

## 7<sup>th</sup> International Conference on Microreaction Technology (IMRET 7)

한국에너지기술연구원

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2003년 9월 스위스의 Lausanne에서 “The seventh International Conference on Microreaction Technology (IMRET 7)”가 개최되었다. 여기에 세계 20개국에서 참가한 약 330명이 120개 이상의 구두와 포스터 발표를 통해 마이크로 반응기 관련 연구 개발에 대하여 논의하였다. 이 중 일부 연구 결과들이 “Chemical Engineering Journal, Vol. 101(1-3)”에 special issue로 출간되었다. 여기에 실린 연구 결과들 중 micro fuel processor와 관련된 8개의 자료들의 초록을 모아보았다.

### **Micro-structured string-reactor for autothermal production of hydrogen**

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#### **Abstract**

Novel micro-structured string-reactor designed as catalytically active wires placed in parallel into a tube was developed. The small diameter of the channels ( $\sim 100 \mu\text{m}$ ) leads to a short radial diffusion time, a narrow residence time distribution (RTD), and a low pressure drop. This reactor was applied for the oxidative steam-reforming of methanol (OSRM) to produce hydrogen in autothermal mode for fuel cells. The heat generated during methanol oxidation at the reactor entrance is axially transferred to the reactor zone of the endothermic steam-reforming. The brass metal wires (Cu/Zn=4/1) were used as precursors for the preparation of string-catalysts. The brass wires have high thermal conductivity (120 W/(m K)) and the chemical composition is similar to the active phase of the Cu/ZnO/Al<sub>2</sub>O<sub>3</sub> traditional catalyst during the steam-reforming of methanol. Brass-based string catalysts are obtained by metal/aluminium alloy formation on the outer surface of wires followed by an acid treatment leaching out aluminium. This treatment leads to an increase of the specific surface area (SSA) due to the formation of porous outer layer on the wire surface. The porous outer layer has the morphology of Raney metals. The catalysts were first tested for the steam-reforming of methanol and showed high activity together with selectivities close to 100% towards hydrogen and carbon dioxide. Then, the optimized catalyst was tested during the methanol partial oxidation (POX) and during OSRM. Oxygen was observed to be totally converted via total oxidation and therefore, higher methanol conversion in the OSRM together with CO<sub>2</sub> selectivity of 99% and H<sub>2</sub> selectivity of 60% were obtained.

## Development of microchannel methanol steam reformer

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### Abstract

Microchannels were patterned on the metal sheets and fabricated to make a reformer and vaporizer unit. Three types of patterned sheets were prepared to construct a base structure. The aspect ratio of microchannels can be regulated by changing the number of microchannel sheets. Catalyst was deposited inside the microchannel of a reformer unit. Electric heaters provide heat for the endothermic reaction and the vaporization of liquid fuel. A vaporizer and a reformer unit were connected serially to make a proto-type microreactor. The dimensions of the reformer and vaporizer unit excluding fittings were about 70 mm×40 mm×30 mm, respectively. The steam reforming of methanol was conducted with the microreactor. The performance of the microreactor was investigated at various operating conditions. The developed fuel processor generates enough hydrogen for power output of 15 We.

## Catalytic conversion of propane to hydrogen in microstructured reactors

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### Abstract

A microstructured reactor has been fabricated from the high temperature alloy Fecralloy (72.6% Fe, 22% Cr, 4.8% Al). The reactor was oxidized at high temperature to form a porous layer of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> on the surface of the channels and subsequently impregnated with Rh. The reactor was tested for partial oxidation (POX) and oxidative steam reforming (OSR) of propane at 1 bar and in the temperature range 500–1000 °C. The results were compared to those obtained from equivalent experiments using reactors made of pure Rh or from oxidized Fecralloy reactors without catalyst or impregnated with Ni. OSR gives higher yields of hydrogen than POX for the Rh/Al<sub>2</sub>O<sub>3</sub>/Fecralloy system. Formation of H<sub>2</sub>, CO, CO<sub>2</sub> and H<sub>2</sub>O by gas phase reactions does not seem to play a dominating role below 1000 °C. Small amounts of methane and ethene were detected at the highest temperatures. The Rh/Al<sub>2</sub>O<sub>3</sub>/Fecralloy reactor gives higher selectivities to hydrogen as compared to the reactor made of Rh, indicating that having the catalyst dispersed on a porous layer in the microchannels is beneficial. Changing the residence time by changing

the feed rate influenced conversions and selectivities differently for Rh/Al<sub>2</sub>O<sub>3</sub>/Fecralloy and for Al<sub>2</sub>O<sub>3</sub>/Fecralloy without any Rh. Deactivating including coke formation was not observed for the Rh/Al<sub>2</sub>O<sub>3</sub>/Fecralloy system under the conditions applied. Results from Ni/Al<sub>2</sub>O<sub>3</sub>/Fecralloy are more ambiguous, and it appears that sintering effects, oxidation of Ni or possibly loss of material occur in this system. Microstructured Fecralloy foils were prepared in the same way as used for the reactors in order to characterize the resulting oxide layer and the impregnated catalyst. XRD, SEM/EDX and XPS measurements confirm the existence of an  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> layer. XPS furthermore confirms the presence of metallic Rh, while Ni appears to be present as both metal and oxide.

