LOW COST, HIGH EFFICIENCY REVERSIBLE FUEL CELL SYSTEMS

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Abstract

Fuel cell technologies are described in the 2001 DOE Hydrogen Program Annual Operating Plan as "cost effective, highly efficient, and critical for overall success in the Hydrogen Program Strategic Plan". *Fuel cells which operate on fossil fuels as well as on hydrogen serve as a* ***transitional technology,*** *as the world moves away from fossil fuels, and as an* ***end point technology,*** *for the production and utilization of Hydrogen.*

The TMI reversible fuel cell – electrolyzer system employs a high temperature solid-oxide based electrochemical process to produce electricity from common hydrocarbon fuels (e.g., natural gas, propane, and bio-derived fuel) as well as hydrogen. In electrolyzer mode, the reversible system uses electricity and thermal energy to convert pure water into fuel (hydrogen and oxygen). TMI's reversible system uses the waste thermal energy produced during electricity generation mode to achieve high systems efficiency during electrolysis mode, ultimately lowering product life cycle costs for the combined system. To further increase systems efficiency, TMI has adopted a 'passive' cell design which minimizes balance of plant components

During this phase, TMI demonstrated passive reversible cells and stacks that met many of the performance targets including reversible efficiency and life. Several conditions were evaluated to understand the sensitivity of performance on operating variables such as temperature and current density. The highest reversible efficiency (DC_{Volts_Out} / $DC_{Voltage\_In}$) measured was 90.8% at 925°C and 50 mA/cm^2. A five cell stack was operated at < 2% / 1000 hours voltage degradation in fuel cell mode for over 1200 hours.

Data from this work was used to inform economic and engineering studies during Phase II. A versatile product line was envisaged, with the capability of delivering 5000 psi pure hydrogen for vehicle uses in addition to electric power and recovered usable heat. The proposed modular systems have high expected reliability and could be sized for either residential or commercial applications (including "power parks") being fed by electric power (renewable or low-cost grid) and/or fuel (fossil or biomass-derived).

The program has met or exceeded all technical objectives (as revised by DOE Hydrogen Program management) on budget and on time. Experimental results have been promising. The economic / engineering studies indicate potential for the TMI fuel cell - electrolyzer reversible system to set new performance standards, applicable to most alternative technologies, for achieving lower cost of H_2 production, lower pollution levels, and potentially serving as an enabling technology for hydrogen fuel cells.

Table 1. TMI projections for 10 kW Systems Producing 5000 psi Pure Hydrogen

Parameter	DOE Goal	TMI Projection*
Pressurized H_2 at refueling station from fossil fuels	$12-15/MM BTU	$14.26
Renewable-based H_2 production	$10-15/MM BTU	$14.41
Electrolyzer cost	< $300./kW	$275.
Electrolyzer efficiency	> 92%	95%

Introduction and Background

In the Fiscal Year 2002 Annual Operating Plan for the Hydrogen Program,[1] various cost and performance goals were cited: pressurized hydrogen from fossil fuels for vehicles delivered at the refueling station for $12-15/MMBtu, hydrogen from renewable fuels for $10-15/MMBtu, water electrolyzer systems having 92% efficiency for under $300. per kW, and reversible (fuel cell-electrolyzer) systems having round trip efficiencies of 70% and costs under $600. per kW.

Phase I of this program, completed in September 2000, included detailed economic and engineering studies for several alternative grid-independent, residential scale, electric power systems[2]. System components studied included engine-generators, storage battery banks, wind turbine-generators, TMI fuel cell systems, and TMI reversible fuel cell/energy storage systems. The suitability of the technology for 'peak shaving' was also examined. The concept is to augment high priced electricity during peak useage periods with lower priced "stored" power produced at off-peak rates. The method is to use reversible fuel cells to convert and store electricity in reactant form until later in time when it can be reconverted back to electricity. The reversible fuel cell systems had projected round trip efficiencies of 73% (thus exceeding the above target). Preliminary laboratory tests on reversible single cells were also performed.

TMI's Phase II modified statement of work sought to expand on the understandings of reversible solid oxide systems and explore a variety of materials and engineering options.[3] TMI's reversible solid oxide fuel cells have been tested primarily between 800° and 1000°C, with recent testing between 850° and 925°C. TMI has repeatedly demonstrated (since 1994) the capability of the solid oxide fuel cell to operate in electrolysis mode, with typical polarization voltages equal to or less than fuel cell polarization voltages at the same currents. In fuel cell mode, cells and stacks have been repeatedly demonstrated to be capable of operation on either hydrogen/steam fuel mixtures or on reformed hydrocarbon mixtures.

Cell Geometry

Figure 1 shows how passive cells operate in fuel cell and electrolysis modes. The hollow cylindrical cells have outside diameters of 56 mm. In fuel cell mode (a), the fuel reactant is delivered through a central fuel plenum and diffuses radially outward. Spent fuel (CO_2 / H_2O) diffuses inward to the fuel plenum and is eventually released as exhaust. At the perimeter, air oxygen diffuses inward until it is transported electrochemically across the ceramic electrolyte. Nitrogen acts as an inert diluent. In electrolysis mode (b), water vapor diffuses from the fuel plenum outward toward the rim. Oxygen is removed via high temperature electrolysis. The stripped hydrogen then diffuses back toward the fuel manifold. The diffusion processes are driven by chemical concentration and not by hydrodynamic pressure gradients.

