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A Review Paper on Heat exchangers used for PROX reactor

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Abstract — *The PROX reaction process occurring in the reactor with 1%(wt) platinum based on iron hydroxyphosphate was analyzed between temperature range of 80 ^0C and 120 ^0C in hysys software. A shell and tube type heat exchanger was designed and analyzed for temperature control for PROX reaction, containing copper tubes. The temperature profile suggests the expected temperature rise in the PROX reactor and temperature drop in the heat exchanger. The heat exchanger was able to bring down the temperature of gases from 210 ^oC to 113 ^oC successfully. The gas composition sent to the PROX reaction process was 40% H_2, 1% CO, 9% CO_2, 0.5% O_2, and 49.5% Ar. The result suggests that the removal of heat from the gases increases the CO selectivity and conversion.*

This paper is a review on different heat exchanger used for PROX reactor.

I. Heat Exchangers for Temperature Control

Holding the temperature in the reactor near the ideal value is the key to achieving peak performance. Proper heat transfer is necessary to remove the heat generation from the reactions. Several methods of heat removal have been investigated with varying success [7, 17-21]. Each case, however, indicates an improvement over a similar reactor without heat removal. The ideal reactor is one where every point inside the reactor is held at the optimal operating temperature. This reactor should be small and light weight relative to the volume flow rate that moves through it, and scalable to the flow rate required for transport vehicles. The reactor should also lend itself to thermal integration with the total fuel cell system so that the heat generated by the reaction can be utilized by another component of the system.

1.1 Isothermal Reactor

A PROX silicon microreactor developed by Ouyang et al. [7] closely simulates an isothermal reactor. The microreactor is a single channel (500 μm width x 470 μm depth x 4.5 cm length) in a silicon chip capped with PyrexTM glass. A thin film of Pt/Al_2O_3 catalyst was deposited on the channel walls. Each point within the reactor was maintained at the same temperature because the generated heat was quickly conducted through the reactor walls. Experimental data shown in Figure displays near 100% CO conversion and 40% CO selectivity achieved at temperatures between 180 and 220 °C. A numerical model was developed to compare the performance of the microreactor and two mini packed-bed tube reactors (m-PBRs). The m-PBRs had a tube diameter of 2 mm and 4 mm. The gas stream was exposed to equal amounts of catalyst by weight in each reactor. The results of the model are shown in Figure. The model predicted severe hot spots in the m-PBRs (up to 300 °C higher than the wall temperature) and a decrease in CO conversion at higher temperatures due to r-WGS activity. The hot spots were attributed to heat transfer limitations. CO conversion in the microreactor remained near 100% at high temperatures. Hot spots were avoided in the microreactor because of the short heat transfer path between the catalyst and the channel wall. The absence of hot spots greatly widened the optimal temperature window for the microreactor by delaying the onset of the r-WGS reaction.

The isothermal condition maintained in the microreactor is dependent on coolant flowing through channels in an interface block pressed against the silicon chip. Practical applications would require many reactant and coolant channels densely packed into a unit volume. The concept could work well if all silicon and glass material was removed from the chip except for material directly around the reactant channels, and a cooling fluid was made to flow around the dense array of reactant channels in the space left open by the removed material. The device would then be a heat exchanger with catalyst deposited on the surfaces seen by the gas stream. Fortunately, the channel walls do not need to be made of silicon. Instead, the walls can be made of metal like stainless steel, nickel, or even the material that is the catalyst. This promising concept presents an interesting manufacturing challenge in the form of fabrication and assembly of the micro channels. Future research should develop a micro channel heat exchanger PROX reactor with the catalyst coated directly on the channel walls.

