Numerical Simulation Techniques for the Efficient and Accurate Treatment of Local Fluidic Transport Processes Together with Chemical Reactions

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DFG Priority Program 1740: Reactive Bubbly Flows

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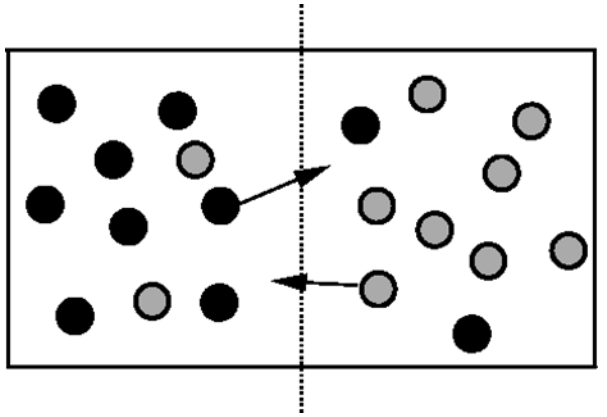
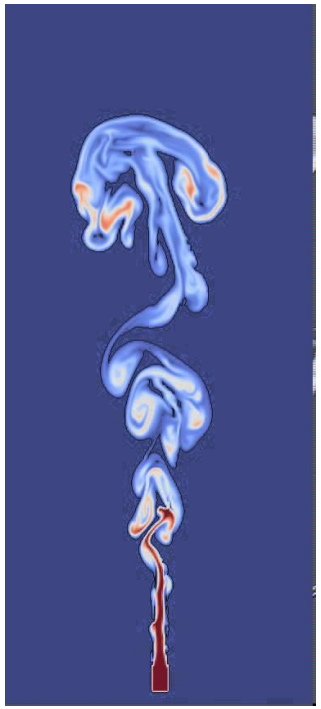
The central of the priority program is to develop new and improved methods for the analysis, modelling and computing

- ❑ local hydrodynamics with turbulence,
- ❑ local concentration distribution with mass transfer / mass transport,
- ❑ reaction progression with transport limitation





TU Dortmund Contribution

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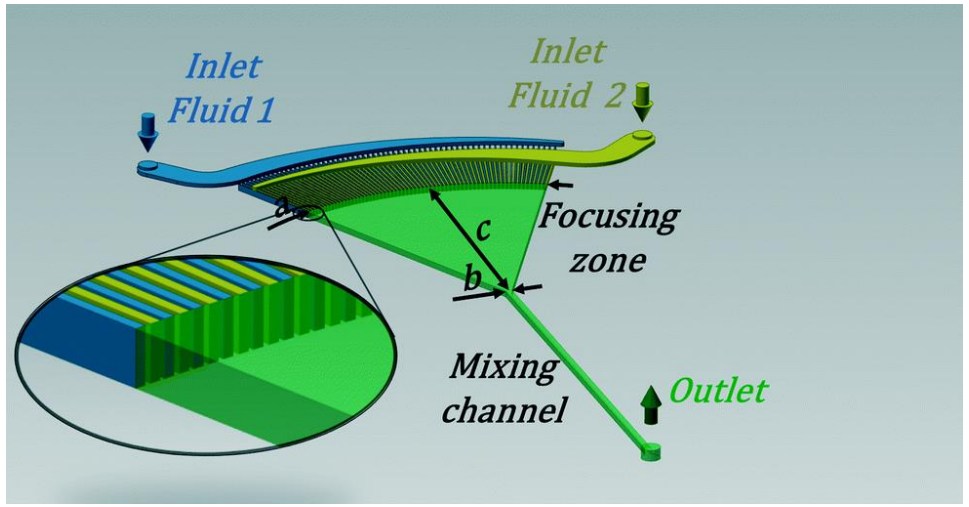
Leading Experiment I **'Superfocus Mischer'**

- Geometrically complex configuration
- Miscible fluids
- Species undergoing chemical reactions.



Leading Experiment II **'Taylor Flow Capillary'**

- Combination of the achieved developments

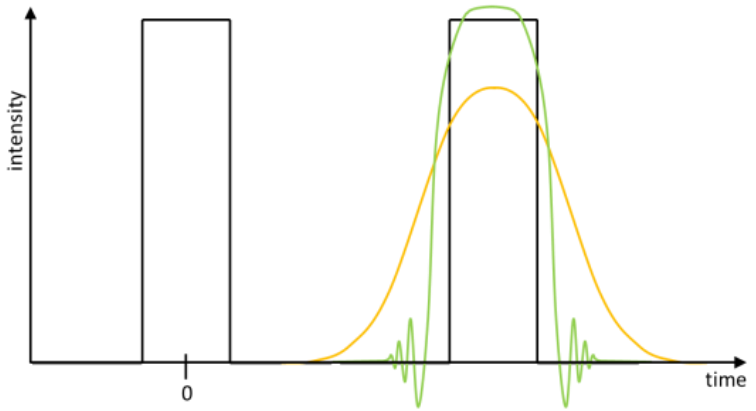
Numerical Techniques for Convection Dominated Transport

High-resolution AFC stabilization

$$\frac{\partial c}{\partial t} + \nabla \cdot (\mathbf{v}c) - \alpha \Delta c = f(c) \qquad \text{with} \quad \alpha \longrightarrow 0$$

Standard discretization face shortcomings for non-smooth solutions

- Low order methods lead to smearing
- High order methods introduce unphysical oscillations



Remedy: Algebraic Flux Correction method [1]

- Local extremum diminishing (+)
- Positivity preserving (+)
- Transforms the linear problem to a non-linear one (-)
- Robust and highly accurate (+)

[1] Kuzmin D.; Turek S.: Flux correction tools for finite elements, Journal of Computational Physics, **175**:525–558, DOI: 10.1006/jcph.2001.6955, 2002.

