

Fuel processor development for a soldier-portable fuel cell system

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1. Introduction

The remarkable recent advances in wireless and portable communications devices (e.g., laptop computers, cellular phones, portable digital assistants) have fueled a need for high-energy-density portable power sources for consumer use. Similarly, interest in portable power sources has increased in the military and intelligence communities. Currently, portable military electronics are dependent on batteries to supply electrical power for long-duration missions. This poses two major problems which result from the low energy density of current battery systems: excessive weight/bulk, and reduced mission duration.

Compact fuel cell systems that operate on liquid hydrocarbon fuels offer an efficient, lightweight alternative to batteries, thus allowing for greater portability and longer mission lifetime. For instance, the energy densities of diesel fuel and methanol are each at least an order of magnitude greater than that of lithium-ion batteries. Another option, hydrogen storage (compressed or chemical), provides an energy density not much greater than can be found in batteries. As shown in Table 1, a hydrocarbon-based fuel cell system operating at just 5% overall efficiency has a higher energy density than a lithium polymer battery and at least equal energy density as a polymer-electrolyte membrane (PEM) fuel cell system operating on stored hydrogen. Clearly, liquid hydrocarbons would be the preferred energy source for a portable power system if a rugged, reliable, and lightweight fuel processor were available to convert hydrocarbon fuels to hydrogen. An appropriate fuel processor would efficiently produce hydrogen of sufficient quantity and purity to drive a PEM fuel cell, and would do so within a small volume.

Table 1. Comparison of energy densities from various sources

Fuel	LHV (kJ/mol)	Energy Density (kW-hr/kg)	Efficiency Required (to match batteries)
Methanol	639	5.5	5.5 %
n-Butane	2650	12.7	2.4 %
n-Octane	5100	12.4	2.4 %
H ₂ storage	242	0.5-1.0	30-60 %
Lithium-ion battery	--	0.3 (projected)	--

Battelle is a leader in the development and demonstration of small-scale, hydrocarbon fuel processors to generate high-purity hydrogen for portable fuel cells. For several years, Battelle has led the development of micro-process technology for various applications and device sizes. These technologies include fuel vaporization,¹⁻⁴ gas conversion,⁵⁻⁷ fuel processing,^{2, 6-12} heat transfer,^{1, 3, 4, 13, 14} mass transfer,^{15, 16} catalytic combustion,^{3, 4, 13, 17} and partial oxidation.¹² In each application, the microchannel architecture drastically reduces the heat- and mass-transfer resistances relative to conventional systems.

Many of these advances are built upon Battelle's aggressive effort to develop catalysts for microreactor applications. Battelle has developed highly active steam reforming catalysts for various hydrocarbon fuels. These catalysts were developed for millisecond contact time ($GHSV = 10^4$ - 10^5 hr^{-1}) applications for use with many different fuels, including methanol, butane, iso-octane, diesel, and JP-8.

Such micro-process and catalyst technology is, thus, a natural fit for portable power systems, where size and weight must be minimized. For instance, the required catalyst volume for a 15-W steam reformer operating at 100 ms contact time is less than 0.5 cm^3 . This translates to correspondingly small device footprints, such that the majority of the weight and bulk of the final system is dictated by the related fuel supply rather than the fuel processor (based on multi-day missions).

2. Objectives

Under this development project, funded by the U.S. Army Communications-Electronics Command, Battelle is developing a 15-W_e fuel processing system suitable for portable power applications. The need arises from a desire to reduce the weight and bulk, and thus increase the mobility and effectiveness of the Army's combat divisions. This portable power source is expected to provide a clean hydrogen stream to a small fuel cell according to the target specifications listed in Table 2.

Table 2. Target specifications for soldier-portable power system

Average Power	15 W
Peak Power	25 W
Volume	< 100 cm ³
Weight (excluding fuel)	< 1 kg

For military use, the system will ultimately operate on diesel or some other logistics fuel. However, for commercial use and the first military applications, the fuel of choice is methanol. Not only can methanol be reformed at lower temperatures than other hydrocarbons, but it also is miscible with water, which is a key issue in fuel storage and delivery. The lower reforming temperatures of methanol allow for lower heat losses, require less insulation, and simplify the thermal management of the integrated system.

3. Experimental

Development of the portable fuel processor begins with the design, fabrication, and testing of individual unit operations. Afterward, these unit operations are integrated at the bread-board* level. This occurs simultaneously with catalyst testing and selection for both the combustor and the steam reformer. The next step, which is beyond the scope of this paper, includes the full integration of all unit operations in a single, rugged device suitable for field testing.

