Microchannel Chemical Reactors for Fuel Processing Applications. II. Compact Fuel Vaporization

Anna Lee Y. Tonkovich,[(a)] Sean P. Fitzgerald, Jennifer L. Zilka,
Mike J. LaMont, Yong Wang, David P. VanderWiel, Robert S. Wegeng

Pacific Northwest National Laboratory,[(b)] PO Box 999, Richland, WA 99352

Abstract

A fuel processor is a critical element for the deployment of automotive fuel cell power systems. One component of the fuel processor, the compact gasoline vaporizer, was demonstrated at full-scale using commercial-grade gasoline. The full-scale process volume was less than 0.3 liters; it vaporized nearly 300 mL/min of gasoline, which is sufficient to support a 50-kW_e fuel cell. The reduction in hardware volume was made possible using a microchannel reactor-based design; the compact process hardware is roughly an order of magnitude smaller than conventional technology.

Introduction

A fuel processor produces hydrogen in sufficient quantity and quality for a fuel cell that produces electrical power. The desired fuel for automotive power applications is gasoline, because it reduces costly infrastructure changes to the world's fuel supply system. Furthermore, the constraint of on-board hydrogen generation requires that a process be compact, lightweight, and inexpensive. The microtechnology-based fuel processor meets each constraint, and will enable the commercialization of automotive fuel cell power systems.

The conversion of gasoline to hydrogen in a fuel processor includes a series of process steps, including vaporization, primary conversion of the fuel to synthesis gas, the water gas shift reaction, and carbon monoxide removal. The full-scale gasoline vaporizer, defined at a capacity of 50 kW_e, will be discussed in this paper.

Microchemical reactors enable miniaturization of the fuel processor because they minimize heat and mass transfer resistance. Thermal conduction and mass diffusion distances in the described microchannel reactor range between ten and several hundred microns. Conduction and diffusion distances vary from ten to

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(a) Author to whom all correspondence should be addressed: aytonkovich@pnl.gov.
(b) PNNL is operated by Battelle for the U.S. Department of Energy.

several hundred millimeters in a conventional reactor. Slow heat and mass transfer dominate the operation of conventional reactor, and thus they will require a larger hardware volume to produce the same quantity and quality of hydrogen for the fuel cell.

Background

Compact fuel cell power systems have potential applications in markets such as residential and commercial power, space exploration and defense. Automotive power systems are one of the most promising applications because of the variety and size of potential markets present in this field. Compared to conventional internal combustion engines, fuel cells are more efficient, can utilize a variety of fuels, result in reduced emissions and can be compact and modular. With the advent of federal and state legislation requiring the implementation of alternative fuels and reduced emissions vehicles into the next century, corporate and government research in this area is rapidly advancing fuel cell power system technologies. Federal initiatives such as the 1992 Energy Policy Act (EPACT) and the 1993 Partnership for a New Generation of Vehicles (PNGV) require the development of efficient and clean technologies that can operate on a wide range of fuels. Adoption of these technologies is required to begin in 2010, with California mandates requiring that 10% of cars sold in the state must be zero-emission vehicles by 2003. Ultimately, the adoption of fuel cell transportation technologies has the potential to decrease the country's dependence on imported crude oil and to decrease nationwide CO_2 emissions, while generating jobs and decreasing consumer fuel costs.

Motivated by this legislation, as well as requirements in foreign countries, the private research sector has responded with a wide range of newly developing technologies. Many automobile manufacturers have invested in electric power systems research, and are preparing to bring fuel cell and hybrid electric vehicles to the market within the next 5 to 10 years. Early fuel cell powered vehicles contained on-board hydrogen tanks, which may not be easily filled by the consumer and which present potential safety concerns. The latest fuel cell vehicles operate on methanol that, while more easily available than hydrogen, does not utilize the existing infrastructure of gasoline distribution and has a much lower volumetric energy content compared to gasoline (or diesel).

