

**Chicken... Meet the Egg!**  
**A Cost Effective Hydrogen Supply Solution**

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## **1. Introduction**

ChevronTexaco Corporation continues to evaluate and monitor advanced energy options and opportunities across the entire value chain through its subsidiary ChevronTexaco Technology Ventures LLC (CTTV). CTTV's Hydrogen Group is actively developing the competencies necessary to become a key participant in the emerging hydrogen economy by building organizational capability in distributed hydrogen production while evaluating and commercializing the best alternatives to produce hydrogen economically. ChevronTexaco believes that producing hydrogen on site, where it is consumed is the most practical and cost-effective approach to producing hydrogen fuel. Development of an economic distributed hydrogen infrastructure is fundamental to the success of a hydrogen economy and the commercialization of fuel cells in both transport and stationary power applications. CTTV's technical expertise is in the onsite reformation of natural gas and other hydrocarbons for economic distributed onsite generation that may be used to supply hydrogen for a hydrogen energy station, thereby providing hydrogen for both vehicle and stationary applications.

ChevronTexaco and Modine Manufacturing Company have developed a compact reformer design which features a mechanical as well as thermal integration of the steam reforming, catalytic oxidizer, and water-gas shift reactions in a single vessel. The design is thermally neutral and requires no external cooling and no control loops. Additionally, process design, system efficiency, and cost estimate studies have been performed leading to the design and construction of a low cost natural gas fed 40 kg/day fuel processor system. Economic analysis on the design shows that it has the potential to surpass the U.S. Department of Energy's (DOE) near term hydrogen cost targets for distributed production of hydrogen from natural gas.

The main objective of this joint development project is to develop a natural gas reformer that will provide ChevronTexaco with a significant product cost advantage in the "hydrogen economy". Key attributes for a cost effective reformer designs are: (1) safe, robust and reliable, (2) low operating costs through improved fuel efficiency, (3) low capital costs through reduced system components and controls complexity, (4) manufacturable in high volumes.

## **2. Technology Selection**

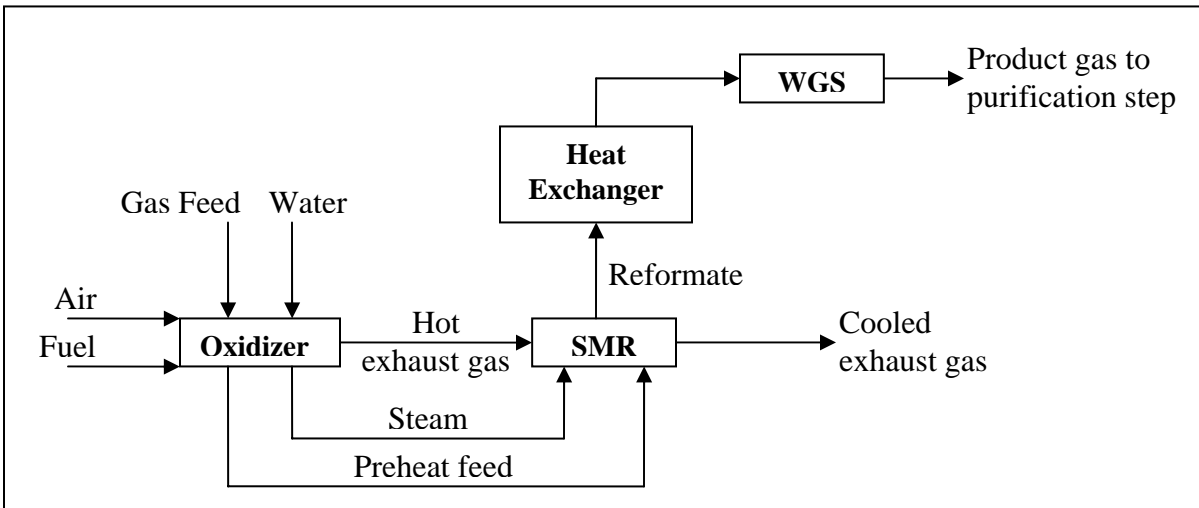
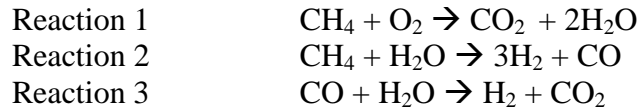
Steam methane reforming (SMR) and autothermal reforming (ATR) are the two process technologies typically considered for distributed hydrogen generation from natural gas. Various studies and reports<sup>[1,2]</sup> have concluded that the most cost-effective option, in terms of cost of hydrogen generated, is steam methane reforming coupled with pressure swing adsorption (PSA) hydrogen purification. Technical and economic analyses, performed by CTTV, have also supported this conclusion. Furthermore, as shown in Table 1, high pressure (HP) SMR operating at PSA pressure is the best technology option because it is far less energy intensive when compared to low pressure (LP) SMR.

