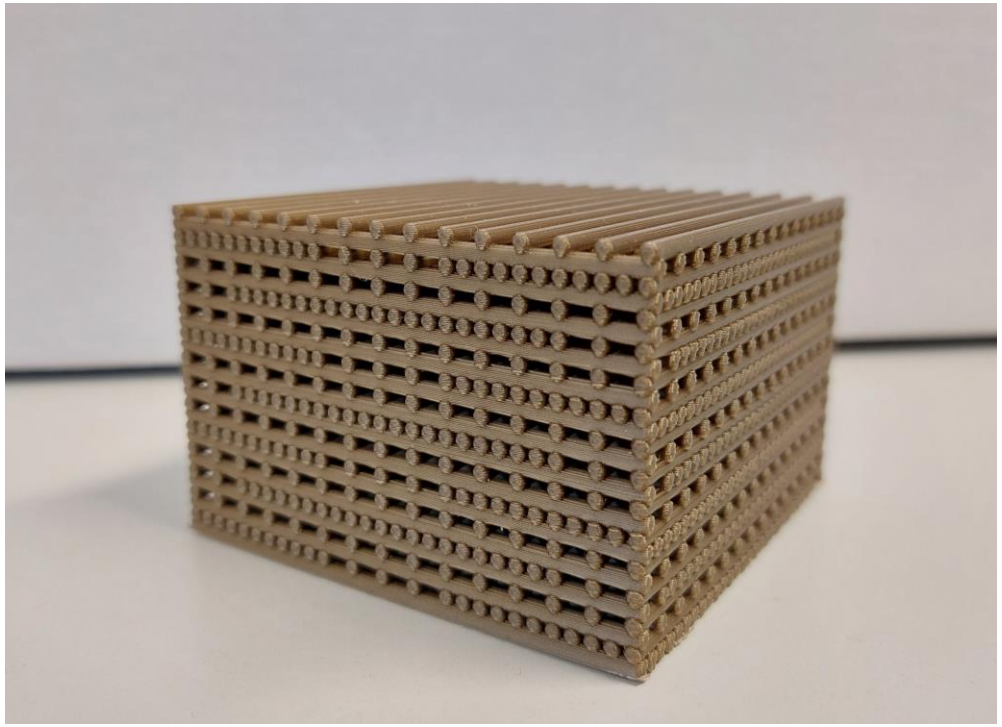
Replotted from: Rosseau, L.R.S. *et al*. *ACS Eng. Au*, 2022, 2:3, pp 236-247.

Local variations in structures

- How to exploit the unique advantages of 3D printing?

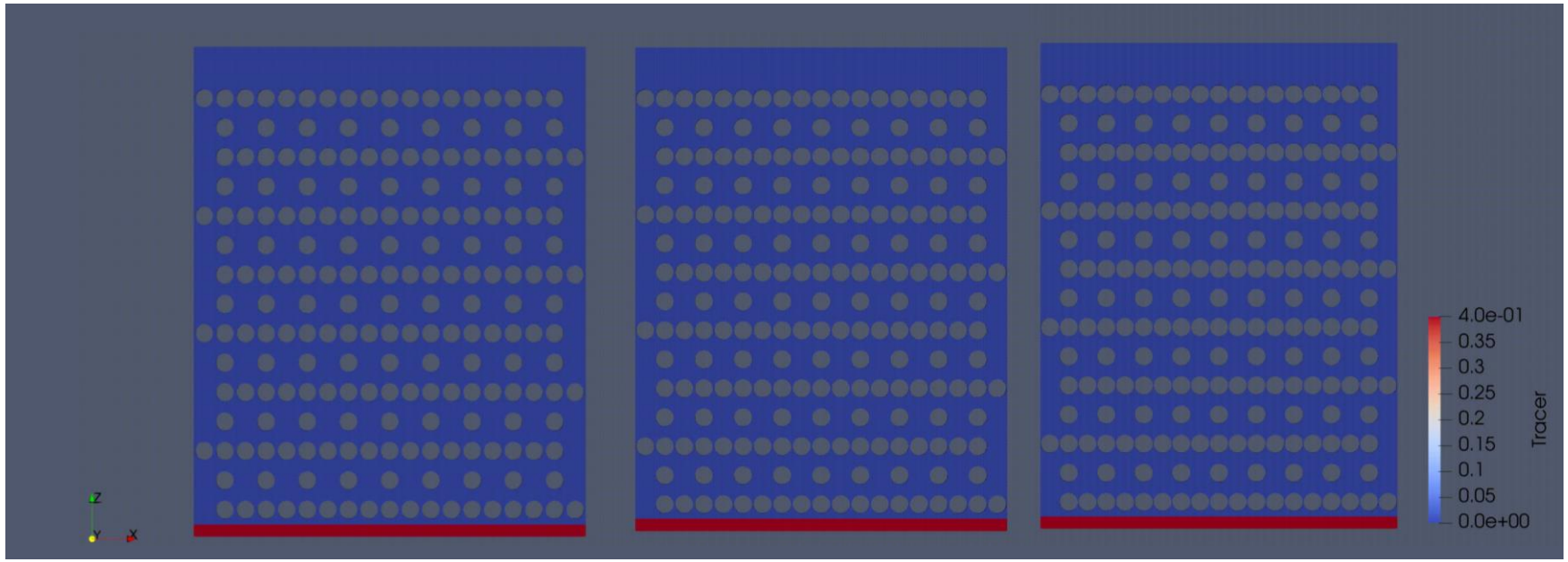
- Local variations
  - Baffle-like structures
  - Guide the flow, without full obstruction

