THE REVOLUTION THROUGH EVOLUTION: DELPHI SOLID OXIDE FUEL CELL FOR APU AND HYDROGEN REFORMATION

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ABSTRACT

For people working on Powertrain technologies and energy technologies, this is one of the most exciting times but also one of the most uncertain. What will be the new Powertrain in the coming years? What will be the new fuel as an energy source for transportation? What will be the next energy storage and generation for the future? How will we bring the car out of the environmental equation? At Delphi, we have tried to prepare that revolution through evolution and herein we introduce two key technologies that address these concerns: introduction of hydrogen in discrete quantity in order to improve engine combustion and emissions through hydrogen on-board reformation, and a fuel cell Auxiliary Power Unit (APU) to supply power to engine-independent accessories with high efficiency and very low emissions.

Our design selection criteria drove us to the following realizations: the Delphi hydrogen catalytic micro-reformer, and the Delphi solid oxide fuel cell (SOFC) APU. We discuss in detail these technologies in this paper.

SOFC-BASED AUXILIARY POWER UNIT

A fuel cell based Auxiliary Power Unit is a high efficiency generator that is used for supplying power to the electrical accessories in the vehicle (Figure 1) (1-4). The SOFC APU has the potential to be a paradigm shift in the supply of electric power in transportation. This fuel cell applied to the electrical system has two main advantages relative to a conventional generator:

- It can be operated independently of the engine. This opens up completely new possibilities for additional functions, even when the engine is off, such as auxiliary air conditioning or the "mobile office."

- The power is supplied with higher total efficiency than the conventional method of "engine, generator, battery." Compared to the low efficiency of an engine-powered generator (10 – 22%), the efficiency of the APU is 35-50% (Fig. 1).

The application of the fuel cell Auxiliary Power Unit (APU) for transportation comes in the context of a significant trend to electrification of vehicle accessories. Many
automotive systems are being converted to electric power to support cost, weight, and packaging objectives. Intermittent accessories, such as heated seats, heated windshield, power steering and brakes have been adopted first.

SOFC is extremely attractive for use as an APU. PEM fuel cells may also be used for application in an APU, but require a complex reforming process for conventional fuels and need on-board water management. Some of the advantages of using SOFCs are:

- Ability to use CO along with H₂ as fuel. Furthermore, the SOFC demonstrates a relatively higher tolerance to fuel impurities such as sulfur.
- Relatively simple reformer technology and compatibility with hydrocarbon fuels.
- Compatibility of temperature between reformer and stack.
- Temperature and water conditions permit internal reforming at the anode of SOFC.
- No noble metal catalysts in the stack.
- Water management is not required.

![Figure 1. Mechanization for a fuel cell based APU.](image)

**FUEL CELL FUNDAMENTALS**

Two types of fuel cells that have received widespread interest in the automotive industry are: proton exchange membrane (PEM) fuel cell and solid oxide fuel cell (SOFC). While the focus of PEM fuel cell development has been for applications in vehicle propulsion, the SOFC is being developed primarily as an Auxiliary Power Unit (APU).

The solid oxide fuel cell is a solid-state energy conversion device that produces electricity by electrochemically combining fuel and oxidant gases across an ion conducting ceramic membrane (Figure 2). As with all fuel cells, SOFC consists of two electrodes (anode and cathode), separated by an electrolyte.
The electrolyte in this fuel cell is a solid non-porous metal oxide, usually Y$_2$O$_3$-stabilized ZrO$_2$. The cell operates at 650°C to 1000°C. Typically, the anode is a Ni-ZrO$_2$ cermet, and the cathode is Sr-doped LaMnO$_3$ or LaFeO$_3$.

The electrochemical reactions (Figure 2) occurring in SOFC utilizing H$_2$ and O$_2$ are based on the following equations:

\[
\begin{align*}
\text{Anode} & : \quad \text{H}_2 + \frac{1}{2} \text{O}_2 + \text{2}e^- \rightarrow \text{H}_2\text{O} + \text{2e}^- \\
\text{Cathode} & : \quad \frac{1}{2} \text{O}_2 + \text{2e}^- \rightarrow \text{O}^{2-}
\end{align*}
\]

The overall cell reaction is:

\[
\text{H}_2 + \frac{1}{2} \text{O}_2 \rightarrow \text{H}_2\text{O}
\]

In SOFC, carbon monoxide and light hydrocarbons such as methane can also be used as fuels.