### **CO selective oxidation in a microchannel reactor for PEM fuel cell**

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#### **Abstract**

It is indispensable to remove CO at the level of less than 50 ppm in H<sub>2</sub>-rich feed gas for the proton exchange membrane (PEM) fuel cells. In this paper, catalyst with high activity and selectivity, and a microchannel reactor for CO preferential oxidation (PROX) have been developed. The results indicated that potassium on supported Rh metal catalysts had a promoting effect in the CO selective catalytic oxidation under H<sub>2</sub>-rich stream, and microchannel reactor has an excellent ability to use in on-board hydrogen generation system. CO conversion keeps at high levels even at a very high GHSV as 500 000 h<sup>-1</sup>, so, miniaturization of hydrogen generation system can be achieved by using the microchannel reactor.

### **Wall coating of a CuO/ZnO/Al<sub>2</sub>O<sub>3</sub> methanol steam reforming catalyst for micro-channel reformers**

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#### **Abstract**

Capillaries of quartz and fused silica were coated with a commercial BASF CuO/ZnO/Al<sub>2</sub>O<sub>3</sub> catalyst. The objective was to create micro channels with reactive coatings for steam reforming of methanol to produce H<sub>2</sub> for proton exchange membrane (PEM) fuel cells. A slurry of catalyst with a boehmite binder was filled in the capillary and then displaced by a stream of inert gas. This gas displacement method allowed us to generate coatings ranging from 1 to 25  $\mu$ m in thickness by varying the slurry viscosity. The 25  $\mu$ m coating thickness was deposited on a range of tube diameters, for example 4.1 mm, 2 mm, and 530  $\mu$ m diameter. The coating was adherent

and did not peel off during catalytic reaction tests which lasted up to 10 days. The activity of the coated catalyst was superior to that of the same catalyst in a packed bed.

### **Experiments and modelling of an integrated preferential oxidation–heat exchanger microdevice**

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#### **Abstract**

Microreactor technology creates opportunities for the development of miniature chemical devices, in which several unit operations are integrated. We describe in this paper the design, experimental, and modelling work concerning a microdevice for the preferential oxidation of carbon monoxide in hydrogen-rich reformat gas. The microdevice consists of two heat exchangers and one reactor, all integrated in a single stack of microstructured plates. Experiments show that the initial carbon monoxide conversion is high. However, the catalyst deactivates rapidly. It takes over one hour to reach the required reactor temperature during startup, which is too long for application in a portable fuel processor. The measured temperature gradients in the heat exchangers are twice as small as predicted by a one-dimensional heat exchange model of the microchannels. A two-dimensional model shows that large differences in temperature exist between channels close to the inlet and channels further from the inlet, causing the one-dimensional model to fail. This paper shows that for an accurate description of heat transfer in a micro heat exchanger, the complete (two-dimensional) plate geometry needs to be considered.

### **Hydrogen production for fuel cell application in an autothermal micro-channel reactor**

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#### **Abstract**

Results concerning the coupling of the steam reforming (SR) and total oxidation (TOX) of methanol in a two-passage reactor are presented. A commercially available copper based catalyst is used for the steam reforming. For the total oxidation, a highly active cobalt oxide catalyst was developed. Both catalysts are used in form of thin layers immobilized on the wall of the micro-channels. Reactor design and operating conditions are based on kinetic models developed under isothermal conditions in micro-structured reactors. For the oxidation reaction, complete conversion of methanol (>99%) at temperatures higher than 250 °C is observed. For the steam reforming, the hydrogen and CO<sub>2</sub> selectivity is higher than 96% for methanol

conversion up to 90%. Besides the steady state, the dynamic behavior of the coupled system is studied. It is shown that the transient behavior is mainly determined by the thermal inertia of the system.

## **MEMS-based components of a miniature fuel cell/fuel reformer system**

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### **Abstract**

The components of a novel miniature fuel cell/fuel reformer system fueled by liquid gases such as butane and propane were prototyped by MEMS technology and tested. In this system, fuel, air and water are supplied to the fuel reformer by utilizing the vapor pressure of the liquid gas for the reduction of power consumption by peripherals and the simplification of the system. The system is composed of a reforming reactor, a catalytic combustor, a polymer electrolyte fuel cell (PEFC), an ejector to supply air to the combustor and other peripherals. The reforming reactor demonstrated the steam reforming of methanol at an equivalent power of 200 mW and a total efficiency of 6%. The combustor had a stable combustion area above 5 W, and the complete combustion of butane was confirmed by gas chromatography. The ejector showed a potential to supply air required for the complete combustion of butane (31 times larger volume than butane). The PEFC worked, but only at low power density of about 0.1 mW/cm<sup>2</sup> due to poor adhesion between a polymer electrolyte membrane (PEM) and catalytic electrodes.