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SYSTEM OPTIMIZATION AND COST ANALYSIS OF PLASMA CATALYTIC REFORMING OF HYDROCARBONS

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Abstract

Thermal plasma reforming offers advantages in hydrocarbon reforming especially in small to medium scale plants and in plants with fast transients. The combination of a thermal plasma reformer operating with partial oxidation with a catalyst bed will be described. Reduced concentrations of CO (1-3% vol) can be achieved, with high hydrogen yields and minimal plasmatron electrical power requirements. A model for the cost of hydrogen production by this method, including hydrogen cleanup utilizing a conventional pressure-swing absorption unit, has been developed. The model uses experimentally determined conversion yields and operational parameters. The conditions that result in system optimization and cost minimization have been determined.

1 Introduction

Manufacturing of hydrogen from natural gas, biofuels and other hydrocarbons, is needed for a variety of applications. Plasma technology could provide important improvements in reforming hydrocarbon fuels for the production of hydrogen-rich gas for fuel cells and other applications. The plasma conditions (high temperatures and a high degree of ionization) can be used to accelerate thermodynamically favorable chemical reactions or provide the energy required for endothermic reforming processes. Plasma reformers can provide a number of advantages:

- economically attractive operation in small scale hydrogen production applications
- operation with a broad range of fuels, including natural gas and biofuels
- decreased problems of catalyst sensitivity and deterioration
- compactness and low weight (due to high power density)
- fast response time (fraction of a second)
- minimal cost
- high conversion efficiencies

Hydrogen-rich gas could be efficiently produced in compact plasma reformers with a variety of hydrocarbon fuels including natural gas, biomass, and others. The technology could be used to manufacture hydrogen for a variety of stationary applications (e.g., distributed, low pollution electricity generation from fuel cells or hydrogen-refueling gas stations for fuel cell driven cars). It could also be used for mobile applications (e.g., on-board generation of hydrogen for fuel cell powered vehicles).

In this paper, the cost issues of a plant that incorporate a plasma reformer are investigated. It is assumed that the plant operates on natural gas, although the plasmatron is capable of operating in a wide range of fuels. The experimental results of plasma reforming of natural gas are briefly reviewed in section 2. A model of a process plant is developed, and a model for determining the hydrogen cost is described in section 3. The sensitivity of the cost to capital cost, cost of natural gas and to manpower requirements are then presented in section 4. The tradeoff between higher conversion and increased electrically energy consumption in the plasmatron is described in section 5. Finally, the conclusions and direction of additional work is described in section 6.

2 Plasma reforming

Plasma devices referred to as plasmatrons can generate very high temperatures (>2000 C) with a high degree of control, using electricity [1]. The heat generation is independent of reaction chemistry, and optimal operating conditions can be maintained over a wide range of feed rates and gas composition. Compactness of the plasma reformer is ensured by high energy density associated with the plasma itself and by the reduced reaction times, resulting in short residence time. Hydrogen-rich gas can be efficiently produced in plasma reformers with a variety of hydrocarbon fuels (gasoline, diesel, oil, biomass, natural gas, jet fuel, etc.) with conversion efficiencies into hydrogen-rich gas close to 100% [2,3].

The plasma conditions (high temperatures, high degree of dissociation and substantial degree of ionization) can be used to accelerate thermodynamically favorable chemical reactions without a catalyst or provide the energy required for endothermic reforming processes.

The technology could be used to manufacture hydrogen for a variety of stationary applications e.g., distributed, low pollution electricity generation from fuel cells [4]. It could also be used for mobile applications (e.g., on-board generation of hydrogen for fuel cell powered vehicles) and for refueling applications (stationary sources of hydrogen for vehicles).

A previous paper [5] presented results of plasma catalytic conversion of methane, using partial oxidation. In this section, more recent results are very briefly described. Figure 1 shows the hydrogen yield as a function of the specific energy consumption (electrical power required for a given hydrogen throughput). The best results are 95% yield at a specific energy consumption of 13 MJ/kg H₂. These results were obtained without the use of a heat exchanger. It is estimated that with a heat exchanger and with improved thermal management, the specific energy consumption can be decreased to 7 MJ/kg H₂ (0.18 kWhr/m³ H₂) at slightly higher yield (97%). Equivalently, 20 kW of H₂ gas are produced per kW electric power into the plasmatron. These numbers will be assumed in the calculations below.