Pde-free Grid Deformation Method

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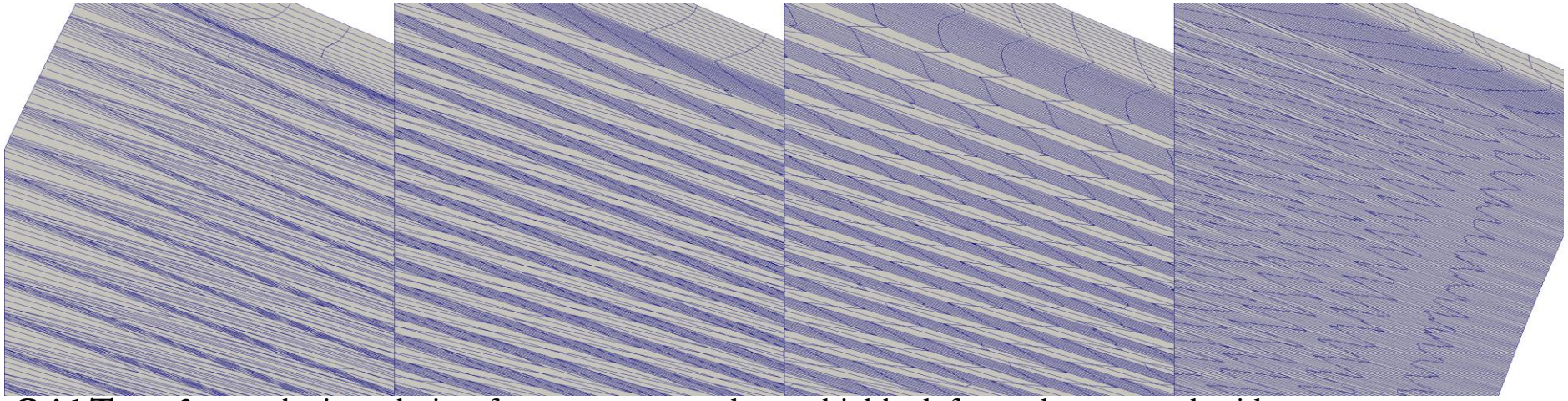
- Construction of (dynamic) monitor function related to distance, curvature, concentration **gradient**, vorticity, etc.
- Anisotropic Laplace smoothening (fast due to the algebraic realization → no PDE) [1]
- Arbitrary Lagrangian-Eulerian Method (ALE) for non-stationary problems
- Handling geometrically complex/dynamic problems in the framework of moving interfaces even on unstructured meshes

SuperFocusMixer application

- Scalar field transport is realized on structured meshes (via automatic mesh generator)
- Monitor function is scalar field related



+ Grid Transfer – velocity solution from unstructured onto highly deformed structured grid

[1] B¨aumler K,, Simulation of single drops with variable interfacial tension, PhD Thesis, 2014, Erlangen, Friedrich-Alexander-University Erlangen-Nurnberg.

Numerical Techniques For Fast Chemical Reactions

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Simulation Strategy For Laminar Steady Flows (one way coupling→ diluted solutions):

1) Compute the steady state velocity distribution on an appropriate grid
2) Compute the steady concentration fields

iterations

- deform the computational grid using the monitor function (concentration gradients)
- interpolate the velocity field from the original grid to the deformed one
- decouple the transport of species in space (diffusion and convection) from the reaction term

1st step PDE in spatial dimension Δt 2nd step ODE in chem. species dimension $M\left(\frac{\Delta t}{M}\right)$

$$\frac{\partial c_i}{\partial t} + \nabla \cdot (\mathbf{v} c_i) - \alpha \Delta c_i = s(c_{j=1,2,\dots N}) \quad i = 1,2,\dots N$$

RHS terms:
Reaction mechanism/constant specific!

According to Reaction Engineering:

$$r_j = k_j \prod_{i=1,n_R} c_i^{|\nu_{j,i}|} \quad for \quad j = 1, n_{CR}$$

$$\frac{dc_i}{dt} = \sum_{j=1,n_{CR}} \nu_{j,i}\, r_j \quad \text{and} \quad k_j = k_{0j} e^{-E_a/RT}$$

n_R : number of reactants
n_{CR} : number of chemical reactions
ν_i : stechiometric coefficients
$k_{0,j}$: reaction rate constants

Possible difficulties

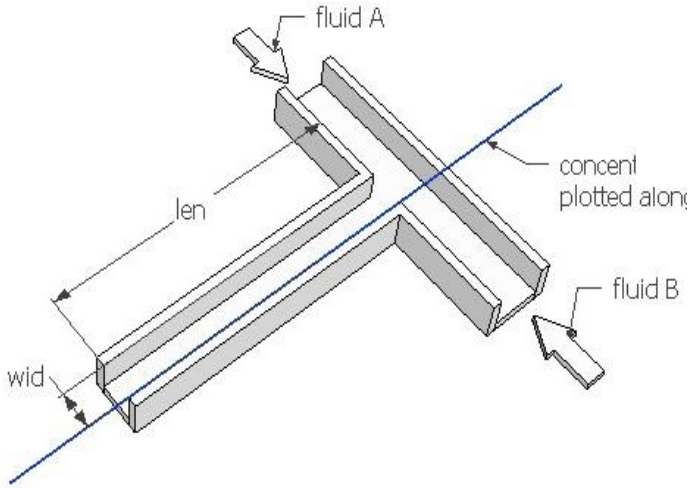
- Extreme resolution requirements of the chemical species → AFC + GD
- Necessity of multiple grids for different transport problems → GT
- Extremely different time-scales (chem. react. vs transport) → OS

