

A light hydrocarbon fuel processor producing high-purity hydrogen

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Abstract

This paper discusses the design process and presents performance data for a dual fuel (natural gas and LPG) fuel processor for PEM fuel cells delivering between 2 and 8 kW electric power in stationary applications. The fuel processor resulted from a series of design compromises made to address different design constraints. First, the product quality was selected; then, the unit operations needed to achieve that product quality were chosen from the pool of available technologies. Next, the specific equipment needed for each unit operation was selected. Finally, the unit operations were thermally integrated to achieve high thermal efficiency.

Early in the design process, it was decided that the fuel processor would deliver high-purity hydrogen. Hydrogen can be separated from other gases by pressure-driven processes based on either selective adsorption or permeation. The pressure requirement made steam reforming (SR) the preferred reforming technology because it does not require compression of combustion air; therefore, steam reforming is more efficient in a high-pressure fuel processor than alternative technologies like autothermal reforming (ATR) or partial oxidation (POX), where the combustion occurs at the pressure of the process stream. A low-temperature pre-reformer reactor is needed upstream of a steam reformer to suppress coke formation; yet, low temperatures facilitate the formation of metal sulfides that deactivate the catalyst. For this reason, a desulfurization unit is needed upstream of the pre-reformer.

Hydrogen separation was implemented using a palladium alloy membrane. Packed beds were chosen for the pre-reformer and reformer reactors primarily because of their low cost, relatively simple operation and low maintenance. Commercial, off-the-shelf balance of plant (BOP) components (pumps, valves, and heat exchangers) were used to integrate the unit operations. The fuel processor delivers up to 100 slm hydrogen >99.9% pure with <1 ppm CO, <3 ppm CO₂. The thermal efficiency is better than 67% operating at full load. This fuel processor has been integrated with a 5-kW fuel cell producing electricity and hot water.

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1. Introduction

A fuel cell is an electrochemical device where a fuel reacts with oxygen generating electricity and producing combustion by-products. Hydrogen gas is the preferred fuel for fuel cells because of its high reactivity on the fuel cell electrode and the low environmental impact of the reaction product, water. Unfortunately, there are no known sources of gaseous hydrogen on planet earth, and it is unlikely that any sizable reservoir of gaseous hydrogen will ever be found. The small size of the hydrogen molecule facilitates its diffusion through solids, and because hydrogen is too light to be retained by the pull of gravity, any gas released in the atmosphere is lost to outer space. It follows that all hydrogen gas has to be generated using available energy sources. Because hydrogen gas is difficult to store and transport, it

should be generated close to the final user. For practical fuel cell systems hydrogen gas has to be generated from locally available fuels, most commonly hydrocarbons. The chemical conversion of a fuel to obtain gaseous hydrogen is carried out in a fuel processor.

A fuel processor used in fuel cell applications must meet specific technical and marketing demands. It must be small and lightweight, processing feeds of varying composition to deliver hydrogen free from CO and other fuel cell poisons over a wide range of flow rates at a cost comparable to current market values. The unit must be sufficiently rugged to withstand frequent shut downs and cold start ups, and it must operate for many years, unattended and with minimum service. Meeting those constraints requires a number of design trade-offs; for this reason, every fuel processor, without exception, is the result a series of compromises. Different designers have made different sets of design compromises, resulting in the many fuel processor designs available today.

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The goal of this communication is to discuss the design process leading to the development of a fuel processor for stationary fuel cell power generation, and present performance data for the fuel processor operating with either natural gas or LPG. The selection of the unit operations is presented in sequential order, and a flow sheet for the fuel processor is discussed. Then, the specific equipment selected for feed desulfurization, fuel reforming, and hydrogen separation is discussed with some detail. Finally, performance data for a prototype fuel processor operating with natural gas and propane is presented.

2. Process design

A sketch of a light hydrocarbon fuel processor is shown in Fig. 1. The hydrocarbon fuel is desulfurized in a bed of solid adsorbent, mixed with steam and fed to the reformer. The feed first contacts the reforming catalyst at low pre-reforming temperatures to react hydrocarbons heavier than methane yielding a mixture of methane, hydrogen, carbon oxides, and water. This stream is then reformed at higher temperature to convert the methane and increase the concentration of hydrogen. After heat exchanging, the reformer effluent is contacted with a hydrogen-permeable membrane. Hydrogen permeates through the membrane, while the residual gases, or raffinate, containing water, carbon oxides, methane, low concentrations of other organic compounds, and some hydrogen, are combusted in a burner to provide the heat of reforming. The hydrogen stream, or permeate, passes over a methanation catalyst to convert traces of carbon oxides that leak through imperfections in the membrane assembly.