Figure 1. Hydrogen yield as a function of specific energy consumption for plasma catalytic reforming of methane.

A schematic diagram of a plasmatron is shown in Figure 2. The arc is generated between the water-cooled cathode with a hafnium tip, and a water-cooled tubular anode. The plasmatron gas has a large vorticity in order to spin the arc, centering the cathode arc root in the hafnium tip, and rotating the anode spot. The natural gas was injected downstream from the arc, as shown in Figure 2.

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Figure 2. Schematic diagram of the plasmatron.

Both gas phase reactions as well as catalytic reactions were studied for air and air/water vapor mixtures injection into an air plasma. In the heterogeneous experiments, nickel based catalyst on alumina support was used. The catalyst (United Catalyst C-11) came in rings, and best results were obtained with crushed catalysts to 5 mm average size.

Drinking water was the source of the steam. The gas analysis was performed using a HP M200D GC, with two columns and with two thermal conductivity detectors. Calibration gases were provided by Matheson Gas.

3 Costing model

The cost of the hydrogen produced through a plasmatron process is investigated in this section. The flow diagram is shown in Figure 3. It is assumed that the hydrogen rich gas is produced at pressure so that it is not necessary to pressurize the hydrogen rich gas prior to the pressure swing absorption section. Air, not oxygen, is used as the oxidizer, which means that the synthesis gas contains a large amount of nitrogen. It is also assumed that methane is the hydrocarbon used, although other hydrocarbons can also be utilized. Further assumptions are that the system has good thermal management, and the power consumed by the plasmatron as well as the exothermic energy released by the partial oxidation process are used to produce the steam required for the system. Steam is required mainly in the water-shift reaction. There is an excess of steam for the steam shifter of 200%.

The capital costs of the system, for a 571 m^3 /hr (20000 scfhr) are shown in Table 1. The corresponding plasmatron power is 90 kW. The syngas is at pressure, and there is no

need for boiler to produce steam since it can be produced by the syngas itself prior to the water shift reaction, which occurs at relatively low temperatures. If the plant is amortized over 15 years, then the amortized capital cost is on the order of \$150k per year.



Figure 3. Flow diagram of plasmatron fuel converter based hydrogen generation system

Air compressor	\$100,000
Reforming Reactor	\$5,000
plasmatron	\$50,000
convertor (power supply)	\$18,182
spares	\$100,000
Boiler package	\$30,000
Shift Reactor/cooler/KOD	\$20,000
H2 PSA	\$200,000
Feed filter/KOD	\$3,500
Controls/Instrumentation	\$90,000
Engg./Skidding/Packaging	\$65,000
Cap cost total	\$681,682
Installation (10% cap)	\$68,168
Plant Cost (Installed)	\$749,850
Cost per year, amortized over 15 years	\$149,970

Table 1. Capital costs for system with 571 m³/hr

The operational costs depend on the number of personnel operating the plant. Since the high power plasmatron is a well-established technology, it is assumed that the system is automated and needs minimal supervision. Plasmatron maintenance, consisting in replacing the electrodes, needs to be performed every 1000 hours of operation, and lasts only a few minutes. During plasmatron maintenance, the system can continue to operate

at reduced level. Several plasmatrons are used in the system, and electrode replacement is carried out one by one.

The power requirements are shown in Table 2. The syngas is at pressure when leaving the plasmatron reactor. Air compression requires more power than the corresponding oxygen compression. The power requirements are dominated by the plasmatron power requirements of 90 kW.

Table 2. Power consumption in 571 m^3/hr plant

Air compressor power, kW	130
Natural gas compressor power, kW	0
Syngas compressor power, kW	0
Plasmatron power, kW	91
Total Power, kW	221

The cost of the natural gas is assumed to be \$2.5/MMBTU, and the electricity cost is 0.04/kWhr. Assuming that 0.5 full time people (all shifts) are required in the plant (at a cost of \$70000/person-year), then the annual costs are given in Table 3, for capacity of $1 \text{ m}^3/\text{hr}$ of H₂ gas.