All catalyst preparation, device fabrication, system testing, and product analysis were performed on site. The current system is composed of stainless steel process units connected by 0.125-in or 0.250-in stainless steel tubing. Thermocouples and pressure transducers are placed strategically throughout the device, and system data is collected through an on-line data acquisition system.

Catalytic combustion of methanol is used to provide system heat which is necessary for reactant vaporization and preheat as well as for heat of reaction for the endothermic steam reforming of methanol (+50 kJ/mol). As illustrated in Figure 1, methanol (from a syringe pump) and compressed air are fed separately to the vaporizer/preheater, from which the combined vapor stream enters the combustion zone. Hot combustion gases are then used to heat the steam reformer. Downstream of the reformer, the combustion gases then provide heat to the two vaporizers that feed the combustor and the reformer feed stream.

A premixed solution of methanol and water (1:1 ratio by weight) is fed to the system using an HPLC pump. The combined stream first enters the reactant vaporizer, then flows through the steam reformer, where it is converted to H_2 , CO_2 , and CO . The reformat is chilled, passes through a vapor-liquid separator to

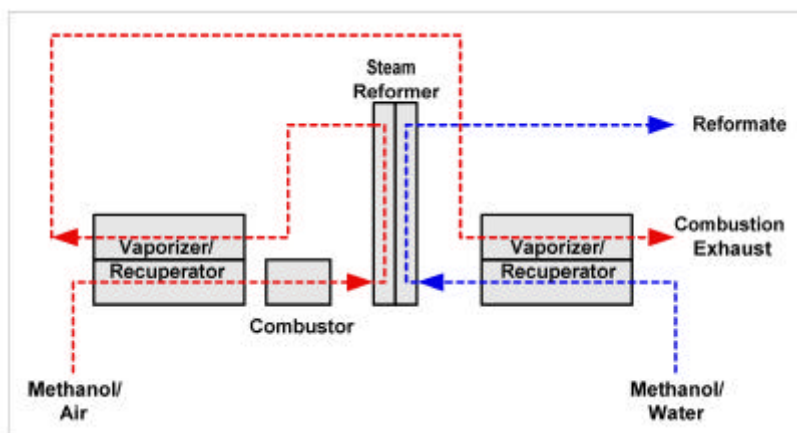


Figure 1. Bread-boarded* fuel processing system for methanol

* The term "bread-board" refers to a process train in which all major components are present and connected, but not fully integrated.

remove residual water and methanol, and then flows to the online gas chromatograph for analysis.

The gas chromatograph used in our investigation is an Agilent Technologies Micro-GC capable of detecting gases and hydrocarbons as large as C₈. However, in the methanol-reforming system, detection of compounds up to C₂ is sufficient. All gases other than H₂, CO₂, CO, and CH₄ remain below the detection limit (100 ppm) of the instrument under the system conditions investigated.

4. Results and Discussion

4.1. Steam Reformer

Figure 2 illustrates the performance of the steam reforming reactor over a range of temperatures and contact times. As can be seen from the plot, the reactor can be operated at temperatures as low as 300 °C, but this requires a contact time of 300 ms. Even at very fast throughput (50-ms contact time), the reaction only requires a temperature of 375 °C.

