Application of Micro-structured Plate Heat-Exchanger Reactor Technology For the Development of Portable Fuel Processors

G. Kolb, Y.Men, H.Pennemann, J.Schürer, D.Tiemann, M.Wichert, R. Zapf, V. Hessel and H. Löwe

Institut für Mikrotechnik Mainz GmbH (IMM)

The introduction of heat-exchange capabilities into chemical reactors is a well-known measure taken to improve the thermal management of numerous processes – hot spot formation may be suppressed and heat losses are minimised, which helps to improve the overall process efficiency. Apart from the safe management of hazardous reactions, the application of micro-structured reactor technology enhances the heat and mass transfer, which reduces the reactor size demand [1]. Especially small scale portable power generation systems require compact hydrogen supply in case fuel cell technology is applied [2]. Fuel processing is a viable option to meet the limited space demands of portable auxiliary power units (APUs) owing to the high energy density of liquid or even gaseous fuels. A fuel processor requires energy supply for both fuel evaporation, steam generation and for the reforming process itself – the latter in case an endothermic process such as steam reforming is applied.

At IMM, all components required for building a fuel processor are addressed. Commercial and home-made catalyst coatings [3] are under investigation for the steam reforming and partial oxidation of propane [4], for methanol [5] and ethanol steam reforming as well as for the catalytic combustion of these fuels, which serves as energy source for steam reforming and evaporation processes. The catalytic CO clean-up has been addressed by catalyst screening for water-gas shift [6], preferential oxidation [7] and methanation reactions based upon noble metal catalyst technology. In selected cases 1,000 hours stability testing is performed on selected catalyst coatings at weight hourly space velocities, which are sufficiently high to meet the demands of future fuel processing reactors.

Fig.1 a to d show 1,000 hour stability tests performed on self developed catalyst coatings. For propane steam reforming, full conversion of the fuel is required, which is demonstrated in Fig.1a. The same applies for the total oxidation of propane, which may be applied for feeding a propane steam reformer with energy (Fig.1b). The catalytic CO-clean-up is addressed by water-gas shift (see Fig. 1c) and the preferential oxidation of carbon monoxide (Fig.1d). For the former case, equilibrium conversion of CO is achieved, whereas a reduction of the content of CO in the product to levels below 50 ppm are required to ensure stable operation of conventional CO-tolerant low-temperature PEM fuel cells. The thermal and mechanical stability of these wash-coatings throughout temperature cycles, exposure to moisture and mechanical shocks has been demonstrated [8]. However, alternative routes of generating catalyst coatings such as sol-gel techniques are under investigation as well.

Integrated plate heat-exchanger reactors for the combined propane steam reforming / propane combustion, methanol steam reforming / anode off-gas combustion (see Fig.2) and for the steam generation fed by hydrocarbon combustion are operated at temperatures exceeding 750°C (in case of hydrocarbon reforming) and up to a size range of 5 kW_{el} of the fuel cells supplied. Fig.3 shows a 2 kW combined diesel steam reformer/catalytic afterburner for fuel cell anode off-gas developed at IMM. Cooling capabilities have been introduced into reactors designed for the partial oxidation of propane, the water-gas shift [9], [10] and the preferential oxidation of carbon monoxide. Fig.4 shows a 5 kW high temperature water-gas shift reactor with cross-flow cooling capabilities, whereas a counter-flow 2 kW water-gas shift reactor is shown in Fig.5. A 100W reactor/heat-exchanger for the preferential oxidation of CO is shown in Fig.6.

Balance-of-plant components such as cross-flow and counter-flow (see Fig.7) heat-exchangers working up to 900°C, evaporators, condensers and pre-heaters for fuel cells and fuel processors do complete this list.

Short system start-up time demand is one of the requirements most difficult to meet in the case of portable systems. Owing to the lack of sufficient buffer battery power, combustion processes need to be applied for start-up.