**Table 1.** Qualitative comparison between SMR and ATR

Parameter (SMR + PSA)	SMR relative to ATR	HP SMR (150 psig) relative to LP SMR (5 psig)
Reforming Temperature	Higher	Higher
H <sub>2</sub> Concentration in Reformate pre-PSA	Higher	Lower
PSA Size (to achieve similar purity)	Smaller	Same
Parasitic Power Consumption	Lower	Lower
Net System Efficiency	Higher	Higher
Operability and Controls	Simpler	Simpler
<b>Cost per kg of H<sub>2</sub> produced</b>	<b>Lower</b>	<b>Lower</b>

### 3. Technical Approach

Traditional SMR employs a combination of direct and indirect heat transfer, where heat is generated oxidizing auxiliary fuel (Reaction 1) to provide the necessary thermal energy for the steam reforming reaction (Reaction 2). As illustrated in Figure 1, product gases (reformate) generated by the steam reforming reaction are then typically cooled, using a heat exchanger, to a desired temperature range necessary for the subsequent water-gas shift (WGS) reaction (Reaction 3) to occur.



**Figure 1.** A simplified schematic representation of the typical SMR steps

Although the process chemistry remains the same for a small scale SMR (50 – 100 kg/day H<sub>2</sub>) when compared to a large scale refinery SMR, there are severe economy of scale penalties related to capital costs for small scale SMR based hydrogen energy stations. In addition, natural gas reforming is a relatively high temperature process. Scaling the process down from larger systems results in greater heat losses that contribute directly to lower production efficiency, higher operating costs, and ultimately higher cost of hydrogen. To address these challenges, the project approach aims at developing a small scale SMR that is: (1) thermally and mechanically

integrated to maximize heat recovery, minimize heat loss, and minimize balance of plant components, (2) able operate at pressure required for purification step to minimize electrical power consumption, and (3) thermally balance to achieve passive temperature control and to minimize the number of process control loops.

#### 4. Conceptual Design

With the project approach in mind, the design team developed an integrated SMR design by combining all process reactions and necessary heat transfer steps into a single, unitized vessel assembly. Internal to the vessel, the design employs concentric fin type heat exchangers where their surfaces are coated with catalysts. Figure 2 shows a cross-section of a concentric geometry where inner fins and the outer fins are brazed onto a cylinder wall.

Subsequently, the inner fins are coated with steam reforming catalyst and the outer fins are coated with oxidizing catalyst. The configuration allows for heat generated by the exothermic oxidizing reaction to be directly transferred to the endothermic steam reforming reaction. In addition by combining separate reactor vessels and heat exchangers into one single unit, the arrangement significantly reduces capital equipment costs.

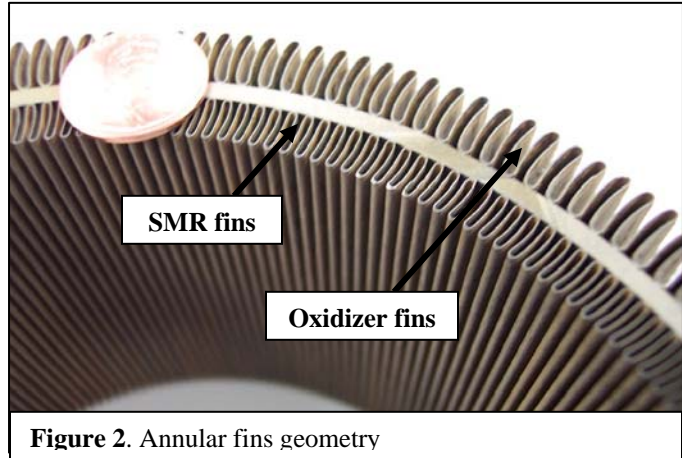


Figure 2. Annular fins geometry

Coating catalyst on a metal surface and the ability to develop highly effective SMR catalyst is not new<sup>[3]</sup> as they been done for many years for various applications. The main challenges associated with coating catalyst on a heat exchanger fin surface are: (1) the ability to evenly apply the catalyst into every fin channel and over the entire length of each channel, and (2) catalyst wash-coat adhesion both initially and over the long period of operation.