The design considerations involved in drafting this flow sheet are discussed next.

2.1. Hydrogen purity

One of the first decisions made in the design process was regarding product quality. The fuel processor product may be a hydrogen-rich stream containing gases that are inert in

the fuel cell environment, such as water, CO₂, and nitrogen; or it may be pure hydrogen. In the first case, the fuel processor can operate at pressures close to atmospheric, but removal of fuel cell poisons is difficult. CO may be removed by selective catalytic oxidation, but organic impurities are not effectively removed by catalytic processes. Also, inert dilution of the hydrogen fed to the cells results in reduced emf and, perhaps more importantly, reduces hydrogen utilization. In contrast, pure hydrogen can be separated from other reformat gases by pressure-driven processes that require no additional chemical reactors, but compressing a gaseous feed such as natural gas is a demanding operation in the tight energy budget of fuel cell power systems. Furthermore, portions of the processor need to be designed as pressure vessels, adding weight to the system and delaying start-up times.

Low-pressure fuel processors could be light and fast starting; however, that goal is not achievable yet mainly because of the large mass of catalyst needed to remove CO from the hydrogen product. In a low-pressure system, a reformer outlet stream passes over a water-gas shift (WGS) catalyst to react CO with water producing CO₂ and hydrogen. Then, any unconverted residual CO is eliminated by reaction with oxygen over a preferential oxidation catalyst. The conversion in the WGS reactor must be high to maximize hydrogen production. Lower temperatures favor high equilibrium CO conversion in the WGS reactor, while high temperatures favor intrinsic kinetics. Typically, WGS units include a high temperature and a low temperature bed operating at 400 and 200 °C, respectively. The WGS catalyst must be highly active to reduce reactor volume, and it must be stable in the fuel processor environment, which may include exposure to air and liquid water when the system is shut down [1]. Commercially available, copper-based low-temperature WGS catalysts are not stable in fuel cell applications. Noble-metal preparations, now under development, can tolerate exposure to air and water, but those materials require large reactor volumes and deactivate rapidly under reaction conditions [2]. Highly active, stable low-temperature WGS catalysts must be available for low-pressure fuel processors to become practical for fuel cell applications. Because such catalysts are not commercial today, we chose to build a fuel processor that would deliver pure hydrogen.

Pure hydrogen gas may be separated from the reformer outlet stream, or reformat, by pressure-swing adsorption (PSA) or by permeation through a hydrogen-selective membrane. These are mature technologies that have been used in the chemical industry for years, and can be scaled down to be used in fuel processors. In pressure-swing adsorption, a pressurized reformat stream passes over an adsorption column that selectively adsorbs all species but hydrogen. Once the bed is saturated, the reformat stream is switched to a fresh bed. The saturated bed is regenerated by reducing the pressure to facilitate desorption of the adsorbed gases and purging with hydrogen. The outcome of the PSA system is a hydrogen-stripped, low-pressure stream, containing

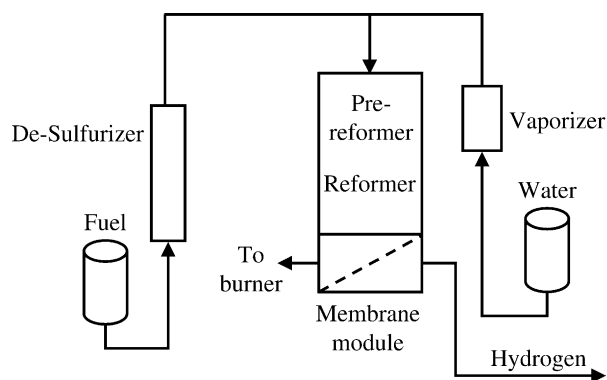


Fig. 1. Flow diagram for a light hydrocarbon fuel processor.

mainly unconverted methane, carbon oxides, and water; and a high-pressure, pure hydrogen stream. PSA systems comprise a number of adsorption columns, solenoid valves, and controls. These systems tend to be bulky and add complexity (i.e. number of parts) to the fuel processor.

