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Additive Manufactured open cell polyhedral structures as substrates for automotive catalysts

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ABSTRACT

Polyhedral open cell lattice catalyst substrates are proposed based on results of numerical simulations and recent advances in Additive Manufacturing (AM) techniques.

Detailed simulations have compared different polyhedral structures in terms of mass transfer (aiming at optimal reactivity in the mass transfer limited domain) and flow through resistance. The simulations have taken into account dimensional limits given by the possibilities of AM techniques. Comparisons with state of art honeycombs have also been used in order to identify the optimal shape.

Substrates with these optimal characteristics have been manufactured out of Al_2O_3 with Stereolithography. Subsequently, these substrates have been coated and used for measurements of C_3H_6 oxidation in a model gas reactor. Measurements have focused in determining oxidation efficiency at different gas hourly space velocities as well as light-off behaviour.

Simulation results show that the optimal open cell structures are comprised by a cubic elementary cell rotated by 45° so that one spatial diagonal of the cube is aligned to the main gas flow. Higher porosities and smaller strut diameters improve the reactivity to pressure drop trade off. However, given the current manufacturing limitations, it is not possible to produce structures with strut diameter lower than 0.5 mm. This results in high porosity but low specific surface area (i.e. $\varepsilon = 0.95$ and $S_v = 400\ m^2/m^3$). Thus, reaching a target conversion requires higher overall catalyst volume. The simulations show that for a series of geometrical parameters the open cell structures can reach identical conversion in respect to the honeycombs with only a fraction of the overall surface area and thus a fraction of the noble metals, while the overall dimensions are in the same order of magnitude and the pressure drop can reach lower levels.

Measurements in the model gas reactor confirm the mass transfer advantages of the polyhedral structures as predicted by the simulations. Measurements also show that the polyhedral lattices have very similar light-off behaviour in spite the four times lower surface area.

1. Introduction

Catalyst technologies for automotive aftertreatment systems require constant developments to comply with the latest regulations concerning real driving emissions.

The current benchmark in catalyst substrates are honeycombs (HCs), while open cell structures represent a new paradigm that is gaining attention because of its promising properties [1]. In HCs the laminar flow in the channels results in low heat and mass transfer. Instead, the network of solid struts of the open cell lattices is characterized by tortuous paths that enhance gas-wall interactions and contribute to lower thermal inertia [2]. The result is higher conversion efficiencies [3,4], lower cold start emissions and higher flow uniformity, which is a key factor for catalyst durability [5–8]. They also allow more flexibility in the geometrical configuration of the reactor [9]. The increased mass and momentum transfer properties of open cell structures, however, result also in a higher pressure drop per unit of length [10,11], decreasing engine efficiency. Thus, to have a fair comparison, the performance index I has been introduced [3], which evaluates catalyst efficiency by relating conversion to pressure drop. CFD analysis suggested that the trade off is in favour of open cell structures when the porosity is high enough [1,12].

In the past, work on open cell catalysts focused on foam structures, with some degree of uncontrollable randomness,

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Nomenclature

A	cross section of catalyst
AM	Additive Manufacturing
CPSI	Cells Per Square Inch, commercial characterization of honeycomb catalyst substrates
Cubic	Additive Manufactured (AM) catalyst substrate consisting of cubes as elementary cells aligned with the main flow
Cubic45	AM catalyst substrate consisting of cubes as elementary cells rotated by 45° so that one spatial diagonal of the cube is aligned to the main gas flow
D_{ij}	diffusivity of specie i in a gas j
d_c	wetted width of a (square) honeycomb channel
D_c	inner width of a (square) honeycomb channel, difference to d_c is the coating thickness
d_s	strut diameter
ghsv	gas hourly space velocity through the catalyst, it corresponds to the ratio between the gas volume flow rate and the catalyst volume
HC	honeycomb catalyst substrate (conventional)
K	mass transfer coefficient
Kelvin	AM catalyst substrate consisting of Kelvin cells (tetrakaidekahedral) as elementary cells
L	length of entire catalyst
L_c	cell length
L_s	strut length (in case of Cubic elementary cells $L_c = L_s$)
MM_i	molar mass of specie i
MM_{CH4}	molar mass of methane
Octet	AM catalyst substrate consisting of octets as elementary cells
PGM	Platin Group Metalls
$\dot{Q}_{in}$	gas volume flow to the catalyst
rc	chamfer dimension at the corner in the HC structure
Re	Reynolds number based on strut diameter
Sc	Schmidt number
Sh	Sherwood number
S_v	specific surface area
S_w	wetted surface area
SLA	Stereo Lithography (Additive Manufacturing technique)
TC	Thermo Couple
V	volume of entire catalyst
v_{in}	mean gas velocity upstream at the catalyst entrance
X_{CH4}	mass concentration of methane
Xi	mass concentration of specie i
Y_i	molar concentration of specie i
Y_{CH4}	molar concentration of methane
ΔP	pressure drop through the catalyst
ε	(macro-) porosity of the catalyst
η	conversion through the catalyst
φ	tube quartz reactor diameter