A PEM fuel cell, on the other hand (Figure 2), operates at low temperatures (80°C) and uses an ion exchange membrane like fluorinated sulfonic acid polymers as the proton (H$^+$) conducting electrolyte. Water management in the membrane is critical for efficient operation.
performance so as to keep the membrane hydrated at all times. The low temperature
operation of a PEM fuel cell requires precious metal catalysts like Pt for both anode and
cathode. Carbon monoxide (CO) acts as a poison at these temperatures and so unlike
SOFC, PEM fuel cells can only use pure H2 as fuel. The primary electrochemical
reactions at the electrodes for a PEM fuel cell are:

Anode \[ \text{H}_2 \rightarrow 2\text{H}^+ + 2\text{e}^- \] (4)

Cathode \[ \frac{1}{2}\text{O}_2 + 2\text{H}^+ + 2\text{e}^- \rightarrow \text{H}_2\text{O} \] (5)

Performance of Fuel Cells: Ideal Voltage and Efficiency

The Nernst equation shown below provides a relationship between the ideal standard
potential \( E^0 \) for the cell reaction and the ideal equilibrium potential \( E \) at other
temperatures and partial pressures of reactants and products in a fuel cell. For equation
(3), the Nernst equation is:

\[
E = E^0 + \frac{RT}{2F} \ln \left[ \frac{P_{\text{H}_2}/P_{\text{H}_2\text{O}}}{P_{\text{O}_2}^{\frac{1}{2}}} \right] \tag{6}
\]

where, \( E^0 = \) standard potential, \( E = \) equilibrium potential, \( P = \) gas pressure, \( T = \) temp, \( F = \) Faraday’s constant, \( R = \) universal gas constant.

The efficiency of a fuel cell is a function of the thermodynamic efficiency, the voltage
efficiency and the fuel utilization (5, 6):

\[
\eta_{\text{FC}} = \eta_T \eta_V U_f \tag{7}
\]

**Thermodynamic Efficiency** is the intrinsic maximum energy that can be converted to
electrical energy in a fuel cell. This is a function of the Gibb’s free energy change.

\[
\eta_T = \Delta G/\Delta H = 1 - T\Delta S/\Delta H \tag{8}
\]

where, \( \Delta G = \) Gibb’s free energy change; \( \Delta H = \) enthalpy change; \( \Delta S = \) entropy change

**Voltage Efficiency:** The voltage efficiency is defined as the ratio of the operating
voltage to the ideal voltage.

\[
\eta_V = \frac{V}{V^0} \tag{9}
\]

In an operating fuel cell, the cell voltage is always less than the ideal voltage as shown in
Figure 3 below. The reduction in cell voltage at operating conditions under current load
is a function of many different factors like temperature, pressure, gas composition and
gas flow.

The difference between the operating voltage and the ideal voltage is called polarization.
A typical fuel cell “voltage versus current” curve shows contributions from three main
polarizations. They are activation polarization, ohmic polarization and concentration
polarization.
The factors affecting the loss of voltage in different fuel cells are specific to the materials and the processes in the fuel cell. Both PEM and SOFC have demonstrated >1.2 W/cm² under ideal conditions and similar voltages.

**Fuel Utilization:** Fuel utilization is the amount of fuel that is being consumed for electrical output in comparison to the total fuel flow in the stack. This is a function of many different parameters within the stack like thermal distribution, flow distribution, cell design and even system mechanization.

Overall, the efficiency of different types of fuel cells depend on various intrinsic and design based factors. For all fuel cells, operating at higher voltages and increased fuel utilizations would overall increase the efficiency of the stack. However, it is extremely difficult to discriminate between fuel cells solely based on the efficiency of the stack alone without looking at the total system efficiency (7). As will be discussed later in the paper, it is the overall system efficiency that would allow for the correct choice of a fuel cell system for a particular application, especially if it is dependent on a certain fuel.

**REFORMER FUNDAMENTALS**

In this section, we discuss some of the fundamentals of reforming hydrocarbon fuels like gasoline as applied to internal combustion engines (ICE) and SOFCs.

