36a Integrated Partial Oxidation and Purification Microsystems for Autothermal Production of Hydrogen from Methanol

Kishori T. Deshpande, Benjamin A. Wilhite, M. A. Schmidt, and Klavs F. Jensen The high efficiency and energy density of miniaturized fuel cells provide an attractive alternative to batteries in the portable power generation market for consumer and military electronic devices [1-3]. The best fuel cell efficiency is typically achieved with hydrogen, but safety and reliability issues remain with current storage options. Consequently, there is continued interest in reforming of liquid fuels to hydrogen. The process typically involves high temperature reforming of fuel to hydrogen combined with a low temperature PEM fuel cell, which implies significant thermal loss. Alternatively, fuel reforming can be combined with solid oxide fuel cells capable of operating at high temperatures.

Methanol is an attractive fuel since it is easily stored liquid and partial oxidation produces 66% hydrogen and the balance being carbon dioxide.

 $CH_3OH + \frac{1}{2}$. $O_2 -> CO_2 + 2H_2 \Delta H^o = -192.3 \text{ kJ/mol}$(1)

However, carbon monoxide is also formed in side reactions, which necessitates cleaning up the generated hydrogen to prevent catalyst poisoning in the fuel cell. We have previously demonstrated hydrogen purification using Pd-Ag membranes [4] and the integration of these membranes with LaNiCoO₃ methanol reforming catalysts for production of hydrogen. At 475°C, the maximum extracted hydrogen yield was 47%, but electrical heating had to be used to heat the palladium hydrogen separation membrane and maintain the reaction temperature. Microreactors possess high surface to volume ratio leading to high heat transfer rates to the environment. These loses have to be compensated by thermal management, specifically isolation and energy input from the fuel. The palladium membrane separation process implies an equilibrium amount of hydrogen remains in the exhaust gas. This hydrogen, along with unconverted methanol and carbon monoxide, can be burned by catalytic combustion to provide the necessary energy to make the overall hydrogen generation from methanol autothermal.

We present an integrated fuel processing microsystem consisting of hydrogen generation and catalytic combustion units. The hydrogen generation unit combines a 200 nm thick palladium-silver film with a methanol reforming catalyst, e.g., LaNiCoO₃. The catalytic combustion units employ a platinum catalyst. Both units are formed in a silicon wafer by bulk silicon micromachining techniques. The energy generated in the combustion unit is efficiently transferred to the hydrogen production unit by the good thermal conduction of silicon. The whole system is thermally insulated. We characterize the performance of this integrated, autothermal hydrogen generation system in terms of energy efficiency and hydrogen production. Startup issues are also addressed.

References:

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