Multi-scale approach for the analysis and assessment of a heat-exchanger monolith reactor dedicated to catalytic gas-liquid reactions

Carine Julcour*, Anne-Marie Billet, Hélène Chaumat, Audrey Devatine

Laboratoire de Génie Chimique, Université de Toulouse, CNRS, INP, UPS, Toulouse, France

**Corresponding author: carine.julcour@ensiacet.fr*

Highlights

- Design and assessment of a heat-exchanger monolith reactor for three-phase hydrogenations
- Combination of CFD & 1D models, local measurements of GL mass transfer & fluid distribution
- Analysis of coupling between hydrodynamics, mass transfer & reaction in Taylor flow at unit cell scale
- Investigation of different gas-liquid systems with or without (catalytic/homogeneous) reaction

1. Introduction

The limitations of conventional three-phase reactors for exothermic and selective reactions, such as hydrogenations, have prompted extensive research on gas-liquid milli-structured reactors, and in particular honeycomb monolith reactors [1,2]. These offer a high surface to volume ratio for mass and heat transfer intensification, more regular and controlled flow patterns, and a high catalyst efficiency due to the short diffusion path in the thin active layer. The following contribution presents the numerical and experimental developments related to a heat-exchanger reactor of monolith type (HExMR) made in aluminum alloy by selective laser melting, with channels alternatively dedicated to the catalytic reaction and to the cooling fluid.

2. Methods

The heat transfer efficiency of the reactor was evaluated using 2D simulations, which showed that the monolith scaffold was uniform in temperature for the considered design. This suggested a hierarchical approach to quantify the capabilities of this innovative reactor. Indeed, each channel having the same thermal environment, the overall reactor behavior could then be described through that of the channels treated separately, by combining their outlet flows and considering the fluid distribution in between them. Thus, the continuous experimental setup used to assess the reactor performance consisted of jacketed capillary tubes coated with a Pd/Al_2O_3 layer for the selected hydrogenation reactions (alpha-pinene and edible oil). In parallel, the fluid distribution, which dictates the flow regime in the different channels, was probed by several techniques, including an original high-frequency multi-channel resistive sensor. This helped to design an optimized gas-liquid distributor granting similar Taylor flow in most of the channels and a high bubble frequency. On the other hand, since hydrogenation reactions are often limited by the gas-liquid mass transfer, the evaluation of the associated coefficient(s) is of primary importance. This task is complicated by the strong and complex interaction between the transport phenomena and the reaction at the scale of the repeated Taylor flow pattern, the so-called unit cell (UC). This results from different mass transport mechanisms, both in series and in parallel [4], and the high dependence of their contributions on the UC characteristics and the reactions (which drive the concentration gradients in the lubrication film). Therefore, local measurements using optical techniques and CFD simulations of the periodic UC were carried out to gain insight into the coupling of the involved phenomena before developing reduced-order (1D) models. Different cases were evaluated: the evaporation of ethyl acetate for the gas-side mass transfer coefficient, the physical absorption of O_2 in water and the reactive absorption of CO_2 in two different solvents (NaOH and MDEA solutions) for the liquid-side mass transfer coefficient, the hydrogenations of alpha-pinene and sunflower oil as examples of catalytic conversion of bio-sourced feedstocks. In the most complex cases, transient hydrodynamic and mass transport equations had to be solved simultaneously. The calculations, performed in a reference frame moving with the bubble, also took into account the change in bubble size and shape, using a moving mesh approach considering the total mass flux and/or the combined effects of viscous and surface tension forces acting which warp the gas-liquid interface.

3. Results and discussion

The direct numerical simulations showed the expected features of the Taylor flow, with strong recirculation vortices in the liquid slugs and quasi-stagnant lubrication film with a velocity opposite to that of the bubble in gas-liquid upward flow, in accordance with Abiev's model [3]. The bubble adopts a typical bullet-type shape with meandering rear cap for capillary numbers exceeding 10^{-3}, and a final volume driven by the solute mass transfer with reaction or by the thermodynamic equilibrium (solvent evaporation). These calculations thus allowed to determine the effects of the bubble shape and UC size (i.e. bubble frequency) on the gas-liquid mass transfer, to quantify the separate contributions of the different interface regions (bubble caps and film), to evaluate the gas-side [5] and liquid-side mass transfer coefficients [6], and to analyze the impact on catalytic reaction of the transport mechanisms of H_2 and of the substrates [7]. Their predictions were found to be in a good agreement with local mass transfer characterizations based on bubble image processing (using ethyl acetate vaporization and reactive CO_2 absorption) [5,6] and with the hydrogenation yields measured at the reactor outlet [7]. Furthermore, the different mass transfer coefficients (gas-liquid, but also liquid-solid) could be obtained from the CFD for comparison with existing correlations and for tuning of the 1D heterogeneous plug flow model of the channel. The latter model gave then very close predictions as compared to CFD and could be used to predict the effect of fluid maldistribution on the overall reactor performance. To this end, the main flow features (bubble frequency and velocity, gas holdup) could be extracted by processing the 84-channel signals recorded simultaneously on a cold mock-up of the HExMR for different gas-liquid distributors [8]. A 3D-printed distributor with two separate chambers for the gas and the liquid and multiple T-junctions was found to outperform the classical 'showerhead' configuration in terms of uniformity of fluid distribution among the channels, range of conditions achieving the desired Taylor flow regime and bubble frequency.

4. Conclusions

3D printing techniques offer new possibilities for the design of efficient milli-structured heat-exchanger reactors for exothermic and selective three-phase reactions, including optimized gas-liquid distributors. The performance of such a reactor can be evaluated using a multi-scale hierarchical approach, from the unit cell scale to the single channel and the multi-channel configuration. CFD simulations at the UC scale are most valuable to gain insight into the intricate coupling between hydrodynamics, mass transfer and reaction. For different selected cases, these calculations were found to match local and/or global experimental data and were used to develop reduced-order models for predicting the overall reactor performance.

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Keywords

three-phase reactors; heat-exchanger monolith reactor; CFD simulations; local metering techniques