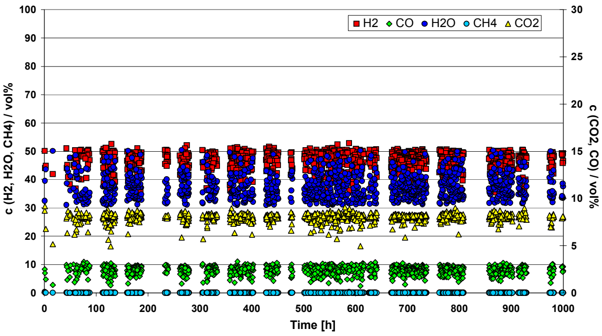

Fig.1a: 1,000 hours stability test for self-developed catalyst for propane steam reforming

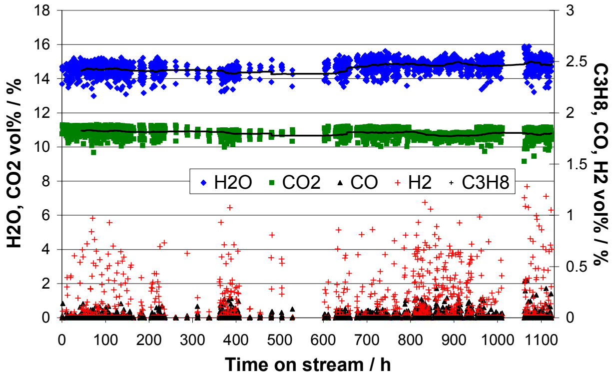

Fig.1b: 1,000 hours stability test for self-developed catalyst for total oxidation of propane (750°C)

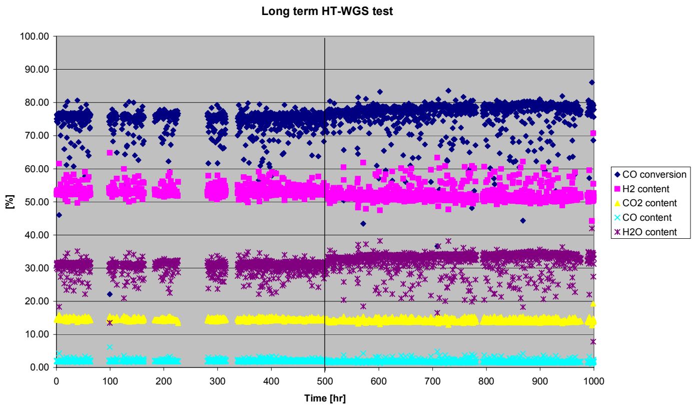

Fig.1c: 1,000 hours stability test for self-developed catalyst for water-gas shift

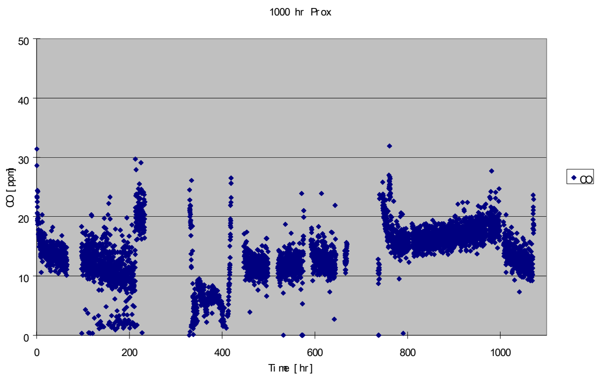

Fig.1d: 1,000 hours stability test for self-developed catalyst for preferential oxidation of CO

