Università di Pisa

Chemical Reactions in microdevices: experimental and numerical analysis

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Introduction

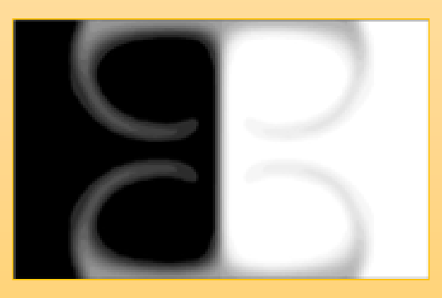
Micro-reactors, constituted by channels with typical dimensions < 1 mm, are receiving an increasing attention in the pharmaceutical and fine-chemistry industries as they enable continuous operation with enhanced heat transfer (due to the high surface-to-volume ratio) and well controlled residence time. This may lead to higher reaction yields (thus lower material and energy consumption, leading to lower environmental impact) with respect to conventional batch processes, the latter often demanding for high dilution levels for safety reasons.

Since the tiny dimensions, the flow is laminar so that special techniques should be adopted to promote the mixing of reactants. Among them, the interest is towards passive micro-reactors that are able to mix efficiently reactants without any external force and just through a special micro-device design, which allows to break flow symmetries and, thus, to enhance mixing through convection.

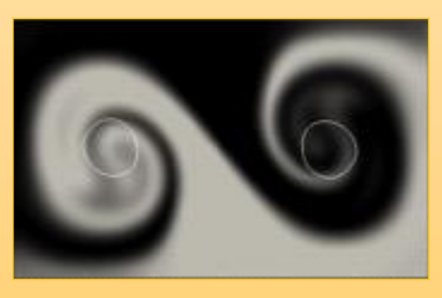
Experiments in micro-reactors are difficult to be carried out because the small dimensions; hence Computational Fluid Dynamics (CFD) appears well suited to address the problem, as it may provide a full characterization of the flow and concentration fields. Numerous CFD investigations have recently analyzed micro-mixers, showing the presence of different flow regimes depending on the Reynolds number (Orsi et al., 2013; Galletti et al., 2015; Siconolfi et al., 2018). For instance, some studies on T-shaped micro-reactors have even pointed out the presence of unsteady time-periodic motions than can improve strongly mixing (Galletti et al., 2017; Mariotti et al., 2018). However, most of the available investigations have been performed in absence of any chemical reaction, thus focusing only on the mixing process. The present work intends to study the dependence of reaction on flow regimes in a T-shaped micro-reactor.

Stratified regime: the two streams remain segregated (mixing only due to diffusion)

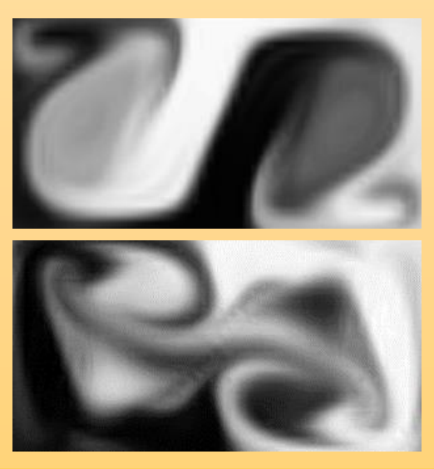
Vortex regime: double pair of steady counter-rotating vortices in the mixing channel (steady symmetric regime)



Engulfment regime: pair of steady co-rotating vortices in the mixing channel (steady symmetric regime)



