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Towards the intensification of methane steam reforming by conductive packed foams: experimental investigation and numerical analysis

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1. Introduction

The increasing demand for exploitation of small methane reservoirs as well as for flexible and distributed hydrogen/syngas production calls for clean, sustainable, intensified and cost-competitive production processes. In this view, the steam reforming of natural gas is one of the most interesting solutions, made highly reliable by the industrial experience and the existing infrastructures. Scale-down of this process from large scale reactors to small and distributed systems requires however alternative reactor configurations with respect to the traditional packed reactors, whose performances, at the small scale, are limited by radial heat transfer [1]. Structured catalysts based on highly conductive metallic substrates have been reported as valuable solutions to enhance heat transfer in fixed bed reactors with respect to conventional packed beds [2]. Metallic supports (i.e. honeycomb monoliths, open cell foams) are usually made catalytically active by depositing a thin layer of catalytic material onto their surface, using the slurry coating technique. Nevertheless, washcoated systems have drawbacks (i.e. a low catalyst inventory, limited washcoat adhesion and catalyst loading/unloading issues), which have so far discouraged the application of this technology at the industrial scale [3]. In this work, a novel fixed bed reactor configuration for the steam reforming of CH_4 is proposed: it consists in filling the void volume of conductive open-cell foams with small catalytic pellets, aiming at enhancing the radial heat transfer thanks to the high thermal conductivity of the solid matrix.

2. Methods

In this work, catalytic pellets in form of egg-shell particles were used as catalytic medium; alumina spheres (D_p = 0.6 mm by Sasol) were activated by the incipient wetness impregnation technique using a rhodium nitrate water solution, aiming at a rhodium content of 0.3 % wt. with respect to the carrier and at an impregnation depth of 35 µm.

In the case of the packed foam layout, three different cellular matrices were tested, namely a 12 PPI FeCrAlY foam (FeCrAl10 in the following; $\varepsilon = 0.92$ and D_{cell} = 5.2 mm, by Porvair) and two copper foams from ERG Aerospace Corporation, with 10 PPI (Cu10 in the following; $\varepsilon = 0.91$, D_{cell} = 4.6 mm) and 40 PPI (Cu40 in the following; $\varepsilon = 0.88$, D_{cell} = 2 mm), respectively. For all samples, diameter and height of the foam were set at 29 mm and 25 mm, respectively. The packed foam configuration was obtained by filling the foam void volumes with catalytic particles. Additionally, tests in a packed bed system were performed and reported as benchmark of a conventional layout. In this case, the catalytic bed was diluted with SiC particles, aiming at the same reactor volume as for the packed foam.

Catalytic tests were performed in a tubular reactor (I.D. = 29.5 mm) externally heated by a tubular furnace set at a temperature in the 600-800 °C range. A steam/CH_4 mixture with S/C ratio of 3.5 was fed to the reactor. Temperature profiles were recorded longitudinally across the catalytic bed in three different radial positions, namely at the centerline, at 8 mm from the center and at the external wall. Water was condensed and separated downstream from the reactor. N_2 was mixed with the dry product stream as an internal standard and the product mixture was analyzed by a µGC.

3. Results and discussion

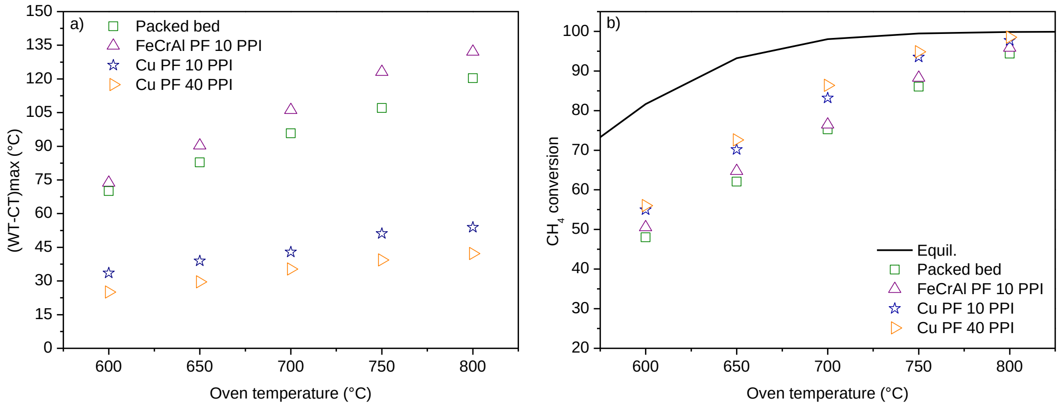

Figure 1 Maximum radial difference between wall (WT) and central (CT) temperature in catalytic bed (a); methane conversion as a function of furnace temperature (b). GHVS = 10000 h^{-1}.

At GHSV = 10000 h^{-1}, the maximum radial temperature difference between the wall and the center of the reactor was significantly reduced thanks to the presence of the highly conductive copper foam matrix (Figure 1-a). Moreover, at any furnace temperature, CH_4 conversions of packed foam configurations were greater than that of the packed bed (Figure 1-b). This improvement can be ascribed to the enhanced heat transfer properties of the copper-based systems, thanks to the conductive heat transfer mechanism enabled by the presence of the metallic structure.

To achieve a rational assessment of the collected data, an equivalent electric circuit was developed to estimate the heat transfer performances of the system: the latter was obtained by incorporating independently estimated effective heat transfer parameters for packed bed reactors and open-cell foams. Catalytic pellets and flowing gas were assumed as a pseudo homogeneous phase exchanging heat with a solid phase, which accounts for the additional contribution of the conductive open-cell foam to intra-reactor heat transfer and inter-phase heat transfer terms. The heat transfer model (accounting for all the convective and conductive contributions associated with the internals) was validated against the experimental results and used to analyze and interpret the observed trends, thus quantifying the beneficial role of a conductive structure. According to the collected evidence, further optimization of the reactor layout can be obtained, aiming either at the maximization of the syngas productivity or at an optimal tradeoff between fixed and operational costs.

4. Conclusions

The packed foam configuration was proved to be a valuable alternative to the traditional packed bed systems to enhance the performances of the steam reforming process. A heat transfer model based on the equivalent electrical circuit approach was found to be in good accordance with the experimental results.

References

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