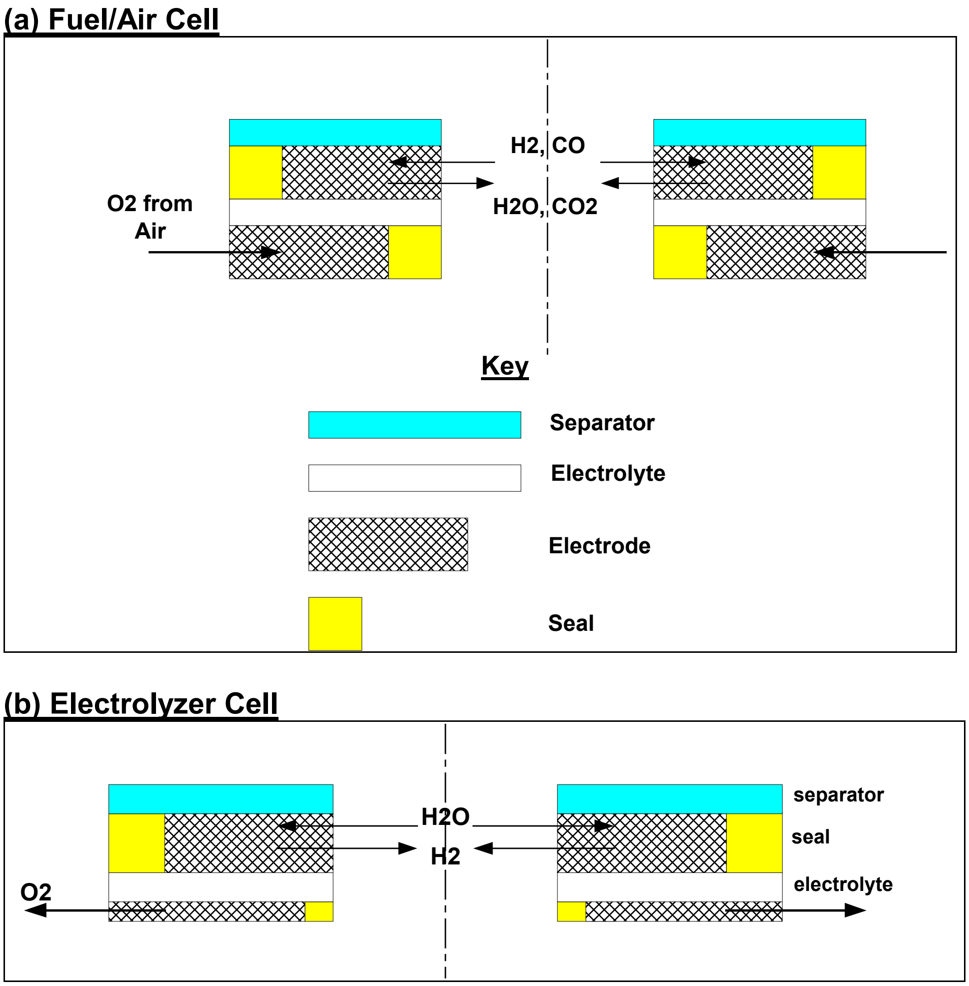

FIGURE 1. TMI PASSIVE CELL SCHEMATICS (radial cross sections not to scale, showing diffusion species)

Heating Values and Efficiency Definitions

Lower heating value (LHV) is the enthalpy change of combustion of a fuel at a reference temperature (usually 60°F) with H_2O remaining as vapor. Higher heating value (HHV) is the enthalpy change with water condensed to liquid. Natural gas is treated as a methane-rich mixture with a variable composition. Heating values may be converted to volts by dividing heating value in J/mole by Faraday's constant (96,485 C/mole) multiplied by electrochemical equivalents. The heating values for hydrogen equals the enthalpy change for decomposition of H_2O.

The efficiency of a fuel cell system is defined as (electric power output)/LHV. Energy efficiency is (electric power output + useful thermal output)/LHV. The efficiency of an electrolyzer is (hydrogen HHV)/electric power input. The "round trip" or reversible efficiency is considered as (energy output)/(energy input). Several different efficiencies are of interest for systems that produce hydrogen from fossil fuels such as natural gas. The ratio of moles of hydrogen delivered per mole of natural gas may be combined with values from Table 1 to give various values. The ratio of hydrogen HHV or LHV to natural gas LHV are relevant. However, since fuel cells operate on electrochemical equivalents, the output/input ratio of these (a smaller value than the heating value ratios) is arguably the most relevant efficiency for these systems.

Experimental Development Work

Cell Component Development

The goal of the cell testing effort was to validate the passive mode of operation and begin characterizing performance, particularly on temperature and gas phase concentration gradients, because the processes are driven by gaseous diffusion.

Evolutionary improvements and modifications were made to existing TMI materials specifications, fabrication methods, and cell dimensions to accommodate the new design. For instance, passive cell testing required thicker electrodes and seals than standard TMI cells and larger diameter seals. Thicker oxygen electrodes were fabricated by laminating multiple layers of standard TMI cathode stock. Minor modifications of standard TMI fabrication procedures were sufficient to produce thick fuel electrodes. Larger diameter, thicker seals were made using methods from internally funded programs. A reformulated ink for the fuel electrode/metallic separator interface was inserted into this program after performance was verified. Fifty-percent thinner electrolytes were procured and used in some of the trials.