Periodic regimes: unsteady asymmetric regime and unsteady symmetric regime

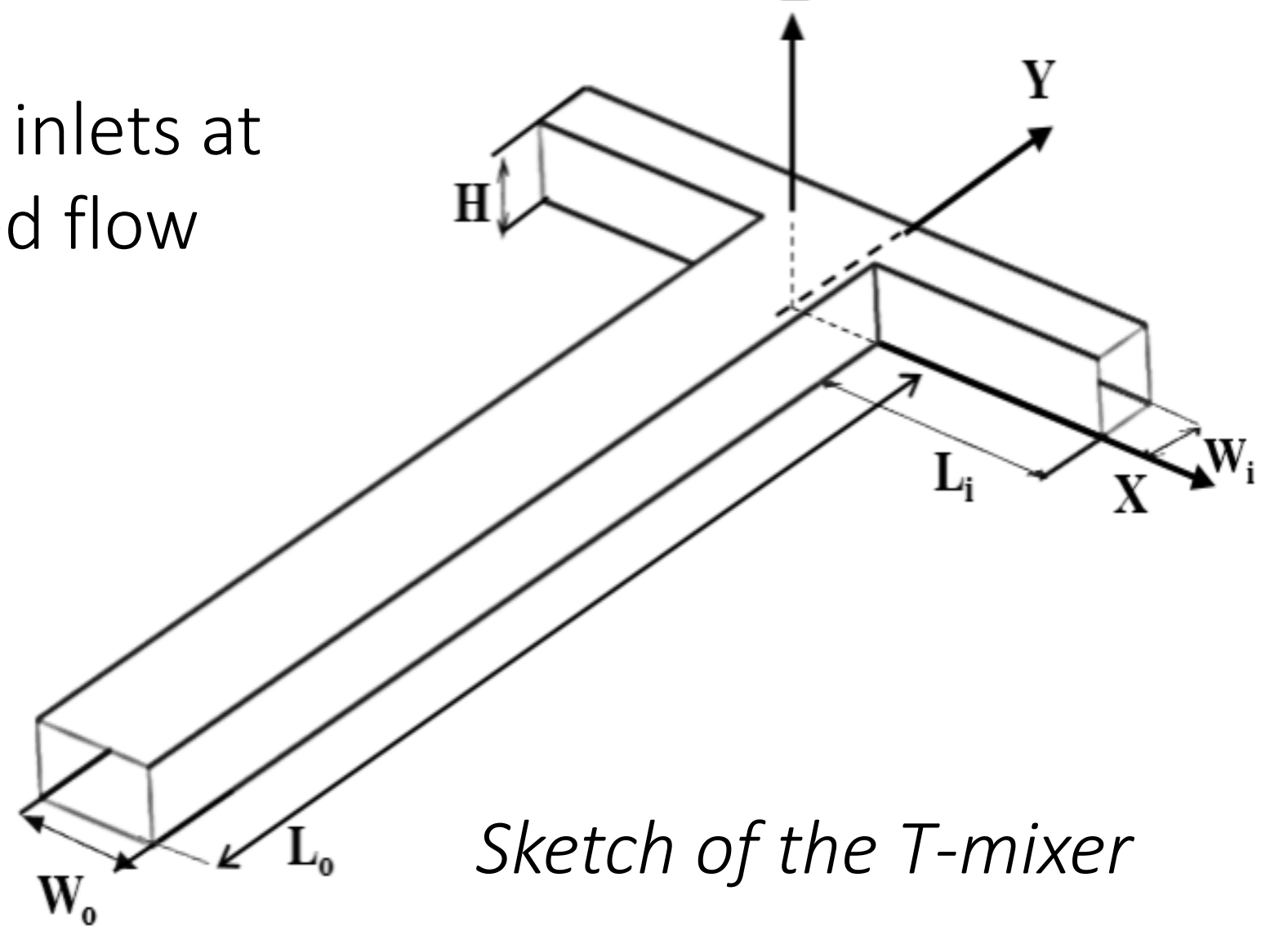


Chaotic regime

Reynolds number

Flow configuration

- Aqueous solutions are fed from the two inlets at the same flow rate with a fully developed flow
- U = average inlet velocity
- D = hydraulic diameter
- Wo = 2Wi
- Li = 30d
- Lo = 45d
- square inlet Wi= H