The development of compact, efficient fuel processors to generate hydrogen is critical to the deployment of PEM fuel cell power systems. These fuel processors must have the ability to convert conventional fossil fuels, for which a supply and distribution infrastructure already exists. Further, the use of fuel cells in automotive applications requires that the fuel processing system be small, lightweight and modular in nature. Figure 1 is a schematic of a typical fuel processing system for a PEM fuel cell. The first step in this process involves vaporization of a liquid hydrogen fuel such as methanol, ethanol, gasoline, diesel, mixtures of these, or others. Once fully vaporized, the hydrocarbon fuel is converted to syngas, a mixture of carbon monoxide and hydrogen. Options for this primary conversion step include partial oxidation, steam reforming or

autothermal reforming. A comparative paper on the attributes of partial oxidation versus steam reforming is reported by Argonne National Laboratory [1]. Since carbon monoxide (CO) poisons the fuel cell, the next step involves secondary conversion of CO to CO_2 in a water-gas shift reactor, which has the added benefit of increasing the hydrogen content of the fuel stream. Additional CO removal to concentrations below 10-ppm is achieved in a third reactor using preferential oxidation or CO absorption. A complete system description of the fuel processor and fuel cell is presented by Delphi [2]. A third fuel processing system configuration is under development at Arthur D. Little [3]. Each of the reported systems is based upon conventional fixed-bed reactor technology, which does not scale linearly with throughput because of inefficient heat and mass transfer. These conventional fuel processors are expected to be at least an order of magnitude larger than the microchannel-based fuel processor. Heat and mass transport limitations, which dramatically increase the size of the conventional technology [1], are mitigated with the emerging technology described in this paper.

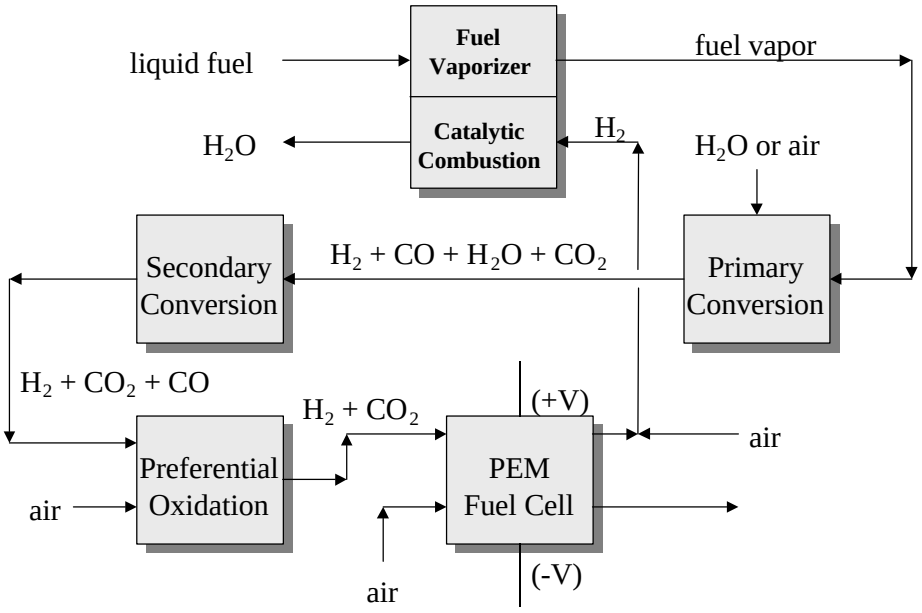

Figure 1. Schematic of Fuel Processor/Fuel Cell Power System

Microchannel chemical reactors [4,5] utilize micron-scale dimensions and integrated engineered catalyst structures to bring reactants, catalysts and heat transfer surfaces into intimate contact. In such systems, the thermal and mass diffusion paths are on the order of a thousand times shorter than those found in traditional packed-bed reactors. The resulting decreased resistances to heat and

mass transfer allow the utilization of more highly active catalysts, which take advantage of intrinsically rapid surface reaction kinetics. The application of these principles has been successfully demonstrated for a water-gas shift reactor [6] and microchannel heat exchangers [7]. The characteristics of these systems result in highly efficient and compact reactors. In addition, these systems can be designed without the large pressure drops commonly associated with small or complex flow structures.

A microchannel fuel vaporizer has been developed and demonstrated to supply gasoline vapor at a rate sufficient to feed a 50-kW_e fuel cell power system. Based on early designs [8, 9], this vaporizer utilizes integrated catalytic combustion of a simulated PEM fuel cell anode effluent to supply heat to vaporize liquid gasoline. Weighing only 1.8-kg and displacing about 0.3 L, this compact unit vaporizes gasoline at nearly 300-mL/min. These results show that this component is more than an order of magnitude smaller than a conventional fuel vaporizer [10]. Based upon our results with this and other component investigations, we project a complete system volume of less than 8 L to produce hydrogen at a sufficient rate and quality to produce 50 kW_e from a PEM fuel cell.