The fundamental reactions that govern reforming with their respective enthalpies (ΔH) are: { For a fuel = C₇H₁₃ (hypothetical molecule); using ΔH at 298.15 K for C₇H₁₃ (estimated value of -80.7 kJ/mol), CO (-110.53), CO₂ (-393.51), H₂O(g) (-241.81)}

**Full Oxidation:**

\[
C_7H_{13} + 10.25 (O_2 + 4 N_2) \rightarrow 7 CO_2 + 6.5 H_2O + 41 N_2 \quad \Delta H_r = -4246 \text{ kJ/mole}
\]

**Partial Oxidation:**

\[
C_7H_{13} + 3.5 (O_2 + 4 N_2) \rightarrow 7 CO + 6.5 H_2 + 14 N_2 \quad \Delta H_r = -693 \text{ kJ/mole}
\]
Steam Reforming:
\[ C_7H_{13} + 7 H_2O \rightarrow 7 CO + 13.5 H_2 \quad \Delta H_r = 1000 \text{ kJ/mole} \]

Dry Reforming:
\[ C_7H_{13} + 7 CO_2 \rightarrow 14 CO + 6.5 H_2 \quad \Delta H_r = 1288 \text{ kJ/mole} \]

Autothermal Reforming:
\[ C_7H_{13} + 2.87 H_2O + 2.07 (O_2 + 4 N_2) \rightarrow 7 CO + 9.37 H_2 + 8.28 N_2 \quad \Delta H_r \approx 0 \]

These equations are all shown at the syngas ratio. In all real systems it is normal to have a surplus of oxidant in order to approach full conversion. Which reaction to employ depends on the fit between reaction attributes and application requirements. Table 1 is a general evaluation of the primary reforming reactions for the various oxidants in use versus their application impact.

For automotive applications related to using reformate in an engine as well in a SOFC APU, POx is the reformer of choice due to its fast start up, small reactor size and system simplicity. As Table 1 implies there are some trade-offs that must be made to gain these benefits. These are discussed below in detail for each of these automotive applications.

| Oxidant: | Air | H2O | CO2 |
|----------|-----|-----|-----|
| Reforming Name: | POx | Steam | Dry |
| Reaction enthalpy: | Exothermic | Endothermic | Endothermic |
| Efficiency (theoretical max; HC=2) | 84% | 124% | 130% |
| Sensitivity to Excess Oxidant | Loss in efficiency and temperature management required | None | None |
| Typical Reformer Out Temperature | 900 | 900 | 1000 |
| Kinetics (speed of reaction) | Fast | Slow | Slow |
| Reactor Size | Small | Large | Large |
| System Complexity | Low | Needs recycle or H2O source | Needs recycle |
| Start up time | Fast (makes its own heat) | Slow (requires heat) | Slow (requires heat) |
| Carbon Formation | Sensitive to mixture preparation. Operate at O/C lean of syngas to avoid carbon | Best oxidant for carbon minimization | Requires high temperature to avoid carbon |

Table 1. Attributes of primary reforming reactions.
Hydrogen/Reformate in SI Engine Combustion Fundamentals

Hydrogen (H\textsubscript{2}) has been widely used in engine research as a fuel for SI engines. It has notable advantages in efficiency and emissions – based on its combustion properties. Increasingly stringent emission control standards and interest in hydrogen as a non-petroleum fuel (with potential renewable sources) have led to recent interest in commercializing hydrogen engines (for example at BMW, Ford). In the same way that on-board fuel reforming is a bridge to hydrogen fuel for fuel cells, on-board reforming applied to internal combustion engines is a step towards using hydrogen in transportation.

