

MICRO-SCALE CATALYTIC REACTOR FOR HYDROGEN PRODUCTION VIA PROCESS INTENSIFICATION

Salvatore Vaccaro*, Luca Malangone and Paolo Ciambelli
Department of Chemical and Food Engineering, University of Salerno, Fisciano, SA,
Italy

Summary

Aim of this work was the study of a micro-scale catalytic reactor for the production of hydrogen via methane steam reforming. In this auto-thermal system the heat to sustain the endothermic reaction is provided by an exothermic reaction occurring in the same device and using the same main reactant. The main advantages are compactness and space and energy saving. The system was studied both through experiments aiming to ascertain the reactor real performance and by modelling aiming to develop a valid tool for the reactor design and for the interpretation of the experimental findings. Promising results were obtained in terms of methane conversion and endothermic and exothermic reactions coupling. The model predicts the reactants conversion quite accurately although differences were found between the calculated and the experimental temperature profiles.

Keywords

Process intensification, Micro-reactor, Hydrogen

1. Introduction

In recent years, micro-scale reactors were developed for process intensification and used to couple exothermic and endothermic reactions for their noticeable temperature control and improved mass transfer [1,2]. In fact, especially for reacting systems where heat and/or mass transfer may limit the overall kinetics, the use of such devices may strongly improve the overall efficiency.

The advantages of such systems are compactness and, then, space and energy saving and often they can make the resulting plant inherently safe allowing the use of much smaller quantities of dangerous reactants.

2. Experimental system

The micro-reactor used in this work consists of three parallel plates in which channels are bored. The central channel (50 mm long, 10 mm wide, 3.0 mm thick) is shaped inside the inner plate while the outer channels (50 mm long, 10 mm wide, 1.5 mm thick each) are located inside the outer plates. The channels are separated by foils onto which different catalysts were deposited on the opposite faces. This configuration allow different flow arrangements such as reforming stream in the central channel and combustion streams in the outer channels or vice versa and for each flow arrangement co-current or counter-current stream flow patterns can be realized.

Graphite foils were used as gaskets between the catalytic foils and the stainless steel plates. 0.5 mm thick aluchrom slabs (70% Fe, 25%Cr, 5% Al), were used as structured metallic supports. They were thermally pre-treated before applying an Al₂O₃ based washcoat: Ni and Pt were used as active species to catalytically promote reforming and combustion reaction, respectively. Reforming catalyst reduction was performed, before testing, in situ with 20 l/h at STP of a 5 mol % H₂ in N₂ stream.

Temperature monitoring inside the channels was accomplished using three K thermocouples for each channel located at 1, 3 (center) and 5 cm from the channel inlet.

2. Experimental results

Tests were performed considering the reactants and the molar flow rates listed in table 1.

Reactant	Molar flow rate (STP)
CH ₄ (combustion side)	0.268 (mol/h)
CH ₄ (reforming side)	0.536 (mol/h)
Air	2.141 (mol/h)
Steam	1.624 (mol/h)

Table 1: Feeding conditions

* To whom all correspondence should be addressed

The O_2/CH_4 ratio was set to 1.68 according to previous tests showing that stoichiometric or excess air negatively affected the methane combustion. The ratio between the CH_4 flow rates fed to the combustion and to the reforming channel was set to about 2 in order to have proper matching of the two reacting systems considering the different kinetics, the different enthalpies of reactions and the heat loss through the external walls.

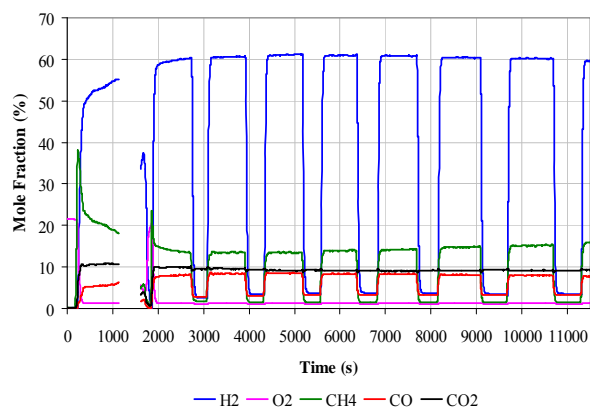


Figure 1: Transient evolution of the molar fraction of the species inside the system for the flow rates of table 1.

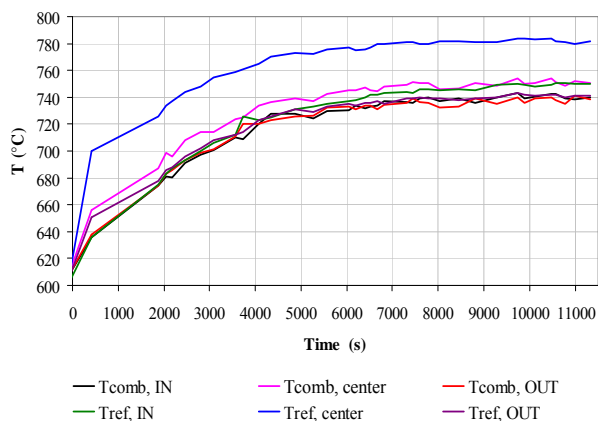


Figure 2: Transient evolution of temperature profiles at the different positions inside the system for the flow rates of table 1 (IN, center and OUT stand for 1, 3 and 5 cm from channel inlet)

Typical results are shown in figure 1 as mole fraction on dry basis of species at the reforming channel outlet as a function of time. The abrupt variations in the molar fractions in figure 1 are due to data acquisition system commutation between the combustion and reforming channels. Calculation carried out with such data showed that when the system reached steady state, the methane conversions were about 88.5 % and 53.3% on the combustion and the reforming side, respectively, and the hydrogen yield 3.46 (mole hydrogen produced/mole methane converted). Figure 2, instead, report the temperature time profiles inside the reactor. Both figures show a relatively long transient of the system. The

thermal transient, however, lasts much longer than the reacting one.

3. Mathematical modelling

Transport phenomena occurring in the micro-scale catalytic reactor were also modelled and a commercial software (Comsol Multiphysics 3.5) was used to carry out simulations [3]. Due to calculation complexity it's not uncommon in literature to consider simplification when formulating models for such systems. In fact, with respect to other works performed with similar reactor geometry and reacting systems, this one does not use simplifying assumptions such as catalyst layers modelled by one-dimensional approach, fully developed laminar flow or transverse heat and mass transfer taken into account through lumped heat and mass transfer coefficients [4]. Multicomponent mass transport was modelled using a Fick-like law instead of more complex approach such as Maxwell Stefan equation or Dusty Gas model in order to limit computational costs.

Because of the entering configuration and the limited length/width ratio of the reactor a simple bi-dimensional modelling appeared inappropriate and, therefore, a 3D geometry was considered for modelling.

For the reforming kinetic the Hou and Hughes model [5] was employed, while for the combustion process a power-law expression was used [6]. The model is able to predict the reactants conversion quite accurately but deviations exist when comparing the calculated with the experimental temperature profiles. Therefore, for a complete validation of the model it's necessary to correctly take into account the thermal dissipation through all the external boundaries.

4. References

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