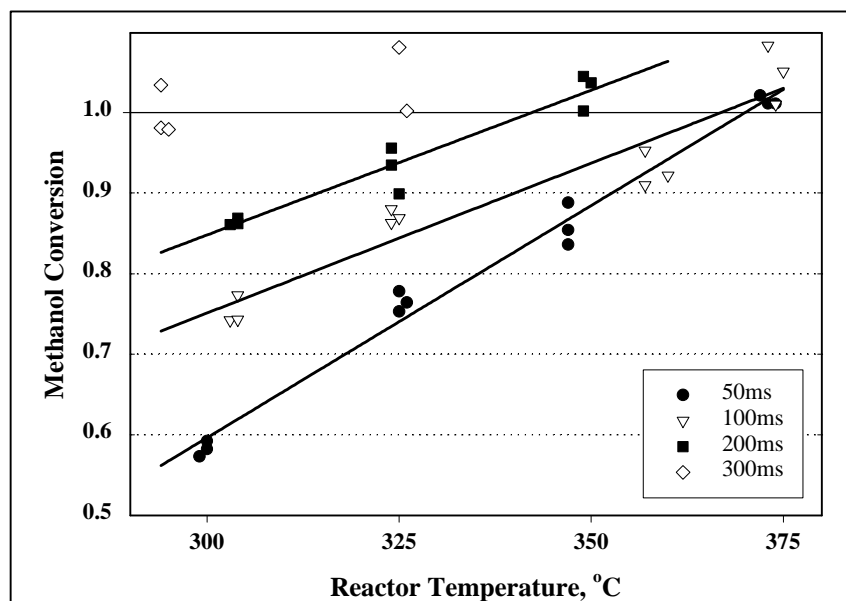
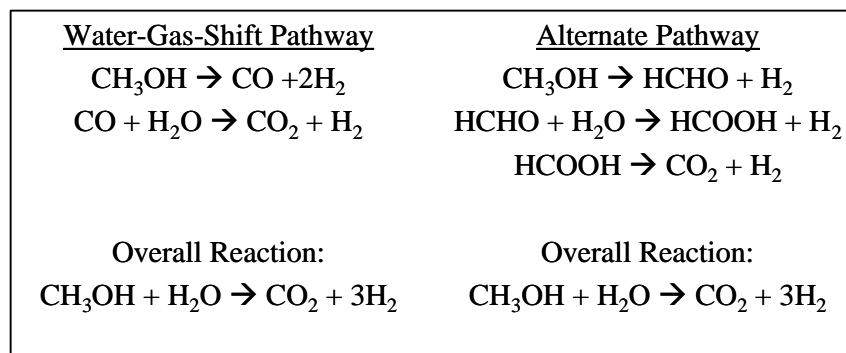


Figure 2. Steam reforming of methanol, reactor performance

Under these conditions, typical dry reformate composition was 73-74% H₂, 25-26% CO₂, and 0.6-1.2% CO. This represents 100% selectivity to H₂ and 95-98% selectivity to CO₂. The low levels of CO (lower than equilibrium) are achievable only with methanol, which does not need to be converted through the water-gas

shift mechanism, as described by Takahashi, et al.,¹⁸ and illustrated in Scheme 1, below. Furthermore, the low level of CO leads to two major advantages in this methanol-reforming system. First, it provides for higher hydrogen production. That is, the theoretical maximum hydrogen production for methanol steam reforming is 3 mol H₂ per mol MeOH converted. Under these conditions, this hydrogen production ratio is 2.7 to 2.8, or 90% to 93% of the theoretical maximum yield. The second advantage of low CO concentrations is the simplification of CO cleanup. Since current PEM fuel cells cannot tolerate even these low levels of CO, a CO cleanup step must eventually be incorporated into the process train. However, such low levels of CO eliminate the need for a water-gas shift reactor.



Scheme 1. Optional pathways for methanol steam reforming

4.2. Bread-board System

The same reactor that produced the results of Figure 2 was integrated into the “bread-board”^{*} system consisting of a combustor, two vaporizers, and a steam reforming reactor. Under self-sufficient operating conditions, this bread-board system yielded hydrogen outputs equivalent to 10 to 64 W_e. The equivalent wattage was calculated assuming a fuel cell with an energy efficiency of 60% and a hydrogen utilization of 80%. Based on these assumptions, the electrical wattage is equal to about half the thermal wattage of the hydrogen produced due to inefficiencies of the fuel cell.

Figure 3 illustrates the operating conditions under which 100% conversion of methanol is attained. As expected, higher contact times (lower throughput) are necessary at lower operating temperatures. Operation at a temperature as low as 325 °C is possible at a contact time of only 270 ms. Conversely, the system can be operated at a very short contact time of 50 ms, but at such high throughput, the

^{*} The term bread-boarded refers to a process train in which all major components are present and connected, but not fully integrated.

reactor temperature must be raised to 440 °C. A system analysis will be performed to determine the optimal balance between contact time and temperature. For instance, higher operating temperatures may allow for smaller reactor volumes, but they also increase the need for insulation. At such small device sizes, bulk may ultimately be more important than weight, so there is likely to be a tradeoff between reactor volume and insulation volume.

Efficiency calculations were performed on the bread-board system using a base case. Heat recovery and insulation issues have not yet been fully addressed, so the current process train represents a worst-case scenario. As listed in Table 3, the base-case operating conditions were 350-°C reactor temperature, 140-ms contact time, and a steam to carbon ratio of 1.8:1. Under these conditions, methanol conversion was 100%, and the system produced 27 W_i of hydrogen. Based on the amount of methanol fed to the system, this translates to a fuel processor efficiency of 45%. Based on the previously mentioned fuel cell efficiency assumptions, the overall fuel processor/fuel cell system would produce 13 W_e , an overall efficiency of 22%.

