Advancing Catalytic Processes for the Energy Transition: Joule-Heated Structured Reactors as a Key Solution for Decarbonization and Intensification.

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Highlights

- Cellular catalyst substrates enable enhanced conductive heat transfer and direct ohmic heating
- SMR over Joule-heated washcoated SiSiC foams promises reduced process footprint and low-C H_2
- Same concept for electrification of CO₂ methane reforming and reverse water-gas shift

1. Introduction

The adoption of thermally conductive structured substrates in tubular reactors allows to shorten industrial multi-tubular reactors for non-adiabatic applications, as well as to operate heat-transfer limited catalytic processes at intensified conditions [1]. In Milano we have studied conductive cellular structures with interconnected cavities (open-cell foams, 3D printed periodic-open-cell-structures, aka POCS) applied to the intensification of e.g. the strongly exothermic Fischer-Tropsch synthesis [2] and the endothermic Steam Methane Reforming [3]. Further to exploiting the continuous matrix of cellular substrates for heat conduction, we are now exploring their electrical conductivity for direct ohmic heating of the catalyst in endothermic heat-transfer limited processes, in view of combined intensification and electrification [4] of key catalytic processes for the energy transition. Renewable electricity can be converted into heat and thermally drive chemical reactors. In this context, electrified steam methane reforming (eSMR) is a promising concept for low-carbon H_2 production, as it offers potential to utilize excess energy to produce either building blocks for chemical processes or clean fuels, while at the same time mitigating CO_2 emissions. Moreover, by bringing the heat source closer to the catalytic sites, it drastically reduces the heat transfer limitations, which are the bottleneck of the conventional SMR process, allowing for process intensification. Wismann et al. [5] have proposed direct Joule heating of a FeCrAl-alloy tube washcoated with Ni-catalyst for SMR. The performance of such a reformer configuration is controlled by external mass transfer limitations, which calls for the adoption of tubes with small diameters, leading eventually to the development of reactor solutions based on either honeycomb monoliths or micro-channel technologies. Compared to laminar flow in wall coated reactors, open cell foam catalysts could greatly reduce external mass transfer limitations. Along these lines, we have demonstrated the feasibility of direct electrification of SiSiC foam structures, washcoated with a Rh/Al₂O₃ catalyst, for the strongly endothermic SMR reaction [6-8]. The application of the same approach to the direct electrification of the CO₂ reforming of methane (eCRM) and to the reverse watergas shift (eRWGS) processes in washcoated structured reactors [9] will be also reported.

2. Methods

In our electrified tubular reactor, a SiSiC foam serves both as the catalyst support and as the Joule heating element connected to an external power supply via steel connectors (Figure 1(a)). 80% of the

foam is washcoated with an active 1% Rh/Al2O3 catalyst formulation, while the 20% uncoated entry region is used to preheat the feed stream. Outlet temperatures (Tdown) and compositions are monitored by a thermocouple and a micro GC [6].

3. Results and discussion

A specific energy demand as low as $1.24 \text{ kWh/Nm}_{H2}^3$ and a high H₂ productivity of approx. 8200 Nm³_{H2}//h/m³_{foam} were measured in eSMR tests on a washcoated SiSiC foam with a catalyst density of 88 g/L (GHSV = 150000 cm³/h/g_{cat}, S/C = 4.1, ambient pressure, Figure 1(d)), accompanied by an unprecedented high energy efficiency of 81%. Preliminary scale up simulations promise high energy intensities and H₂ productivities in compact eSMR reactors.



Figure 1. (a) Electrified methane steam reforming (eSMR) reactor layout; (b) CH4 conversion vs. input power at different space velocities, 2.2 g of 1% Rh/Al2O3 washcoated (4/5) on SiSiC foam; (c) CH4 conversion vs. Tdown at different pressures, 2.5 g of 1% Rh/Al2O3 washcoated (4/5) on SiSiC foam; (d) Methane conversion vs. Tdown during runs at intensified conditions, 5.5 g of 1% Rh/Al2O3 washcoated (4/5) on SiSiC foam.

4. Conclusions

Direct Joule heating of cellular reactor internals removes the heat and mass transfer limitations affecting conventional packed-bed reactors for the SMR process and reduces drastically its footprint by eliminating the firebox. If electricity from RES is used, low-carbon hydrogen can be produced. The same concept paves the way for the intensification of other endothermic catalytic processes which are highly relevant for the ongoing energy transition.

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Keywords

Process electrification, Joule heating, structured catalysts.