SYSTEM CONTROL AND EFFICIENCY MEASUREMENTS FOR A PORTABLE HIGH TEMPERATURE PEM FUEL CELL SYSTEM WITH ONBOARD FUEL PROCESSOR

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Abstract
A small 1W reformed hydrogen fuel cell system was built and tested. It consists of a fuel processor for steam reforming methanol to supply hydrogen gas for a high temperature PEM fuel cell stack. The fuel processor was integrated with an elevated temperature fuel cell stack and enclosed in a 1 cm thick insulation package. Performance and control issues of the integrated system are discussed. Energy inputs and outputs for the integrated device were estimated from the measured flow rates of methanol liquid input, the output gas measurements, and the fuel cell power output. Using this information, efficiency estimates under optimum operating conditions were made for this particular integrated fuel processor and the fuel cell system design. Issues with the current control scheme and designs for a better control scheme are discussed.

Introduction:
On board generation of hydrogen fuel using methanol steam reforming in a miniaturized reactor is one option for supplying fuel to portable fuel cells (1-7). The output from the methanol steam reformer typically contains 1-2%CO and is unsuitable for low temperature (25-100°C) PEM fuel cell use. Additional water gas shift and preferential oxidation reactors or a H2 separation membrane are required to reduce CO levels for low temperature PEM fuel cell use. These approaches require more controls and increase the system complexity. An alternative approach is to use a high temperature PEM fuel cell operating at 160-200°C which can use the output from the methanol steam reformer directly (8), which simplifies the fuel processor design. At Motorola we are using this alternative approach to develop reformed hydrogen fuel cell systems for portable power applications (3,9-12). We have designed and built a prototype 1W reformed hydrogen fuel cell system by integrating a high temperature PEM fuel cell stack with a small methanol fuel processor. These two units are thermally integrated in an insulating package in order to achieve good system efficiency. Figure 1 shows a schematic diagram of the integrated reformed hydrogen fuel cell system. The fuel processor consisted of a fuel vaporizer, methanol steam reformer, fuel/air mixer and a catalytic combustor. The planar fuel cell consisted of four cells connected in series electrically. The design, fabrication, and performance of the individual components prepared using a multilayer ceramic technology were reported previously (3,9-12). In this paper we report on the control issues and efficiency measurements of a small 1W design integrated reformed hydrogen fuel cell system.

As seen from the schematic system diagram, a catalytic combustor provides the heat input required for fuel vaporization and steam reforming reactions. Output from the
fuel cell anode consisting of unused hydrogen, CO, unreformed methanol vapor, water vapor and CO2 is routed through the catalytic combustor before exhausting into the environment. This will oxidize CO and methanol as well as capture the calorific value of the unused fuel for increased system efficiency. During operation of the fuel cell, the chemical energy of the fuel is directly converted into electrical energy and the inefficiencies of the energy conversion are dissipated as heat. By thermally integrating the fuel cell and the fuel processor this waste heat can be captured for increased system efficiency. For the integrated fuel cell system with an on-board fuel processor, careful balance of fuel processor and fuel cell outputs, and thermal integration of these two units are required for better system efficiency.

![Diagram](image)

**Figure 1. Schematic outline of an integrated reformed hydrogen fuel cell system.**

**Experimental:**

Design and fabrication of the fuel processor and the high temperature fuel cell components was accomplished using multilayer ceramic technology, which allowed the fabrication of monolithic 3D ceramic structures with a high degree of integrated features and functionalities. Details of the processing are described elsewhere (3,11).

**Fuel Processor:** A small fuel processor measuring 5cm x 5cm x 0.06 cm capable of producing sufficient hydrogen for a 1W size fuel cell was fabricated using multilayer ceramic technology. Fig 2 shows a picture of the fuel processor used in this experiment. The fuel processor consists of four major sections: a fuel vaporizer, a packed bed steam reforming section, a mixer for mixing the fuel and air before feeding into the combustor, and a catalytic combustor, which is located below the reformer and vaporizer sections.

