

## Design and test of a miniature hydrogen production integrated reactor

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### Highlights

- Thermally coupled reforming-combustion reactions in hydrogen production is investigated
- Micro-reactor efficiency increases with catalyst loading and reforming fuel flow rate
- Micro-combustor conductivity is able to maintain the MSR reaction practically isotherm

### 1. Introduction

The complete reformer-fuel cell unit is proposed as an alternative to conventional portable sources of electricity due its ability to provide an uninterrupted supply of electricity as long as supply of methanol and water can be provided. Methanol steam reforming (MSR) is an endothermic reaction, and heat source is necessary to maintain reaction temperature. The heat supply to the reformer may be achieved in various ways. In most of the cases, the reformer is heated externally by wrapping an electric heater around the reformer wall or internally by placing a heat source inside the reformer. However, several studies recently proposed integrating a micro-reformer with catalytic micro-combustor as heat supply [1]. The thermal coupling becomes an important part of developing compact units for process intensification using micro-channel reactors. Furthermore, the micro-reactor design and operating conditions have a great importance for optimal thermal management. It is therefore of great interest to determine the optimal conditions to provide design guidelines for hydrogen production using coupled combustion-reforming catalytic micro-reactors. A detailed study of the experimental issues involved in the design and operation of methanol steam micro-reformer is presented in this work. Micro-milling technology was utilized to fabricate metallic micro-channel block coupling the exothermic and endothermic process. Calculations involving the heat loss from the reformer device, the endothermic heat effect of the reforming reaction and the exothermic heat effect of the methanol combustion reaction were carried out to estimate the total power requirement for continuous operation of the reformer.

### 2. Methods

Microchannel blocks were manufactured using Al-alloyed ferritic stainless steel (Fecralloy®). Micro-channel plates were fabricated by micro-milling 1 mm-thick Fecralloy® plates. In each 20 × 20 mm plate, 10 square channels of 700 μm were milled with separations of 300 μm. Final blocks were composed of 20 micro-machined plates alternately stacked at 90° resulting in 100 micro-channels for each direction (Figure 1). Catalysts were deposited on micro-channel block by washcoating with 2.5 wt. % Pd/ZnO (reforming catalyst) and 1 wt. % Pd/Al<sub>2</sub>O<sub>3</sub> (combustion catalyst) [2]. The catalytic tests were carried out at atmospheric pressure in a Computerized Microactivity Reference Catalytic Reactor from PID Eng&Tech. 6 thermocouples were used to monitor temperature (Figure 1). The shells used to connect the micro-channels block also included 4 heating cartridges for the catalyst reduction step. This auxiliary electrical heating system is also used to compensate the heat losses of the micro-channel reactor. For the methanol steam reforming, the catalyst was reduced at 773 K for 2 h, under a flow of 50 NmL min<sup>-1</sup> of 10 % H<sub>2</sub> in N<sub>2</sub>.

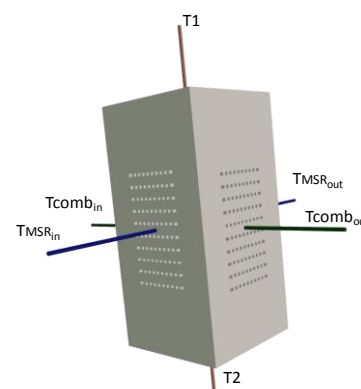


Figure 1. Micro-channel block

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### 3. Results and discussion

First of all, the influence of the reforming conditions (temperature, catalyst loading and reforming flow) on the system efficiency was studied heating the block by electric cartridges. The methanol reforming conversion was 98 % at 623 K and stable selectivity was obtained, producing 60  $\text{LH}_2/\text{h}\cdot\text{g}_{\text{cat}}$  with a very low CO content ( $<1\%$ ) and consuming 47.4 W of electric power. However, this results shows that the power provided to keep the reaction temperature at 623 K is much higher than that for the reforming ( $Q_{\text{React}} = 8.76\text{ W}$ ). The responsible for this difference are the heat losses: 79.3% for heat loss and 20.7% for the methanol reforming. This difference is related to the laboratory scale setup, since the dissipation surface to the volume ratio is very large. At the studied conditions, increasing temperature from 598 to 648 K, the heat loss increased from 79 to 83%. However, the system efficiency increases with increasing catalyst loading and reforming fuel flow rate. Increasing catalyst load to 425 mg (15  $\text{mmol}_{\text{MeOH}}/\text{min}\cdot\text{g}_{\text{Cat}}$ ), the heat lost decreased to around 60% of applied total electrical heat (Figure 2). The CFD simulations performed by Uriz et al. [3] show that heat losses are critical for micro-reactor operation mainly at low space velocity. The energy input decreases with the space velocity requiring higher energy to compensate the heat losses.

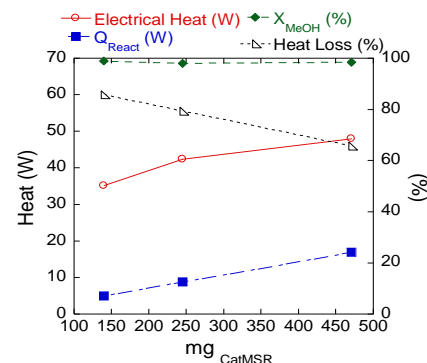


Figure 2. The effect of catalyst loading

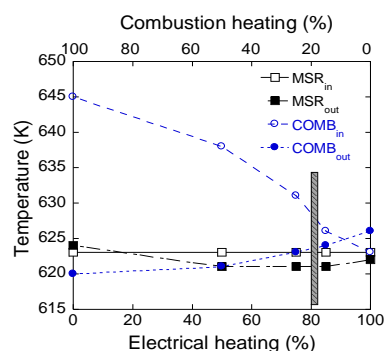


Figure 3. Block temperature profile

The measured  $\Delta T$  in the block were very small ( $T \leq 5\text{ }^\circ\text{C}$ ) heating the system only electrically (Figure 3). Based on the results, micro-reformer coupled to the combustor was investigated. In subsequent experiments the power of the cartridges was fixed at decreasing values (from 47 to 0 W) and the temperature (623 K) was controlled through the methanol combustion with the cascade flow temperature controller. As the electrical power was reduced, the combustion power increased (increased methanol/air flow) producing an increase in combustion entry temperature (Figure 3). This increase was due to the extremely fast combustion, which occurs in the first 10% of the micro-channel. Nevertheless, the excellent heat conduction characteristics of the micro-channels block homogenized the temperatures.

### 4. Conclusions

A micro-channel reformer/combustor was designed for methanol steam reforming. The obtained results show that the combustor performance could be controlled and allowed maintaining the micro-reformer temperature at 623K. Methanol combustion is rapid and takes place near the micro-combustor inlet. However, the micro-combustor conductivity was able to maintain the methanol steam reforming reaction practically isotherm.

### References

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### Keywords

Integrated micro-reactor, MSR, hydrogen production, Catalytic combustion.