DEVELOPMENT OF ADVANCED REACTION SYSTEMS FOR HYDROGEN GENERATION

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Natural gas as a global energy source has been gaining widespread use in recent years as a result of the high oil prices, the need for energy diversification and supply security, the growing global awareness of environmental issues, and the development of new gas-related technologies. Methane steam reforming (SR) keeps being the main method for large scale syngas and hydrogen production. Besides, the miniaturization of this process is still a technological challenge [1]. Moreover, for remote or off-shore applications, process intensification to obtain hydrogen from fosil fuels and biomass, as a way to increase the eficiency, is also a big challenge [2].

In this work hydrogen production by methane SR in microreactors is studied. For this purpose, Ni-based (over MgO and calcium modified Al_2O_3) and noble metal based (Pd and Pt over Al_2O_3) catalysts were tested.

The platelets with a channel width of 500 μm and a channel depth of 250 μm were first cleaned with isopropanol for 10 min in an ultrasonic bath and after drying calcined at 1273 K for two hours.

Catalysts were impregnated filling manually the microchannels with a suspension.

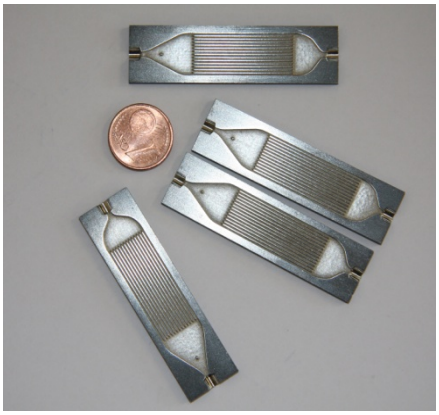

Fig. 1

All the catalysts were tested in the methane SR reaction. The feed mixture included CH_4, H_2O and inert (N_2) gases whose flows were adjusted by electronic controllers. The CH_4:H_2O:N_2 = 1:1:1,88 (molar ratio) mixture was fed into the reactor at atmospheric pressure. Steam to carbon ratios of 1.5 and 2 were also tested to obtain higher conversions. Analyses were carried out by GC equipped with FID and TCD detectors. The temperature was varied between 973 and 1073 K. Finally, the activity is compared to a fixed bed system with the same space velocity.

For the Cat2 (Ni over MgO) and Cat3 (Pd) systems some fissures were detected on the surface by SEM analysis. For the Cat1 (Ni over Ca-Al_2O_3) and Cat4 (Pt), the results are presented in Figures 2 and 3, respectively. In Fig. 2 homogeneous and uniform distribution of the catalytic grains is shown.

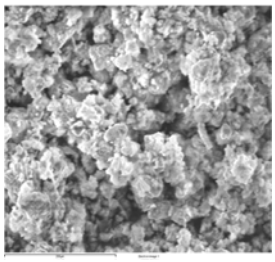

Fig. 2

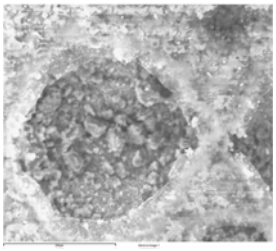

Fig. 3

In figure 4, activity results for the Cat2 from 973 to 1073 K are presented (WHSV = 40-430 h^{-1}). As it would be expected methane conversion and hydrogen production increase with temperature. During the time on stream for these tests, no catalyst deactivation was detected.

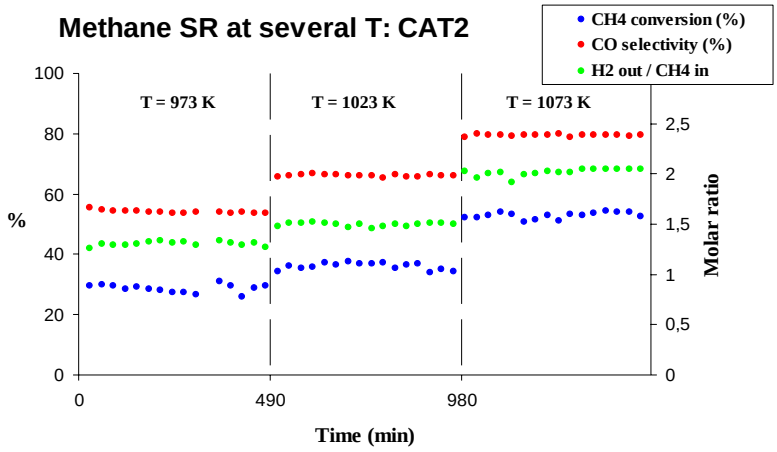

Fig. 4

Natural gas SR activity tests were also performed with similar activity results. Afterwards, differences between structure before and after reaction were analyzed by SEM.

References

[1] D.A. Castelo Branco, A. S. Szklo, R. Schaeffer. CO_2e emissions abatement costs of reducing natural gas flaring in Brazil by investing in offshore GTL plants producing premium diesel. *Energy* **2010** 35(1) 158-167.

[2] M. O'Connell, G. Kolb, R. Zapf, Y. Men, V. Hessel. Bimetallic catalysts for the catalytic combustion of methane using microreactor technology. *Catal. Today* **2009** 144(3-4) 306-311.

Acknowledgements

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