Table 3. Annualized costs for a 571 m³/hr (20000 scfh) hydrogen plant

Labor	\$35,000
Catalyst	\$10,000
Power	\$73,636
Natural gas	\$221,952
Total Op. + Util.	\$340,588
Total cost Op+util+cap	\$490,558

With an annual hydrogen production of 60k MMBTU, then the cost of the hydrogen is about \$8.5/MMBTU. This is a very preliminary look at the cost, and more detailed calculations will be presented at a later time.

4 Sensitivity of hydrogen cost

The dependence of the hydrogen costs on the plant capacity, capital cost, the methane cost and the required personnel is described in this section.

The model can be used to determine the cost of the hydrogen as a function of the plant capacity. Figure 4 shows the results from this calculations, assuming that the flow diagram remains constant (i.e., no changes in energy recovery or in specific energy consumption by the plasmatron fuel converter). It is assumed that the cost of the compressor, the reformer reactor, the plasmatron and its power supply, the shift reactor, the PSA unit and the feed filter scale as the 0.7 power of the plant capacity. The control and instrumentation scales as 0.2 power of the plant capacity, and the engineering/skidding/packaging scales as the 0.3 power of the plant capacity. In addition,

it is assumed that the personnel requirement scales as the 0.3 power of the plant capacity. The cost of hydrogen decreases from about \$8.5/MMBTU to about \$6.8/MMBTU in the range considered. The cost of the hydrogen, in the range considered, scales inversely as the 0.1 power of the plant capacity. Above 3500 m³/hr, the cost model needs to be revised.



Figure 4 Cost of hydrogen as a function of the plant capacity.

The sensitivity of the cost of hydrogen to the cost of natural gas in shown in Figure 5. The cost of the hydrogen is about \$5.5/MMBTU more expensive than the cost of the natural gas. For free natural gas, the cost of the hydrogen is about \$4.5/MMBTU.



Figure 5. Hydrogen cost as a function of natural gas cost, for a 571 m³/hr (20000 scfh) plant capacity

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From Table 3, it can be seen that about half the cost of the hydrogen is due to the natural gas, with the electrical power about 1/3 of the natural gas cost. Increasing the cost of the capital equipment by a factor of 2 increases the hydrogen cost from \$8.2/MMBTU to \$10.7 /MMBTU, for 571 m³/hr plant capacity. For the same plant capacity, increasing the cost of electricity to \$0.08/kWhr increases the cost of hydrogen to \$9.5/MMBTU.

5 Tradeoff between methane conversion and specific energy requirement

As mentioned in the sections above, there is a tradeoff between the methane conversion and the hydrogen yield. A simplified curve illustrating this tradeoff is shown in Figure 6. For a specific energy consumption less than about 0.17 kWhr/m^3 there is a sharp drop in the methane conversion. At higher specific energy consumption, the methane conversion does not increase much.



Figure 6. Methane yield and cost of hydrogen as a function of the specific energy, for $571 \text{ m}^3/\text{hr}$ plant capacity

Figure 6 also shows the hydrogen cost as a function of the specific energy consumption. All the other parameters are the same as in the previous sections. There is a well-defined minimum in the cost. To the left of the minimum, the hydrogen yield is decreasing because of insufficient methane conversion, and to the right of the minimum the electrical cost is increasing faster than the hydrogen yield. It is important to determine experimentally the minimum cost.

6 Discussion and conclusions

Plasma reforming of natural gas has been briefly described. A process involving plasma reforming of natural gas for the production of hydrogen is then modeled to determine the

cost and the cost-sensitivity of hydrogen produced by the process. Although preliminary, the costs indicate that plasma reforming may be a means for hydrogen production at competitive costs, even in small capacity hydrogen generating units.

Several assumptions need to be demonstrated experimentally. It is necessary to demonstrate the lower specific energy consumption assumed in the paper, by the use of better thermal management and heat recovery. It is necessary to operate the plasmatron at high pressure, to minimize the required pumping power. This could have substantial effect in the case of liquid feedstock, since it requires little power to pressurize the liquid but substantial power to pressurize the reformate. If electrode life or other effects limit the plasma reformer operation at high pressure, then a reformate compressor needs to be included in the system. Heat recovery requires a heat exchanger that was not included in the cost calculations.

The lifetime of the catalyst is another unknown. Sulfur in the feedstock is readily transformed into H_2S which needs to be removed from the system but which does not affect the catalyst. The costing model is continuously being improved.

Acknowledgements

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