ECONOMIC FEASIBILITY ANALYSIS OF HYDROGEN PRODUCTION BY INTEGRATED CERAMIC MEMBRANE SYSTEM

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Abstract

Praxair has completed a technoeconomic feasibility analysis of a small scale hydrogen production system based on oxygen transport membrane (OTM) and hydrogen transport membrane (HTM). This system has a potential to significantly reduce the cost of hydrogen for use in the transportation sector for fuel-cell vehicle fueling stations and in the industrial sector as a small, on-site hydrogen supply. This paper updates the results obtained from an economic feasibility evaluation, as well as the future plan for the HTM development.

Introduction

Hydrogen is expected to play a vital role in the transportation sector for the fuel cell vehicles (FCVs). One of the crucial factors for the successful introduction of FCVs on the U.S. roadways is a low-cost supply of hydrogen. The demand for hydrogen at fueling stations for FCVs is projected to be less than 10,000 scfh. To be competitive with the untaxed gasoline, the cost of hydrogen delivered to a vehicle must be below $20/MMBtu. A key challenge for the on-site plant is to reduce capital costs. The approach taken in this program is to reduce capital costs by reducing the complexity of the process and thus reducing the equipment needed to generate hydrogen.

Process Design

Two process options were evaluated. In the first process option, both the OTM and the HTM were integrated into a single unit such that various processing steps (syngas generation, shift conversion and hydrogen purification) necessary for hydrogen production occur in a single reactor (Shah, 2001). Since the OTM reactor operates at high temperatures (800 to 1100 ^{o}C), it is necessary to have the HTM operating at high temperatures. The ceramic proton conducting membranes can operate at temperatures up to 900 ^{o}C and they were considered as HTMs for this process option. The Pd (palladium) alloys are not suitable for high temperature operation. In the second process option, OTM and HTM are placed in two separate reactors (Figure 1). By decoupling these two membranes, the temperature constraint for the HTM is removed and the HTM reactor can be operated at much lower temperatures (e.g. 300 to 600 ^{o}C) than the OTM reactor. The Pd alloy membranes were considered for this process option.

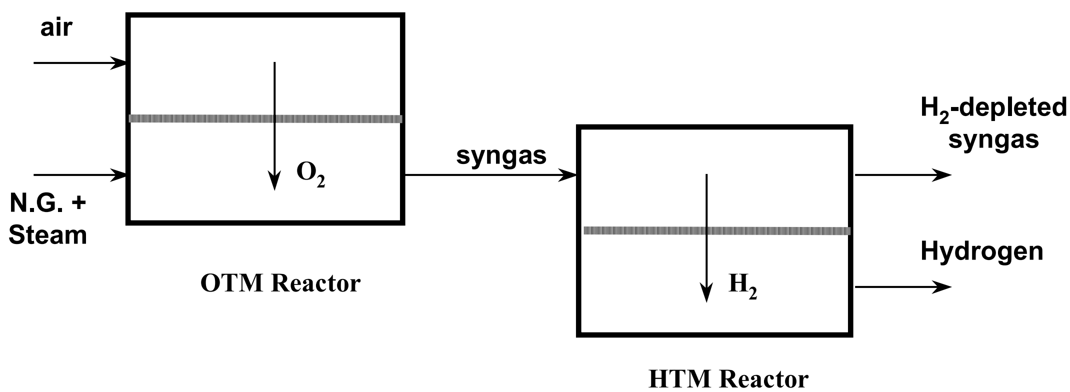

Figure 1. Sequential OTM and HTM Reactor System

Air at low pressure (~25 psia) is passed to the retentate side of the OTM and compressed natural gas (200 - 300 psia) and steam are passed to the permeate side of the OTM. Oxygen is transported across the OTM to the permeate side, where it reacts with natural gas to form syngas. A portion of natural gas also reacts with steam to form syngas. Additional hydrogen is formed by the water-gas shift reaction:

$$CH_4 + 1/2O_2 \rightarrow CO + 2H_2 \quad \text{(Partial Oxidation)}$$
$$CH_4 + H_2O \rightarrow CO + 3H_2 \quad \text{(Reforming)}$$
$$H_2O + CO \rightarrow H_2 + CO_2 \quad \text{(Shift)}$$

A catalyst is incorporated in the reactor to promote the reactions. The syngas from the OTM reactor is cooled and then fed to the retentate side of the HTM reactor. In the HTM reactor, the shift reaction and the hydrogen separation through HTM take place. Hydrogen is transported to the permeate side of the HTM by the partial pressure difference driving force. Due to removal of hydrogen from the reaction zone, more hydrogen is formed by the shift reaction. As much hydrogen as possible is recovered from the reaction zone by transport through the HTM to the permeate side. Eventually, a partial pressure pinch between the reaction zone and the permeate side is reached, limiting the amount of hydrogen that can be recovered.