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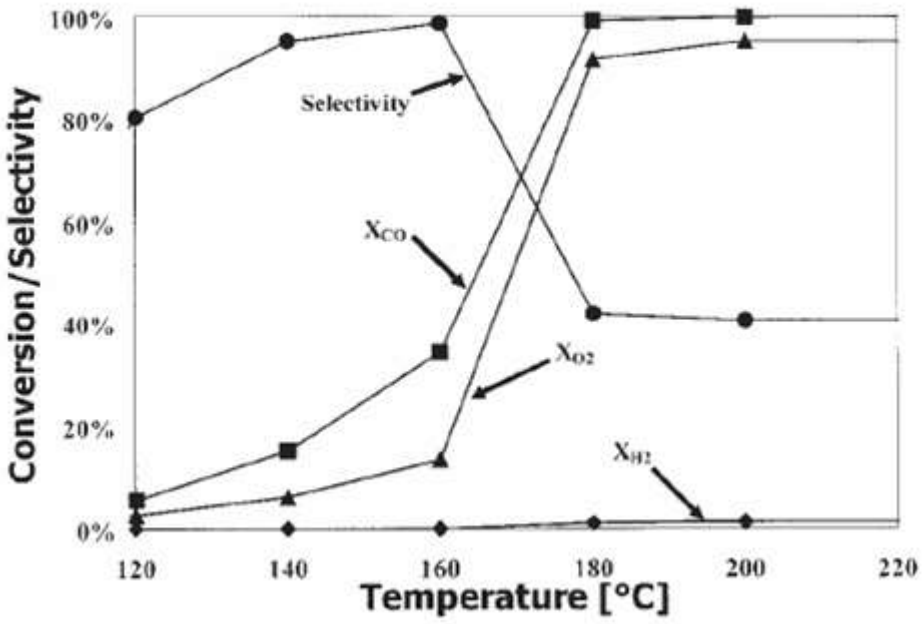

Figure 1.1.1: Experimental results for the silicon microreactor, conversion of CO, O_2, H_2, and CO selectivity vs. reactor temperature [7].

1.2 Multi Stage PROX Reactor with Plate Fin Heat Exchangers

Heat exchangers have been used to control the temperature of the gas stream by removing the heat generated during the PROX reaction. A working multistage PROX reactor reported by Pan et al. [18] demonstrates the feasibility of thermal integration and high thermal efficiency. The researchers cite the importance of temperature control and use plate fin heat exchangers to remove heat from the reactions. Catalyst was wash coated on ceramic monoliths in the reactor sections. The reactor layout and a diagram of the plate fin heat exchangers are shown in Figure. The temperature rise in each stage was independently controlled by regulating the oxygen input to each catalyst bed. Heat generated in the PROX reactor was used to preheat the methanol fuel (used as coolant in the heat exchangers) before the methanol was sent to the reformer upstream of the PROX reactor. The PROX system displayed the saw tooth axial temperature profile illustrated in Figure. CO was reduced from 1.5% to concentrations below 50 ppm which was the minimal requirement for the CO tolerant PEMFC used in the study. CO selectivity was not reported but the overall fuel processor (the reformer and the PROX reactor) efficiency was maintained above 75% throughout 1000 hours of operation. The researchers defined efficiency as the LVH (lower heating value) of H_2 exiting the system divided by the LVH of methanol entering.

Based on the reported 34.15 ml/min liquid methanol inlet flow rate, and 75% efficiency, the fuel processor should produce approximately 0.04 moles/sec of H_2. The exact flow rate of gasses though the PROX reactor was not reported. Coupled with a PEMFC with 50% efficiency the electrical power output would be 5 kW. The complete fuel processor was 68 cm x 50 cm x 40 cm and weighed 40 kg. The total PROX reactor volume was 1.2 liters not including the heat exchangers but the weight was not reported. This successful fuel processing system validates the incorporation of heat exchangers in a PROX reactor.