Sketch of the T-mixer

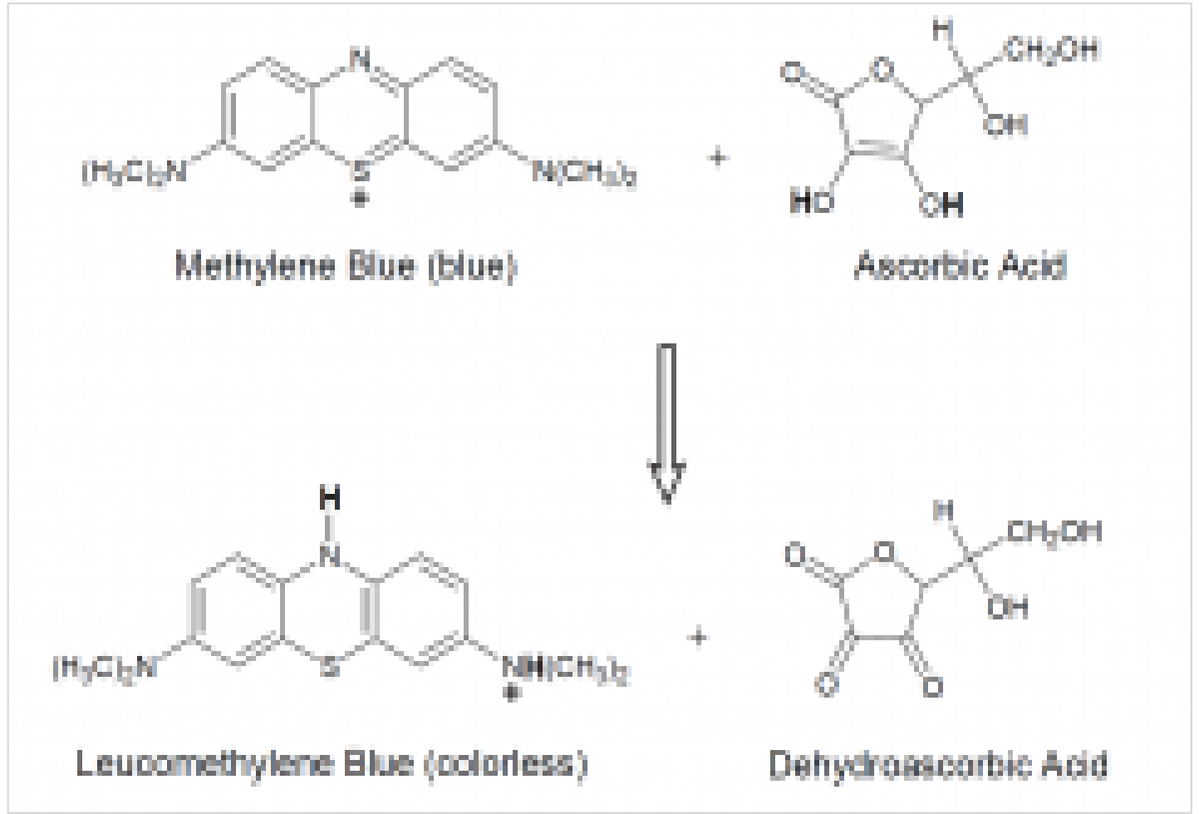
Governing equations and reaction

$$
\begin{cases}
\nabla \cdot \mathrm{u} = 0 \\
u \cdot \nabla u + \nabla P = \mathrm{Re}^{-1} \nabla^2 u \\
u \cdot \nabla c_i = \mathrm{Pe}^{-1} \nabla^2 c_i + \varepsilon_i \dfrac{d}{L} \mathrm{Da}_I c_i^n
\end{cases}
$$

$$\mathrm{Re} = \mathrm{Ud}/\nu, \quad Pe = Sc \cdot \mathrm{Re} = \rho \mathrm{Ud} / D$$

$$\mathrm{Da} = k c_i^{n-1} \tau, \quad \varepsilon_\mathrm{i} = \text{stechiometric coeff.}$$

