Novel Microreactor Design for Balancing Heat and Mass Transfer

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Introduction

The water-gas shift (WGS) reaction is a member of the class of reversible, exothermic reactors where reaction kinetics are faster at high temperature but conversion is favored at lower temperature. As a consequence, removing heat as the reaction proceeds using microreactors can substantially reduce the amount of catalyst needed and the size of the reactor. Integrating heat exchange into microchannel reactors not only makes it possible to remove heat of reaction but also progressively reduce the reaction temperature along the reaction pathway. The inlet of the reactor can be maintained at higher temperature to initially favor kinetics, while the outlet end is kept at lower temperature to favor conversion. Appropriate management of the temperature profile can substantially reduce the amount of catalyst required, the residence time, and the size of the reactor, resulting in a greater degree of process intensification than what is achievable through adiabatic or even isothermal operation in a microreactor.

A novel design is presented that provides an effective method for balancing heat and mass transfer within a microreactor. The WGS reaction is only mildly exothermic, so the standard approach of interleaving planar layers of reaction channels and heat exchange channels [1] tends to provide too much heat transfer. This problem becomes accentuated when trying to design for a desired temperature profile. A unique microreactor geometry has been developed that effectively decouples the areas for heat and mass transfer. Not only does the geometry provide versatility in reaction engineering and reactor design, but results in a much higher fraction of the volume being occupied by engineered catalyst, which is typically only 20-25% in an interleaved microreactor. A reactor will be presented that contains over 50% by volume of engineered catalyst structure.

Materials and Methods

A microchannel reactor was designed and fabricated to achieve 90% CO conversion in steam reformate at a flow capacity to support a 2 kW polymer electrolyte membrane (PEM) fuel cell. The reactor was loaded with Sud-Chemie PMS-5B precious metal low temperature water-gas shift catalyst. A simulated reformate stream is use to feed the WGS reactor based on steam reforming of gasoline at a 3:1 steam to carbon ratio. Noncondensible gases in the reformate are fed from a premixed gas cyclinder—comprised of dry gas having at composition of 12% CO, 14% CO2, and 74% H2—and mixed with steam and fed to the reactor at a controlled temperature. Coolant air is also metered to the reactor and preheated to a specified temperature. Temperatures and pressures are measured at the inlet of the reactor, and samples are extracted from the outlet header of the reactor and from a midpoint, dried, and then analyzed for composition using a gas chromatograph. Compositions are used to calculate CO conversion and to evaluate the approach to equilibrium based on the measured temperature.

Results and Discussion

The prototype WGS reactor was operated for over 50 hours at flow rates equivalent of 1 up to 2.5 kW power levels assuming 44% efficiency in a PEM fuel cell. The CO level was reduced to less than 1% (dry) at 90% CO conversion at the highest flow corresponding to 2.5 kW power level. At this level, the catalyst GHSV was 80,000 for the overall reactor. Parameters that were varied during testing included the feed flow rate and temperature, and flow and inlet temperature of the cooling air. Results are summarized in Figure 1 showing the outlet CO concentration as a function of outlet temperature for both the midpoint and at the exit of the reactor. Reactor performance was not strongly dependent on the inlet temperature or the cooling air flow and temperature.

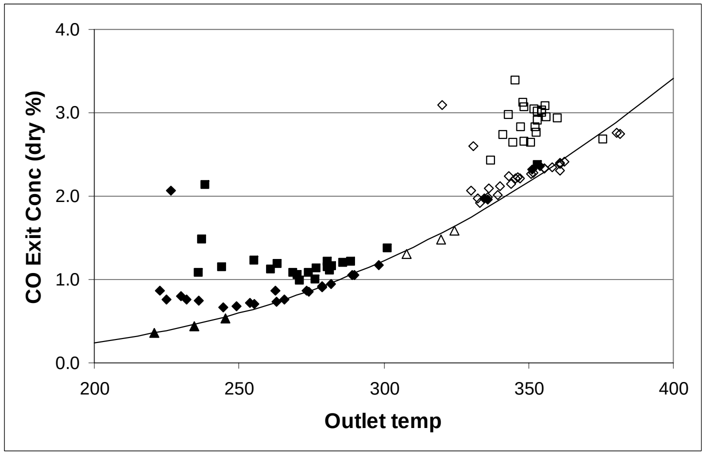

Figure 1. Performance of the prototype WGS microreactor operating with a steam reformate feed showing CO conversion at a midpoint in the reactor (open symbols) and from the reactor outlet (filled symbols) versus temperature of the stream at a 1 kWe equivalent flow rate (▲), a 2 kWe rate (◆), and a 2.5 kWe rate (■). The curve indicates the equilibrium conversion.

Significance

Size and weight estimates for a full-scale 50-kWe WGS microreactor are 3.4 L and 7.2 kg, respectively, which correspond to 14,700 We/L power density and 6,900 We/kg specific power. The size of a comparable conventional 2-stage WGS system with intermediate heat exchanger [2] utilizing the same precious metal catalyst is estimated to be 8.9 L

References

1. Holladay, J.D., Y. Wang, and E. Jones *Chem. Rev.* **104**, 4767 (2004).
2. Petterson, L.J. and R. Westerholm *Int. J. Hydrogen Energy* **26**, 243 (2001).