Numerical Modelling of the Original and Advanced Version of the TEMKIN-Reactor for Catalysis Experiments in Laboratory Scale

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Introduction

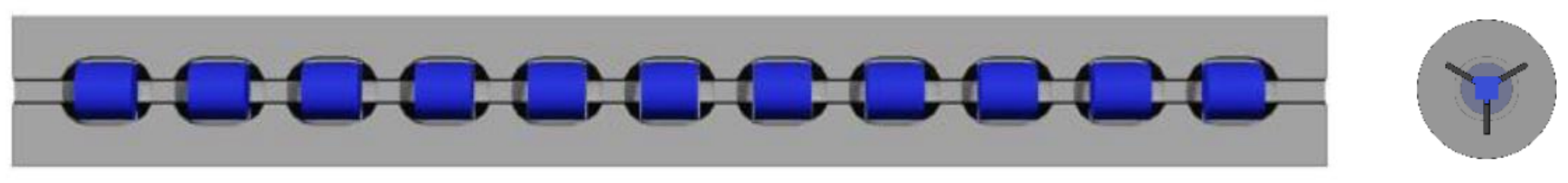
Many industrial, especially heterogeneously catalysed, processes are characterised by a strong interaction between the reaction kinetics and transport phenomena. Because experiments in laboratory scale can be very time- and cost-intensive, Temkin and Kul'kova developed a new reactor design for the direct testing of industrial catalysts.[1] Based on this concept of linearly alternating catalyst and inert pellets inside a small tube, our working group developed an advanced version of this reactor where the catalyst pellets are aligned in the centre of separate small cavities.[2] The performance of the two TEMKIN reactor designs regarding catalysis experiments is evaluated and compared by using COMSOL Multiphysics®.[3-4]

Overview

TEMKIN reactor design

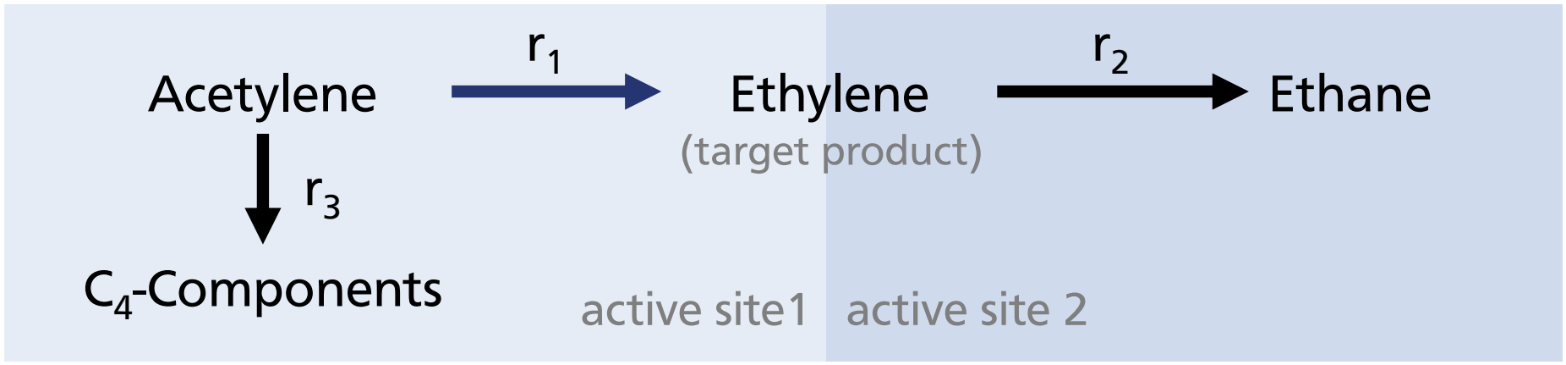
- Original version

- Advanced version



Selective hydrogenation of acetylene

- Removal of acetylene traces in the C_2 cut of a steam cracker
  - Industrial tail-end conditions
    - T = 45 °C, p_{abs} = 11 bar, GHSV = 4000 h^{-1}
    - Hydrogen, acetylene, propane (standard): 1 Vol-% each; Ethylene: 30 Vol-%; Argon: 67 Vol-%,
  - Cylindrical Pd-Ag/Al_2O_3 egg shell catalyst
- Kinetics based on PFR experiments[5]
  - Reactions occurring at two different active sites s_1 and s_2