Experimental

A fuel processing system requires a vaporizer to prepare liquid fuels for primary conversion to hydrogen and carbon monoxide. The latent heat of vaporization required for a given fuel is provided through combustion of a dilute hydrogen stream from the fuel cell anode effluent.

H_2 (g) + O_2 (g) $\rightarrow$ H_2O (g) ΔH_r = -13.4 kJ/kg

gasoline (l, STP) $\rightarrow$ gasoline (v,215 C) ΔH_v = 293 kJ/kg

Catalytic combustion of the dilute hydrogen (6 to 8%) remaining in the anode effluent stream is a facile reaction using palladium. No pre-heating is required because hydrogen combustion initiates at room temperature.

The full-scale vaporizer consists of the body (an integrated reactor and heat exchanger) and one cover plate. The body measures 9.2-cm x 9.9-cm x 2.54-cm (or 3.6” x 3.9” x 1”). The device weighs 1.8-kg. The body is fabricated using a laminated design (patent pending). The reactor section has four parallel engineered-catalysts. The inlet flue-gas stream (anode effluent plus air) enters through the top of the unit and is separated into four parallel catalyst chambers. Each of the four streams of the hot combustion gas is then directed through a slit in the monolith footer to the header of the heat exchange cell below. A cell is defined as an array of parallel microchannels that operates in parallel to another array of parallel microchannels. After passing through the heat exchanger cell, the flue-gas stream exits the top of the vaporizer. Heat conducts through the metal web that separates the two arrays of microchannels. The cooling fluid (evaporating gasoline) enters the unit through the bottom of the vaporizer. The

gasoline moves countercurrent to the hot combustion gas stream in the heat exchange cell, before exiting through a port on the bottom side of the unit.

Each catalyst monolith has a cross section (normal to the direction of flow) of 1.61 cm^2 and the heat transfer area between the gasoline vaporization cell and the flue-gas cell is 7.2 cm^2. The channels on the flue-gas side are 254-microns wide and have an aspect ratio of 18:1. The channels on the vaporizing side of the heat exchanger plate are 254-microns wide and have a 10:1 aspect ratio.

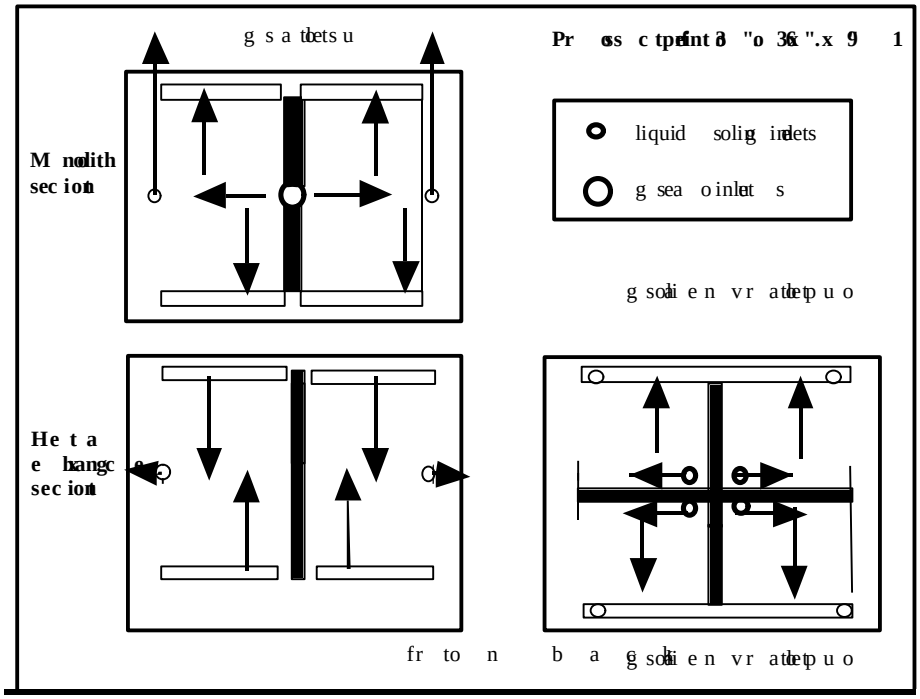

Figure 2. Full-scale gasoline vaporizer (four parallel reactor and heat exchanger cells)

Results and Discussion

The full-scale microchannel vaporizer was evaluated with consumer-grade gasoline. The device occupies a volume less than 0.3-L and is sufficient to vaporize gasoline for a full 50-kW_e fuel processing system. Performance data is given in Figure 3.

