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

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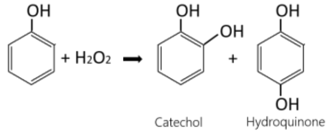
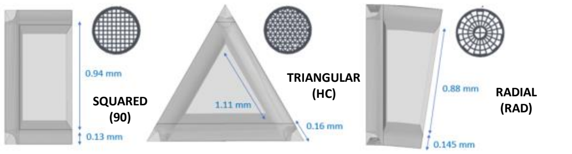
INTRODUCTION

- ✓ **3D Fe/SiC monoliths** with interconnected channels printed by direct ink writing (Robocasting).
- ✓ 3D Fe/SiC monoliths are feasible catalytic reactors for **phenol hydroxylation with hydrogen peroxide** in water .



- ✓ **CFD** to study the effect of **channel geometry** on the oxidation performance (fluid dynamics, mass transport and kinetics).



COMPUTATIONAL METHODS

COMSOL MULTIPHYSICS® → **CFD MODULE**

1. Mass transfer - *Transport of diluted species*

- **Channel**: convection + external diffusion
- **Porous wall**: effective internal diffusion + reaction

$$\nabla \cdot (-D_i \nabla c_i) + \mathbf{u} \cdot \nabla c_i = R_i \qquad \mathbf{N}_i = -D_i \nabla c_i + \mathbf{u}\, c_i$$

$$-(r_{H2O2}) = \frac{k_1\, C_{H2O2}}{1 + k_2\, C_{H2O2}} \left[\frac{mol}{g_{cat} \cdot h}\right]$$

$$-(r_{phenol}) = \frac{k_{phenol}\, C_{phenol}\, C_{H2O2}}{1 + k_2\, C_{H2O2}}$$

	k_1 (L/g_{cat}·h)	k_2 (L/mol)	k_{phenol} (L^2/mol·g_{cat}·h)
80ºC	$1.66 \cdot 10^{-2}$	11.84	$2.31 \cdot 10^{-2}$
85ºC	$1.54 \cdot 10^{-2}$	8.13	$3.05 \cdot 10^{-2}$
90ºC	$1.50 \cdot 10^{-2}$	3.81	$2.85 \cdot 10^{-2}$

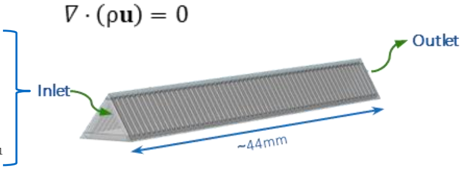
2. Fluid flow – *Creeping flow interface (Re<1)*

Laminar flow with inertial term neglected

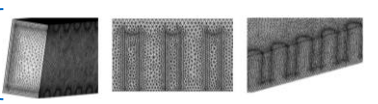
$$0 = \rho(\mathbf{u} \cdot \nabla)\mathbf{u} = \nabla \cdot \left[-p\mathbf{I} + \mu\left(\nabla\mathbf{u} + (\nabla\mathbf{u})^T\right)\right] + \mathbf{F}$$

$$\nabla \cdot (\rho\mathbf{u}) = 0$$

$C_{phenol,0}$ = 0.3M
$C_{H2O2,0}$ = 0.3M
T=80-90°C
Q=0.25-2mL/min
W=3.81 g
τ =W/Q=31.75-254 (g·h)/L[-1]



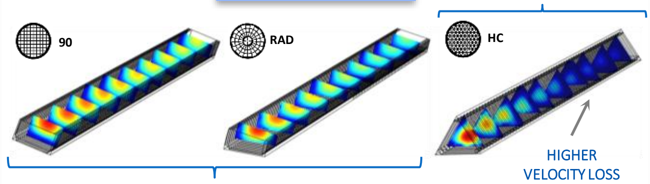
Free Tetrahedral mesh with Normal element size



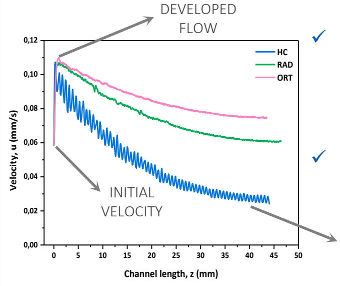
RESULTS

FLUID DYNAMICS

Maximum velocity position varies along the channel



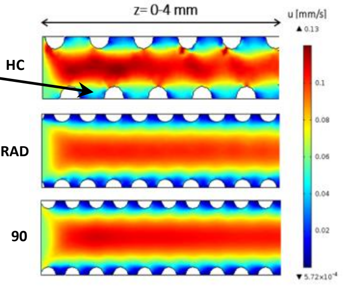
Maximum velocity along the centreline of the channel



OSCILATING MOVEMENT DUE TO TURBULENCE

- ✓ **Oscillating movement** in **HC geometry** caused by the **angular contact** of the interconnexions (which are not faced, as in the other geometries).
- ✓ Velocity loss in HC geometry **increases residence time** of the fluid in the interior of the channel

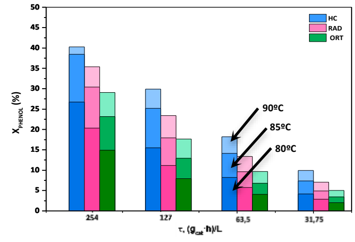
- ✓ Flow of the reactants through the interconnexions only in **HC geometry**
- ✓ HC geometry provides better contact between the catalyst and the reactants



- ✓ Pressure drop (ΔP): 90<RAD<HC; but still negligible (max. 83 mPa)

MASS TRANSPORT AND CHEMICAL REACTION

- ✓ External and internal mass transport are not limiting (data do not shown) → ***Chemical reaction controls the overall rate***



Triangular (HC) geometry provides superior performance

Exhibits the **highest phenol conversion**

CONCLUSIONS

- ✓ Triangular shaped-channel monoliths provide the best performance for phenol hydroxylation. Therefore, triangular cells are selected as prototype to be robocasted in the 3D printer.
- ✓ Work is now on 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.