characterized by statistical averaged quantities [13]. Some effort has been spent on their modelling. Computing capabilities have allowed performing computational analysis of real CT foam scans [2,14–17]. Foams can also be accurately reconstructed with elaborated algorithms based on Voronoi tessellations [18–20]. However, these techniques increase the computational load. To simplify the procedure, in literature, an alternative has been to model foams as regular cell structures. Some works have been conducted by numerical simulations of open cell foams consisting of regular cells [3,21,22]. Periodic open cellular structures were also studied experimentally: [23] investigated the effects of porosity and cell orientation on pressure drop. Regular structures are easier to handle because they require only two independent parameters (for example the characteristic pore dimension and its ratio to the strut diameter) and mathematical expressions allow obtaining all the other geometrical properties [1]. The Kelvin cell has often been used as a typical elementary cell. A flow resistance comparison between Kelvin cells and foams has been performed by [24]. The flow pattern inside real and ideal foams at various Reynolds numbers has been investigated [25,26]. It has been demonstrated that regular open cells perform better than randomized foams [2]. In fact they always show a better trade off between mass transfer and pressure drop in respect to the equivalent real foams. So, regular cell lattices should not be seen only as a geometrical model, but as catalytic structures that outperform foams.

In the last years, advances in AM techniques have provided new possibilities for manufacturing [27–29]. Latest improvements have even allowed the direct manufacturing of ceramic lattices [30–32]. AM comprises several processing techniques where parts are fabricated by physical or chemical consolidation layer by layer, starting from a CAD file. A core concept, intrinsic in these AM techniques is the rapid and seamless transition between a computer model and the physical realization thereof [33].The revolution consists in the new approach of components' design by function and no longer by manufacturability [32]. Considering catalyst applications specifically, design is performed through an intensive campaign of numerical simulations, while AM stereolithography proved to be the most suitable process, allowing the manufacturing of extremely thin and complex structures with reduced flaw size [34]. The shape of conventional catalysts is determined to a large extent by the manufacturing methods, instead, the AM techniques have introduced a significant flexibility and a higher freedom in design. Big efforts have been spent in using additive fabrication tools for structured monoliths for improving mass and heat transfer characteristics with a higher geometric surface area [29]. In [35], with the support of CFD computations, robocasting is performed to create a ceramic face-centered-cubic structure with high surface to volume ratio able to convert approximately six times more methane than extruded monoliths. Robocasting is also used to fabricate Ti-alloy and stainless steel monoliths with well-controlled contact times [36]. 3D printing has also enabled to fabricate complex ceramic supports for use in packed beds [37], or reaction vessels and microreactors with integrated catalysts [38]. In all cases AM has led to improved performances through more complex structures, difficult to be produced by extrusion.

Thus, AM closes the gap between theory and experiment, by enabling accurate fabrication of a great variety of geometries, optimized through computational fluid dynamics and evaluated through experiments [29].

The present study adopts this new combined approach for the design of an open cell polyhedral lattice catalyst substrate for automotive applications. It identifies the optimal open cell polyhedral catalyst lattice in terms of shape and dimensions through detailed numerical simulations. The performance of the open cell lattice structures has been evaluated by the mass transfer coefficient, by the pressure loss per unit of length and by the dimensionless performance index I [3].

Based on simulation results, the polyhedral ceramic substrates have been manufactured with stereolithography, coated in house and then tested in a gas reactor. C_3H_6 oxidation has been measured