$$\text{Acetylene} \xrightarrow{r_1} \text{Ethylene} \xrightarrow{r_2} \text{Ethane}$$

$$\text{Acetylene} \xrightarrow{r_3} C_4\text{-Components}$$

(target product)

active site1 active site 2

$$r_1 = \frac{k_1 p_A p_{H2}^{\frac{1}{2}}}{\left(1 + K_{A,13}\, p_A\right)^2} \quad r_2 = \frac{k_2 p_{Ey} p_{H2}}{\left(1 + K_{A,2}\, p_A\right)^2} \quad r_3 = \frac{k_3 p_A p_{H2}^{\frac{1}{2}}}{\left(1 + K_{A,13}\, p_A\right)^2}$$

↳ Reaction rate scaling factors for varying specific numbers of the two active sites (⇨ see validation)

Computational Method

Mass, energy and momentum balances

- Distinguishing between different domains:
  - Original version
  - Advanced version

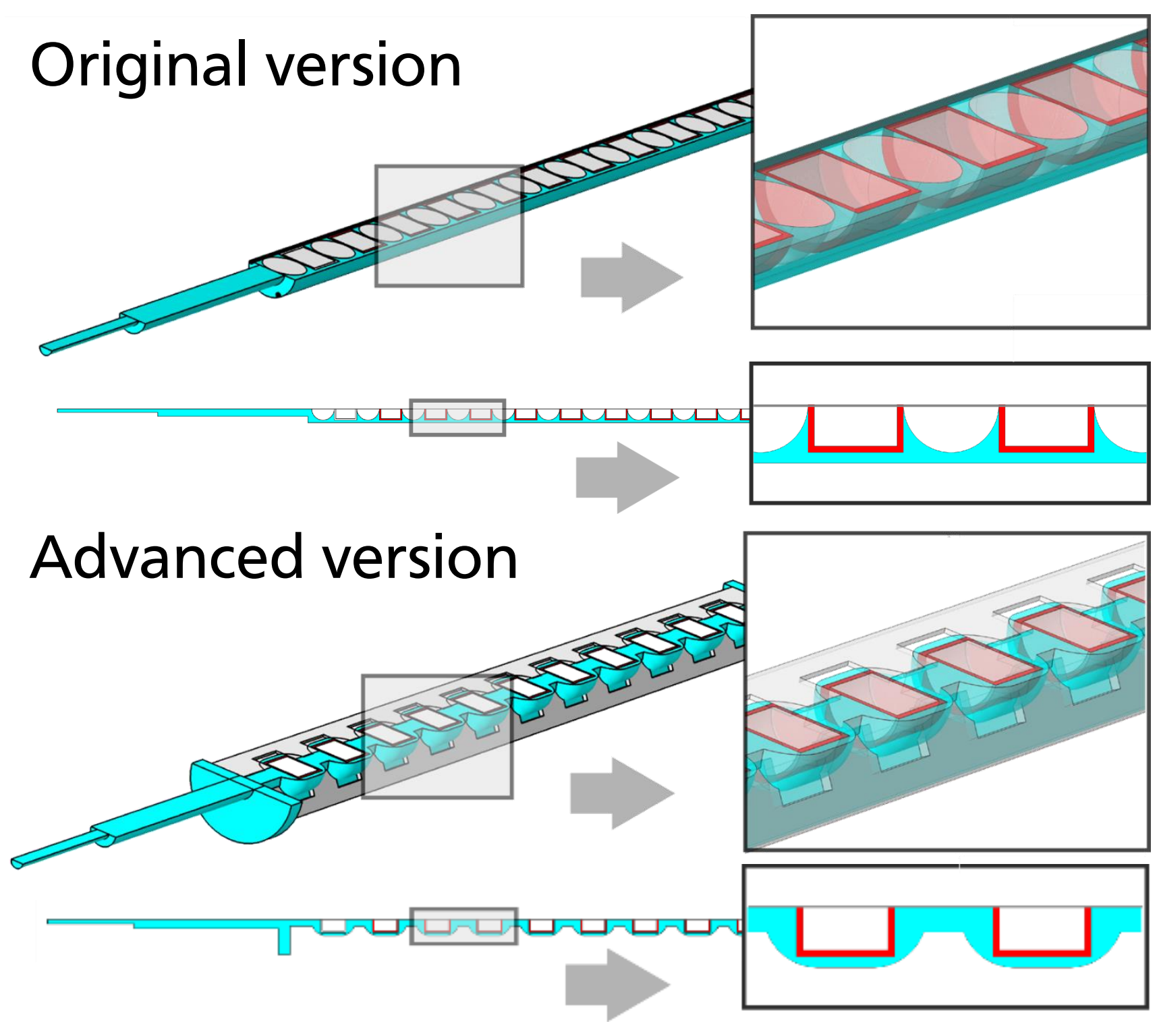


Figure 1 Different balancing domains in the two- and three-dimensional models.