For the combustion catalyst, a platinum based thick film catalyst paste was applied by screen printing process during the fabrication of the ceramic fuel processor. After sintering the multilayer ceramic fuel processor, about 500 mg of a commercial CuO/ZnO/Al2O3 steam reforming catalyst powder (200-250 um particles) was filled into the reformer section of the fuel processor and sealed with an alumina cement followed by an epoxy coating. For the inlet and outlet connections, stainless steel or fused silica tubes were joined to the ceramic fuel processor using the same sealing method. The fuel
processor was packaged within a 1 cm thick microporous insulation material (Microtherm, \( k = 0.023 \text{ W/m-K} \)) for characterization.

**Fuel Cell Stack:** A ceramic substrate measuring 5.7cm x 5.7cm x 0.06cm, for assembling a planar stack of four fuel cells connected in series was designed and fabricated using the multilayer ceramic technology. Fig 3 shows the picture of the ceramic substrate used for the fuel cell assembly. As seen from Fig 3.1, screen printed thickfilm gold conductors were used for current collection. Parallel gas flow fields for distributing the anode and cathode gases were built into the substrate. High temperature PEM fuel cell MEAs from PEMEAS were sandwiched between the two ceramic substrates (Fig 3.2). Active area for each cell was 2.5 cm\(^2\). A PTFE based gasket was used to achieve a gas tight seal around the MEAs between the two ceramic substrates. Fig 3.3 shows an assembled fuel cell stack. Series electrical connections were made externally and PTFE tubes were connected for the anode and cathode gas inlet and outlet connections.

![Fig 2: Fuel processor fabricated using the multilayer ceramic technology.](image)

![Fig 3: Ceramic substrates for fuel cell housing. 1) shows flow fields and current collectors 2) MEA assembly with PTFE gaskets 3) assembled PEM fuel cell](image)

**Integrated Fuel Processor and Fuel Cell:** For the integrated unit, two separate components consisting of a fuel processor and a fuel cell were assembled together and enclosed in a 1 cm thick insulation package. Fig 4 shows a picture of the integrated fuel processor and fuel cell system in the insulating package with the top cover removed. The output from the fuel processor is connected to the anode inlet of the fuel cell, and the output from the fuel cell anode is connected to the catalytic combustor. The size of the integrated unit after insulation is approximately 8.9cm x 8.9cm x 5cm.
Characterization: Initially the fuel processor was tested separately without integrating with the fuel cell. For startup, hydrogen gas and air were combusted in the catalytic combustor to bring the fuel processor to the operating temperature of 230°C. After stabilizing the fuel processor at the operating temperatures, controlled amounts of liquid fuel consisting of methanol/water mixture in 1:1.1 mole ratio was fed into the fuel processor using a small peristaltic pump. Inside the fuel processor, the liquid fuel mixture will first flow through the vaporizer section and then over the steam reforming catalyst bed to generate a hydrogen rich gas. The output from the steam reforming reaction consists primarily of H₂ and CO₂ and small quantities of CO, unreacted methanol vapor and water vapor (excess water from the input feed). The output gases were passed through a condenser to remove the methanol and water and the remaining total output gases were measured using a flow sensor. Air was supplied using a small diaphragm pump, and the air flow into the combustor was measured using a flow sensor. Several thermocouples were attached to the top surface of the fuel processor at different zones and temperature was monitored during the operation. The fuel cell was later added to the fuel processor, and the influence of thermal integration of the fuel cell with the fuel processor was studied. A programmable power supply was used to control the pumps and valves. While the system is in control, the power inputs (current and voltage) to the pumps and valves were monitored for energy balance and efficiency calculations. A LabView™ control program with a PID controller was written to control the temperature of the fuel processor and to monitor all the process variables (temperatures, gas flows, power inputs to the pumps and valves). A current source meter (Keithley 2400) operating in the sink mode was used to draw a constant current from the fuel cell and to monitor the stack voltage.

Figure 4. Integrated fuel cell and fuel processor in an insulation box with top cover removed.