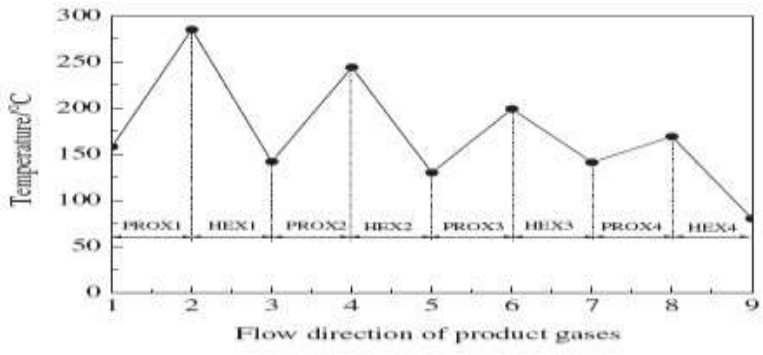

Figure 1.2.1: Axial temperature profile in the four stage PROX reactor

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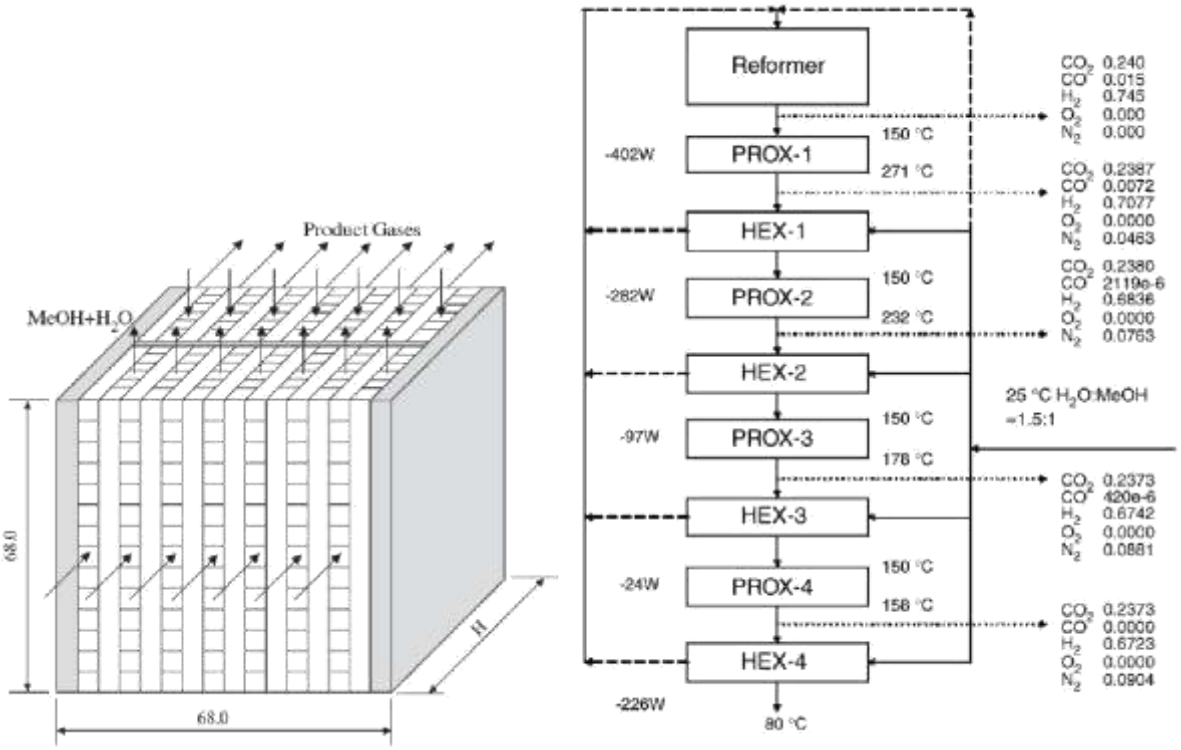

Figure 1.2.3: Diagram of the plate fin heat exchangers and a simplified layout of the fuel processor

1.3 Plate Fin Heat Exchanger PROX Reactor

Dudfield et al. [19] compared the performance of three PROX reactor designs that all feature active heat removal from the reaction zone. Photographs of the reactors are shown in Figure. A shell and tube heat exchanger was filled with porous micro spheres coated with catalyst. The shell and tube reactor had the advantage of being easy to recharge with fresh catalyst. The second reactor was made by compressing and sintering irregular stainless steel granules around a heat removal element. Characteristics of the elements were not reported. The sintered metal was then washcoated with catalyst. The third reactor was a plate fin heat exchanger fabricated from thin aluminum. The reactor design provided a surface area to volume ratio of 1500 m^2/m^3 in the reaction chambers. The walls of the reaction zone inside the heat exchanger were washcoated directly with catalyst. Heat generated by the reaction was therefore quickly transferred to the coolant on the other side of the wall.

The plate fin reactor proved superior to the other designs in terms of lower pressure drop, greater temperature control, and higher CO conversion. A second set of tests found that by combining two plate fin reactors in series (doubling the reactor volume), CO concentrations could be reduced from 7000 ppm to less than 12 ppm. Results are displayed in Figure. The total reactor volume was 4 liters and was capable of processing enough gas for a 20 kW PEMFC. The performance relative to reactor volume is comparable to the system developed by [18] except lower exit CO concentrations were achieved. The performance reported by [19] is due to an active catalyst, high surface area to volume ratio, and thermal management. Volume could be further reduced by increasing the surface area to volume ratio.