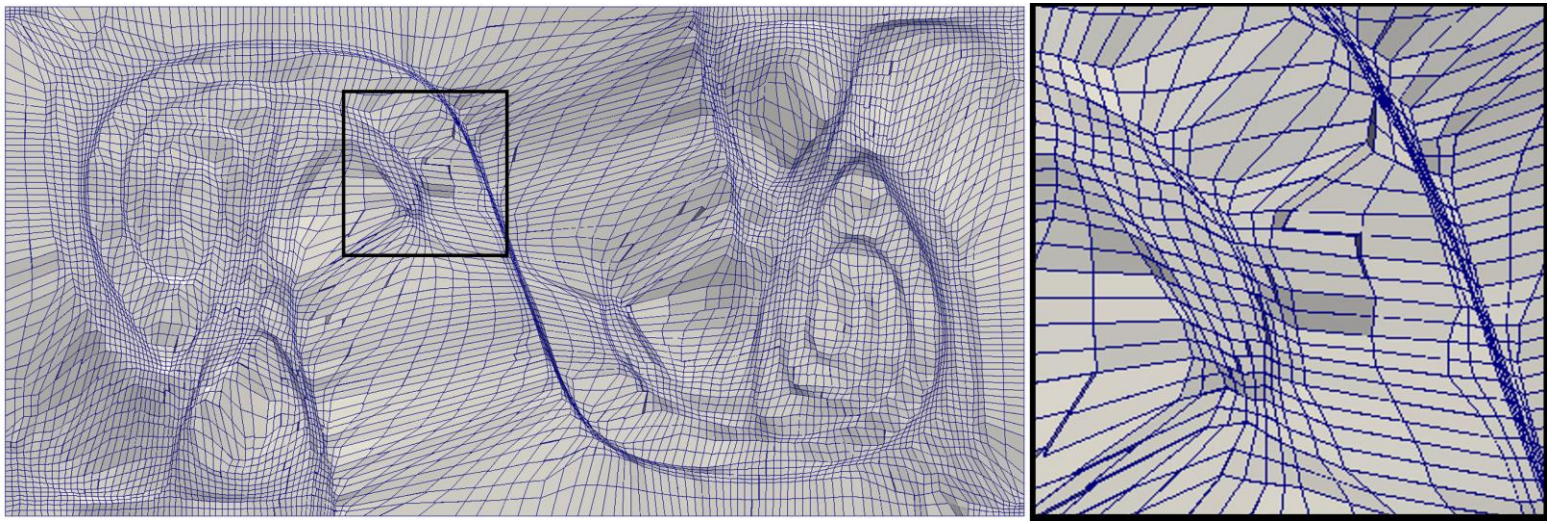
T-Mixer Validation

$$Na\,OH + HCl \rightarrow NaCl + H_2O$$

$Re = 186$ Engulfment regime $Sc \approx 300$ for the pair H^+/Cl^-
$Sc \approx 470$ for the pair Na^+/OH^-

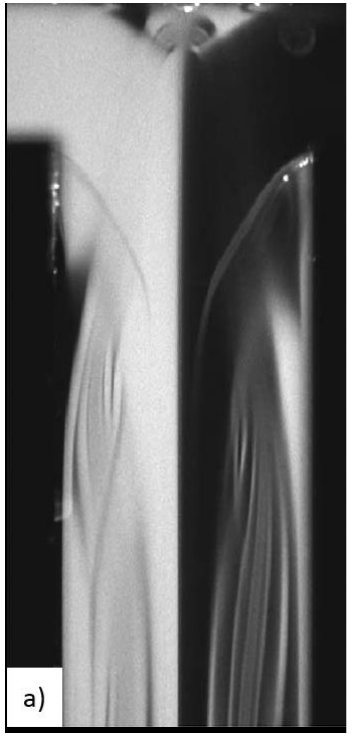
Reaction rate $k = 10^{11}\ L\ mol^{-1}\ s^{-1}$





[1] Bothe D., Lojewski A., Warnecke, H.-J., Computational analysis of an instantaneous reaction in a T-microreactor, AIChE Journal, **56**, pp: 1406–1415, 2009, DOI: 10.1002/aic.12067.

T-Mixer Validation



TUHH[1] - Bothe et.al.[2] –TUDO[3]

[1] Hoffmann, M.; Schlüter M.; Räbiger N.: Experimental investigation of liquid-liquid mixing in T-shaped micro-mixers using µ-LIF and µ-PIV, Chemical Engineering Science, 61(9), pp. 2968-2976, 2006, DOI: 10.1016/j.ces.2005.11.029.

[2] Bothe D., Lojewski A., Warnecke, H.-J., Computational analysis of an instantaneous reaction in a T-microreactor, AIChE Journal, **56**, pp: 1406–1415, 2009, DOI: 10.1002/aic.12067.

[3] Mierka, O.; Munir, M.; Spille, C.; Timmermann, J.; Schlüter, M.; Turek, S.: Reactive liquid flow simulation of micromixers based on grid deformation techniques. Chemical Engineering & Technology, 2017, accepted article, DOI: 10.1002/ceat.201600686.

SFM-Mixer Validation

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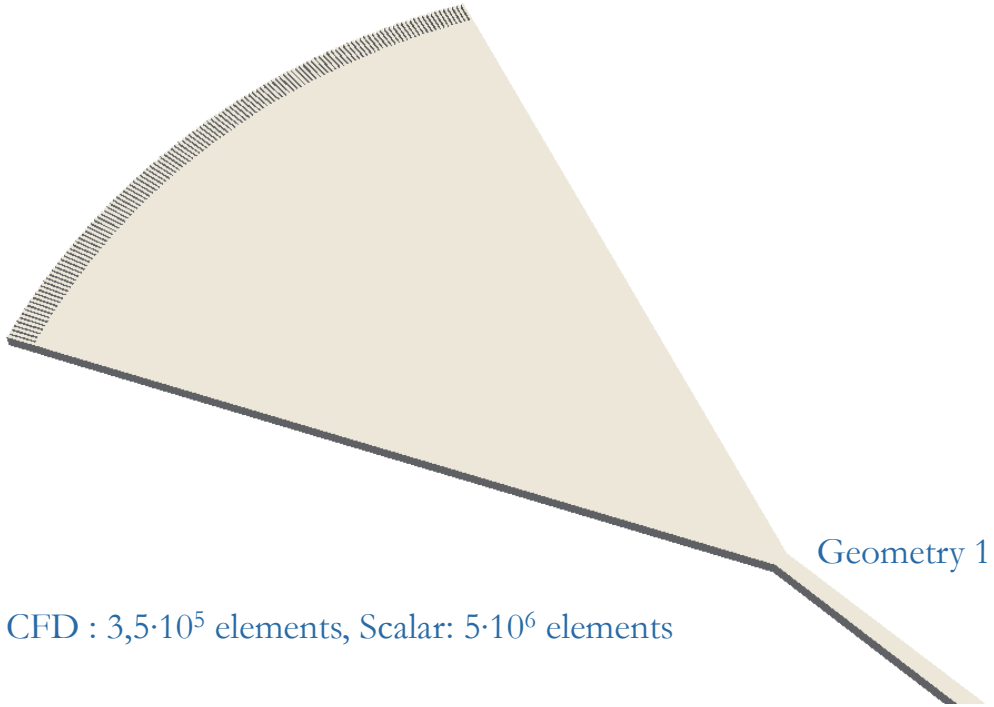
Investigation of different levels of geometrical complexity
- Influence of velocity distribution on mixing and chemical reactions