Under these same operating conditions, the dry gas composition was 74% H_2 , 25% CO_2 , and 0.8% CO . Any methane production during base-case operation was below the detection limit of the gas chromatograph. The very low CO concentration of the reformat eliminates the need for a water-gas shift reactor, significantly simplifying the overall system. In fact, at these low CO concentrations, a water-gas shift reactor would facilitate the reverse-water-gas shift reaction, resulting in higher CO levels in the product gas and consuming some of the product H_2 .

The power density of the bread-board system is 720 W-hr/kg, a level that is already several times higher than the best lithium-ion battery. There are several obvious areas which, if addressed, would significantly raise the efficiency and power density of the system. For instance, line losses and exhaust heat were found to account for more than 40% of the combustion heat produced in the system. Eliminating these losses alone would increase the fuel processor efficiency to 60% and the power density to 870 W-hr/kg.

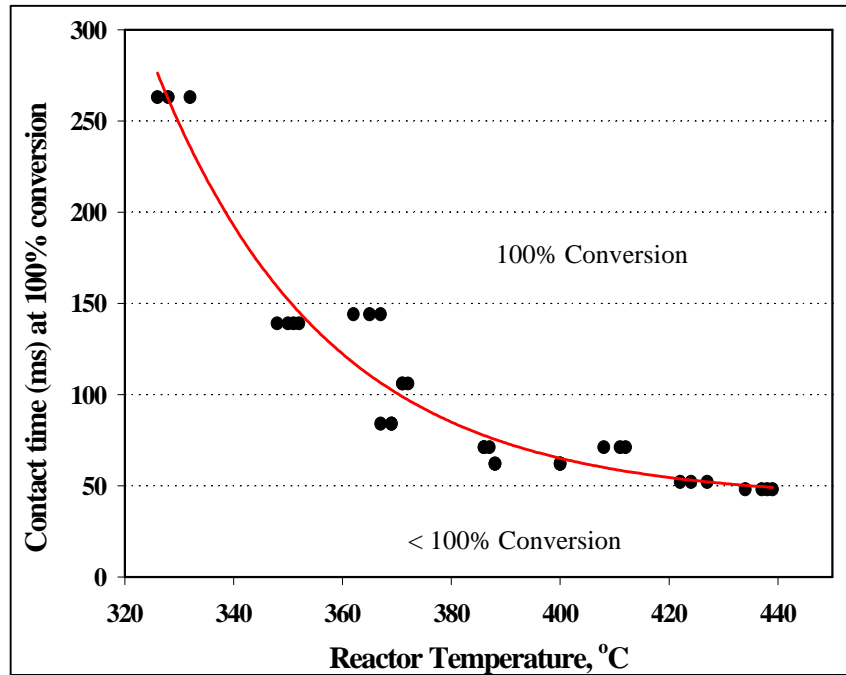


Figure 3. Contact time versus temperature at 100% conversion of methanol

Table 3. Base case demonstration results

Operating Conditions	
Reformer Temperature	350 °C
Contact Time	140 ms
Water/MeOH molar ratio	1.8
Fuel Processor Results	
MeOH conversion	100 %
Gas Composition	
H ₂	75 %
CO ₂	24 %
CO	0.8 %
Hydrogen Production	27 W _t [*]
Methanol Fed to System (reformer and combustor)	60 W _t [*]
Fuel Processor Efficiency	45 %
Fuel Cell Power Estimate [#]	13 W _e [*]
System Efficiency [#]	22 %
Estimated System Power Density [†]	720 W-hr/kg

*W_t = watts thermal, W_e = watts electric

[#]Assuming fuel cell efficiency of 60% with 80% H₂ utilization

[†]Assuming 14-day mission and 1-kg fuel processor/fuel cell system

5. Conclusions

A bread-boarded 15-W_e methanol fuel processor has been developed for portable power applications. The current reactor train includes a combustor, two vaporizers, and a steam-reforming reactor. The device has been demonstrated under thermally self-sufficient conditions over the range of 10 to 64 W_e.

Assuming a 14-day mission life and a 1-kg fuel processor/fuel cell assembly, a base case was chosen to illustrate the expected efficiencies. Operating at 13 W_e, the system has a fuel processor efficiency of 45% and an estimated overall efficiency (including fuel cell) of 22%. This translates to an energy density of 720 W-hr/kg, which is several times the energy density of the best lithium-ion batteries. Some immediate areas of improvement in thermal management also have been identified.

6. Acknowledgment

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