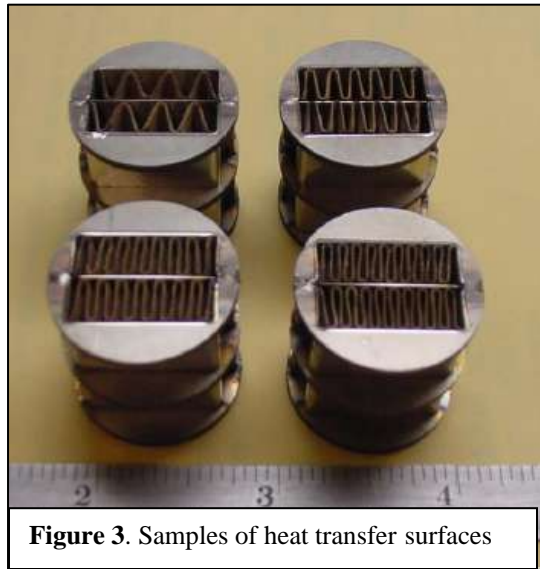


Figure 3. Samples of heat transfer surfaces

The issue with adhesion is not in regards to the metal fin material but rather in the brazing material that is used. The design team was able to successfully overcome the first challenge by modifying the fin structure and catalyst coating technique. To address the second challenge, sample heat transfer fin surfaces were developed and coated with steam reforming catalyst and oxidizing catalyst, separately. Figure 3 shows a picture of these samples containing fin surfaces with fin density ranging from 9 to 25 fins per inch (fpi).

The samples coated with steam reforming catalyst were tested in a high temperature reactor furnace at design operating conditions (720 – 760°C, 120 psig, 3.5 steam to carbon ratio, 28000 hr<sup>-1</sup> gas hourly space velocity). After the initial reforming cycle, all

samples maintained adhesion. As shown in Figure 4, long term testing of a 15.5 fpi SMR fin sample yielded consistent conversion over a period of 750 hours with 6 startups/shutdowns. Conversion was 2 – 5% lower than equilibrium as the reactor furnace was temperature limited to approximately 740<sup>0</sup>C. Conversion is expected to improve at designed temperature of 760<sup>0</sup>C.

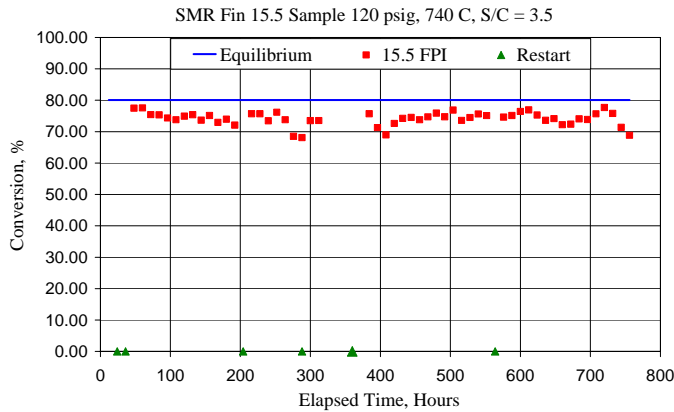


Figure 4. Long term adhesion testing of SMR fin sample

The coating and testing trials demonstrated successful proof of concept and provided the necessary data to proceed to a full scale design. The highly integrated design is capable of oxidizing fuel for heat generation, taking in natural gas and water and convert them into hydrogen-rich reformat product without requiring any active cooling and/or control in any place along the processing train. Figure 5 illustrates a 3-dimensional representation of the final integrated design. The vessel houses a series of fin-type heat exchangers and coated fin heat

exchangers/reactors as discussed previously. Air and PSA off-gas are introduced to enable the oxidizing reaction to take place. As the oxidizer exhaust travels up the vessel, heat is transferred to the SMR region first and the majority of the remaining thermal energy is utilized to generate steam and preheat natural gas. As the exhaust exits the vessel, most of the thermal energy has been extracted by the SMR reaction, natural gas preheating and steam raising process.