- Geometrical description from **TUHH!**
- Mesh resolutions guaranteeing mesh convergence
- Timings for Geometry 1 and 2 :
  <1h CFD, <1d Scalar transport on 16 cores



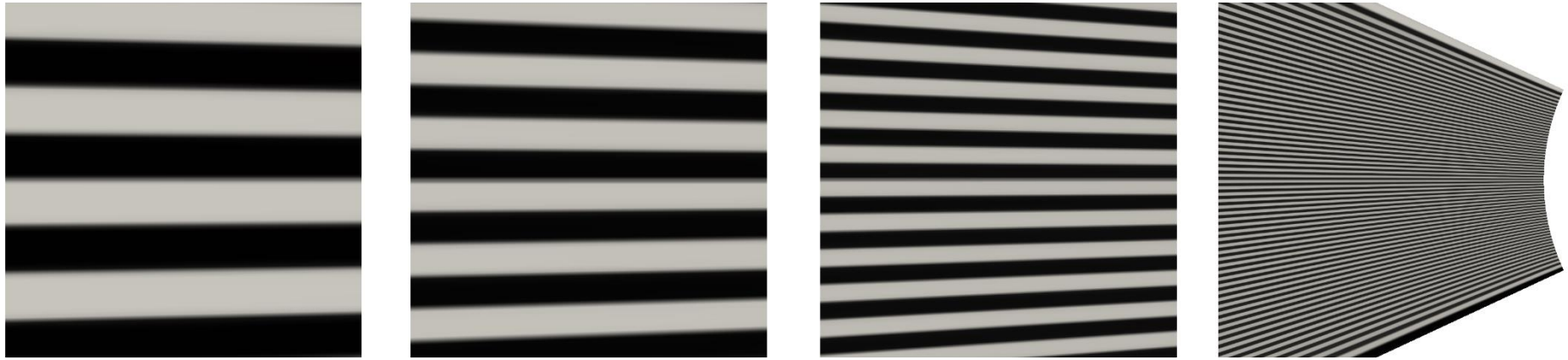
Geometry 1

CFD : $3{,}5 \cdot 10^5$ elements, Scalar: $5 \cdot 10^6$ elements

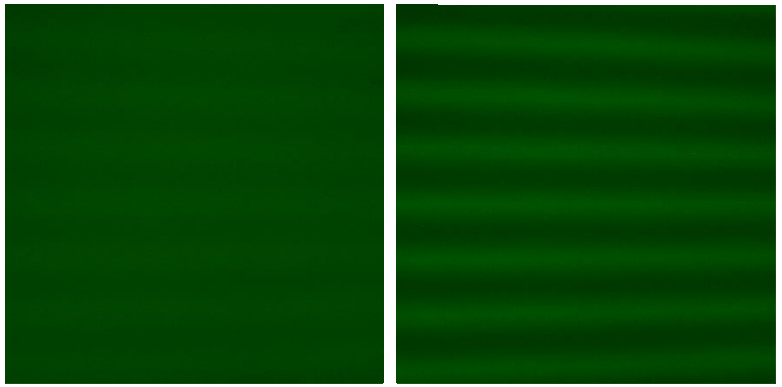
Pure Mixing Without Chemical Reactions

– experimental validation with **TUHH** Geometry 1

$D_{A,B} = 3.0 \cdot 10^{-7}\ \mathrm{cm}^2/\mathrm{s}$

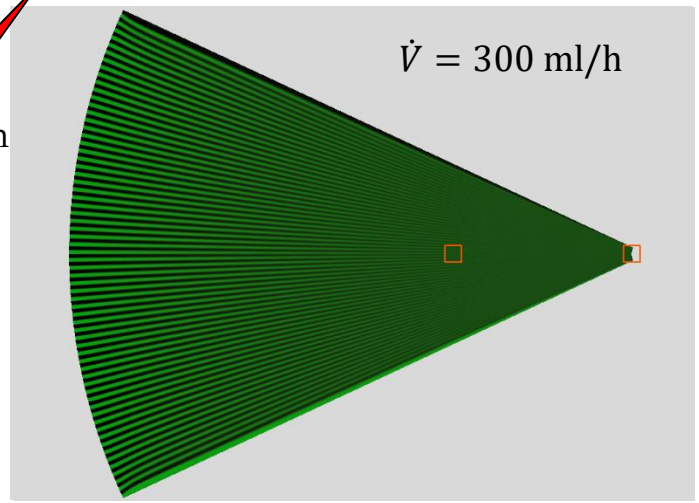


$\dot{V} = 150\ \mathrm{ml/h}$ $\dot{V} = 300\ \mathrm{ml/h}$ $D_{A,B} = 2 \cdot 10^{-5}\ \mathrm{cm}^2/\mathrm{s}$



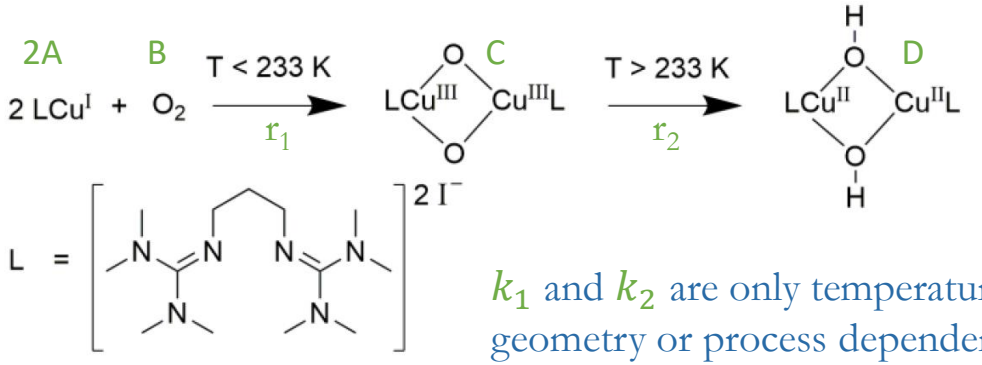
Simulation TU Dortmund

Passive scalar validation on two flowrates.



Pure Mixing With Chemical Reactions

– experimental validation with RWTH Aachen



k_1 and k_2 are only temperature but not geometry or process dependent!