Test Stands

Two highly versatile "three-quadrant" test circuits were designed, built, calibrated, and installed capable of testing single cells and stacks up to 12 cells. Table 2 shows the verified modes of operation. The circuits employ active current regulators with power MOSFETs. Several improvements were made to the humidifier assemblies on all five stands to permit more accurate control of composition of hydrogen/steam mixtures.

Table 2. Three Quadrant Test Circuit

Quadrant	Function	Voltage	Current
1	Normal Fuel Cell	Positive	Positive
2	Electrolysis	Positive	Negative
4	Forced Fuel Cell	Negative	Positive

Cell and Stack Development

This task was a major part of the work during Phase 2. The program included the fabrication, testing, and analysis of conventional forced-flow cells as well as passive cells and stacks. All measurements were made while operating on hydrogen-steam mixtures plus air. Operating conditions were fuel cell, electrolysis, alternating fuel cell/electrolysis, and forced fuel cell modes.

Figure 2 shows data from cells fed with 65% hydrogen/35% steam at two different temperatures. Operation was primarily in electrolysis mode with daily intervals in fuel cell mode. The choice of composition was made to correspond to the approximate midpoint in oxygen potential in a potentially complete system. Characterization at extreme conditions was not considered during this phase.

The lack of any offsets or slope changes near zero current shows that no significant activation polarization exists in these cells (in marked contrast to PEM cells). The plot also shows that practical electrolysis current densities (100-200 mA/cm^2) required electrolysis voltages under

1.1 Volts --- far lower than PEM electrolyzers (which typically operate near 1.9 Volts) -- thus enabling higher electrolysis efficiencies when using TMI solid oxide cells.

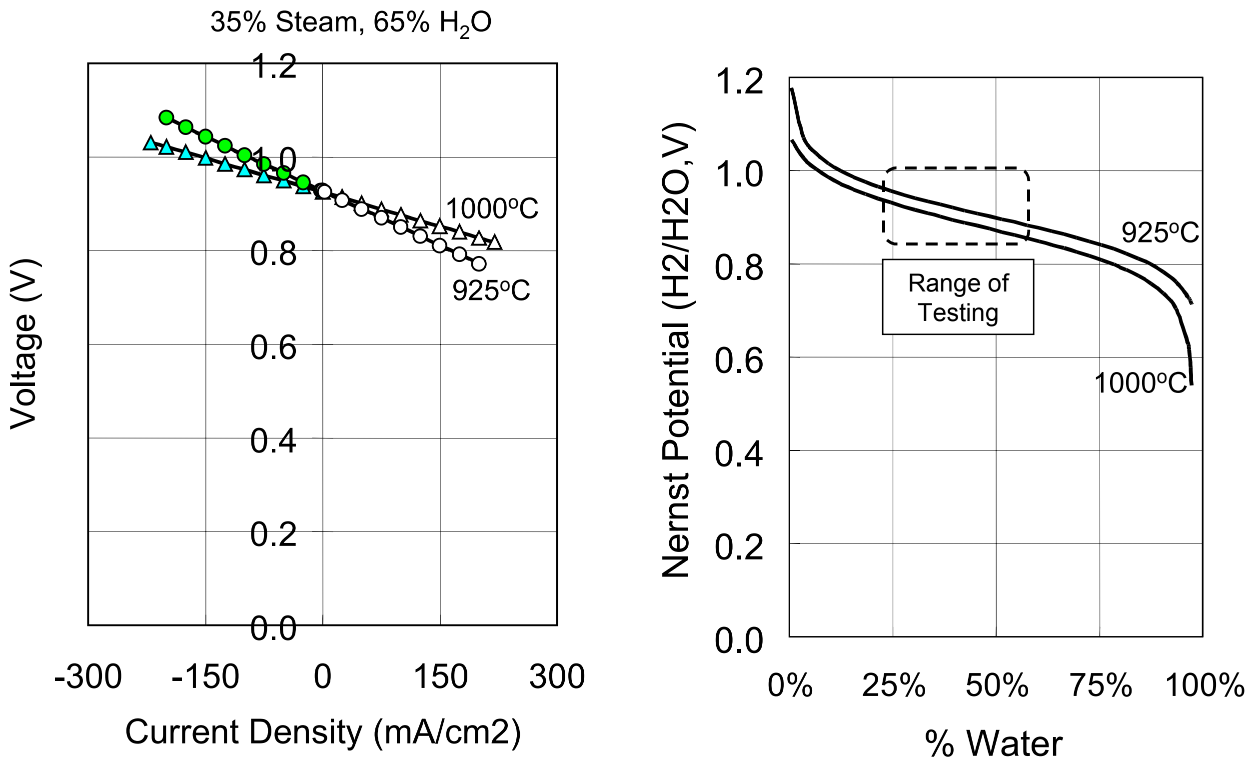

FIGURE 2. REVERSIBLE CELL VOLTAGE VS CURRENT DENSITY

Notwithstanding considerable variation among tests, trends were consistent for similar materials and construction. The most likely source of variability was fabrication quality and processing parameters. Because of the short program duration, only a few sets of fabrication conditions were explored. Finding an optimal set of fabrication conditions is essential for reproducible results.