Simplicity of operation and separation efficiency are key features of membrane separation systems. Membranes operate as molecular-scale filters separating high-purity hydrogen, or permeate, from the reformat stream. Industrial hydrogen generation processes use polymeric membranes having a variety of permeation and selectivity specifications. Producing high-purity, fuel cell grade hydrogen with those membranes may require two or more steps. Alternatively, more expensive but highly selective palladium-based membranes produce high-purity hydrogen in only one step. Because fuel cells require CO-free hydrogen, and simplicity is a highly desirable feature in fuel processors, palladium membranes are an attractive option despite their cost.

Membranes made of palladium and palladium alloys have extremely high hydrogen selectivities because they operate by catalytic dissociation of hydrogen on the reformat face, and recombination on the permeate face. Hydrogen permeation includes three rate processes in series: dissociative adsorption on the reformat face, diffusion of the hydrogen atoms through the bulk metal, and recombination of those atoms on the permeate face. Under conditions typical of membrane separations, diffusion through the bulk metal is the rate-limiting step. The rate of hydrogen permeation is then proportional to the difference in concentration of hydrogen atoms on both sides of the membrane, and it is inversely proportional to the membrane thickness. Assuming that an adsorption–desorption equilibrium is established on both sides of the membrane:

$$K = \frac{[\text{H}_{\text{surface}}]^2}{p_{\text{H}_2}} \quad (1)$$

where p_{H_2} and $[\text{H}_{\text{surface}}]$ are the hydrogen gas partial pressure and hydrogen atom surface concentration, respectively. The rate of hydrogen permeation (mol permeated/(m² h)) can be written, after solving (1) for $[\text{H}_{\text{surface}}]$:

$$r_{\text{permeation}} = k_{\text{permeation}} \frac{p_r^{1/2} - p_p^{1/2}}{\delta} \quad (2)$$

where $k_{\text{permeation}}$ is a permeation rate constant, p_r and p_p the hydrogen partial pressures in the raffinate and permeate, respectively, and δ is the membrane thickness.

2.2. Fuel reforming

The hydrogen purification technique used in a fuel processor influences the selection of a fuel reforming technology. There are three main types of fuel processing technologies: steam reforming (SR), autothermal reforming (ATR), and partial oxidation (POX) reforming. A steam reformer processes mixtures of hydrocarbon fuel and steam

using heat from an external source to drive the reforming reaction. In an autothermal reformer, air is added to the feed to burn a portion of the fuel to generate the heat required by the chemical reaction. Partial oxidation reforming is essentially fuel-rich combustion; the feed to the reformer contains only fuel and air. ATR and POX systems require bringing the combustion air to the pressure of the process stream; therefore, they are energy inefficient in the high-pressure environment needed for pure hydrogen separation. In contrast, the burner in a SR system can operate at atmospheric pressure, requiring no air compression. For this reason, SR is the preferred hydrogen generation technology used in high-pressure fuel processors. Steam reforming is the established process for large-scale conversion of natural gas and higher hydrocarbons into synthesis gas. A comprehensive review of this technology has been presented by Rostrup-Nielsen [3].

2.3. Coke formation

Coke formation is a pervasive problem in steam reformers. When exposed to high temperatures, typically over 300 °C, hydrocarbon fuels tend to dehydrogenate to form solid species with very low H/C ratios. Those species, known generally as coke, may deactivate the catalyst by depositing on the active sites, blocking the catalyst pores, or causing the metal to separate from the support. Also, coke may grow in the interparticle spaces, increasing the pressure drop in the catalyst bed and eventually blocking gas flow.

The formation of coke can be depressed by contacting steam and fuel with the reforming catalyst at relatively low temperatures (~500 °C), and allowing the reaction to proceed to equilibrium [4]. This has several beneficial effects: steam is a coke inhibitor, the catalyst provides a path for the hydrocarbon to react forming hydrogen instead of coke, and the hydrogen formed further inhibits coke formation. Also, because heavy hydrocarbons react rapidly to give methane, hydrogen, and carbon oxides, the outlet of the pre-reformer contains only trace concentrations of those hydrocarbons. This pre-reformer outlet stream can be fed to the high temperature (>800 °C) reformer with little risk of forming coke. Furthermore, because the pre-reformer operates at temperatures significantly lower than those of the reformer, it can be heated using lower-quality convection heat. This results in improved fuel efficiency [5].