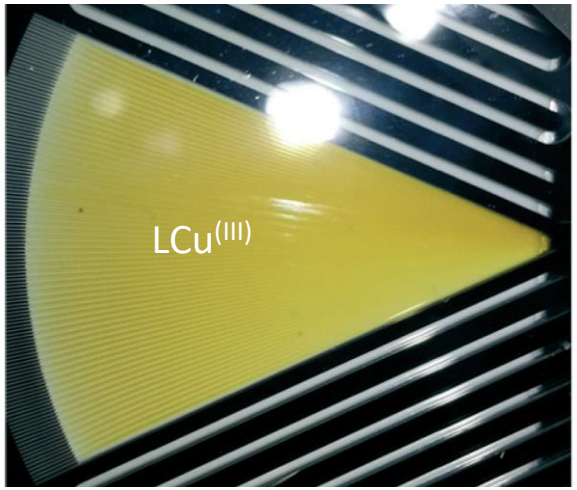
$$r_2 = k_2 c_C^1 = -\frac{dc_C}{dt} = \frac{dc_D}{dt} \quad r_1 = k_1 c_A^2 c_B^1 = -\frac{dc_A}{2dt} = -\frac{dc_B}{dt} = \frac{dc_C}{dt}$$

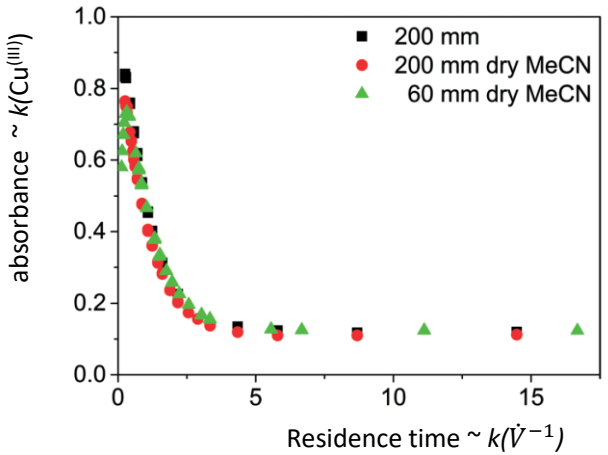
$$k_2 \cong 1\ s^{-1}$$

$r_1 : r_2 = \mathrm{f}(T)$ but in general $r_1 \gg r_2$

"For determining the reaction kinetics … a setup with a fast and close to ideal mixing of the Cu(I) complex and O_2 and in-situ spectroscopy is required."

→ Reaction rates in general strongly depend on the hydrodynamics (mixing)!





[1] Schurr D., Strassl F., Liebh¨auser P., Rinke G., Dittmeyer R., Herres-Pawlis S., Decay kinetics of sensitive bioinorganic species in a SuperFocus mixer at ambient conditions, Reaction Chemistry & Engineering, **1**, pp.:485-493, 2016, DOI: 10.1039/C6RE00119J.

Parameter Study Of Mixing With Chemical Reactions

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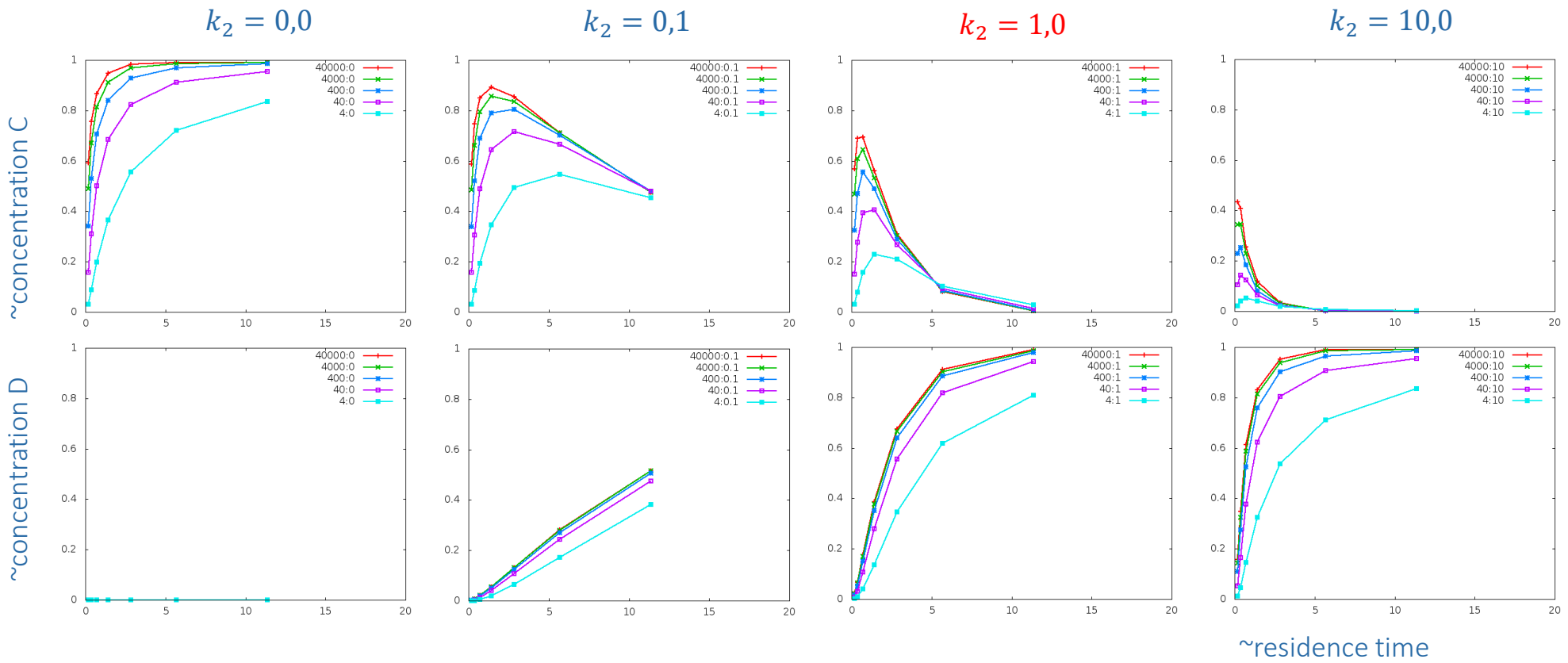
Geometry 0