The ratio of fuel cell to electrolysis voltage at the same cell current and hydrogen/steam feed composition is sometimes called reversible cell efficiency. It is a measure of the maximum possible energy storage efficiency for a reversible system using the cell (system efficiencies will be lower than cell efficiencies). The highest reversible cell efficiency observed was 90.8% (at 925°C and 50 mA/cm^2).

Development using Passive Cells and Stacks
Testing of passive cells and stacks was performed using cells and stacks with large seals to isolate the fuel and air circuits from each other. The majority of the tests were conducted in fuel cell mode, since experimental results indicated that ASR requirements are similar (figure 2 above) in either case and tests in fuel cell mode were more easily completed.

Both single cells and 5-cell stacks were tested, with encouraging results. The best of the trials exhibited ASR values comparable to TMI forced flow cells. Notably, the best tests showed a rate of performance decline with time comparable to current TMI forced flow cells. One of the 5-

cell stacks has operated for over 1200 hours with measured open-circuit voltages approaching theoretical EMF values for the particular hydrogen/steam ratio and cell temperature. Figure 3 shows data measured from the five-cell stack currently operating on 35% H_2O/65% H_2 at 925°C and 50 mA/cm^2. The figure also includes a one-cell stack running cyclically at the same conditions.

Leak testing of selected cells during operation detected minor seal leakages; however, the low differential gas pressure across seals led to minimal adverse effects. Differential shrinkage of seals versus electrodes is currently believed to be responsible for increased ASR. Future development work on seals is expected to produce improvements in both areas.

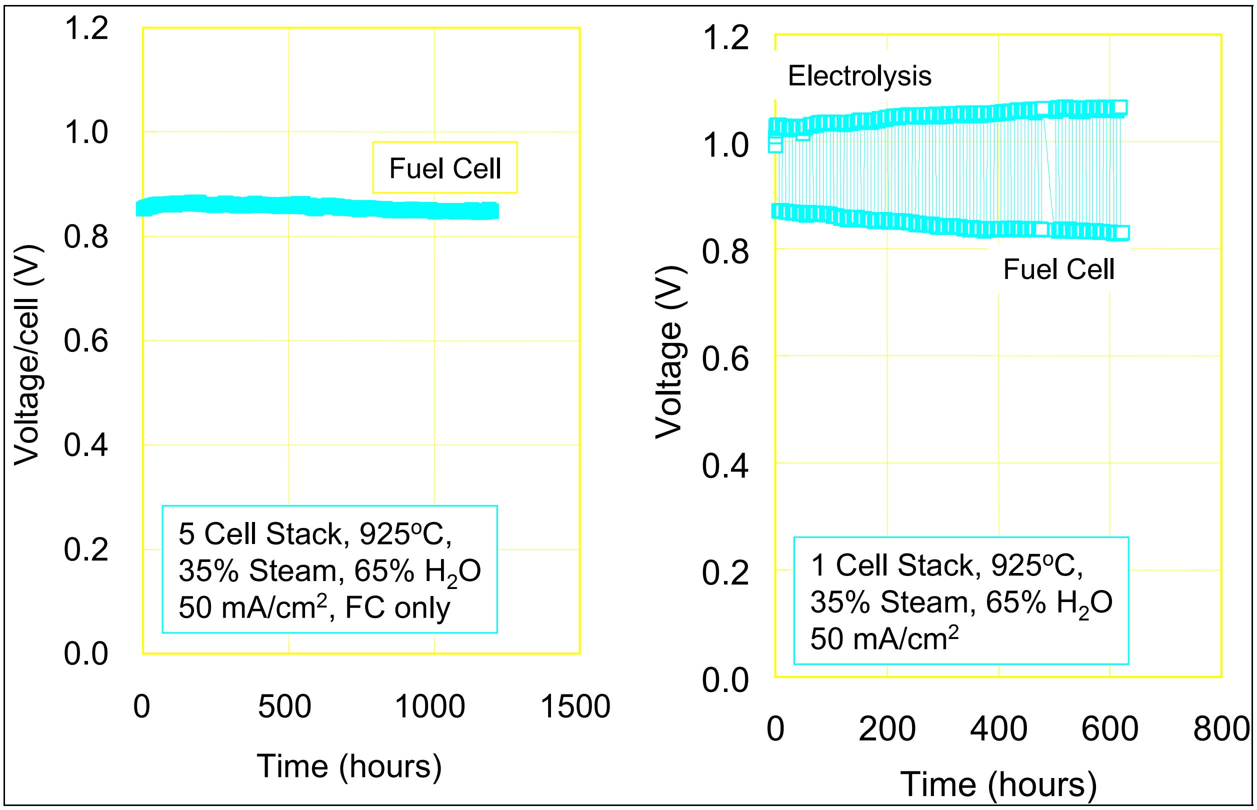

FIGURE 3. LONG TERM OPERATION IN FUEL CELL AND REVERSIBLE MODE.