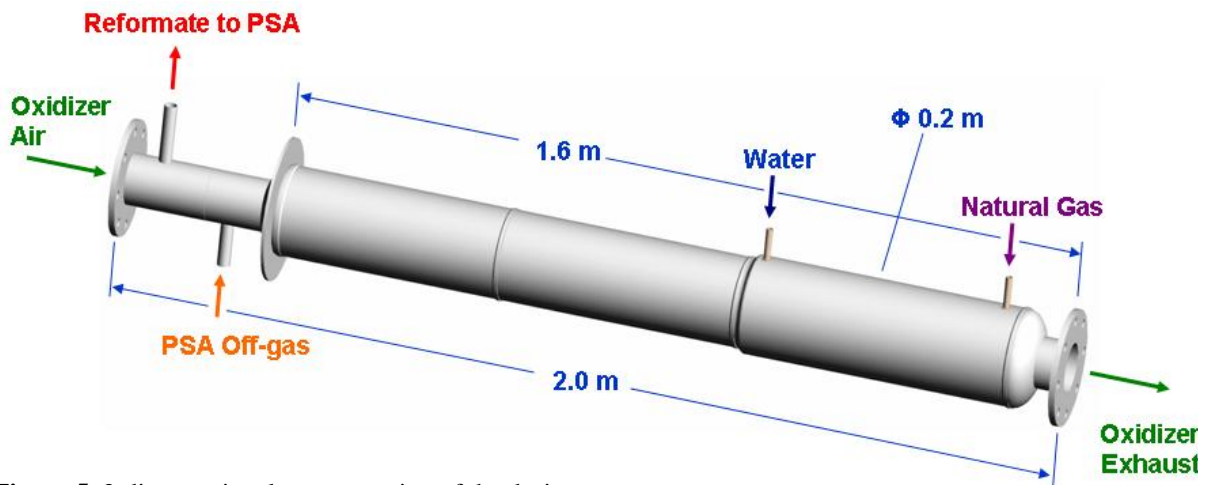


Figure 5. 3-dimentional representation of the design concept

In addition, the vessel houses a WGS monolith reactor which operates adiabatically to further reduce CO concentration and increase hydrogen product in the reformat stream. The high-temperature reformat exiting the WGS reactor is cooled down by the oxidizer air and off-gas mixture in another fin heat exchanger prior to exiting the vessel. Essentially, this thermally and

mechanically integrated design has left “no stones unturned” or more accurately, no usable heat un-recovered.

The integrated process, operating in conjunction with a PSA unit, is thermally neutral where additional supplemental fuel (natural gas) is no longer needed in order to achieve high reforming efficiency and conversion. This directly translates to lower operating costs. Effectively, this design eliminates excess piping throughout the system, reducing capital equipment cost. Furthermore, the heat exchangers’ surface areas and flow configurations are designed such that they serve dual purposes - heat recovery and passive temperature control of process streams. This thermal pinching/passive temperature control technique not only simplifies and adds robustness to the process controls but eliminates control valves and various other moving parts throughout the process. As a result, the only active control parameters of the process are oxidizer air, natural gas, and water flows to the steam reformer. The uniqueness in the process and mechanical design significantly drives down the capital cost of the system.

## 5. Performance and Cost Targets

Performance targets for each integrated reactor vessel are listed on Table 2. These targets are projected based on extensive system modeling and proof of concept testing. The hydrogen production rate refers to the hydrogen rate out of the reactor vessel at operating pressure (120 psig) prior to entering a PSA unit. The total gas consumption assumes off-gas utilization by the oxidizer. The electrical power consumption includes all electrical power required to operate the reforming system but excludes electrical power for the PSA system. The reformat delivery to the PSA does not contain liquid condensate. The reforming pressure is supplied by the compression of natural gas from line pressure and pumping of de-ionized water from a separate purification system.