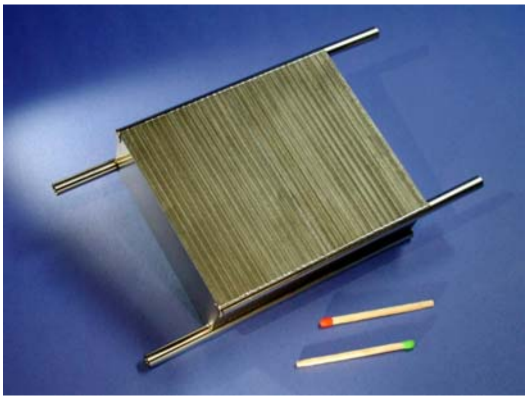

Fig.2: 100 W combined methanol steam reformer/catalytic afterburner

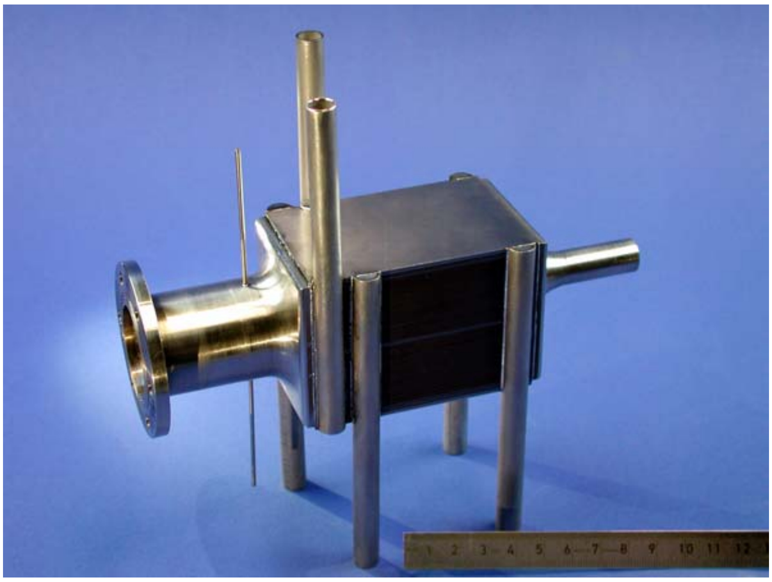

Fig.3: 2 kW combined diesel steam reformer/catalytic afterburner

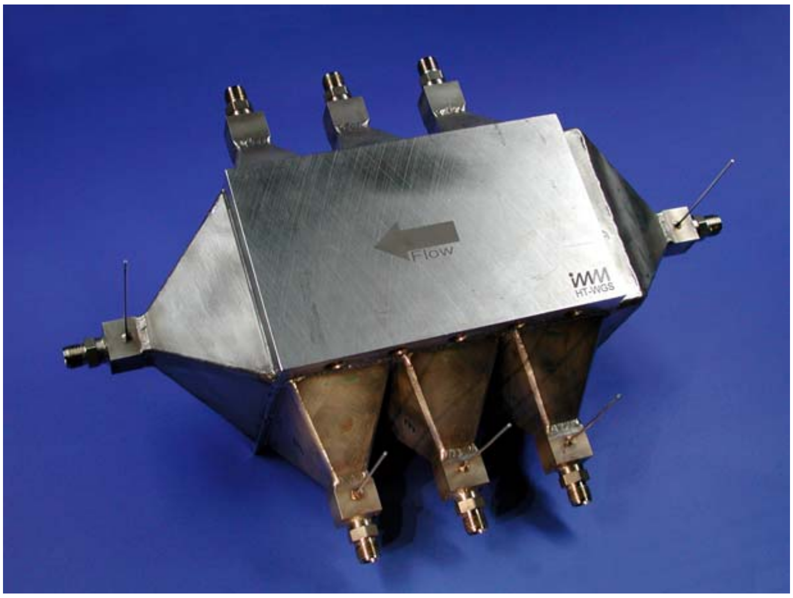

Fig.4: 5 kW high temperature water-gas shift reactor with cross-flow cooling capabilities