A process model was developed for the process with sequential OTM and HTM reactors. The Hysys simulation was used to evaluate the performance of the process.

The overall efficiency of the plant is defined as follows:

$$H_2 \text{ Efficiency} = \frac{\text{Energy Recovered in } H_2 \text{ (HHV) x 100}}{\text{Energy Input in Natural Gas (HHV)}}$$

Table 1 summarizes utility consumption and the H_2 efficiency for the sequential reactor process.

Table 1. Performance Summary of the Sequential Reactor Process

Hydrogen capacity, scfh	1,000
N.G., scfh	404
Power, kW	4.6
Water, gpm	1.1
H_2 Efficiency, %(HHV)	79 %

The H_2 efficiency of the process with sequential reactors was estimated to be 79% (HHV or higher heating value). This compares with ~76% efficiency (HHV) for the process with the integrated OTM-HTM reactor. The higher efficiency for the process with sequential OTM and HTM reactors was due to lower temperature, which is favorable for shift reaction equilibrium.

Economic Feasibility Analysis

The cost estimate developed for the process based on the integrated OTM-HTM reactor was used as a baseline cost and the cost estimate for the process with the sequential reactors was developed by extrapolation. The hydrogen plant capacity was fixed at 1,000 scfh for the cost estimation. The capital costs for 2,000 and 5,000 scfh were estimated by using appropriate scale-up factors. For each capacity, costs were estimated for 10, 100, and 1,000 plants built/year. After reviewing the results, it was clear that the plants with capacities of 1,000 and 2,000 scfh will not be economically viable, because the cost of hydrogen from such plants is either comparable to or higher than the cost of liquid hydrogen. Therefore, the results for only 5,000 scfh plants are presented here.

The cost estimate presented last year for the integrated reactor process was subjected to internal review and revised. The cost components with significant revisions were costs of reactor, instrumentation, natural gas and capital recovery. In addition, the costs related to contingency and safety were added. To estimate capital recovery costs, the method described in the Hydrogen Infrastructure Report (Thomas 1997) was used. The financial parameters listed in Table 2 were used.

Table 2. Financial Parameters

15% after–tax rate of return
38% corporate tax rate
15-year plant life
0% inflation rate

These parameters lead to capital-related charges of 23.5% of capital investment/year. In addition, the assumptions listed in Table 3 were made.

Table 3. Cost Estimation Assumptions

Natural gas	$4/MMBtu (HHV)
Power	$0.05/kWh
Water	$0.1/1000 gal.
M & R	3% of capital investment/year
Capacity Utilization	80%

Figures 2 and 3 show the revised capital and product costs for the process with the integrated reactor. Figures 4 and 5 show capital and product costs for the process with the sequential reactors. The range in capital costs for any given point is due to the level of uncertainty in the cost estimate, especially in the costs of membrane reactors.

