Anisotropic POCS: Unlocking Mass Transfer Potential

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Significance and Relevance

The study demonstrates that modifying the POCS diamond lattice geometry by decreasing the strut angle enhances the trade-off between gas-solid mass transfer and pressure drop, achieving up to a two-fold improvement. This topological modification, developed through detailed CFD investigations, enables superior performance over conventional POCS substrates overcoming honeycomb monoliths as well even in terms of pressure drops. Validation through dedicated experiments further highlights their potential for process intensification, offering a novel degree of freedom to design intensified catalytic reactors based on advanced POCS designs.

Preferred and 2nd choice for the topic: Automotive and stationary emission control/ Fundamental advances in understanding catalysis

Preferred presentation: Oral preferred or Short Oral

Introduction and Motivations

Periodic Open Cellular Structures (POCS) are gaining considerable interest as advanced structured materials for process intensification of catalytic reactors[1]. Ceramic-based POCS are particularly attractive for environmental applications, such as exhaust after treatment, due to their ability to optimize the trade-off between mass transfer and pressure drop. In these applications, they present a promising alternative to conventional honeycomb monolith supports. POCS consist of regularly arranged unit cells with interconnected struts forming open windows, creating a permeable 3D structure suitable for fluid flow in all directions. Previous studies identified the diamond lattice as the most promising structure for maximizing the trade off between gas-solid mass transfer and pressure drop. Moreover, motivated by the potential of additive manufacturing to produce customizable lattice materials, in previous works, we have further modified the cell topology[2] by adjusting the angle between the struts and the fluid flow, resulting in a novel geometry with anisotropic permeability in both streamwise and transverse directions[2]. To validate the CFD-derived correlations[2], different POCS were 3D-printed, washcoated and catalytically activated to perform external mass transfer tests under rich H_2 oxidation[3].

Materials and Methods

In a previous work[2], a comprehensive investigation of gas-solid mass transfer and pressure drop, in terms of Merit Index (MI)[4] (Eq.1), in the modified diamond lattice was carried out through detailed CFD simulations. The analysis showed an improvement of MI by decreasing the angle between the struts' axis and the streamwise direction (α), thus obtaining a stretched unit cell along the streamwise direction and shrunken in the transverse directions. An experimental study on the external mass transfer properties was conducted on 4 different POCS characterized by the same porosity ε, same strut diameter d_s, but different angles α. The samples were realised using a lab scale SLA 3D printer and each sample was washcoated with Pt/CeO_2. The probe reaction adopted was the rich-H_2 oxidation[3] (4% H_2, 1% O_2, 95% N_2 v/v) since it is active enough to be operated under mass transfer controlled conditions at temperatures below the resin HDT and the outlet composition of the gas mixture was analyzed with a GC (Agilent HP 6890).

$$MI = \frac{-\ln(1-\chi_{O_2})}{\frac{\Delta p}{\rho u^2}} = \frac{d_s \cdot S_v \cdot Sh_{d_s}}{\phi_{d_s} \cdot Re_{d_s} \cdot Sc} \qquad \text{Where } \phi_{d_s} = \frac{\Delta p}{L}\frac{d_s}{\rho u^2} \qquad \text{(Eq.1)}$$

A 1D PFR mass transfer model has been adopted to interpret the experiments aiming at deriving dimensionless mass transfer coefficients. Due to the high dilution of the system and the isothermal condition verified experimentally, it is possible to adopt the hypothesis of constant volumetric flowrate and constant diffusivity through the length of the reactor. In this way, it is possible to correlate the experimental O_2 conversion with the average Sh number of the system ($Sh_{avg,ds}$):

$$u\frac{d\chi_{O_2}}{dz} = k_{m,O_2} \cdot S_v \cdot (1-\chi_{O_2}) \longrightarrow Sh_{avg,d_s} = \frac{-ln\,(1-\chi_{O_2}) \cdot u \cdot d_s}{L \cdot \mathcal{D}_{O_2,mix} \cdot S_v} \qquad \text{(Eq.2)}$$

Results and Discussion

To validate the experimental procedure, a catalytic bundle of four monoliths with a circular channel geometry was used. This allowed to compare the Sh number obtained experimentally with the well-known theoretical one of circular ducts. Due to the presence of entry effects (Gz>2), the comparison was made using the Tronconi-Forzatti correlation[5] which accounts them. The tests were performed at flowrates between 10 and 15 NL/min. The experimental conversions were used to evaluate the Sh_{exp} number of the system, and they were compared with the ones obtained from the Tronconi-Forzatti correlation[5] demonstrating a good agreement (Fig. 1a) and validating our experimental methodology.

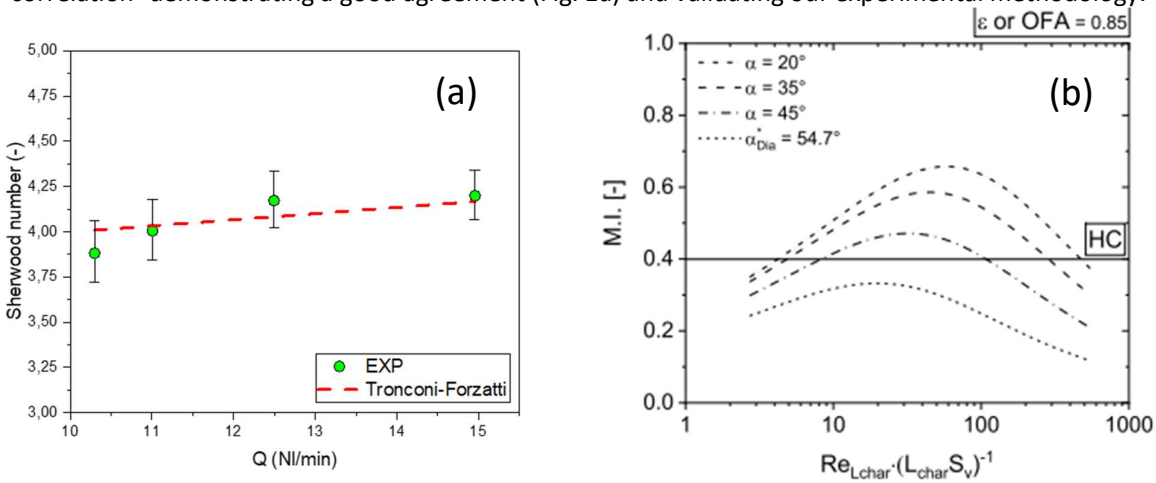

Figure 1: (a) Sh_{exp} (•) and Sh_{avg} T-F(---) VS operative flowrate for monolith bundle with circular channel geometry, (b) Merit Index of the modified diamond lattice as a function of α at constant porosity ε = 0.85

The same procedure has been applied to the evaluation of the external mass transfer properties of the modified diamond lattice. Fig. 1b compares the overall performance of the modified diamond lattice with that of the honeycomb monolith as a function of the Reynolds number. The modified diamond lattice offers up to twice mass transfer coefficients of honeycomb structures at the same pressure drop, demonstrating its strong potential as an enhanced structured substrates for process intensification.

References

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