Reaction scheme

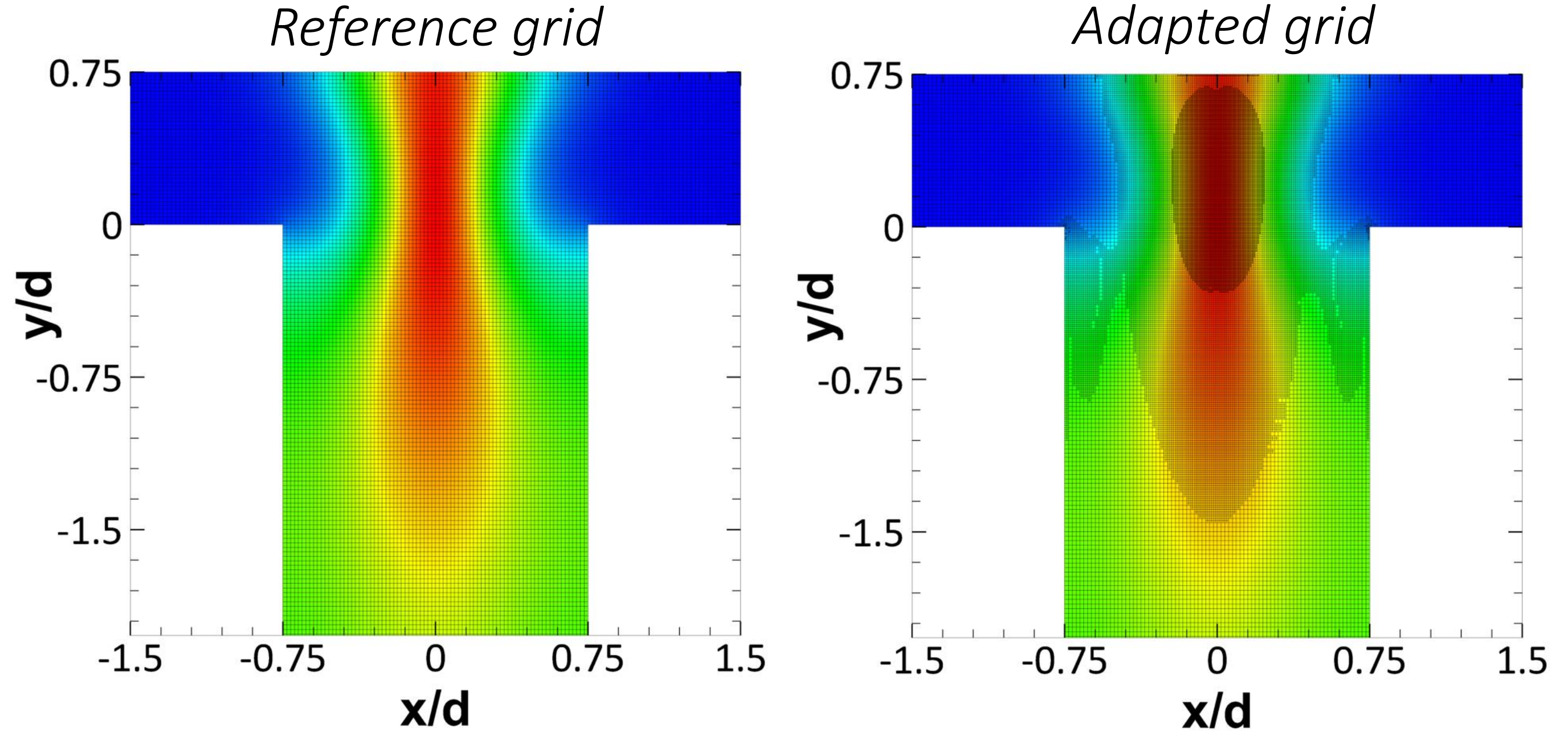


Numerical model and grid adaption

ANSYS® Fluent

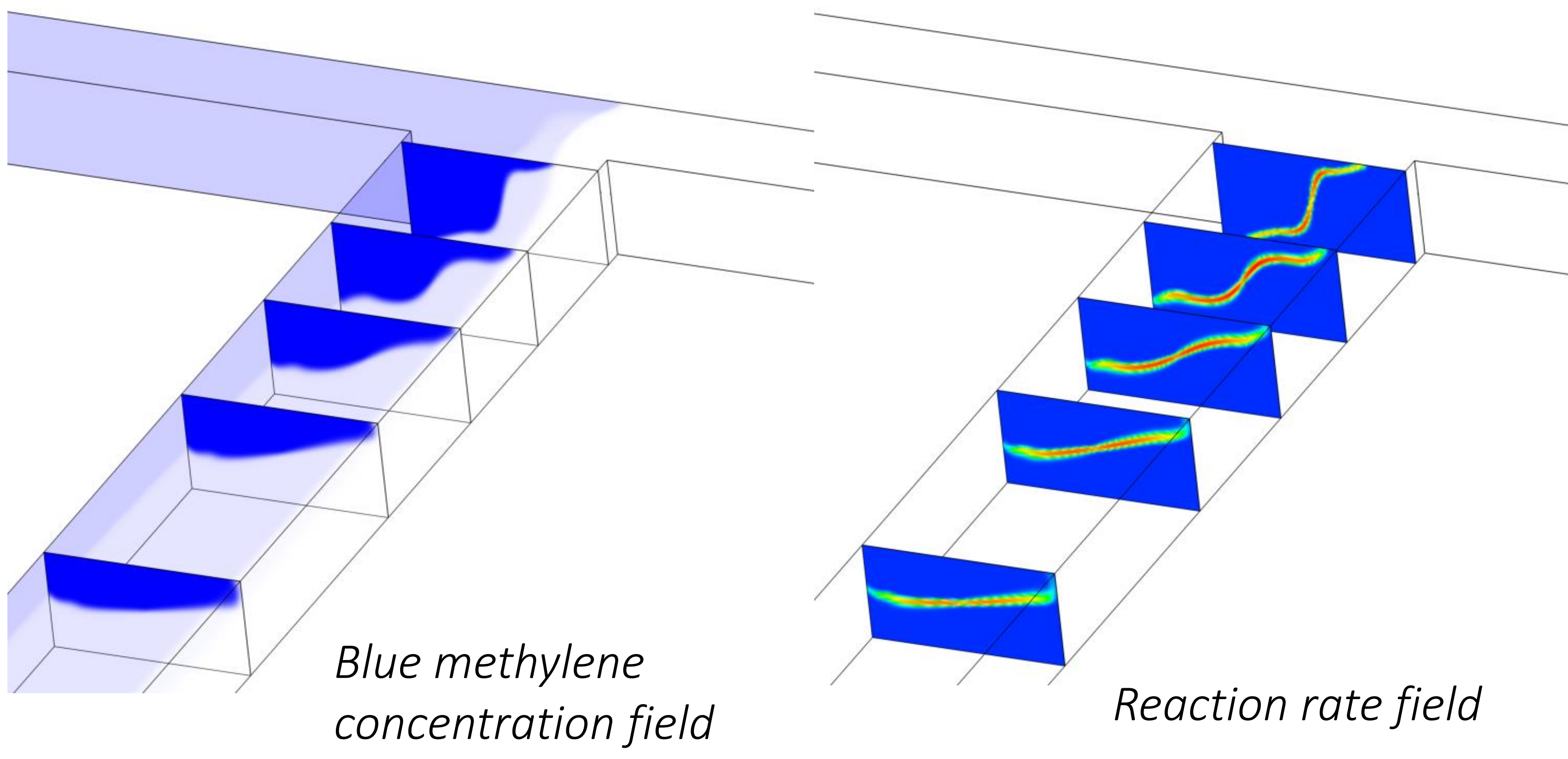
- Commercial finite volume code
- Grid of cubic elements with H/20 edge
- Steady state simulations
  - ✓ second order discretization scheme
  - ✓ SIMPLE algorithm

A preliminary analysis is devoted at understanding the required numerical resolution. The spatial resolution should be able to resolve the mixing of reactants; hence it should be considerably finer than the grid used for the velocity field. Hence, grid adaption technique on the concentration gradient is applied in an iterative manner to refine the grid in the reaction region.