2.4. Sulfur management

All catalysts used in fuel processors for fuel cells, as well as the electrocatalysts in fuel cell electrodes, are susceptible to deactivation by sulfur poisoning. Refining processes remove a significant fraction but not all of the sulfur present in crude oil; hence, all petroleum-based fuels contain some level of sulfur. In addition, safety regulations mandate gaseous fuels to be odorized with sulfur compounds. Sulfur is an undesired component of the fuel processor feedstock because it adsorbs on the active metals in the reforming and

fuel cell catalysts to make surface metal sulfides, which are inactive for steam reforming. The strategy for mitigating coke formation described above, which is to pre-reform at low temperatures, facilitates the formation of surface metal sulfides. In contrast, sulfur has little effect on the reformer reactor when it is operated at temperatures high enough that those metal sulfides are unstable, but without a pre-reforming stage the reforming catalyst would deactivate by coke formation. It follows that the feed to the pre-reformer must be thoroughly desulfurized to avoid deactivation of the pre-reforming catalyst. The most common technology used in oil refineries to remove sulfur from hydrocarbon is hydrodesulfurization, which uses elevated temperatures and pressures, and high hydrogen-to-feedstock ratios, to react sulfur compounds with hydrogen in the presence of a catalyst to yield H_2S . This technology is impractical to use in fuel processors for fuel cell applications mainly because of the energy cost of compressing hydrogen.

A common approach to sulfur management when processing gaseous fuels is to capture the sulfur species in an adsorbent bed at the front end of the processor. A number of adsorbent materials are commercially available. Those adsorbents are generally metal-impregnated high-surface area supports like activated carbon, zeolites, or alumina. The size and composition of a given adsorbent bed depends on the composition of the fuel and the servicing period required in the particular application. IdaTech's fuel processor includes an adsorbent bed to remove sulfur species from the hydrocarbon feed upstream of the reformer.

2.5. Fuel variability

The variability in fuel compositions complicates coke and sulfur management. Common infrastructure fuels are specified for their heating value and macroscopic properties such as density, viscosity, vapor pressure, etc. Chemical composition is not dictated in most of those specifications; hence, the chemical composition of fuels can vary over wide ranges as long as their macroscopic properties are met. For example, typical natural gas compositions include 87–98% methane, 3–13% ethane, 1–3% propane, and smaller amounts of heavier hydrocarbons and inert gases. In some markets, propane and inert gases are added to natural gas to meet excess demand in the wintertime. This practice, known as peakshaving, can result in propane levels in natural gas as high as 23%. The fuel commercialized as propane or LPG, may contain 76–99% propane, 0–22% propylene, 0.5–4% ethane and smaller amounts of methane and hydrocarbons heavier than propane. Also, sulfur content in fuel varies widely. Odorized natural gas might contain 4–6 ppm sulfur, while LPG typically contains 30 ppm sulfur, although levels as high as 100 ppm are not uncommon. Not only the level but also the composition of these odorants varies widely. Because some sulfur molecules adsorb more readily than others, variability in odorant composition adds complexity to the desulfurization operation.

Fuel variability is addressed by designing the pre-reformer to convert the highest levels of hydrocarbons heavier than methane expected to be found in the fuels. This is particularly important when processing LPG, because the rate of carbon formation from olefins is several orders of magnitude higher than from other hydrocarbons [4]. Similarly, the desulfurization unit must be designed to remove the most difficult molecules to adsorb, and its total sulfur capacity must be enough to adsorb the highest concentrations of sulfur expected in the fuel. It follows that, as a consequence of fuel variability, the fuel processor becomes bigger and more costly than needed if fuel composition was constant.

3. Component design

The main components in the fuel processor are the pre-reformer, the reformer, and the hydrogen separation unit. The auxiliary equipment that is required to ensure that the fuel processor can behave as a reliable hydrogen source (pumps, flow meters, solenoid valves, and heat exchangers) is known generically as the balance of plant (BOP). The BOP components may represent a significant fraction of the cost of the fuel processor, and they may be problematic if not selected properly. However, because those components are generally commercially available products, most frequently off-the-shelf items not necessarily built exclusively for fuel processor applications, we will concentrate only on the components that need to be designed specifically for fuel processors: the hydrogen separation unit and the reformer and pre-reformer reactors.