Engineering Studies

A residential scale, grid independent application, using fuel cell and reversible fuel cell systems, was studied during Phase I. High-pressure, pure hydrogen and batteries were assumed as the energy storage mechanism. The small scale and low capacity factors for this contributed to the relatively high unit installed costs and a high cost of electricity. The reversible system projected round trip energy storage efficiency of 73% exceeds the 70% goal of the Hydrogen Program.

The study also considered alternative approaches that de-emphasize the scale limitations in the residential application. The low capacity factor was a major challenge therefore alternate concepts were considered that included options to improve capacity factor and considered producing hydrogen as a commodity useful for other applications.

Alternate Reversible System Concept

During Phase II, a modular system design concept was studied. The block diagram in Figure 4 shows two thermally integrated devices operating in steady state. The first is a passive solid-oxide fuel cell system operating on natural gas producing heat and electricity. The second is a solid-state electrolyzer that consumes electricity and waste heat from the SOFC system while producing hydrogen from pure steam. Supplemental input electric power (when available) could come from wind, photovoltaic, or other sources (e.g., inexpensive grid power). Natural gas (or propane) fuel would be used as needed. Ambient air and water from an automatic purifier would comprise the remaining inputs.

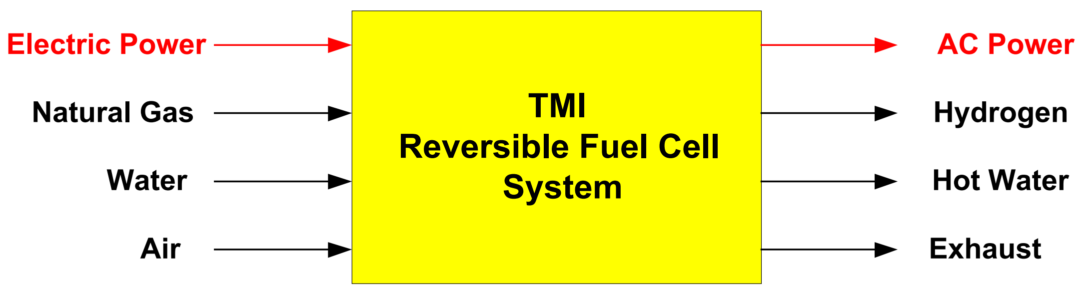

FIGURE 4. SYSTEM BLOCK DIAGRAM

It is assumed the AC power is being produced for local needs, with possible sale of surplus power to the grid. It is assumed high pressure (e.g., 5000 psi) pure hydrogen is being produced for later use in vehicles after temporary storage in adjacent tanks. Hot water would be produced as a useful byproduct when operating from natural gas fuel. The exhaust would be extremely clean and CO_2 emissions would be minimized during operation due to the high efficiency of the system.

Complete systems assembled from two or more identical modules would have the added benefits of redundancy and ease of service. In multiples, overall system capacities could range from an individual residence to large vehicle filling stations or “power parks”. Energy storage is supplanted by producing hydrogen at steady state for later use in vehicles instead, thereby eliminating “round trips” and increasing overall efficiency.

The technology has the potential for serving “premium power” requirements, defined as a power source having very high availability and power quality. For example, a system with 99.999% availability (“five nines”) will average only five (5) minutes per year power outage. High power quality includes good voltage and frequency regulation and low harmonic content (electrical noise). Availability can be tailored by varying the number of redundant modules and optimizing installation details.

Individual module sizes are being considered from 1 to 30 kW. For each kilowatt of capacity, a module could produce up to 1.0 AC kilowatt or up to 12.1 scfh of hydrogen (enough to power expected fuel cell cars for 21,200 miles per year) or a proportion of both. The fuel cell subsystem would operate at atmospheric pressure and the electrolysis subsystem would operate up to 5000 psi. The battery pack would provide high power when needed for load following and surges, but requires only a small energy storage capacity. Mechanical auxiliary devices include a high-pressure water pump and compact heat exchangers.

For some applications, an electrolyzer-only module could be offered, operating on DC input power plus water and delivering high pressure pure hydrogen. Such a module would have much lower installed cost than the above but the same DC to hydrogen efficiency given in Table 4. Both the fuel cell and electrolyzer subsystems would use TMI's passive solid oxide stacks (patent pending), which are capable of achieving very high efficiencies in both fuel cell and electrolysis modes.

Projected Performance

Efficiencies cited in the Table 3 use the lower heating value (LHV) of natural gas as customary for reporting fuel cell efficiencies. Hydrogen production efficiencies use the standard enthalpy of formation of liquid water, equal to the higher heating value (HHV) of hydrogen. The energy efficiency value includes both electrical and thermal outputs (thermal outputs produced only when natural gas is being used). The values cited are current estimates of mature system efficiencies.

Table 3. Projected System Efficiencies

Operating Mode	Electrical Efficiency at 60% Output	Electrical Efficiency at 100% Output	Energy Efficiency
Renewable DC to AC	96%	96%	
Natural Gas to AC	72%	65%	97%
Renewable DC to Hydrogen	95%	95%	
Natural Gas to Hydrogen	83%	75%	97%

The natural gas to AC efficiency is higher than assumed in Phase I due to updated fuel cell subsystem design concepts and experimental data. The Renewable DC to Hydrogen (electrolyzer only) efficiency of 95% exceeds the Hydrogen Program goal of 92%. The Natural Gas to Hydrogen mode employs both fuel cell and electrolyzer subsystems. The proposed modules would be capable of mixed mode operation for both inputs and outputs, i.e. both natural gas and electric power may be input in varying proportions with simultaneous outputs consisting of AC power, hydrogen, and hot water. A computer based controller would dynamically adjust operation based upon operating sensors, hydrogen inventory level, and user preferences.