- Wide range of design variables



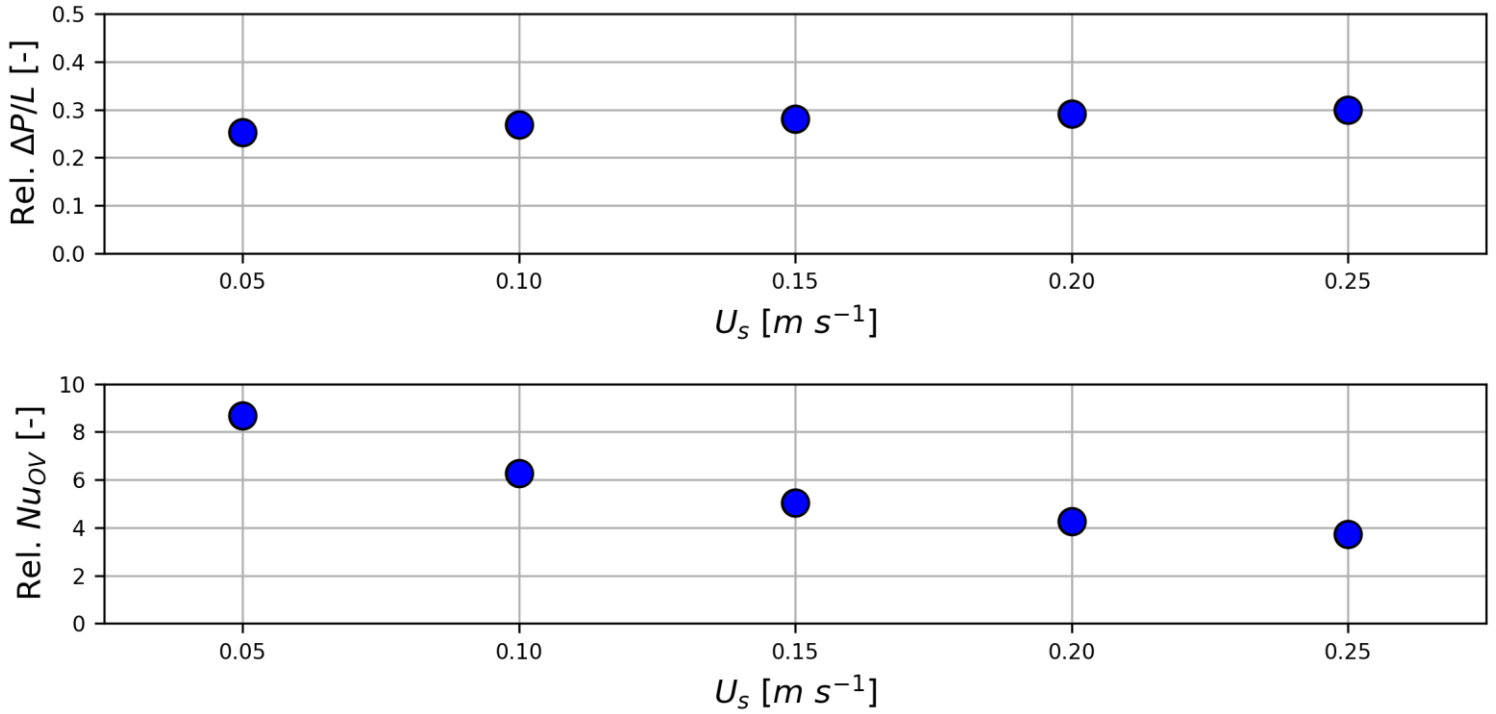
Visualizing cross flow

- Baffle length should be high
- Spacing/pore size can be used as a tuning variable



The pressure drop-heat transfer trade-off

- Comparison with the conventional packed bed shows clear advantage!



Outlook for experimental proof-of-concept

- Relatively brittle samples
- Dimensional accuracy
- Wall-coupling

- Finding a fitting test reaction
  - Highly endo/exothermic
  - Significant equilibrium shift
  - Heat/mass transfer limitations should be main issue

Outlook for experimental proof-of-concept

- Methane dehydroaromatization
  - Complex reaction network
  - Rapid deactivation
- CO_2 hydrogenation (to dimethylether)
  - Multiple reactions with contradicting heat effects
  - Sorption is likely preferred process intensification
- Steam Methane Reforming
  - Strong heat effects
  - Large influence of temperature on equilibrium

THANK YOU

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