3.1. Hydrogen separation and purification

The design of the membrane module is an example of the many engineering compromises needed to develop a commercial fuel processor. Palladium is the metal of choice because of its high permeability and selectivity to hydrogen, but because palladium is a rather expensive metal, it is critical that the mass of the membrane be kept at a minimum. The permeation rate increases with the hydrogen partial pressure differential across the membrane, and it is inversely proportional to the membrane thickness. As the thickness of the membrane is reduced the permeation rate increases, hence less surface area is needed for a given hydrogen production. Reducing the thickness of the membrane has a strong impact on the cost of the fuel processor because both the membrane area and the mass per unit area are reduced. Yet, the trade-off is that thinner membranes tend to have more defects where reforming gases may leak through. Those gas leaks contaminate the product stream with carbon monoxide, a poison to the fuel cell electrocatalyst. A methanation reactor is added downstream from the membrane to convert traces of carbon oxides to methane, an inert gas in the fuel cell environment. Because the methanation reaction consumes hydrogen, the reformat leaking into the

product stream reduces the thermal efficiency of the system (less hydrogen is produced) and lowers the product purity (methane in the product stream). The thickness of the membrane is chosen as a compromise between the cost of the separation unit and product quality.

The membrane surface area can be reduced by increasing the driving force for hydrogen permeation, $p_r^{1/2} - p_p^{1/2}$. The driving force for hydrogen permeation from a reformat stream containing 50% hydrogen at 10 bar to a permeate stream at 1.3 bar, is $1 \text{ bar}^{1/2}$. Raising the reformat pressure to 15 bar increases the driving force to $1.6 \text{ bar}^{1/2}$. A similar effect can be achieved by reducing the pressure in the permeate side to 0.35 bar. Both gas compression and vacuum generation are energy-intensive operations; choosing one over the other depends on the integration of these operations with the rest of the fuel processor. Other variables affecting the membrane surface area required are the temperature of operation and the amount of hydrogen recovered.

IdaTech's hydrogen purifier contains a number of thin, planar membranes made of a hydrogen-permeable palladium alloy mounted on supports designed to provide mechanical stability to the thin membrane. The alloy is designed to provide high hydrogen permeability, eliminate mechanical stresses resulting from metal-hydride transitions, and provide tolerance to sulfur. The membrane is supported on metal screens designed to sustain high-pressure differentials. Details of the membrane assembly are discussed in references [6,7].

3.2. Reformer

The reactor choice is another of the compromises that the fuel processor designer must face to balance competing demands on the system. Packed bed reactors are well suited to satisfy a number of design conditions: operate over wide ranges of flow rates and temperatures, can be cycled to room temperature, and can be designed to operate for long periods of time even in the presence of catalyst deactivation. In addition, they represent one of the least costly reactors, especially for high-pressure service. The main disadvantage of packed beds is poor heat transfer through the walls and within the bed itself. This is particularly severe in steam reformers, because the heat transferred through the walls can not match the duty imposed by the strongly endothermic reforming reaction. Hydrogen production becomes then a function of the wall surface area; for this reason, packed bed steam reformers are much larger than reactors using structured catalysts such as plate reactors [1]. Furthermore, because of poor heat transfer, packed bed tubular reformers tend to generate cold spots that result in inefficient catalyst utilization. However, the advantages of packed beds regarding cost and easy to change catalyst outweigh the drawbacks listed above in applications where the size of the fuel processor is not a major factor.

The reactors of choice for industrial steam reformers are tubular packed beds. Unfortunately, scaling down those

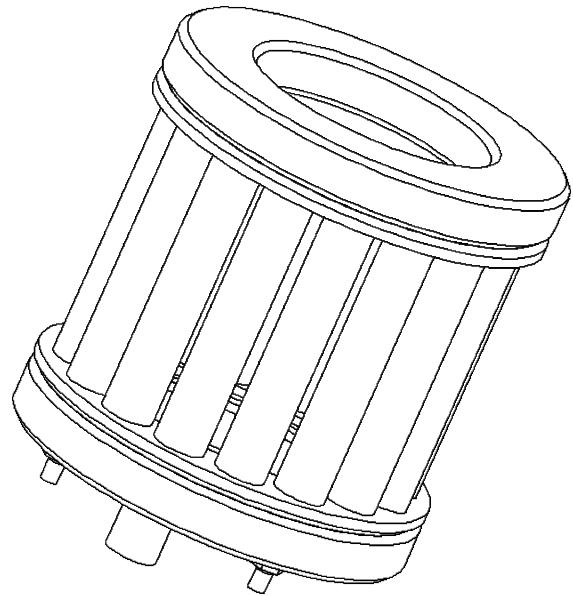


Fig. 2. IdaTech's multi-fuel processor tube bundle. Fifteen 3/4-in.-diameter tubes are arranged around a central burner (not shown). The tubes are heated by radiation from the flame and conduction from the flue gases exiting the assembly through the gaps between the tubes. The inlet manifold uniformly distributes the flow among all tubes.