- Free gas flow (cyan)
  ⇨ *Modelling of laminar fluid flow coupled with heat and species transport*
- Inert support (white)
  ⇨ *Modelling of species and heat transport in porous media (no convection)*
- Catalytically active shell (red)
  ⇨ *Modelling of species and heat transport in porous media including reaction kinetics*
- Reactor body (not shown above)
  ⇨ *Modelling of heat transport*

Validation

Pulse tagging experiments

- Fast pulse detection using a thermal mass flow meter

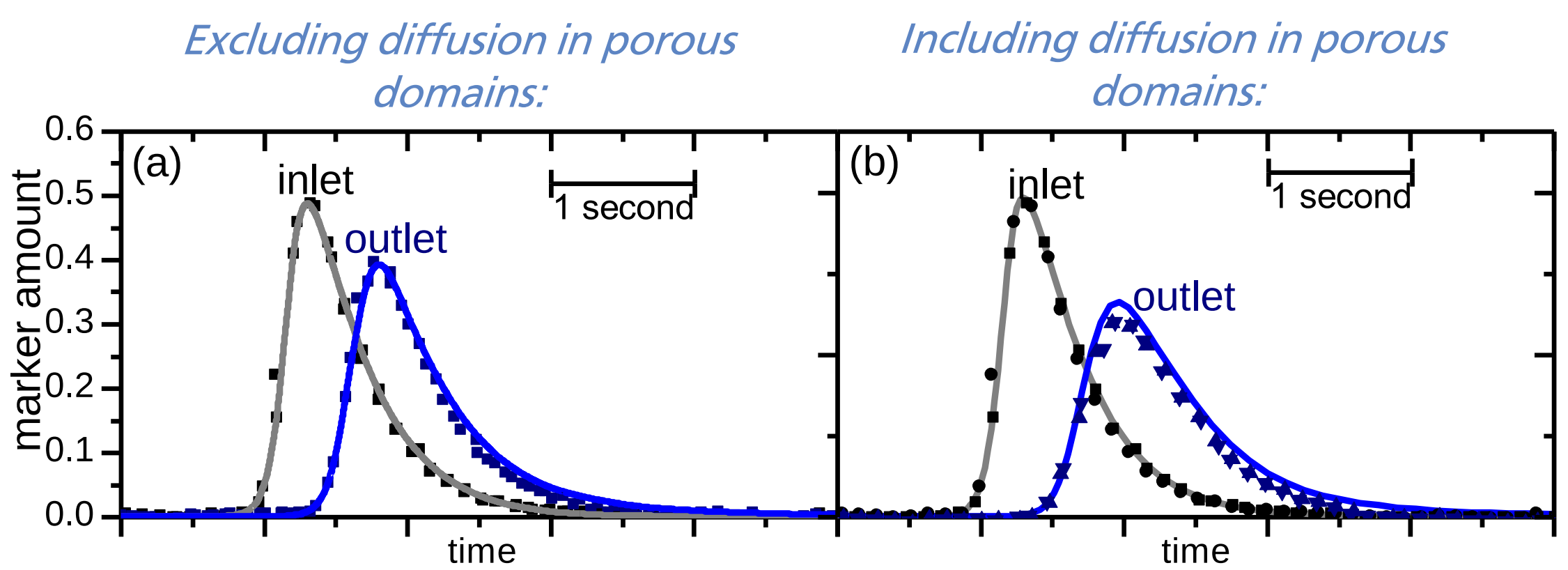


Figure 2 Measured and predicted pulse signal shapes of the advanced TEMKIN reactor. (3D simulations, Carrier flow: 176 mL/min)

