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- **3D printed logpile catalyst structures provide flexible operating trade-offs**
- **Local densification is a unique advantage of 3D printing, providing a cross-flow regime**
- **The choice of reaction system is critical in demonstrating the potential of these structures**

The shaping of catalytic material by means of additive manufacturing (or 3D printing) has increased its technology readiness level over the past decade and this presents unprecedented freedom in the design of catalytic reactor internals. These developments potentially enable intensified chemical processes, as structures can be designed to possess a high catalyst holdup, favourable heat transfer properties and relatively low pressure drop [1]. Thus far, the research scope in published work has mostly been concerned with so-called logpile structures in either an aligned or staggered configuration [2]. The geometry of these structures is similar to that of a honeycomb monolith, but previous work from our group has shown that geometrical variations present a significant variability in transverse dispersion properties, unlike the honeycomb monolith [3]. Whilst presenting some interesting operating characteristics, it is unlikely that such structures exploit the unique advantages of the additive manufacturing technology to the fullest degree, due to their relatively simple geometry.

In this conference contribution, our recent work on 3D printed baffled logpile catalyst structures will be presented. These structures are based on the conventional logpile structures, but include baffle-like densified zones. These effectively represent porous baffles within the structure, enhancing the convective radial heat transfer by cross-flow. The porous character of the baffle suppresses stagnant volume and limits the pressure drop. The trade-off between pressure drop and heat transfer performance can be tailored by tuning the baffle geometry, and this has been extensively studied in a pseudo-2D OpenFOAM Computational Fluid Dynamics study.

The modelling study has led to concrete design guidelines for optimal structures; structures with densified zones spanning nearly the whole diameter of the structure, using the 'baffle pore size' as an optimization variable (in the order of tens to hundreds of micrometers). Six selected designs were also modelled in higher detail, evaluating the contribution of solid phase conduction on the overall heat transfer coefficient. This is an important consideration for these structures, as they consist fully of catalyst, which generally possesses a low thermal conductivity (< 1 W m^{-1} K^{-1}). It was found that for these limited solid thermal conductivities, the fluid-phase convective heat transfer is still the major contribution to the overall heat transfer coefficient. Detailed conclusions and suggestions to improve the solid-phase contribution will be presented.

Motivated by these findings, experimental demonstration was undertaken. This has been done with varying rates of success, due to several reasons:

1) It is challenging to obtain catalyst structures with adequate dimensionality and mechanical stability;
2) The coupling of the structure to the (heated) reactor wall is critical to ensure proper wall-to-bed heat transfer;
3) Enhanced performance thanks to the novel geometry needs to be carefully decoupled from other factors, such as binder presence, internal mass transfer limitations and altered catalyst porosity;
4) The improvement in performance needs to be significant enough to be measured.

Especially the last point requires careful attention, since some reaction systems may yield a sub-percentage performance increase thanks to enhanced heat transfer, which is near the measurement uncertainty of online analysis equipment. In this conference contribution, the choice for the highly endothermic Steam Methane Reforming will be motivated. In addition to this, special attention will be given to the importance of matching the catalyst geometry to the challenges of the chemical process in order to achieve process intensification; there is no one-size-fits-all optimal structure.

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