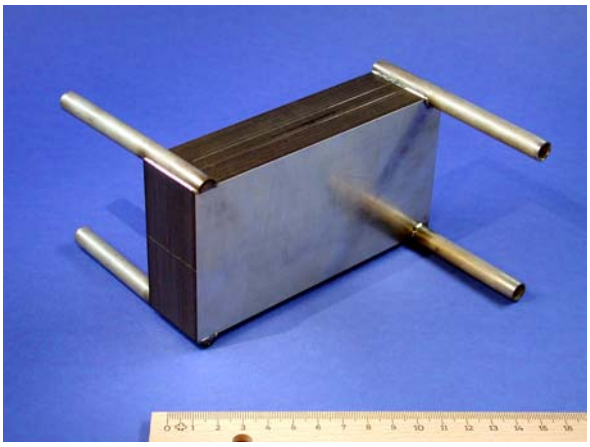

Fig.5: 2 kW Counter-flow 2 kW water-gas shift reactor/heat-exchanger

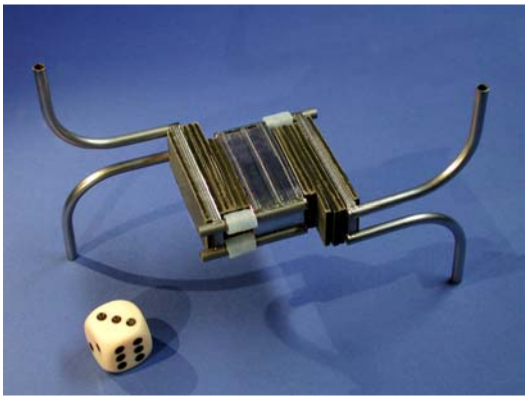

Fig. 6 Reactor/heat-exchanger for the preferential oxidation of carbon monoxide

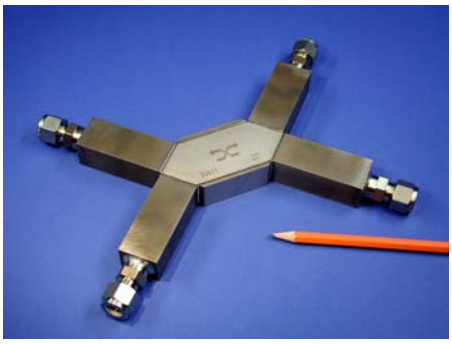

Fig.7: 2 kW Counter-flow heat-exchanger for operating temperatures up to 900°C

Because micro-structured plate heat-exchangers bear the potential of multi-task design, unique features may be introduced into the fuel processor components, which reduce the start-up time demand.

System assembly is the final challenging task. At IMM, a complete fuel processor capable of feeding a 5 kW_{el} fuel cell and working with autothermal reforming of iso-octane as a model substance for gasoline has been assembled and successfully put into operation. Other, smaller fuel processing systems in the power range below 1 kW_{el} are under development.

An overview of the work focussing on integrated components such as heat-exchanger reactors for combined fuel combustion / fuel reforming and integrated complete fuel processor solutions working with and without catalytic CO-clean-up will be presented.

References

[1] Kolb G., Hessel V, „Review: Micro-structured reactors for gas phase reactions" Chem. Eng. J. 98, (2004) 1.

[2] Hessel V., Löwe H., Müller, A., Kolb G., 2005, 'Chemical Micro Process Engineering- Processing, Applications and Plants', Wiley, Weinheim, ISBN-13 978-3-527-30998-6, p.281 ff.

[3] Zapf R., Becker-Willinger C., Berresheim K., Bolz H., Gnaser H., Hessel V., Kolb G., Löb P., Pannwitt A.-K., Ziogas A., Trans I Chem E Part A, 81 (2003) 721.

[4] Kolb, G.; Zapf, R.; Hessel, V.; Löwe, H., 2004, "Propane Steam Reforming in Micro-channels – Results from Catalyst Screening and Optimisation", Appl. Cat. A: General, 277, 155-166.

[5] Cominos V., Hardt S., Hessel V., Kolb G., Löwe H., Wichert M., Zapf R., Chem. Eng. Comm. 192 (2005) 685.

[6] Kolb G., Pennemann H., Zapf R., Catal. Today 110 (2005) 121

[7] Cominos V., Hessel V., Hofmann C., Kolb G., Zapf R., Ziogas A., Delsman E.R., Schouten J.C., Catal. Today 110 (2005) 140
[8] Zapf R. and Kolb G., Chem Eng. Techn., submitted for publ.
[9] TeGrotenhuis, W.E. King, D.L., Brooks, K.P., Holladay B.J., Wegeng, R.S. 6th International Conference on Microreaction Technology, 2002, AIChE, NY, 2002, p. 18
[10] Baier T. and Kolb G., Chem Eng. Sci., submitted for publ.