$$\dot{V} = 0.6 \Leftrightarrow 40.0\ ml/min$$



Mixing With Chemical Reactions Geometry 1

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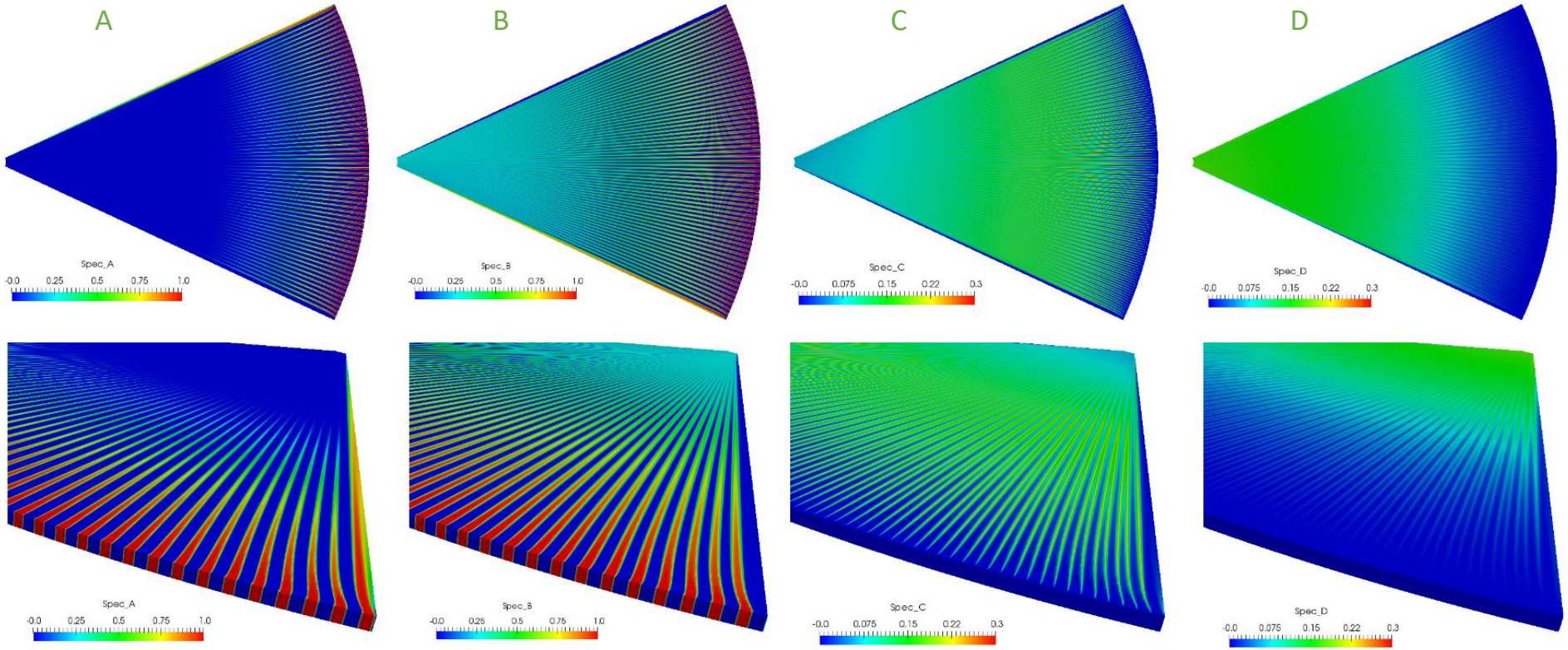
$\dot{V} = 0{,}6\ ml/min$

$k_1 : k_2 = 40000 : 1$



100% conversion (w.r.t. A), (22% C, 78% D)

"selectivity"

Mixing With Chemical Reactions Geometry 2

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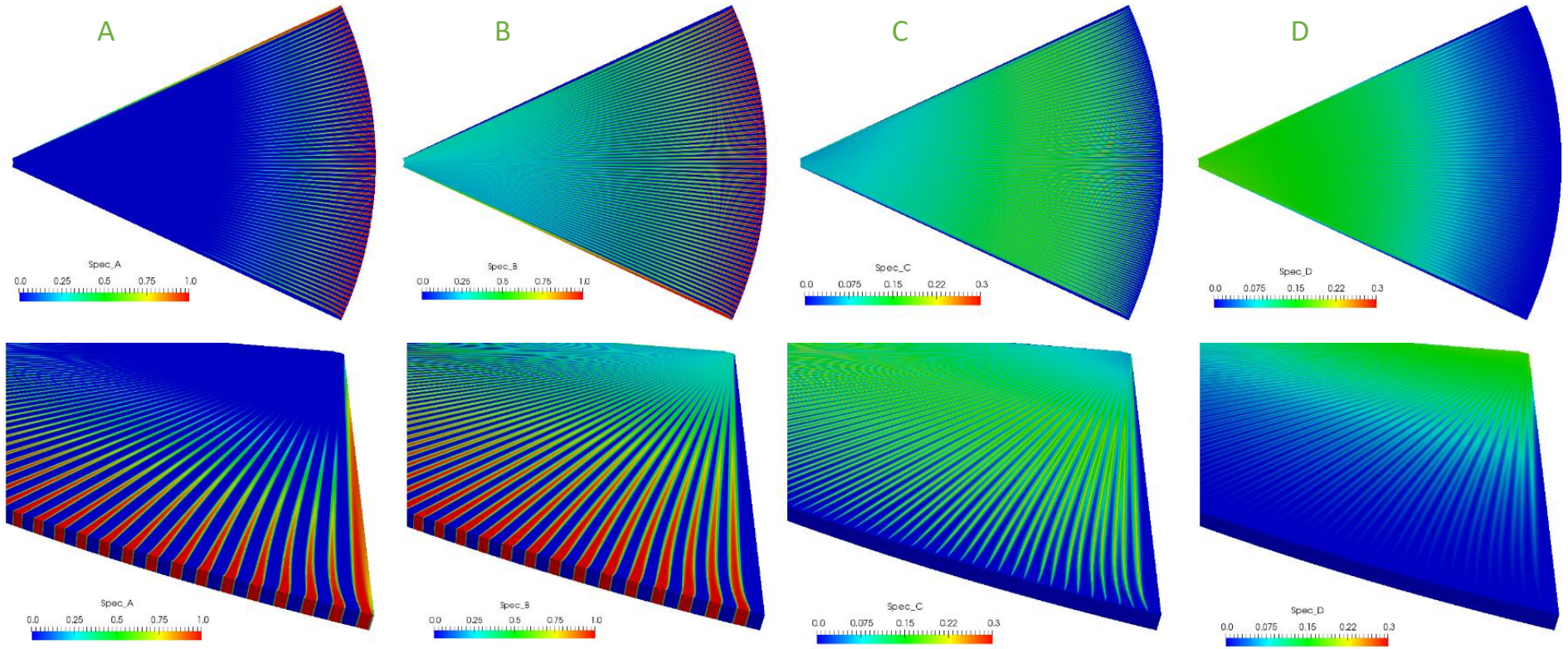
$\dot{V} = 2{,}5\ ml/min$ $k_1 : k_2 = 40000 : 1$