units to sizes compatible with fuel cell applications is not straightforward. Rostrup-Nielsen [8] has shown that the average heat flux through the wall of a 2-cm-diameter reformer is only 20% the heat flux in a 10-cm-diameter industrial reformer. However, because the surface/volume ratio increases as the diameter of the tube decreases, the heat transferred per unit reactor volume is about the same for both reactors. The reactors differ in length; for any given reactor volume the 2-cm-diameter reactor will be 25 times as long as the 10-cm-diameter reactor. It follows that a fuel processor for fuel cell applications can not be built using a single small diameter packed bed reactor because the reformer would be just too long; instead, those reactors may be assembled in bundles of shorter tubes as in IdaTech's multi-fuel processor shown in Fig. 2. Other packed bed reactors include plate reformers, where the catalyst pellets are packed in a plate heat exchanger, and annular or bayonet type reformers. Plate reformers are generally not practical because they do not hold pressure well and may be difficult to heat directly. Annular reformers have been discussed in the patent literature [9] and have been proposed for fuel cell hydrogen production. Fig. 3 shows a compact "heat exchanger reformer" where the heat from burning raffinate is transferred by convection and radiation to the reforming catalyst. After exchanging heat in the reforming section, the cooler flue gases heat the pre-reformer catalyst. This design is suited for small-scale operation; it may operate at high pressures, allows for a wide load turn down, and minimizes wall temperatures [10].

Steam reformers should be designed to avoid severe temperature gradients because, even after pre-reforming,

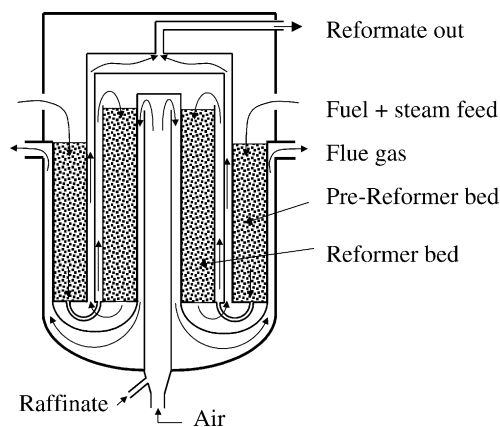


Fig. 3. Heat exchange reformer [10]. The heat of reforming is provided by combustion in a central burner of a stream containing unreacted fuel, such as the residual gas from the hydrogen separator, or raffinate. Fuel and steam are fed through the outer catalyst bed, where it is heated to pre-reforming temperature by the flue gases and the reformed product. The pre-reformed feed is transferred to the inner catalyst bed, where it is heated to reforming temperature by convection from the flue gases and radiation from the burner tube.

methane and CO mixtures may form carbon deposits by the Boudouard reaction:



Carbon formation by this reaction may be suppressed by operating in excess steam, so that thermodynamic equilibrium shifts to carbon oxides and hydrogen. Still, carbon may be formed in hot zones within the reactor. Carbon accumulates on the catalyst and between catalyst particles in overheated zones, deactivating the catalyst and blocking the gas flow. This situation results in the formation of hot zones, creating a positive feedback condition resulting in further overheating of the reactor and more carbon formation. It is necessary then to evenly distribute the heat flux along the reformer to avoid strong temperature gradients [3].

Catalyst composition has a strong effect on carbon formation. Nickel is widely used in industrial steam reformers because of its high activity and low cost. Yet, this metal presents major coking problems because of the formation, diffusion, and dissolution of carbon in the metal. Rhodium is as active as nickel for steam reforming and it does not dissolve carbon to the same extent; hence, carbon formation is less of a problem in rhodium-catalyzed steam reforming. Yet, rhodium is many times more expensive than nickel, but since the catalyst expense represents only a small fraction of the total cost of the fuel cell power system, it is commonly used in fuel processors for fuel cell applications.