Analysis of the main results

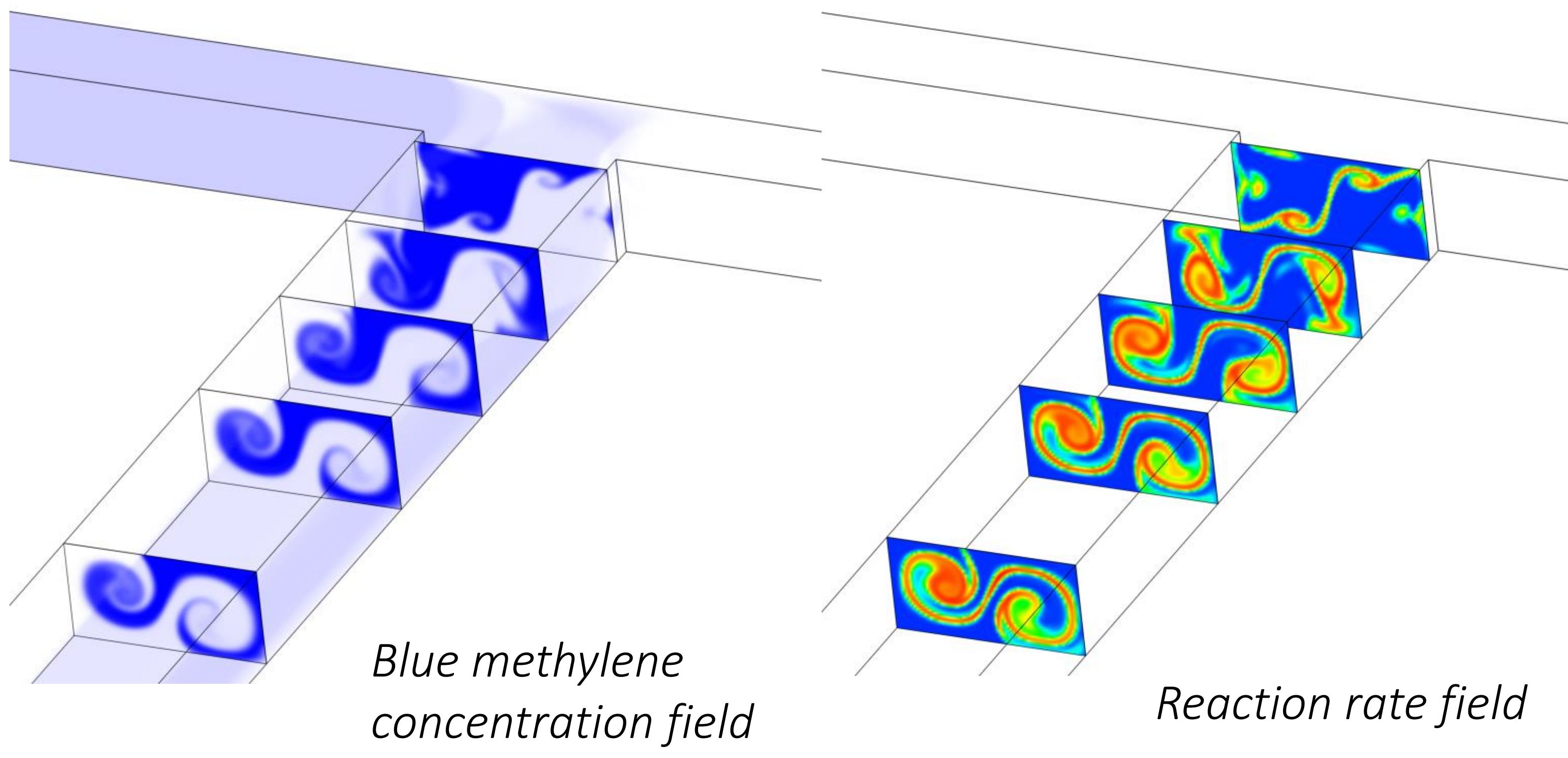
Reynolds number **Re=50**



Blue methylene concentration field

Reaction rate field

Reynolds number **Re=180**



Blue methylene concentration field

Reaction rate field

At low Reynolds number, i.e. Re=50, the reactants flow side by side along the mixing channel and have enough time to stratify because they have different densities. Here reaction occurs in a very limited region, i.e. where reactants mix by diffusion. Increasing the Reynolds number, i.e. Re=180, we observe the engulfment regime with the typical S shaped concentration patterns in the mixing channel cross sections. Such patterns are due to the presence of a pair of strong co-rotating vortical hydrodynamic structures that extend in the mixing channel. These structures push fluid elements at the confluence of the two inlet flows to reach the opposite side of the mixing channel, thus largely increasing the degree of mixing. As a consequence, the reaction region occupies a large portion of the mixer.

Conclusions

A preliminary numerical analysis of a chemical reaction in a T-shaped micromixer has been carried out to understand how flow regimes affect reaction yield. A grid adaption strategy was adopted to well resolve the mixing (and reaction) of reactants. In principle, since the Schmidt number is Sc = O(1000), to accurately predict the concentration field we would need a grid about Sc^1/2 times smaller than the grid used to resolve the flow field. Grid adaption provides a good chance to refine the reactive regions, even though it should be based on an iterative procedure. Results have shown how the engulfment regime can improve reaction rate, by promoting mixing of reactants. Ongoing flow visualization experiment will validate the numerical method in next future.

Acknowledgments

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References

- Galletti C., Brunazzi E., Mauri R. (2017) Unsteady mixing of binary liquid mixture with composition-dependent viscosity. Chem. Eng. Sci. 164, 333-343
- Galletti C., Arcolini G., Brunazzi E., Mauri R. (2015) Mixing of binary fluids with composition-dependent viscosity in a T-shaped micro-device, Chem Eng Sci, 123, 300-310.
- Orsi G., Roudgar M., Brunazzi E., Galletti C., Mauri R. (2013) Water–ethanol mixing in T-shaped microdevices, Chem. Eng. Sci., 95, 174-183
- Mariotti A., Galletti C., Mauri R., Salvetti M.V., Brunazzi E., (2018) Steady and unsteady regimes in a T-shaped micro-mixer: Synergic experimental and numerical investigation, Chem. Eng. J., 341, 414-431
- Siconolfi, L. and Camarri, S. and Salvetti, M.V. (2018) T-mixer operating with water at different temperatures: Simulation and stability analysis, Phys. Rev. Fluids, 3(3), art no. 033902