System Control: In an integrated reformed hydrogen fuel cell system, the temperature control of both the fuel processor and the fuel cell are important. Based on our previous experiments using this particular steam reforming catalyst in our reactors; the optimum
operating temperature for the fuel processor was 230±5 °C in the steam reforming zone. At higher operating temperatures there may be lifetime reliability issues with the catalyst and at low temperatures the methanol conversion may not be complete and also there is a danger of caking the catalyst powder if condensation occurs within the reforming section. Similarly for the fuel cell stack, the optimum operating temperature range is 160-200°C. At low temperatures CO adsorption on the catalyst is a problem affecting the performance, and at high temperatures there are reliability concerns due to possible MEA structure and polymer degradation.

As shown in the schematic diagram (Fig.1), anode effluent from the fuel cell is fed to the combustor to provide the heat for the fuel processor. Thermal integration with the fuel cell allows for the utilization of the waste heat generated from the fuel cell inefficiency, for the vaporization of the liquid fuel (methanol and water mixture) feed into the fuel processor. To control the temperature of the fuel processor according to this scheme requires either controlling the hydrogen flow between the fuel cell and the combustor or controlling the combustor with a supplemental fuel supply as needed. Fig.5 shows one control scheme where the flow of hydrogen is controlled between the combustor and the fuel cell using a two way control valve. As discussed later, this is not the preferred method to control the system. Control of fuel processor temperature with a supplemental fuel to the combustor as needed, using a separate methanol fuel pump, is the preferred method.

Results and Discussion:
Fuel Processor Control: In the initial experiment, only the fuel processor enclosed in an insulating package was tested (no fuel cell). The control scheme shown in Fig 5 was used to maintain a constant temperature in the reforming section. Since no fuel cell was attached in this experiment, one of the outputs from the two way valve, which controls the reformed hydrogen rich gas flow between the combustor and the fuel cell, was exhausted through a condenser and the total gas output was measured. In order to start the fuel processor, hydrogen (24sccm) and air (~100 secm) were combusted in the catalytic combustor to bring the fuel processor to the operating temperature of 230°C. Then methanol/water (1:1.1 mole ratio) liquid fuel feed at 1.35ml/hr flow rate was introduced into the fuel processor using a small peristaltic pump. As the liquid fuel is fed into the fuel processor, it vaporizes and the vapors then go through the catalyst packed bed to the steam reforming reaction. Once the fuel processor has started generating hydrogen gas from the steam reforming reaction, the reformate output gas is fed into the combustor, via the two way valve, to “self sustain” the reforming reaction in the fuel processor with just the liquid methanol/water fuel input from the peristaltic pump. The two way control valve at the outlet of the fuel processor was used to direct the reformate flow into the combustor or to an exhaust (through a condenser to measure the total gas output), which maintained a constant temperature at the reforming section of the fuel processor. Fig 6 shows the results from this experiment while the fuel processor is operating with constant temperature control from the two way valve diversion of excess hydrogen rich reformate output gas. Temperatures from three different zones on the fuel processor are plotted. The control temperature was set at 230°C. The temperature at the reformate gas outlet is 220°C, and the temperature at the liquid feed inlet is 145°C while the system was in control. Gas flow rates are also plotted in the same figure. As seen in the figure, when the reformate flow was going through the combustor, no gas flow was detected after the condenser as expected, and when the two way valve is directing the flow away from the
combustor to maintain a constant temperature, gas flow after the condenser was detected. The average total output gas flow after the condenser was about 31 sccm. The small pump used for the air supply maintained an average air flow rate of ~100 sccm. As seen from the data, this control scheme maintained the temperatures well, but the temperature fluctuations around the set point are larger. These fluctuations can be reduced further by fine tuning the PID controller.

\[ \text{Fig. 5. Schematic diagram of the integrated fuel cell and fuel processor control to maintain a constant temperature for the steam reforming reaction, using a 2-way valve to direct the reformate flow between the combustor and the fuel cell anode. Another method of control is to add methanol fuel to the combustor as needed, using a different pump.} \]

\textbf{Methanol Conversion Efficiency and Energy Balance:} For a methanol/water input flow rate of 1.35 ml/hr, at 100% conversion efficiency, the expected total output dry gas flow rate is 33.5 sccm. As seen from the experimental data in Fig 6, when the two way valve is directing the flow through the condenser, the average total output gas measured was 32.9 sccm. This flow rate corresponds to ~98% methanol conversion in the fuel processor. In this experiment, the composition of the output gas was not analyzed. However, based on previous experiments in our lab on a similar system, GC analysis of the gas composition showed about 1-2% CO, 73-74% H₂ and 24-25% CO₂ (11). Assuming a similar gas composition mix; for a 32.9 sccm total reformate output flow, the expected H₂ flow was about 24 sccm.