A relatively simple one step reforming process can generate a hydrogen rich gas for use in engine combustion and emission control. Table 2 compares selected combustion related properties of H\textsubscript{2} and CO to those of gasoline. It can been seen that properties of CO are often between those of hydrogen and gasoline – so reformate, which contains roughly equal amounts of H\textsubscript{2} and CO, will retain attractive properties compared to gasoline. These properties may be viewed for their impact on engine efficiency and exhaust emissions and aftertreatment. First, let us consider the impact on engine efficiency. The high flame speed and low ignition energy of H\textsubscript{2} and reformate relative to gasoline, permit the use of very lean or high EGR operation. With lean operation, the engine can have high thermal efficiency because of an increase in the ratio of specific heats of the charge closer to that for air (k = 1.4). The higher flame speed of H\textsubscript{2} and reformate helps to maintain combustion as near to constant volume as possible even with lean mixtures. Pumping losses are lower with H\textsubscript{2} or reformate relative to gasoline due to the lower energy density of H\textsubscript{2} and CO (in addition to the high dilution tolerance). This contributes to better efficiency at part load. The high spontaneous ignition temperature of H\textsubscript{2} and CO (coupled with the high flame speed) permits the use of higher compression ratios to boost the thermal efficiency further (13). Second, let us consider the impact on emissions. Ultra lean or highly dilute combustion results in low peak cycle temperatures and very low NOx emissions. Lean operation can also result in low CO emissions. H\textsubscript{2} or reformate fueled engines have low and non-reactive HC emissions (related to light hydrocarbons that survive the reforming process and to lubricating oil). Furthermore, smoke (particulate matter) should also be extremely low. When blended with gasoline, H\textsubscript{2} and reformate can help reduce the HC emissions by decreasing the quench distance for better flame propagation in crevices. In terms of aftertreatment, H\textsubscript{2} and reformate are attractive for catalyst heating and lightoff, because they can be more easily and complete reacted on a catalyst. Thus, a rapid catalyst lightoff with minimal emission breakthrough should be possible with reformate added to the exhaust during startup.

Table 2. Comparative properties of gasoline, hydrogen and carbon monoxide.

| Property                        | Gasoline | Hydrogen | CO   |
|--------------------------------|----------|----------|------|
| Lean limit equivalence ratio in air | 0.58     | 0.1      | 0.34 |
| Flammability limits (volume %)   | 1.2-6    | 4-75     | 12-74|
| Laminar flame speed (m/s)        | 0.37-0.43| 1.9 – 2.7| -0.4 |
| Spontaneous ignition Temperature (°C) | 280-400  | 574      | 609  |
| Net energy density (MJ/m\textsuperscript{3}) | 202      | 10.3     | 12.6 |
| Quench distance (mm)             | ~ 2.0    | 0.64     | -    |
| Minimum ignition energy (mJ)     | 0.24     | 0.02     | -    |
| Lightoff Temperature (°C) over platinum based catalyst | ~ 270    | ~ 30     | ~ 220|

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Reformer-Assisted Internal Combustion Engine Operation

The above section has described fundamental differences in hydrogen-fueled combustion compared to gasoline. We have documented Delphi's reformer-assisted internal combustion engine work in a number of publications describing our hydrogen enrichment strategy for meeting SULEV emissions (11-17). The strategy uses POx reforming based on its ability for fast start-up, compact size and lower complexity compared to other reforming methods (see Table 1). Hydrogen-rich reformate has a number of attributes that make it an attractive fuel for very low emissions. It offers a fuel source that is very low in hydrocarbons. Reformate also promotes low temperature conventional exhaust catalyst lightoff, and it has very wide flammability limits. Our hydrogen enrichment strategy with an on-board reformer uses attributes of reformate to augment (not replace) a 3-way catalytic aftertreatment system by dramatically lowering cold start emissions and by avoiding the extreme precious metal loadings which would otherwise be required to meet SULEV. Briefly, the hydrogen enrichment cold start strategy consists of:

- fueling with 100% reformate during cold start for near-zero engine-out HC and NOx;
- reacting reformate and air in the exhaust during cold start for rapid exhaust catalyst lightoff.

Basic elements of the system are shown schematically in Figure 4. Figure 5 shows an early-generation hydrogen enrichment reformer system mounted under-hood in a vehicle. A recent paper (12) describes progress in fast reformer start-up, a key requirement for our hydrogen enrichment strategy. The current-generation reformer system starts in less than 5 seconds; our ultimate goal is a reformer that starts within one second.

Engine performance as measured by net mean effective pressure (NMEP) during start-up is shown in Figure 6. NMEP is a measure of net engine work output. Several engine cycles go by before the engine starts to fire for the reformate data. The initial delay in NMEP (comprising 5 engine cycles or roughly 460 msec for the 1300 Engine RPM test condition) for the reformate-fueled test is due to the transport time required for the reformate to reach the engine from the reformer. The engine starts up as soon as reformate enters the cylinder and NMEP was quite stable almost immediately after start-up. Overall, engine performance is similar between the gasoline and reformate-fueled tests.