The full-scale microchannel vaporizer evaporated nearly 300-mL/min of consumer-grade gasoline through the use of four parallel arrays of microchannels, also referred to as 4 parallel cells. The maximum available flue-gas (anode effluent stream plus air for combustion) for the vaporizer was roughly 1400-

SLPM for a 50-kW_e system. The hydrogen content of this stream was roughly 6.7%. The heat released from the combusted hydrogen was contained within the flue-gas stream, approaching temperatures of 600 C. The heat was then transferred through four parallel arrays (cells) of microchannels to vaporize the gasoline feed stream. Countercurrent flow of flue-gas and gasoline was employed. The pressure drop in the microchannels on the flue-gas side was roughly 2 psi, and well below 1 psi on the gasoline side.

The microchannel gasoline vaporizer was also evaluated under part-load conditions, as shown in Figure 3. The diamond symbol represents the gasoline flow as a percent of the total capacity (~300-mL/min). The device was operated at 25% load, 50% load and 100% load. In all cases, complete vaporization was achieved (as denoted by the exit temperature of the gasoline – square symbols). The highest measured boiling point for the selected commercial-grade gasoline was 209 C. For testing purposes, complete vaporization was taken at 20 C above the highest boiling point, or at a temperature of 229 C.

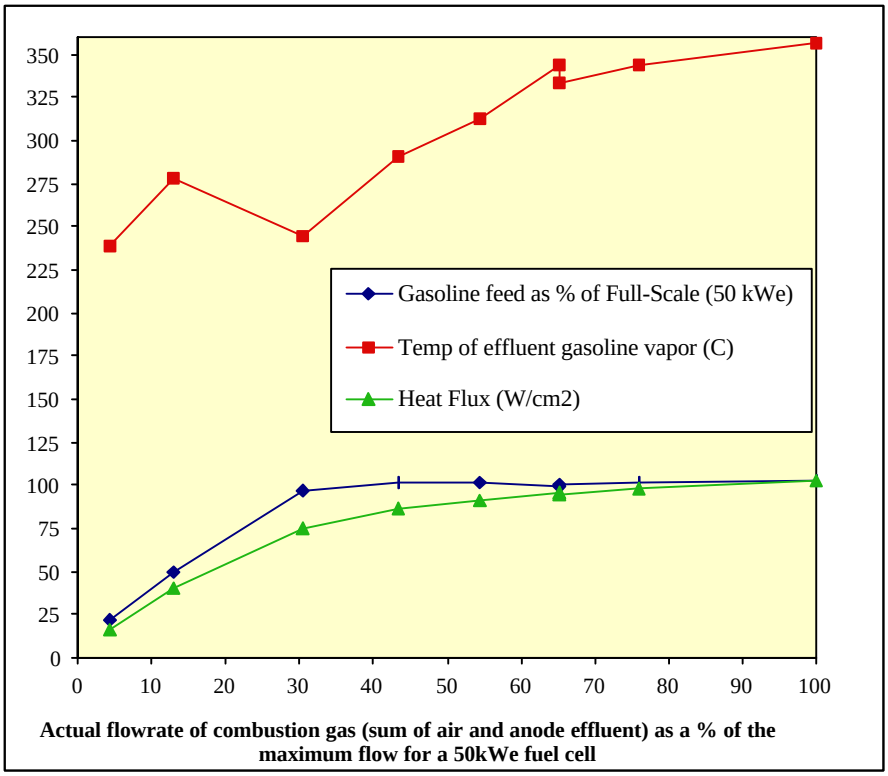

Figure 3. Full-scale gasoline vaporizer performance data

The total amount of flue-gas fed to the vaporizer was also varied between the maximum (~1400-SLPM) and a partial load (5% of maximum). For example, the first data point on Figure 3 reflects a flue-gas feed rate of ~70-SLPM (5% of full load) and a gasoline feed rate of ~75-mL/min (25% of full load). At these conditions, complete vaporization occurred, and a heat flux of roughly 20 W/cm^2 was measured between the arrays of microchannels (shown with the diamond symbols). The addition of extra flue-gas (beyond that needed to vaporize gasoline) superheated the vapor stream. In general, the heat flux during the superheating stage asymptotically approached 105 W/cm^2.

Conclusions

A full-scale gasoline vaporizer was demonstrated for subsequent use in an integrated fuel processing system. The process hardware vaporized sufficient gasoline for a 50-kW$_e$ fuel cell in a volume less than 0.3-L and weighed 1.8-kg. A complete microchannel-based fuel processing system for automotive applications is expected to be less than 8-L.

Acknowledgments

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