Catalysis experiments

- 4 reactor modules in tap-connection arrangement

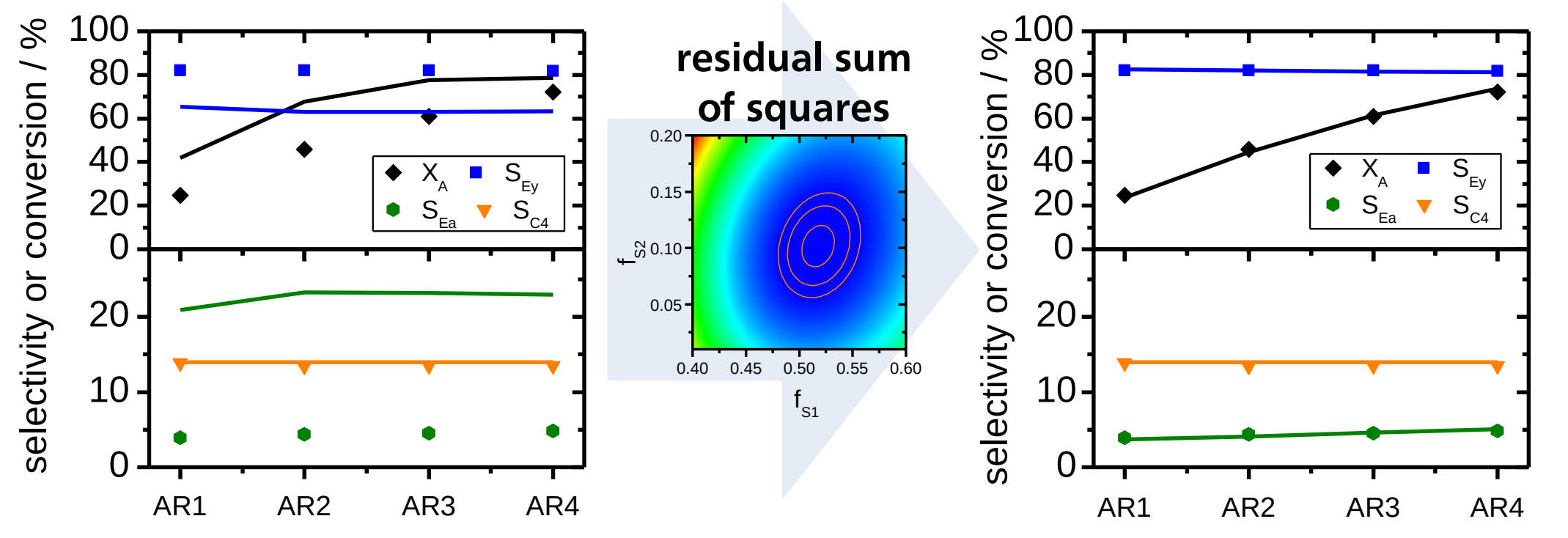


Figure 3 Predicted (lines) and measured (dots) conversion and selectivities when optimising active site rate scaling factors (contour plot).
(2D simulations, tail-end conditions)