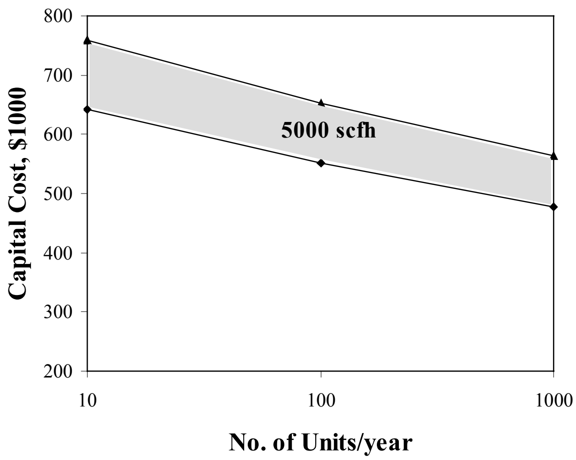

Figure 2. Capital Costs for the Integrated OTM-HTM Reactor Process

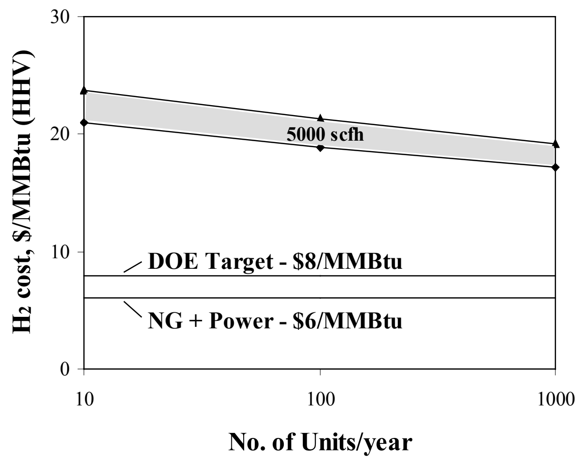

Figure 3. Product Costs for the Integrated OTM-HTM Reactor Process

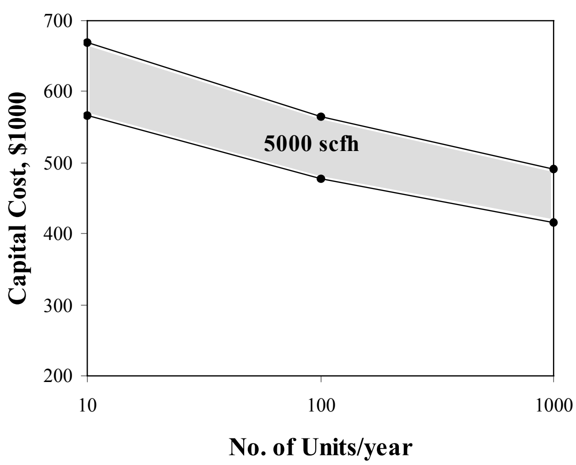

Figure 4. Capital Costs for the Sequential OTM and HTM Reactors Process

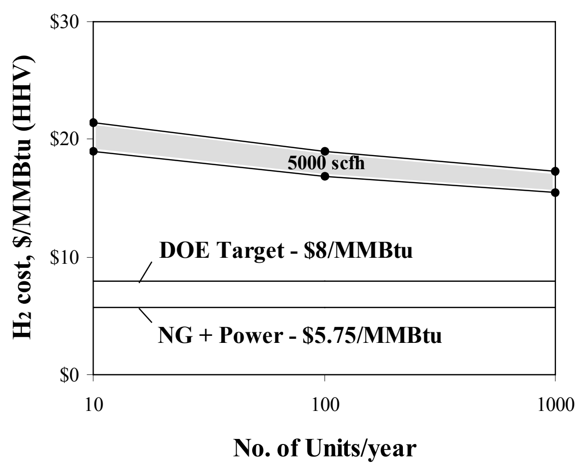

Figure 5. Product Costs for the Sequential OTM and HTM Reactors Process

Comparing the costs for two process options, the capital costs for the sequential reactor process are ~12% lower than the costs for the integrated reactor process. Several factors contributed to lower costs for the sequential reactor process. The complexity of the reactor design is significantly reduced when there are two separate membrane reactors. The operation of HTM reactor at lower temperature makes it possible to use cheaper materials of construction. The OTM reactor still operates at a high temperature, however, its size is significantly smaller than the size of the integrated OTM-HTM reactor. Finally, the hydrogen flux through Pd alloy membrane (which is used in the sequential reactor process) is much higher than the flux through proton conducting ceramic membrane (which is used in the integrated reactor process). As a result, less membrane area is required, which in turn reduces size and the cost of the HTM reactor.

The product hydrogen costs for the sequential reactor process were ~10% lower than the integrated reactor process. The cost of hydrogen production (at 15 psia) ranged from $15 to $21/MMBtu (HHV) depending on the number of plants built per year. These numbers do not include the costs of compression, storage and dispensing. At low production volume (10 units/year), the cost of hydrogen will be $19 to $21/MMBtu (HHV). With mass production (1000 units/year), the cost of hydrogen will drop down to $15 to $17/MMBtu (HHV). The capital cost reduction at higher production volume results from the volume discounts for the equipment purchases and reduction in assembly costs due to experience in building multiple identical plants.