**Table 2.** SMR performance targets per single reactor vessel

<b>Parameter (Single Reactor Vessel)</b>	<b>Unit</b>	<b>Value</b>
Max reformer H <sub>2</sub> production rate	kg/day	55
Max gas consumption rate	kg/day	145
Max electrical power consumption	kW	1.5
H <sub>2</sub> concentration in reformat	%	75
Reformat delivery temperature	°C	60
Operating pressure	psig	120

As previously mentioned, one of the key drivers for this development project is the final design must demonstrate significant product cost advantage. The design team conducted extensive economic analysis to determine the projected cost of hydrogen produced. A Piping and Instrumentation Diagram (P&ID) and a Bill of Materials (BOM) were generated to allow for determination of the actual capital equipment cost to build the first prototype SMR system. This capital equipment cost, along with the performance characteristics shown in Table 2, were then used to determine the overall hydrogen cost. It should be noted that the economic study looked at the reforming system, but the PSA system was not included. Table 3 lists the projected hydrogen production costs for the SMR system and the Department of Energy (DOE) 2005 cost

targets. The results from the economic analysis demonstrated the SMR design has the potential to surpass DOE's near term hydrogen cost targets for distributed natural gas reforming.

**Table 3.** CTTV SMR hydrogen cost targets compared to DOE's draft targets for reforming

Characteristics		Unit	2005 DOE Targets*	CTTV SMR
Reforming	Capacity	kg/day	690	40
	Natural Gas Cost	US\$/kg H <sub>2</sub>	0.62	0.72
	Other Costs	US\$/kg H <sub>2</sub>	1.36	1.12
	<b>Total Cost</b>	<b>US\$/kg H<sub>2</sub></b>	<b>1.98</b>	<b>1.84</b>

\* DOE Technical Plan – Hydrogen Production – Draft (6/3/03)

The basis and variables for calculating the overall reforming hydrogen cost are as follows:

- 248 days of operation per year (5 days per week, 1 day per month downtime)
- Fuel is natural gas
- PSA recovery is 75% (off-gas is utilized for preheat feedstock)
- Natural gas price is \$4/MMBTU
- Electricity buying price is \$0.07/kWh
- Water price is \$0.05/ft<sup>3</sup>
- Depreciation period is 10 years
- Method of calculation depreciation is double declining rate (DDB/SL)
- Inflation rate and hydrogen cost inflation rate is 2%
- Quantity of product is less than 10 units per year
- Annual operating and maintenance cost is assumed at 2% of total capital cost
- Site related property tax is assumed at 1% of total capital cost
- Contingencies are assumed at 20% of capital equipment cost
- Capital recovery is set at 11% (per DOE 2005 target)

## 6. Future Directions

The design team has fabricated the first prototype reactor assembly and the construction of the system is in the final phase. Initial testing of the first prototype is scheduled for May 2005. Each reactor vessel is capable of producing up to 40 kg/day H<sub>2</sub> when coupled with a PSA unit. By combining multiple vessels together, capacity of up to 120 kg/day H<sub>2</sub> can be achieved without significant efficiency loss. This range of capacity allows for various potential market applications in limited volumes. The potential market applications include, but are not limited to (1) low volume industrial applications, (2) hydrogen infrastructure/energy station, and (3) fuel cell distributed generation. With successful testing of the first prototype in the coming months, the design will be ready for initial field demonstration by mid-2006.

## 7. Conclusions

The design team has completed design and construction of the first prototype high pressure SMR reactor, capable of producing 40 kg/day of hydrogen at a competitive production cost of \$1.84/kg. This design meets our project objectives with the following key attributes:

*High Efficiency*

- Thermal integration minimizes heat loss and maximizes heat recovery by employing catalyst-coated heat exchangers.
- Reduced parasitic load with passive temperature control (no external cooling)
- High pressure (120 psig) reforming improves overall process efficiency when coupled with a PSA unit

#### *Low Capital Equipment Cost*

- Manufacturable in high volumes using standard production methods
- Hydrogen capacity scalable by combining multiple modules – “economy of scale”
- Mechanically integrated design minimizes component interconnects

#### *Reduced Balance of Plant*

- Thermal integration eliminates parasitic auxiliary heating and cooling equipment
- Reduction in number of control loops

#### **References**

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3. Ahmed, S, and Krumpelt, M., *International Journal of Hydrogen Energy*, Elsevier, 26, p291, 2001.