ENGINEERING INTERCONNECTED-CHANNEL MONOLITHIC REACTORS: FROM COMSOL TO 3D PRINTING

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Abstract

Three-dimensional (3D) printed 0.52 wt.% Fe/SiC monoliths with interconnected channels, fabricated by direct ink printing or Robocasting, are feasible and robust catalysts for the hydroxylation of phenol to dihydroxybenzenes using hydrogen peroxide as oxidant in aqueous media [1].

Herein, computational fluid dynamics (CFD) simulation was used for a better understanding of the catalytic performance of these interconnected-channel monolithic reactors. The CFD model integrated the fluid dynamics, mass transport and chemical reaction rate (experimentally obtained) to design a prototype that assures the overall efficiency of the 3D-Fe/SiC monoliths prior to its real fabrication by Robocasting. Three different 3D geometries were studied: square, non-uniform square (radial) and triangular channels, with interconnected channels [2].

Simulations were accomplished using the CFD Module of COMSOL Multiphysics®. Particularly, due to the small channel dimensions (d_h<1mm, L≈44 mm and δw=0.26-0.32 mm) and fluid velocities (with Reynold's number, Re<1) of the studied system, Creeping Flow interface was selected to simulate fluid flow on the monolith channel. The evolution and transport of chemical species was modelled using Transport of Diluted Species interface, considering chemical reaction takes place at the channel porous wall domain. Initial considerations showed that the complete geometry could be accurately resolved and within acceptable computation times using predefined Free Tetrahedral mesh with Normal element size.

Fluid dynamic calculations allowed obtaining radial and longitudinal velocity profiles in the channels. As can be seen in Figure 1, the liquid velocity slows down along the channel, being this decrease more significant for triangular geometry. The square and radial geometries show the maximum velocity position at the centre of the channel, while this maximum position varied along the channel in the triangular geometry. This oscillating movement is caused by the angular contact of the interconnexions (which are not faced, as in the other geometries). It provides higher turbulence, pressure drop (but still negligible), velocity loss and, most important, allows the flow of the reactants between channels, since the liquid is not stagnant in the interconnections. Further simulations considering more than one channel are needed to examine interconnection influence on triangular geometry.

Transport phenomena analysis allowed concluding that at the reaction conditions ($C_{inlet,PHENOL}$ = 0.3M, $C_{inlet,H2O2}$ = 0.3M, τ= 31,75-254 g h L-1 and T=80-90ºC), internal and external mass transfer are not limiting and chemical kinetics controls the overall rate. The best performance is obtained with the triangular geometry, which demonstrates that higher residence time inside the monolith produced by the enhanced turbulence, has a beneficial effect on the chemical performance. Therefore, triangular cells are selected as prototype to be robocasted in the 3D printer. Work is now in progress to experimentally validate the reactor modelling.

References
[1] Vega G., et al 3D-PRINTING STRUCTURED CATALYSTS FOR THE SUSTAINABLE PRODUCTION OF DIHYDROYBENZENES ANQUE-ICC3 Conference. Santander (Spain), June 17-18 2019.
[2] A. Quintanilla et al. 3D-Printed Fe-doped silicon carbide monolithic catalysts for wet peroxide oxidation processes. Applied Catalysis B: Environmental, 2018. 235: p. 246-255.

Figures used in the abstract

Figure 1 : a) 3D velocity profile across different channel geometries; b) Development of velocity profiles at the beginning of the channels (T=85ºC; QL= 0.5 mL/min)