IdaTech's reformer was designed to provide a large surface to volume ratio to compensate for the poor heat transfer properties of small packed beds. The heat of reaction is provided by burning the raffinate stream from the membrane module. The reformer is located in the radiant heat zone, and the pre-reformer is heated by convection. Temperature

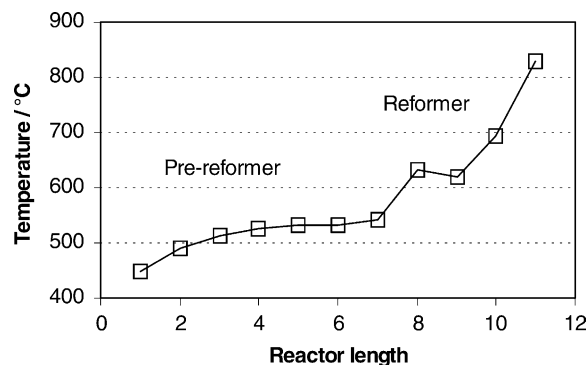


Fig. 4. Temperature profile in IdaTech's light hydrocarbon fuel reformer. The shallow temperature slope in the pre-reformer facilitates complete conversion of hydrocarbons other than methane.

profiles in the pre-reformer and reformer packed beds are shown in Fig. 4. The reactants are gradually heated to a pre-reforming temperature of 550 °C, and kept at that temperature to convert all higher hydrocarbons. Then, the temperature is progressively increased to 850 °C. This temperature profile suppresses carbon formation for two reasons; the process stream is exposed to the highest temperatures when the concentration of hydrogen is the highest, and the contact time at high temperatures is kept to a minimum. This fuel processor can operate with a steam to carbon ratio close to two because of the mild temperature gradients, gradual temperature increase, and the use of a catalyst composition that minimizes the formation of carbon.

4. Heat exchange and thermal integration

The fuel efficiency of a fuel processor depends on high fuel conversion, efficient hydrogen recovery, and adequate thermal integration. All streams need to be heat exchanged to minimize heat losses that would negatively impact the thermal efficiency of the unit. Essentially, it is necessary to use the heat value in the residual gases (raffinate) to provide the heat of reforming and the heat of vaporization of process water. Also, the product hydrogen must be brought to a temperature compatible with that of a fuel cell. To meet these goals, the burner, vaporizer, pre-reformer, reformer, and membrane separation module are assembled according to their operating temperatures within a thermally insulated enclosure, as shown in Fig. 5.

The thermal efficiency of a fuel processor can be defined as the ratio of the lower heating value of the product hydrogen to lower heating value of fuel consumed. An ideal system with no heat losses, fed with reactants at pressure and delivering product at 40 °C and 1.5 bar, could obtain 81% efficiency when reforming a 3/1 steam/methane stream if the reformate is at equilibrium at 800 °C. Real systems operate with lower efficiencies for a number of reasons. Not all hydrogen produced in the reforming reaction can be separated in the membrane module. A hydrogen partial pressure

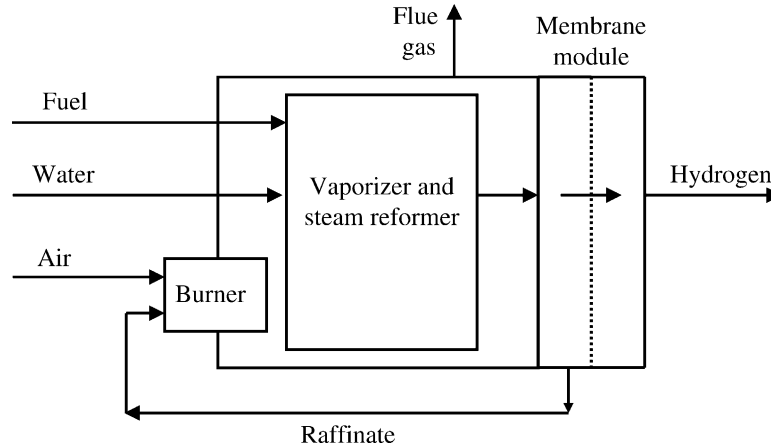


Fig. 5. Sketch showing the heat integration in the fuel processor. The burner, water vaporizer, reformer, pre-reformer, and membrane module are enveloped by a thermally insulated enclosure. The components are arranged to reduce the temperature gradients between them.