System efficiencies will vary with module size, with smaller modules being slightly lower than the Table 3 values. Tradeoffs will also occur between efficiency and capital cost, which will be analyzed further once more data is available. As energy costs rise, higher efficiencies are more easily cost justified.

Cost Calculations

Unit costs for producing electric power and hydrogen were calculated using a greenfield build-up assuming the following factors:

- Installed equipment cost
- Life cycle maintenance cost
- Average efficiencies
- Natural gas cost
- Input electric power cost
- Grid power buy/sell prices (if connected)
- Inputs and outputs versus time
- Accounting assumptions

These are discussed in the following subsections.

Installed Capital Cost

There is a high level of inconsistency regarding capital costs of fuel cell systems in published fuel cell literature. This is due to either ambiguous terminology or to oversights. TMI uses the following cost buildup:

Manufacturing Cost
+ Gross Margin
Factory Selling Price
+ Distribution Cost
User Equipment Cost
+ Installation Cost
Installed Cost

Gross margin, distribution cost, and installation cost all include applicable overhead and profit. Distribution costs include shipping. All costs include applicable sales taxes.

TMI has interpreted the cost targets quoted by the DOE Hydrogen and SECA programs to mean Manufacturing Costs. These targets are $300. per kW for electrolyzers from the Hydrogen Program and $400. per kW for natural gas 3 to 10 kW fuel cell systems (having 40% LHV efficiency) from SECA. Module and accessory manufacturing costs calculated by TMI will depend upon the scale of annual production (of these and related products) and module size, declining with increases in either. This report assumes a module size of 10 kW nominal (capable of producing enough hydrogen to fuel about fifteen typical fuel cell cars) and a manufacturing volume of all products of 1000 MW/year.

TMI has internally projected manufacturing costs for 3 kW natural gas modules having AC output and up to 50% LHV efficiency at 1000 MW/yr to be $354. per kW. The added costs to achieve higher efficiency is expected to offset the unit cost savings from larger module size. The preliminary manufacturing cost estimate for a 10 kW natural gas fuel cell module with 70% LHV efficiency (AC output basis) at 1000 MW/yr is $455. per kW.

At the same annual production volume (of all products), the projected manufacturing cost of a complete stand alone 10 kW water electrolyzer module (with water pump, controls, etc.) operating at 95% efficiency producing 5000 psi pure hydrogen is approximately $265. per kW. Such electrolyzers would meet the Hydrogen Program targets for both cost (< $300.) and efficiency (> 92%). The lower cost compared with a fuel cell module reflects an expected smaller cell count and balance of system required for electrolysis which more than compensates for the cost of the water pump and pressure vessel.

Life Cycle Maintenance Cost

Maintenance requirements assume scheduled replacement of electrochemical stacks and wear parts on rotating equipment. Over time, the mean time between major service is expected to increase while the cost for replacement parts is expected to decrease. A small percentage of modules will suffer breakdowns (leading to automatic shutdown of that module but not affecting the others). Both scheduled and remedial maintenance are accomplished by field exchange (”hot swap”) of a replacement module and returning the removed unit to the factory for refurbishment or repair. The estimated average annual maintenance costs (including labor) for these modules are $85. per kW per year.

Natural Gas Cost

An example commercial natural gas cost of $5.00 per mcf was used, with 926 BTU/scf lower heating value. Actual natural gas costs vary widely within the U.S. and with time.

Input Electric Power Cost

DC power from on-site renewable generation (wind, photovoltaic, or small hydroelectric) can vary widely in cost depending on accounting assumptions. The total cost per kWh will depend on installed equipment cost, annual capital charge percentages, maintenance costs, tax credits (if any), and annual capacity factor (ratio of average annual power to rated power: typically a low percentage for renewable systems). Where equipment cost is treated as “sunk”, the annual capital charge percentage is zero, leading to very low power cost. In some cases, low cost off-peak power may be available from a utility. The example below uses an input power cost of 2.0 cents per kWh, which could equal the maintenance cost of a renewable system.

Grid Power Buy/Sell Prices

Utility cooperation and buy/sell prices for distributed generation sites vary widely with individual utilities. It is assumed many users will choose to operate independent of the grid to minimize installed costs, maximize safety, and avoid grid disturbances from compromising premium power quality. In some cases, utility purchase prices are so low they would not cover the incremental cost of natural gas fuel to generate export power. Where net metering provisions exist, monthly or annual electric energy imports and exports are offset against each other, with the difference being billed or paid. Conversely, a utility interconnection would permit a smaller capacity to be installed, provide enhanced peak power, and produce incremental revenues where utility buy prices are favorable. The extremely high 65-72% natural gas to AC efficiencies of the systems leads to low marginal generation costs, especially when fuel costs are moderate.

