Micromonoliths coated with Pt/Al_2O_3 catalyst for the simultaneous combustion of H_2, CO and CH_4: Effect of the catalytic layer thickness and thermal conductivity

Oscar. H. Laguna[1*], Oihane Sanz[2], Miguel A. Centeno[1], Mario Montes[2], José A. Odriozola[1]

1 Instituto de Ciencia de Materiales de Sevilla (Centro Mixto CSIC – Universidad de Sevilla) Avda. Américo Vespucio 49, CP:41092 – Seville (Spain)

2 Applied Chemistry Department, Chemistry Faculty of the University of the Basque Country (UPV/EHU), Donostia-San Sebastián (Spain)

*olaguna@us.es

Highlights

- The micromonoliths achieve the total conversion of the mixture: $H_2/CO/CH_4$.
- Heterogeneity of the axial thermal is controlled by catalytic layer thickness.
- The thermal conductivity decreases with the loading of catalyst.

1. Introduction

Within the fuel processors technology, the enhancement of the thermal efficiency is one of the challenges that have to be overcome in order to make this a competitive technology [1]. Among the different approaches for designing more efficient fuel processors, the use of the off-gas of the fuel cells for producing heat through a combustion reaction results interesting not only from an energetic point of view, but also from the environmental one because the use of an usually discarded stream that is released to the atmosphere, would reduce the environmental impact of this technology. The produced heat by the combustion of the off-gas of the fuel cell can be transferred by means of a heat-exchanger to the endothermic steps of the overall process.

In the present work we study the combustion of a ternary mixture of fuels (H_2, CO, and CH_4) usually present in the fuel cell off-gas. For this a Pt/Al_2O_3 catalyst is used. This catalyst has been structured in metallic micromonoliths (stainless steel FeCrAlloy®) with 1350 cpsi (cells per square inch), modifying the amount of loaded catalyst. The catalytic performance of the monoliths during the combustion has been tested and compared with that of the powder catalyst. Additionally, axial thermic profiles under specific conditions of the combustion reaction have been measured for the more active monoliths, in order to establish the relationships between the amount of catalyst and the creation of hot spots in the structured systems.

2. Methods

Firstly, for the synthesis of the catalysts, alumina PURALOX® (SCFa140/L3 - Sasol) was calcined at 900 ºC during 6 h. Then, a 2 wt.% of Pt was deposited over the calcined alumina by the incipient wetness impregnation method. The obtained solid was dried at 120 ºC during 12 h and finally calcined at 500 ºC during 6 h. As for the structured systems, cylindrical micromonoliths were manufactured (diameter =1.7 cm; height = 3.0 cm, and geometric exposed area 540 cm^2, using FeCrAlloy® [2]. Different loadings of catalyst were deposited over micromonoliths using the washcoating method (50, 100, 250 and 550 mg of catalyst that correspond to 0.1, 0.2, 0.5 and 1.0 mg/cm^2 of coating densities respectively). Monoliths have been labelled as M0.1, M0.2, M0.5 and M1.0 respectively. The catalytic activity measurements were carried out in a Microactivity Reference PID Eng&Tech® system over the dried slurry (the solid obtained after the drying and calcination of the slurry) and the micromonoliths. Concerning the mixture feed, 200 and 500 mL/min total flows (composed by 2 Vol.% H_2, 2 Vol.% CO, 2 Vol.% CH_4, balanced with air) were used. The products of the reaction were followed by on-line gas chromatography (Varian® CP-4900). For the measurement of the temperature profiles, a K-type thermocouple was placed at different axial positions along the center of the monoliths for measuring the temperature differences (ΔT) between the axis and the external reactor wall due to the combustion. An additional K-type thermocouple was employed for the control of the

temperature, placed in the center of the oven in contact with the external reactor wall. The axial thermal conductivities of the monoliths have been calculated according to Tronconi et al. [3].

3. Results and discussion

The 100% combustion of every component in the ternary mixture occurred at different temperatures in the following order (H_2: <100 ºC; CO: < 200 ºC; CH_4: > 600 ºC). Therefore, although the three fuels were fed simultaneously, the combustion of CH_4 was selected for comparing the studied systems because this fuel required the highest temperatures. In this sense, the calcined slurried catalyst presented a slight decreasing of the catalytic performance respect of the parent catalyst. After H_2-chemisorption studies, this behaviour has been associated to the loss of dispersion of the Pt in the calcined slurried catalyst.

The catalytic activity measurements demonstrated that the catalytic performance of the monoliths was superior as the loading of catalysts increases as can be observed in Figure 1 where the T90 is presented as a function of the catalytic load (layer thickness).

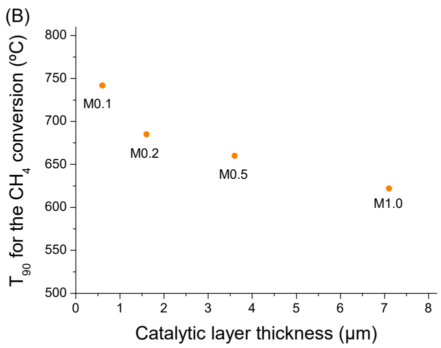

Figure 1. T90 for the CH4 conversion of the prepared micromonoliths

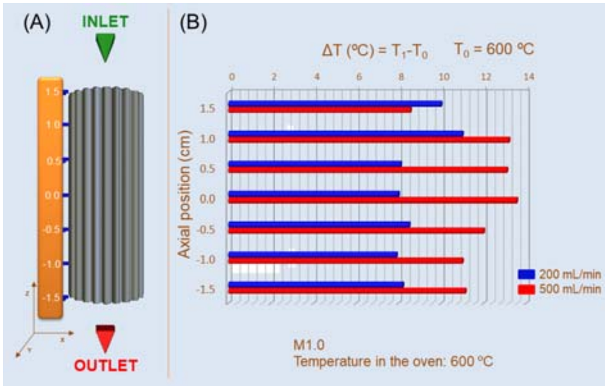

Figure 2. Axial thermal profiles micromonolith M1.0: A) Axial position for the T measurements; B) ΔT at the different axial positions into the micromonolith

However, the studies of the axial thermal profiles during the reaction demonstrated that the thermal control was more irregular with the increment of the total flow of the feed-stream (Figure 2). In fact, the axial thermal profile varies for different space velocities, resulting in the generation of hot spots migration from the entrance of the monolith (for 200 mL/min) towards the middle of the monolith (for 500 mL/min), as can be observed for the M1.0 micromonolith in the Figure 2. Furthermore, the modification of the thermal profile was more evident as the catalytic layer thickness increased. Concerning the axial thermal conductivity, this decreases as the layer thickness increases although mass transport limitation should not be discarded, because the amount of catalyst increases in the monoliths. It also important to note that the total amount of heat released also increases when the total flow rate increases.

4. Conclusions

The obtained micromonoliths achieve the total conversion of the mixture: H_2/CO/CH_4, however the increase of the catalytic layer thickness results in the decrease of the axial thermal conductivity. Furthermore, the heterogenization of the thermal profile was produced with the increase of the space velocity.

References

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Keywords
Metallic micromonoliths; combustion; thermal conductivity.