1.4 PROX Reactor with Flat Panel Heat Exchangers between Metal Foams

Brooks et al. reported a multi section reactor similar to the one developed by where heat exchangers are placed between catalyst sections for temperature control. A total of four catalyst chambers where used with flat panel microchannel heat exchangers inserted between each bed. A drawing of the reactor design is shown in Figure. The report did not describe details about the heat exchanger geometry or performance other than that they provide excellent temperature control per unit volume and low pressure drop. The sequence of catalyst beds and compact heat exchangers allowed for efficient use of space with the catalyst region occupying 70% of the reactor volume (excluding the reactor shell and piping).

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Additional control was achieved by adding oxygen to each stage rather than adding all oxygen at the reactor inlet. The catalyst bed length was shortest in the first bed where heat generation was greatest and increased in each bed downstream. Similarly, catalyst loading was lowest in the first bed and increase in each following beds because activity decreased along the reactor length as the total O2 concentration decreased. An otherwise unidentified non-precious metal catalyst was used in the first three beds to reduce CO concentrations from 1% to 300 ppm. A precious metal catalyst was used in the last stage to bring CO concentrations below 10 ppm. Metal foam was used as a catalyst support because of its high thermal conductivity. The foam was in direct contact with the heat exchangers so that conduction heat transfer through the metal as well as convection through the gas stream could aid the temperature control effort in the reactor. Oil was used as coolant in the heat exchangers. The reactor performed well in experiments by successfully reducing CO from 1% down to below 10 ppm while maintaining selectivity at around 50%. The CO concentration as well as the O_2/CO ratio in each section is plotted in Figure. GHSV was 93000 for the whole reactor. The flow contained the equivalent of 2 kW in hydrogen (the power released by oxidizing the hydrogen stream). The temperature profile in the reactor is plotted in Figure. Temperature variation in the reactor is about +/- 10 °C.

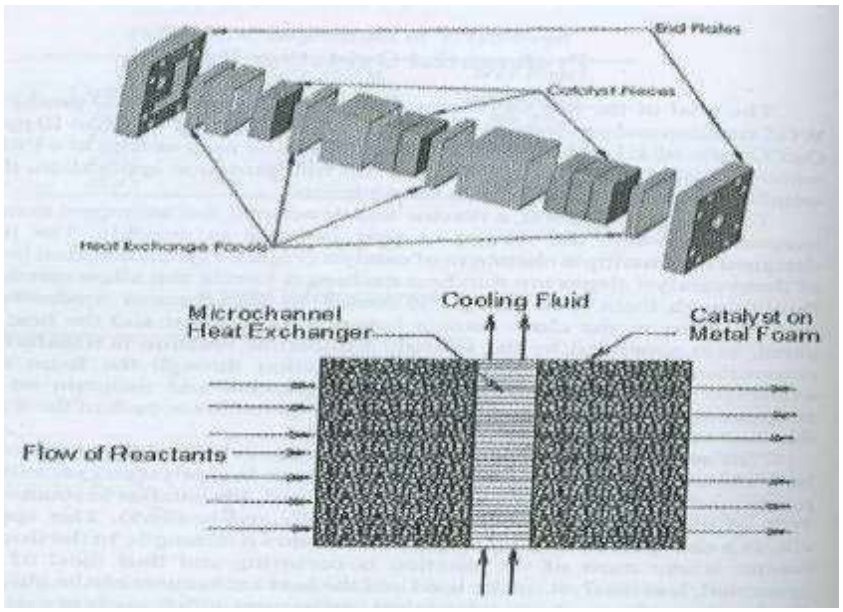

Figure 1.4.1: Multi step PROX reactor. Flat panel heat exchangers between metal foam sections for temperature control

1.5 Comparison of Reactor Designs

Reactor Design	Multi stage with plate fin heat exchanger	Plate fin heat exchanger	Microchannel Heat exchanger	Multi stage with microchannel heat exchanger
Catalyst	Platinum	Pt-Ru	Pt-Ru/Al_2O_3	Precious metal
Catalyst Support	Monolith	Heat exchanger wall	Channel walls	Metal foam
Oxygen addition	At each stage	At all inlet	At all inlet	At each stage
Reactor Volume (L)	>1.2	4	0.06	7
Reactor mass	-	-	0.15	6.6
Inlet CO Conc. (ppm)	15000	7000	5000	10000
Outlet Co Conc.(ppm)	50	12	10	10
CO Conversion (%)	99.7	99.8	99.8	99.9
CO Selectivity (%)	-	-	-	50

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