Inputs

Systems are expected to be capable of operation on natural gas (or propane) and electric power inputs in any proportions. Efficiency from propane is the same as natural gas on an LHV basis. The calculations below were performed for 100% natural gas and 100% electric power. When both are used, costs are in proportion.

Outputs

The three economically valued outputs are AC power, hydrogen, and hot water. Hot water is produced by the system only when natural gas or propane is being used. AC power and hydrogen may be simultaneously produced in any desired proportion.

The average annual load factors (percentage of maximum capacity actually used) depend greatly upon the nature of the business (or residence) where the equipment is used and whether grid connection is used. In the example below, 99% primary output load factor is used:

this is based upon the average availability of each individual module and the use of grid connection to permit full capacity operation.

The credit for hot water usage is calculated using avoided fuel cost.

Accounting Assumptions

Accounting assumptions include the annual capital charge rate (percentage of installed cost), property taxes and insurance, other taxes and credits not otherwise included, and other allocated costs. The annual capital charge rate often reflects the expected useful life of the equipment (design life will be 20 years), forecast net salvage value, and sometimes financing costs.

Actual prices for fuel and utility power are assumed to vary (sometimes markedly) with time. Annual maintenance costs, taxes, insurance, and allocated costs may also be time dependent. General inflation as well as evolutionary technology advances could play a role in some of these costs. It is customary to speak of "levelized" cost of electricity, meaning the cost calculated using predicted 20-year averages for each quantity. The examples below all calculate levelized costs, with sensitivities of selected parameters. An annual capital charge rate of 7% of installed cost per year is used (based on 20 year life, no salvage value, straight line depreciation, plus property taxes and insurance).

Cost Calculation Examples

The example in Table 4 shows very attractive costs for hydrogen (as low as 2.0 cents per mile total and 1.0 cents variable) and for AC power (4.5 cents per kWh total and 2.2 cents variable cost). The cost of hydrogen from renewable power is strongly dependent upon the cost of this power: the fixed costs of hydrogen production from renewables are only 1.0 cents per mile. The Table 4 example costs compare with the DOE Hydrogen Program goals[1] as follows:

Table 4. Costs and Efficiency Comparisons

Parameter	DOE Goal	TMI Projection*
Pressurized H_2 at refueling station from fossil fuels	$12-15/MM BTU	$14.26
Renewable-based H_2 production	$10-15/MM BTU	$14.41
Electrolyzer cost	< $300./kW	$265.
Electrolyzer efficiency	> 92%	95%

*using $5.00/mcf natural gas cost, 2 cents/kWh renewable power cost and other assumptions

The exact balance of grid input and output power as used in the above table may be impractical in many applications. Where utility attitudes and power buying prices are favorable, net power sales to the utility will generate revenues and hence lower effective costs for hydrogen and/or power production.

Since each cost component in the table above varies in the expected manner with the assumptions used, sensitivity effects may be obtained using different proportions. For example, both fuel cost and thermal credit vary directly with natural gas price. Annual capital cost is proportional to annual capital charges rate and installed cost. Figures 6-8 show the effect of input cost upon AC power and hydrogen cost.