Since we were able to detect gases at the condenser exhaust at the 1.35 ml/hr rate of liquid fuel input into the fuel processor, the fuel processor was generating more than enough hydrogen fuel required to “self sustain” the steam reforming reaction and maintain the required 230°C operating temperature. Reducing the flow rate to 1.3 mL/hr, the fuel processor maintained steady state operation for a long time without the need for two way valve control to exhaust excess hydrogen rich gas. So for this particular fuel...
processor, the energy inputs required to "self sustain" were: 1.3 ml/hr flow of MeOH/H2O (1:1.1 mole ratio) or 0.87ml/hr methanol input (balance water). This was equivalent to 4.097 W of energy (using the ΔH of methanol oxidation reaction in air). Other energy inputs were: electrical power input to the liquid fuel pump and the air pump, which were 0.06 and 0.107W, respectively. Thus the total energy input required to "self sustain" the fuel processor of this particular size and design is 4.264 W.

![Fuel Processor Control Experiment](image-url)

**Fig 6: Fuel processor control experiment without fuel cell integration, showing the temperature control, air flow into the combustor and the total output gas flow through the condenser after the control valve.**

**System Efficiency Projections:** In order to generate 1W electrical power using a high temperature fuel cell operating at 30% efficiency with the hydrogen generated from this fuel processor, it would require the generation of 3.33W equivalent of H2 gas or approximately 18.7sccm. To generate this amount of hydrogen from the fuel processor at 98% methanol conversion rate, 1.03 ml/h of MeOH/H2O (1:1.1 mol) feed into the fuel processor would be required. The total liquid 1:1.1 mol MeOH/H2O fuel input to the fuel processor for 1W net electrical operation would be 2.33ml/hr (1.3ml/hr + 1.03ml/hr). The energy content of this fuel is 7.25W, and with the expected electrical output of 1W, the projected system efficiency in this case would be 1/7.25 = 14%. In this analysis the additional heat input required for the fuel vaporization and the steam reforming reaction for the 1.03ml/h fuel input, and the heat generated at the fuel cell due to the inefficiencies of energy conversion were not considered. This gives a rough estimation of the expected efficiencies of this system. Miniaturizing the fuel processor and increasing the efficiency of the fuel cell would increase the overall system efficiency. These first generation
prototypes were not built for optimum performance but they were designed slightly bigger to understand the integration and system control issues.

**Integrated Fuel Processor and Fuel Cell Control:** In the previous experiment only the fuel processor was tested. In the next experiment, a planar high temperature PEM fuel cell stack consisting of four fuel cells connected electrically in series (Fig 3) was integrated with the fuel processor as shown in Fig 4. The output from the fuel processor was connected to the anode side of the fuel cell stack and the output from the fuel cell anode was connected to the catalytic combustor in the fuel processor. A separate air pump was connected to supply air to the cathode side of the fuel cell stack. Initially the fuel cell was left in an open circuit condition. After allowing the fuel processor and the fuel cell to reach stable operating temperatures, the fuel processor was controlled at the desired operating temperature of 230°C, using the control scheme shown schematically in Fig 5. Similar to the method described earlier in the fuel processor control, methanol/water liquid fuel input into the fuel processor was controlled to maintain a stable operating temperature of 230°C for the steam reforming reaction with the excess hydrogen rich gas (reformate gas) flow from that reforming reaction. In this case since the extra size (thermal mass) of the fuel cell is added to the fuel processor, it is expected that a higher liquid fuel input would be required compared to the experiment where only the fuel processor was controlled at 230°C operating temperature.

