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# Experimental and numerical study of mass transfer performances in trickle bed reactors

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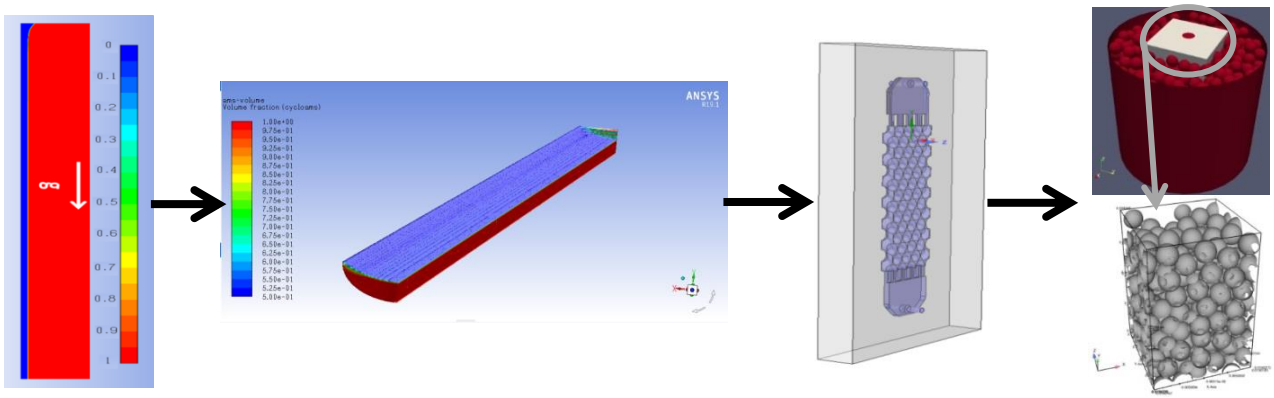
## 1. Introduction

Trickle-bed reactors (TBR) are widely used in various industries (petrochemicals, refining etc.). In this type of reactors, gas and liquid flow co-currently downward through a packed catalyst bed, creating a complex multiphase flow (flow patterns, phase distributions, catalyst wetting surface, etc.). The transfer phenomena and thus the performances of such reactors are often scale-dependant, difficult and expensive to measure in large scale vessels, and globally complex to predict. In this context, CFD is a very interesting tool to simulate and track accurately the fluid-fluid interface and predict mass transfers. Several numerical methods have been developed or implemented for this purpose, including the VOF "Volume Of Fluid" model used in this work. Despite numerous studies conducted on trickle-bed reactors, few contributions have been made towards the direct numerical simulation of gas-liquid flow in these reactors, and to the best of our knowledge, no study has been completed on the efficient coupling of hydrodynamics, transfer phenomena and reaction(s) taking place in the reactor.



(a) (b) (c) (d)

**Figure 1.** "Step by step" approach for validation from simple to complex geometries and study-cases

In this work, a numerical model has been developed to predict mass transfer in two-phase flows over solid catalyst particles or surfaces. The two-phase flow is modelled using the VOF method, which is coupled with mass transfer and heterogeneous catalytic reaction. Initially, the model has been tested considering a simple two-phase flow over an infinite 2D vertical plane (fig. 1-a), and was validated for the corresponding available analytical solutions for hydrodynamics and G-L mass transfer [1,2]. Afterwards, a single channel of a G-L-S pilot falling film micro-structured reactor has been simulated in 3D for a G-L flow over a catalytically functionalized wall (fig. 1-b). The comparison with available experimental results for liquid profile and overall external mass transfer limitation [3] demonstrate a good agreement. Subsequently, to go a step further in the complexity of real TBR with acceptable computing times, a 3D-printed 2-plates milli-reactor similar to a TBR with a perfect tetrahedral array of 50 hemispheres (d=4mm, coated with a thin catalyst layer) has been developed and studied both experimentally and numerically (fig. 1-c). After this last

validation step, the objective is to be able to perform predictive simulations on various representative catalyst bed volumes extracted from a TBR loading (fig. 1-d). As a result, the characteristics of the liquid film flow over the catalyst are obtained. Moreover, the mass fluxes are computed and used to calculate the local and global mass transfer coefficients $k_l a_l$ and $k_s a_s$ from gas to liquid and from liquid to solid respectively.

## 2. Methods

In all the study, hydrodynamic simulations are carried out using ANSYS Fluent, the multiphase flow being modelled using the "Volume Of Fluid" approach discretized with the high resolution scheme CICSAM [3]. For the final geometries, Catalyst beds are generated using "Discrete Element Method" simulation software.

Mass transfer is solved using advection-diffusion model for the transport of hydrogen from gas to liquid and from liquid to solid surface where the reaction occurs. The species transport equation is modified to take into account the thermodynamic equilibrium between gas and liquid phases at the interface, and is defined on a mixture concentration $C^*$ [4]. Using the local exchanged flux at the interface, the mass transfer coefficients are determined at local and global scales.

The numerical model is applied on the cases previously presented in figure 1, and validated for the available analytical solutions [1,2] and experimental results [3]. Except for the 3D-printed plate milli-reactor (fig. 1-c), which was studied experimentally with the very fast $\alpha$-methylstyrene catalytic hydrogenation to cumene in order to obtain data in full external mass transfer control, and simulated numerically to validate the model.

## 3. Results and discussions

The numerical model described in the previous section was first applied and validated on a falling liquid film over a 2D-plane surface. VOF model was able to simulate the liquid flow (interface velocity and film thickness), likewise, solute concentration profile and mass transfer coefficients were in agreement with the corresponding analytical solutions [1,2].

Based on the work published by Tourvieille et al. [3], a further validation of the numerical model has been done in a 3D geometry for half of a single channel of a falling film micro-structured reactor (FFMR). The VOF model was able to reproduce the experimental liquid film thickness profiles inside the domain for different liquid flow rates. Regarding the external mass transfer, the $\alpha$-methylstyrene catalytic hydrogenation reaction that has been studied experimentally, was also introduced in the numerical model. The latter has been able to reproduce realistic overall external mass transfer coefficients in acceptable agreement with the experimental results.

Afterwards, reactive experiments of fully mass transfer limited catalytic hydrogenation and the corresponding 3D simulations have been carried out on the 3D printed plate reactor. The mass transfer measurements were compared to the predicted ones and helped to validate the model. Finally, a representative catalyst volume containing up to 500 particles is extracted from a TBR loading. Simulations have been performed on the extracted volume using the developed model with the idea to compute local and global mass transfer performances as well as other global information.

## 4. Conclusion

Direct numerical simulations using VOF approach have been carried out in order to predict gas-liquid patterns and transfers in a representative volume of a trickle bed reactor. Starting from hydrodynamic field, mass transfer of the considered species is investigated at local and global scales. Simulations are compared to experimental data in terms of global transfer coefficients and the validity of the classical two-film transfer theory is discussed.

## References

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