![Figure 4. Schematic of the on-board reformer system.](image-url)
Figure 5. On-board reformer system mounted underhood in a test vehicle.

Figure 6. Engine NMEP comparisons for reformate vs. gasoline fueling under representative cold start conditions in single-cylinder engine dynamometer tests.

Engine-out HC and NOx emissions comparing start-ups with reformate and PFI gasoline discussed above are shown in Figures 7 and 8. With PFI gasoline the HC emissions rise quickly during start-up and reach a steady value of roughly 1500 ppm. By comparison, the reformate-fueled exhaust emissions are much lower. Over the first 20 seconds of engine operation, reformate fueling offers a 75% reduction in HC mass emissions and a 98% in NOx mass emissions compared to baseline gasoline fueling. Adding reformate to gasoline significantly reduces engine-out NOx emissions by increasing the dilution tolerance of the engine.
Figure 7. Cold Start Engine-out HC emissions for reformate vs. gasoline fuel.

Figure 8. Cold Start Engine-out NOx emissions for reformate vs. gasoline fuel.

Reforming for SOFC APU

An SOFC system lends itself to several inherent benefits for the POx reforming process. The fact that both H₂ and CO are fuels for the stack drastically reduces reformer system complexity by eliminating large and costly water gas shift and preferential oxidation catalysts found in PEM systems. For SOFCs, a POx system without the need for water can be the fundamental reforming method.

System Efficiency Implications for SOFC APU

Overall system efficiency (\(\eta_{\text{SYS}}\)) for an APU which uses hydrocarbon fuels is a function of the fuel cell stack overall efficiency (\(\eta_{\text{FC}}\)), fuel reforming efficiency (\(\eta_{\text{R}}\)), and parasitic losses (\(P_p\)). The relationship can be generally expressed as:
\[ \eta_{\text{SYS}} = \left( \eta_{\text{FC}} \ast \eta_{\text{R}} \ast P_{\text{FUEL}} - P_{\text{P}} \right) / P_{\text{FUEL}} \]

where \( P_{\text{FUEL}} \) is the energy content of the fuel used by the system. To increase system efficiency, the cell can be operated at a higher voltage, but this will decrease the operating power density of the cell. A typical high load operating point for the SOFC stack has an efficiency \( \eta_{\text{FC}} \) of 38\% (from equation 7, with \( \eta_{\text{T}} = 78\% \), \( \eta_{\text{V}} = 70\% \) and \( U_{\text{f}} = 70\% \)).

The next major component in the SOFC APU system efficiency is the reformer efficiency. For the purposes of this discussion, reformer efficiency is defined as the ratio of the lower heating value of the reformer outlet fuel (reformate) to system input hydrocarbon fuel: \( \eta_{\text{R}} = \text{LHV}_{\text{Reformate}} / \text{LHV}_{\text{Fuel In}} \).

An SOFC system with a practical POx reformer, having an efficiency of 77\%, would have a typical gross system efficiency of 29\% \( (\eta_{\text{FC}} \ast \eta_{\text{R}} = 0.38 \ast 0.77) \). However for an SOFC system a portion of the stack anode exhaust gas can be fed into the reformer inlet along with the incoming hydrocarbon fuel. This recycled anode exhaust gas contains H\(_2\)O, CO\(_2\), and unused H\(_2\) and CO. In this way H\(_2\)O and CO\(_2\) can be provided to the reformer for endothermic reforming without an external source of H\(_2\)O. In addition, the actual fuel utilization of the system is greater than the stack internal fuel utilization by recycling unused H\(_2\) and CO. A further benefit is that H\(_2\) and CO concentration in the reformate is increased, as compared to POx reforming (due to less N\(_2\) dilution), which increases the fuel cell power density. As a result, effective reformer efficiency in a system with high recycle can approach 150\%, which would raise gross system efficiency to 55\%! The benefits of stack anode exhaust gas recycle reduce the necessity for high stack fuel utilization for good overall system efficiency as illustrated in Figure 9.