⇨ Validation successful

Performance Evaluation regarding Catalysis Experiments

Residence time distributions

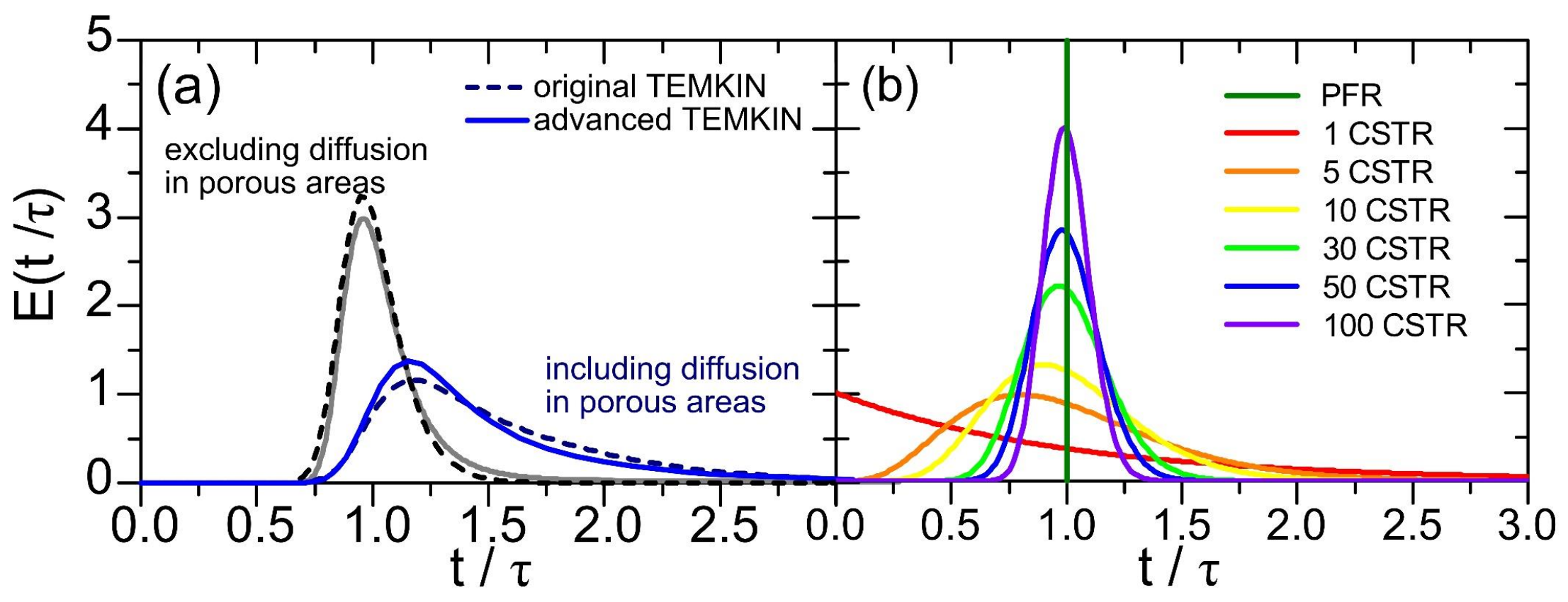


Figure 4 Simulated residence time distributions of acetylene in comparison to simple CSTR cascade models.