100% conversion (w.r.t. A), (22% C, 78% D)

"selectivity"

Mixing With Chemical Reactions Geometry 2

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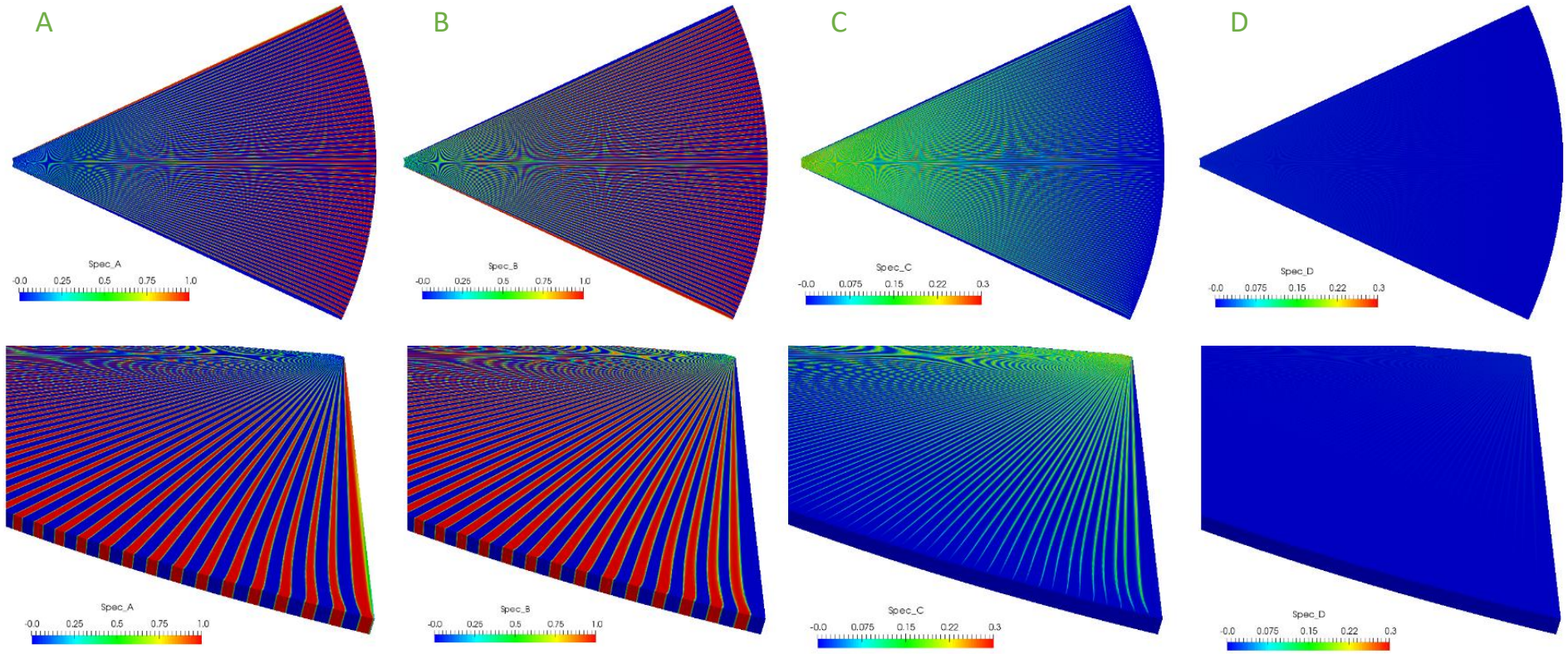
$\dot{V} = 25\ ml/min$ $k_1 : k_2 = 40000 : 1$



91% conversion (w.r.t. A), (84% C, 7% D)

"selectivity"

Mixing With Chemical Reactions Geometry 2

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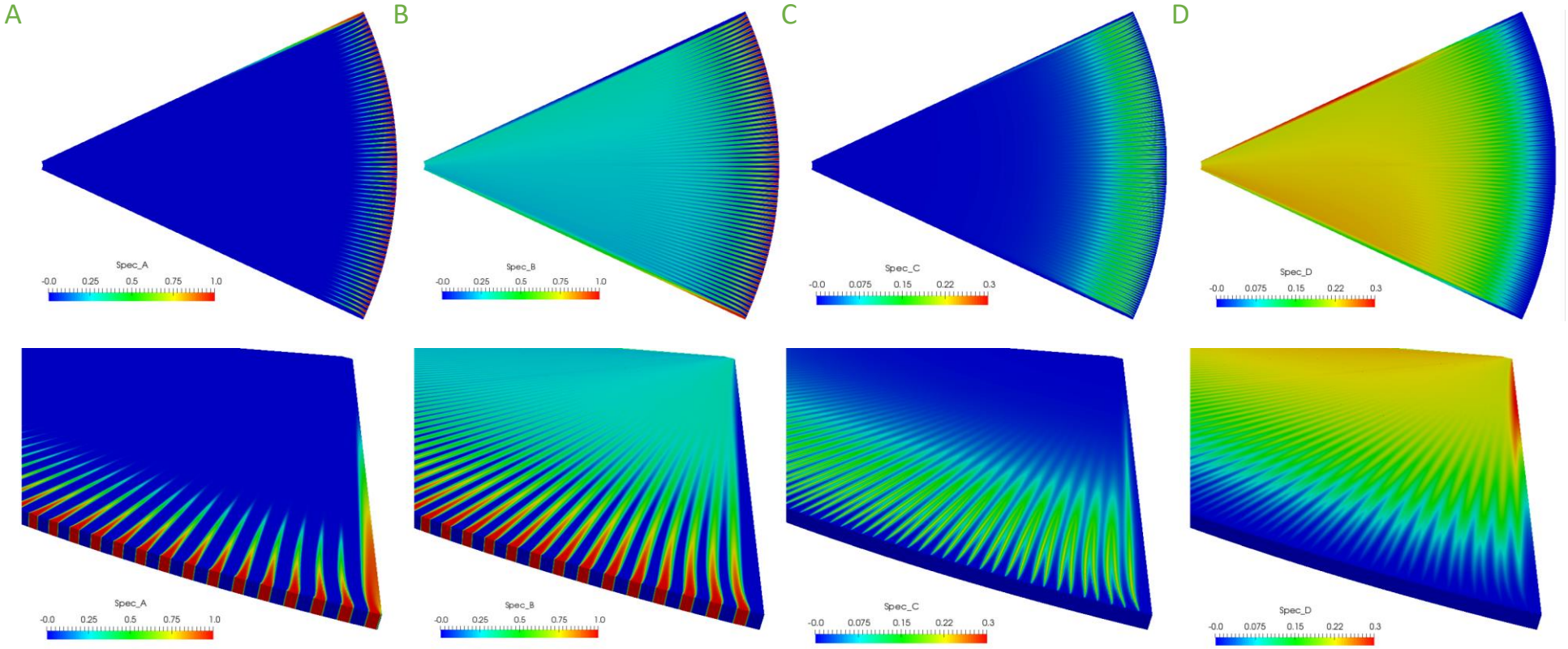
$\dot{V} = 0{,}6\ ml/min$ $k_1 : k_2 = 40000 : 1$