The projected cost of hydrogen production is much higher than the DOE target of $8/MMBtu (HHV). The utilities alone account for ~72% of the DOE target. The capital costs will have to be reduced by 80-85% from the current projections to achieve the DOE target. It is not feasible to achieve such reductions. It may be possible to reduce capital costs by 20 to 35% by reducing membrane reactor costs and using DFMA (design for manufacturing and assembly) approach for mass production. The corresponding hydrogen cost will range from $13 to $ 16/MMBtu (HHV).

Although the DOE target is not achievable, it must be emphasized that the projected costs of hydrogen from OTM/HTM technology are lower than the costs of liquid hydrogen and electrolytically produced hydrogen. Liquid hydrogen costs $30 - $45/MMBtu (HHV), depending on the consumed volume, location and contract length (Chemical Marketing Reporter 2001). The projected capital and M & R costs for mass produced electrolysis equipment (Thomas 1997) and power costs assumed in this study results in the cost of electrolytically produced hydrogen to be $25/MMBtu for a 5,000 scfh plant.

Phase II Plan

Phase I indicated that the two-step reactor system with OTM reactor followed by integrated HTM shift reactor as the preferred approach for an economical hydrogen production system. Significant efforts are required in two areas for successful commercialization: development of cost-effective HTM and development of mass production approach to reduce capital costs. The total duration of the program could be five to six years. The market studies indicate that mass production of fuel cell vehicles (FCVs) may be 10 years away. To initiate a rigorous program that will lead to mass production facility for hydrogen plants in five to six years does not appear to be warranted. Therefore, we are proposing a program with lower level of effort in the initial years with the emphasis on advancing the hydrogen separation technology. Any effort related to mass production will be undertaken when market for FCVs is more clearly visible.

The experimental work on the proton conducting materials based HTMs indicates that the hydrogen flux is not sufficient for commercial viability in the foreseeable future. Therefore, we have decided to focus on the Pd alloy based HTMs for further development efforts. The basis for the future work is the technology established by Research Triangle Institute (RTI) to deposit thin, uniform, defect-free, Pd alloy membrane layer on the ceramic substrate and Praxair's ceramic membrane manufacturing technology.

Phase II of the program is expected to last three plus years and it will be divided into two parts. The first part of Phase II will focus on developing a commercially viable HTM and it will last two plus years. We expect to develop HTM and its sub-components during the first year and test a bench-scale membrane reactor (containing multiple membrane elements) with shift conversion in the subsequent year. The second part of Phase II will involve testing of critical balance of plant components such as OTM reactor, steam generator and high temperature heat exchangers.

In the first year of Phase II, Praxair will develop a low-cost ceramic substrate with desired porosity and mechanical strength. RTI will develop a suitable membrane material with resistance to syngas and repeated thermal cycling. Initial experiments for the substrate and the membrane screening will be performed with disks. Once the appropriate materials (membrane and support) are selected, the composite membrane elements (e.g. tubes) will be fabricated and tested.

The second year effort will focus on the development of HTM reactor with shift conversion. A multi-tubular bench-scale membrane reactor will be assembled. Suitable sealing techniques will be developed as needed. The performance of the reactor will be evaluated with simulated synthesis gas compositions. A long-term test will be carried out to test durability. At the end of Part I of Phase II, economic and business analyses will be updated and a go/no go decision to proceed will be taken.

In Part II of Phase II, the focus will be to test other critical components of the process and address any development issues related to those components. Currently, the OTM technology is under development in a separately funded program at Praxair and is expected to be available by the time the HTM development efforts are completed. A bench-scale OTM reactor will be tested in the hydrogen production mode. Steam generator and high temperature heat exchangers will also be tested during this Phase of the development.

Conclusions

The efficiency of the sequential reactor process is estimated to be 79% (HHV). The sequential OTM and HTM reactors process with HTM reactor operating at lower temperature will result in lower hydrogen costs compared to the integrated OTM-HTM reactor process. The cost of hydrogen is estimated to range from \$15 to \$21/MMBtu (HHV) depending on the number of hydrogen plants built per year. The projected hydrogen costs from the proposed system are lower than the competing supply options, such as electrolysis and liquid hydrogen.

Phase II plan has been defined. The sequential reactor process has been selected as a preferred process option. The first objective of Phase II will be to develop a low-cost HTM (based on Pd alloy) with high hydrogen flux and tolerance for syngas components and thermal cycling. The next task will be to design and test a bench-scale membrane reactor to carry out shift conversion and hydrogen separation. Finally, other critical components such as OTM reactor, steam generator and high temperature heat exchangers will be tested.

Acknowledgments

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