![Figure 9. APU efficiency as a function of stack fuel utilization and anode recycle from system models.](image)

Parasitic power losses are the last factor, which impacts SOFC APU system efficiency. Overall system parasitic losses for a SOFC system can be less than 10\% of the stack power output – so net fuel to electric efficiency of nearly 50\% is possible.
Delphi is currently developing a 5 kW Solid Oxide Fuel Cell APU running on gasoline (1-4). A SOFC stack and an integrated (closely coupled) catalytic partial oxidation (POx) reformer and Energy Recovery Unit (ERU) has been designed allowing for a compact, low mass system. The APU is being tailored for increased efficiency by lowering parasitic losses, increasing fuel utilization and optimizing thermal losses. Development is in progress to validate the functioning of this system. Figure 10 shows a picture of the Delphi’s Generation 2 prototype system under development.

Figure 10. Generation 2 SOFC APU System under development.

INTEGRATION OF SOFC AND LEAN INTERNAL COMBUSTION ENGINES IN A COMBINED CYCLE

As has been discussed in this paper:

(1) the SOFC APU is a highly efficient method of generating direct current electric power on-board a vehicle – especially from conventional hydrocarbon fuels like gasoline, diesel or natural gas.

(2) A small, fast start-up reformer has significant leverage in reducing start-up emissions and improving low emission robustness of gasoline EMS.

But how do we integrate these strategies for maximum benefit?

Because of the high operating temperatures, SOFCs can use waste heat from the stack to drive an isothermal endothermic reforming process. The energy required to endothermically reform gasoline is about 27% of the lower heating value of the fuel.

For example, an endothermic reforming process could be used to operate the APU with the following energy flows (expressed in the percentage of lower heating value of the gasoline input):
Gasoline (100%) $\rightarrow$ Reformate (127%) $\rightarrow$ Electricity (48.5%)
      Anode Tailgas (38.1%)
      Waste Heat in Stack (40.4%)

The waste heat is surplus to the minimum required to drive the endothermic reforming reaction – so that, in fact, additional gasoline can be processed in the SOFC – yielding a larger by-product of reformate, as follows:

Gasoline (149%) $\rightarrow$ Electricity (48.5%)
          Anode Tailgas (100.9%)

The additional fuel results in lower fuel utilization, which will result in slightly higher voltage in the stack and higher system efficiency (but this effect is ignored here).

With efficient, endothermic reforming, substantial lean burn (at light load) and heavy EGR (at high load) dilute combustion modes are enabled by the SOFC – with a large increase in powertrain efficiency. In fact, in some cases the lean limit can be extended so much that NOx levels are reduced to a few ppm. This is SULEV emissions without NOx aftertreatment and with efficiency approaching diesel!

**FUTURE AUTOMOTIVE SOFC MECHANIZATIONS**

While it is likely that the first APU mechanizations will be low volume options on specific types of vehicles, the combined cycle approach leads to several attractive future mechanizations:

**SOFC - ICE Combined Cycle Systems**

The next generation automotive SOFC systems will involve integration of the fuel cell into the engine management system (EMS). A parallel innovation at Delphi is the development of fast-start-up reformers for use in emission control of gasoline and diesel engine systems (the reformer only mechanizations will not be discussed in this paper).

**Heavier hybrid and Series Hybrid FCEVs**

In these types of vehicles, the same combined cycle SOFC/ICE system will be attractive, but the electric propulsion component will increase and the relative size of the engine compared to the fuel cell will decrease. An 80 - 100% gain in efficiency will be possible compared to a conventional baseline (in low speed city driving). While many new low emission city bus models are being developed with natural gas, this approach will give gasoline and diesel fuel comparable high efficiency and super low emissions.

**Range Extender Electric Vehicles**

To make the EV practical, an APU would be a welcome addition to allow extended range and full vehicle utility. Of course this could be a small ICE generator, a microturbine, a PEM fuel cell or an SOFC. It could also run on liquid fuel or propane or hydrogen. But as in the first APU applications, Delphi believes that SOFC is uniquely attractive –
because of its high efficiency and low emissions, its compact size and weight and its compatibility with conventional fuels.

**Figure 11. SOFC / ICE combined cycle architecture.**

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