Table 1
Performance of natural gas and LPG fuel processors

	Fuel	
	Natural gas	LPG
Hydrogen production (slm)	25	25
	80	85
Thermal efficiency (%)	55	54
	69	67
Outlet temperature (°C)	715	750
	800	785
Fuel conversion (%)	65	73
	83	80
Approach to equilibrium (°C)	10	10
	15	15

differential across the membrane is needed to provide the permeation driving force; for this reason, there is always some residual hydrogen in the raffinate. Also, the flue gases from the burner must be hotter than the atmosphere to provide buoyancy; thus, some heat is necessarily lost to the environment. In addition, real heat exchangers have less than ideal efficiencies because the temperature approach is never zero. Finally, there is always some heat loss in all components because insulation materials are not perfect. For all those reasons, the thermal efficiency of fuel processors is lower than the ideal maximum. Table 1 shows that IdaTech fuel processors operate with full output fuel efficiencies greater than 67%.

5. Fuel processor performance

Two similar fuel processor prototypes were built to operate with natural gas and propane, respectively. The pre-reformer, reformer, and membrane modules in both systems were identical, but some BOP components, and

the BOP design, were different. The LPG processor was designed to operate with liquid LPG; hence, the BOP included a fuel vaporization unit. Other BOP components that could have been similar in both fuel processors were specified differently to gain experience with BOP products from a variety of vendors. After shakedown and debugging, the systems were run for a few hundred hours under different operating conditions. Feed flow rates, steam to carbon ratios, and reformer pressure and temperatures were varied. Results from some of those tests are shown in Table 1. The maximum hydrogen production observed in both processors was slightly over 100 slm; however, because those units were intended to supply hydrogen to fuel cells operating with a power output between 2 and 5 kW, most tests were limited to a maximum production of 80 slm. The minimum production in both reformers was 25 slm, corresponding to a turndown ratio of 3. Lower levels of hydrogen production resulted in unstable (oscillatory) behavior.

The thermal efficiency varied between 55 and 69% for the low and high limits of hydrogen production tested. The efficiency was lower at the lower flow rates mainly because the pressure in the fuel processor decreased with load, reducing the permeation driving force in the membrane module. Consequently, less hydrogen was recovered in the permeate stream. The residual hydrogen was consumed in the raffinate burner, increasing the temperature of the flue gases without contributing to the fuel efficiency of the system.

The reformer outlet temperature at full load was approximately 800 °C in both processors. At partial output, the outlet temperature decreased because the heat generated at the burner diminished while the heat lost through the insulation remained relatively constant. Lower exit gas temperatures resulted in lower methane conversions because the equilibrium concentration of hydrogen decreases with decreasing temperature.

The methane content of the exit gases was somewhat higher than expected from equilibrium calculations. The

approach to equilibrium of the reforming reaction, defined as the difference between the measured outlet temperature and the temperature that would yield the measured methane conversion at equilibrium, varied between 5 and 15 °C. This is within the range observed in industrial reformers [3].

The natural gas fuel processor has been integrated with a fuel cell in a combined heat and power system delivering 5 kW electric and 2.5 kW heat. This unit is undergoing testing prior to shipping for field tests.

6. Conclusions

We have discussed the design process and the performance of a fuel processor intended to meet previously identified market needs. The design path followed and the component choices made are just one set of the multiple options available to the fuel processor developer. While other designs may serve to meet similar goals, we believe that our fuel processor satisfies performance requirements while keeping cost to a minimum.

The design process involved two decision levels. At the system level, we chose to build a fuel processor delivering pure hydrogen largely because of concerns about the performance of catalysts used for low-pressure product purification. Simplicity and cost considerations dominated the choices at the component level. While choosing components individually to meet those constraints was relatively straightforward, decisions regarding component interactions required market information not generally available. For example, a control system could have been designed to keep the pressure in the reformer, and thus the thermal efficiency, independent of feed flow rate. Would the fuel savings at low

loads compensate for the extra cost of the controls? This type of question will not be answered until after customers gain experience using fuel cell power systems. At present, fuel processor design may be characterized as multiple-parameter optimization in which the objective function is poorly defined because it must be defined largely from market inputs. Hence, only heuristic methods are available, and these are the age-old engineering skills of experience and intuition.

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