![Figure 7](image.png)

**Fig 7:** Temperature control of the fuel processor for the integrated fuel processor and fuel cell system. Temperatures and gas flows while the system is in control are shown in this figure. Fuel cell was in the open circuit condition for the first 150 mins, and the excess reformate gas was exhausted out. After 150 mins, slowly increasing load was applied to the fuel cell consuming the hydrogen in the reformate gas.

Fig 7 shows the results from the integrated fuel cell stack and fuel processor control experiment. Three representative temperatures measured on the fuel processor at

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different zones and the gas flows were plotted in this figure. Under steady state conditions, the fuel processor control temperature was maintained at 230°C. The temperature at the reformate gas outlet was 200°C and the temperature at the liquid inlet flow was 90°C. Air flow into the combustor was ~ 100 scm, and the average total output gas flow through the condenser was 52 scm. Liquid fuel (methanol/water mixture) input into the fuel processor under these conditions was 2.1 ml/hr which was equivalent to 6.62W of energy. Since some of the reformate gas was exhausting out at this flow rate, it was expected that a slightly lower liquid fuel input would be needed to maintain the thermal mass of the integrated fuel cell and fuel processor at the required 230°C operating temperature.

After the system was allowed to stabilize for 150 mins, a slowly increasing load was applied to the fuel cell. Fig 8 shows the fuel cell stack voltage data at 120 mA/cm²

![Fig 8: Integrated fuel processor and fuel cell control experiment. After stabilizing the fuel processor, slowly increasing loads were applied to the fuel cell. Fuel cell stack voltage at 120 mA/cm² current density is shown in this figure.](image)

![Fig 9: Fuel processor control with the methanol pump which supplies the fuel as needed to the combustor to maintain a constant fuel processor temperature.](image)
the thermal integration of the fuel cell with the fuel processor. Thermal contribution from the energy conversion inefficiency of the fuel cell stack (~0.97W in this case) was expected to change the fuel processor temperature profile. However sometime in the middle of the test, while the fuel cell stack was under load, the software control program malfunctioned and it was difficult to separate the influence of this thermal contribution on the temperature profile of the fuel processor. This particular method of system control is not preferred because the valve control will interrupt the flow of anode gas into the fuel cell when it is diverting the flow into the combustor. Cutting the fuel supply off to the anode while the fuel cell is under load could degrade the performance of the fuel cell and should be avoided. A better method of control would be supplying additional methanol fuel into the combustor as needed using a different pump. This will not interrupt the anode gas flow to the fuel cell stack, but it will add an extra pump to the balance of plant. Fig 9 shows the results for a fuel processor control with this method. In this method, the control program tripped the power supply to the pump on or off as needed to maintain a constant fuel processor temperature. Integrated fuel cell and fuel processor experiment was not run with this control scheme, but it is planned for future experiments.

Energy Balance and System Efficiency Projections: For the integrated system, the energy inputs were: liquid fuel input 6.62 W pumps 0.17W, giving a total energy input of 6.79W. Output was 0.51W of electrical energy. Thus the system efficiency was 7.5% in this case. If we were to generate 1W electrical power as expected for this fuel cell, then the system efficiency would be 15%. By reducing the physical size of the unit and by increasing the fuel cell power output and its efficiency, the overall system efficiency can be increased further. Miniaturization and good insulation are very important in improving the system efficiency. Our estimations showed that with optimum designs and good insulation it is feasible to achieve 25-30% efficiencies for these reformed hydrogen fuel cell systems. The first generation prototype designs tested in these experiments were not optimized for high efficiency, they were designed for understanding the performance and integration of different components and the control issues of the reformed hydrogen system for portable power applications.

Conclusions:
The feasibility of a small 1W reformed hydrogen fuel cell system was demonstrated. Control issues for the fuel processor and the integrated system were explored and steady operation of the system under controlled operating conditions was shown. For these early prototype samples the overall measured system efficiency is 7.5%. However, continued miniaturization of the components and improving the performance of the fuel cell stack are expected to increase the system efficiencies significantly, making it feasible to design a competitive small power source for portable power applications.

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