⇨ Simple CSTR cascade models fail due to complex intraparticular mass transport

Influence of mass transport

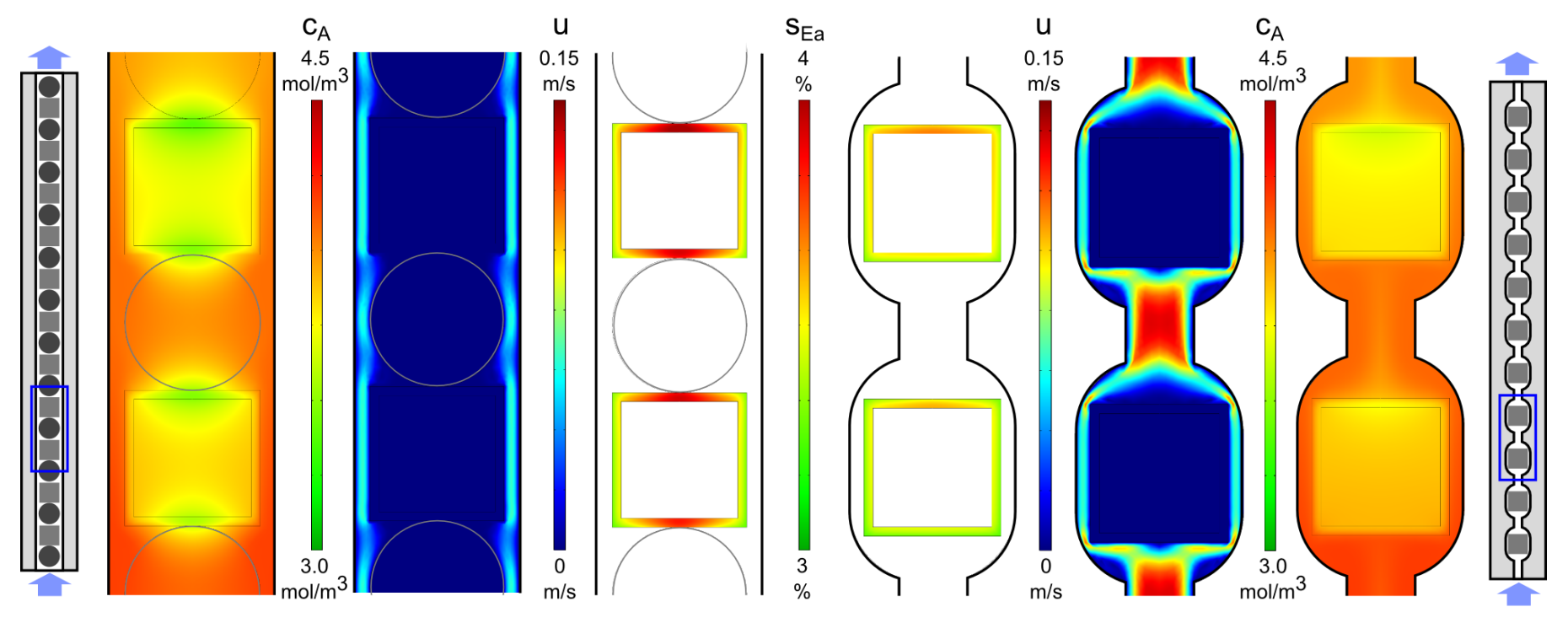


Figure 5 Colour coded values of the acetylene concentration c_A, gas velocity u and differential ethane selectivity s_{Ea}.

⇨ Minimising transport limitations by reducing dead zones

Thermal conditions

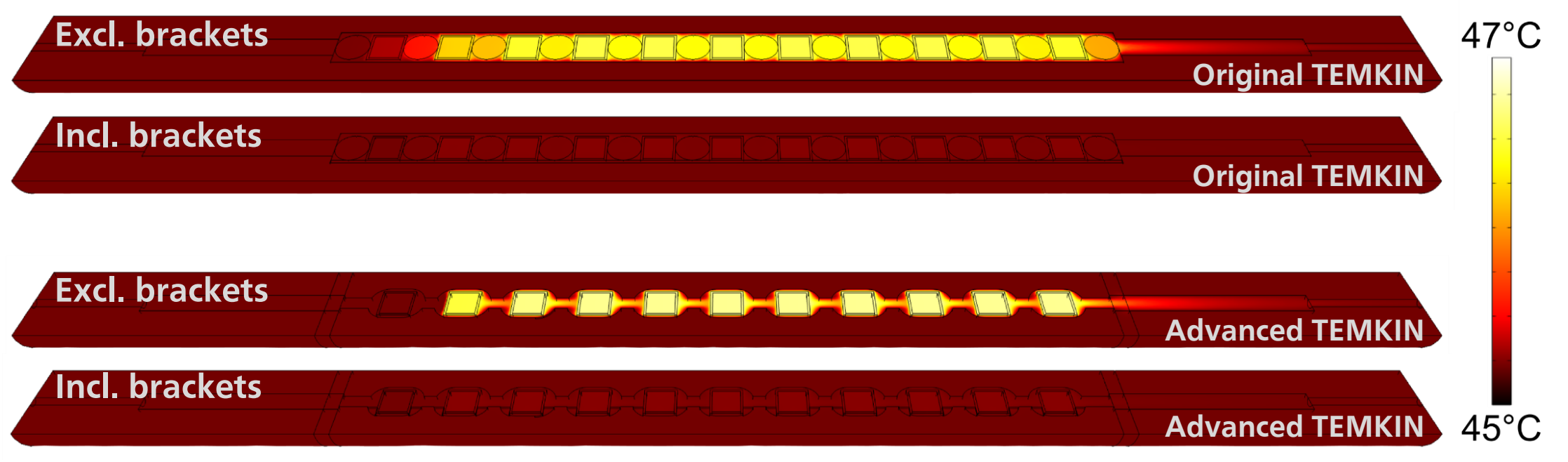


Figure 6 Temperature distribution in the reactors either including or excluding pellet brackets assuming a typical reaction heat in the active shell under tail-end conditions.

⇨ Isothermal behavior in both reactor types

References

[1] M. Temkin, N. V. Kulkova, *Kinet. Katal,* **1969**, 10, 461-463.
[2] a) Patent DE200920003014, **2009**.
b) T. Schulz, Thesis, TU Darmstadt, in preparation.
c) M. Kuhn, M. Lucas, P. Claus, *Chemie Ingenieur Technik*, submitted.
[3] COMSOL Multiphysics, Version 4.3b, COMSOL AB, Stockholm.
[4] D.Götz, M.Kuhn, P.Claus, *Chemical Engineering Research and Design*, submitted.
[5] A. Pachulski, R. Schödel, P. Claus, *Applied Catalysis A: General* **2012**, 445–446, 107– 120.

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