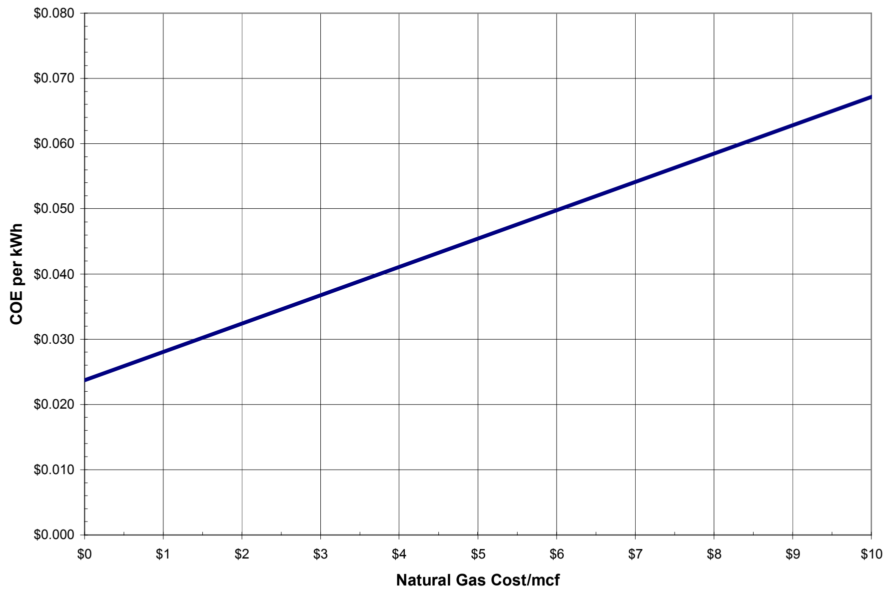

FIGURE 6. AC POWER COST VS NATURAL GAS COST

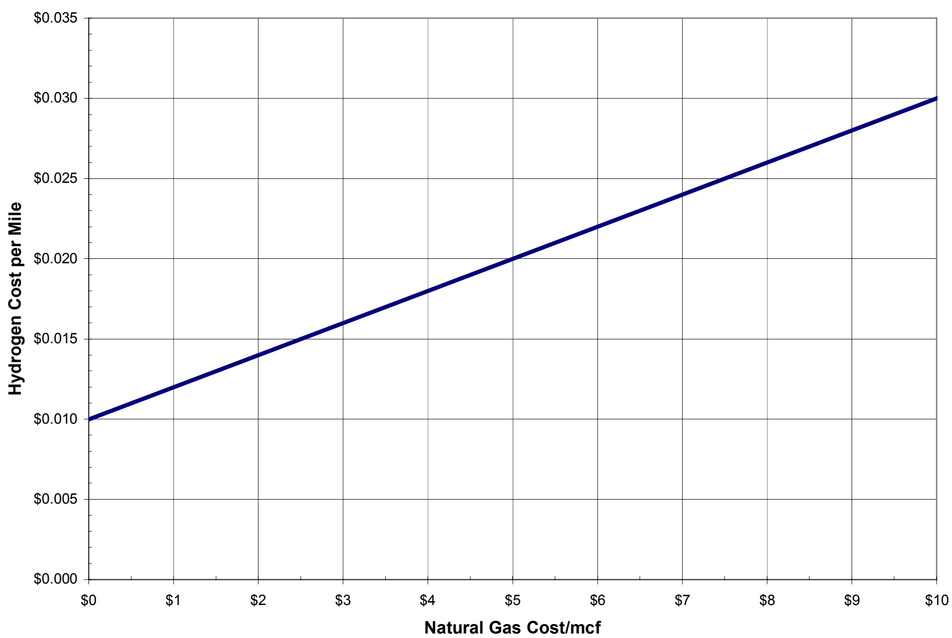

FIGURE 7. HYDROGEN COST VS NATURAL GAS COST

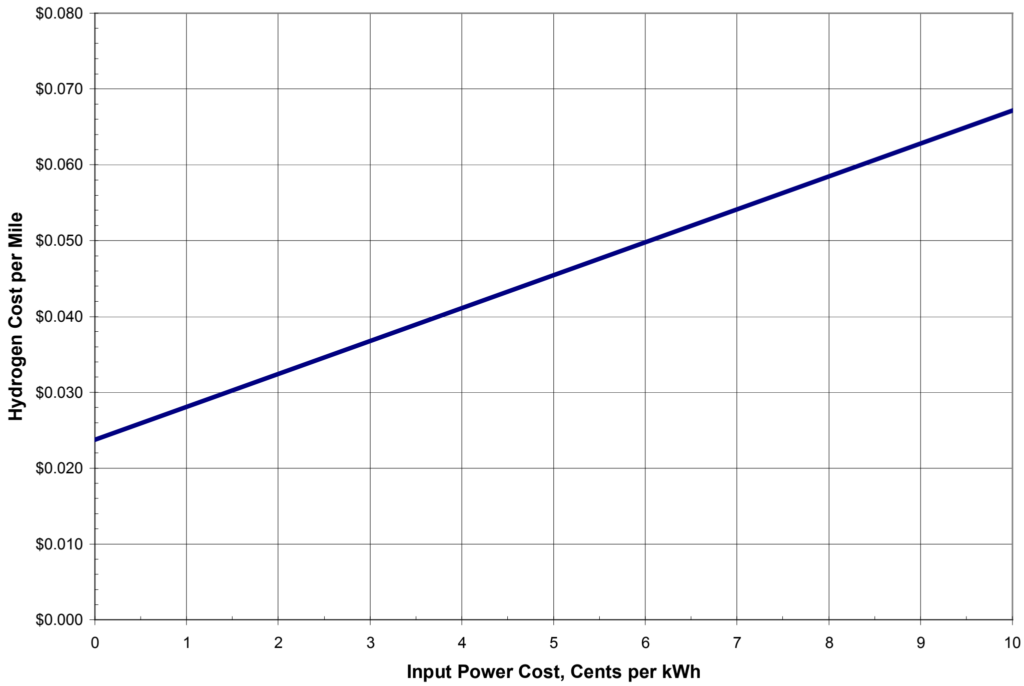

FIGURE 8. HYDROGEN COST VS INPUT POWER COST

Conclusions and Recommendations

The program has met or exceeded all technical objectives (as revised by DOE Hydrogen Program management) on budget and on time. Experimental results have been promising. The economic / engineering studies indicate the potential for reversible systems to set new standards of performance, achieving lower cost of H_2 production, lower pollution levels, and potentially serving as an enabling technology for hydrogen fuel cells.

Future work should build upon the results from this phase and strive toward a larger scale technology demonstration. At the stack level, technical goals include achieving: 1.) negligible gas leakage rates, 2.) reduced polarization voltages, 3.) improved microstructures, and 4.) reproducible fabrication techniques. While low gas leakage rates have been demonstrated, additional work is required to improve reproducibility. Reduced polarization voltages will require optimizing electrode microstructure and continuing to improve the interface with the separator. Improved microstructure can be addressed through materials and processing improvements. Reproducible fabrication can be achieved through a combination of enhanced inspection techniques, data analysis, and mechanized production methods.

At the system level, a demonstration reactor of sufficient size to demonstrate technology proof-of-concept should be designed, built, and operated. The design of the hot subassembly will be based upon engineering designs developed by TMI over the past 10 years and will include the understandings from the work presented above and new learning from advanced stack development.

Acknowledgement

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