100% conversion (w.r.t. A), (0% C, 100% D)

"selectivity"

SPP 1506 Taylor Bubble Benchmark

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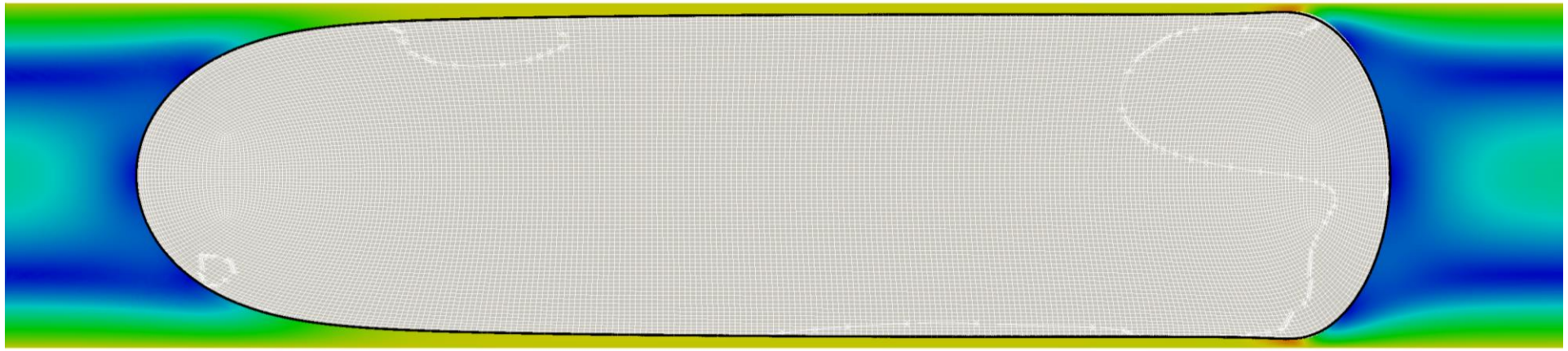
Synergical effect of special numerical ingredients: Laplace Beltrami, Isoparametric FEM, Arbitrary Lagrangian Eulerian Method with Mesh Deformation

Longitudinal cut – experimental (black solid line) vs computational (grey area) results



Levelset-free Method!

No smoothening distance for physical properties and for surface tension, no reinitialization. Mesh needs to be aligned with the interface. Dramatically improved mass conservation.

Benchmark measures		DROPS	FS3D	TURBIT	OF	FFP-l2	FFP-l3	EXP
bubble length	[mm]	7,230	7,197	7,110	7,202	7,199	7,197	7,200
min. film thickness (diagonal cut)	[mm]	0,4392	0,4362	0,4420	0,4566	0,4329	0,4325	0,4331
min. film thickness (longitudinal cut)	[mm]	0,0490	0,0477	0,0280	0,0590	0,0472	0,0473	0,0505
liquid velocity	[cm/s]	-	13,82	-	12,61	13,82	13,82	-
rise velocity	[mm/s]	206,9	197,5	207,8	205,8	197,7	199,6	?203,4?
discretization (el,dof)	[-]	320,000 v_{dof} 26,000 p_{pres}	2,100,000 el	1,200,000 el	300,000 el	8,500 el	68,000 el	-

Marschall H., Boden S., Lehrenfeld Ch., Falconi C. J. D., Hampel U., Reusken A., W¨ orner M., Bothe D., Validation of Interface Capturing and Tracking techniques with different surface tension treatments against a Taylor bubble benchmark problem, Computers and Fluids, **102**, pp.:336-352, 2014. DOI:10.1016/j.compfluid.2014.06.030

Conclusions

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From The Running Period:

- The necessary numerical components have been successfully incorporated into the CFD solver [1]
- Validation of the developed methods has been realized on the basis of the experimental results of TU Hamburg-Harburg (AG Schlüter)

- The reaction module has been qualitatively validated based on the experimental results of Schurr D., Strassl F., Liebhäuser P., Rinke G., Dittmeyer R., Herres-Pawlis S. Quantitave validation is being in progress with (AG Schlüter)

- A novel highly accurate Level Set-free approach has been developed for simulation of multiphase flows which has been validated on the SPP 1506 Taylor bubble benchmark

Outlook To The Next Period:

- To exploit the developed CFD based methodology in a framework of an optimization environment for determination of reaction parameters for subsequent use in CFD related environments (AG Herres-Pawlis)

- Extension of the current scalar-transport-reaction module with an "Electromigration model" being suitable for simulation of ion-related chemical systems (AG Bothe)

[1] Mierka, O.; Munir, M.; Spille, C.; Timmermann, J.; Schlüter, M.; Turek, S.: Reactive liquid flow simulation of micromixers based on grid deformation techniques. Chemical Engineering & Technology, 2017, accepted article, DOI: 10.1002/ceat.201600686.