A Modeling and Experimental Study of CO_2-Selective Water-Gas-Shift Membrane Reactor for Fuel Cells

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Abstract

Improving water gas shift (WGS) reaction for hydrogen purification is of particular interest for the application of fuel cells. As a reversible and exothermic reaction, the conventional WGS reactor is not efficient, resulting in a high concentration of unconverted CO (about 1%) in the H_2 product and a bulky, heavy reactor. For CO cleanup, current known approaches include methanation and preferential oxidation. But, both consume a significant amount of hydrogen and add additional steps. However, using a CO_2-selective membrane reactor shifts the WGS reaction towards the product side, which enhances the conversion of CO and increases the purity of the H_2 product at high pressure. In addition, air can be used as the sweep gas to remove the permeate, CO_2, on the low-pressure side of the membrane to achieve a high driving force for the separation. This presentation will report on a modeling and experimental study of the membrane reactor.

In the modeling study, the WGS membrane reactor was configured to be a hollow fiber membrane module with catalyst particles packed inside the fibers. A one-dimensional non-isothermal model was developed to simulate the reaction and transport process with countercurrent gas flows. The reaction rate equation for the Cu/ZnO catalyst from literature was incorporated into the model. The synthesis gases with different CO concentrations from autothermal reforming of gasoline with air were used as the feed gas, while heated air was used as the sweep gas. The modeling results showed that the exit CO concentration of less than 10 ppm was achievable. The effects of several important system parameters including inlet feed temperature, inlet sweep temperature, feed-side pressure, catalyst activity, and feed inlet CO concentration were investigated. As the inlet feed temperature increased, the membrane area requirement decreased. Fig. 1 and 2 show the effects of inlet feed temperature on the required membrane area and the corresponding feed-side temperature profiles, respectively. The increase of inlet sweep temperature resulted in more significant reduction of the required membrane area because the feed-side temperature was affected over a longer reactor length. However, overly high temperatures would be unfavorable to the exothermic and reversible WGS reaction. The effects of inlet sweep temperature and the related feed-side temperature profiles are demonstrated in Fig.3 and 4, respectively.

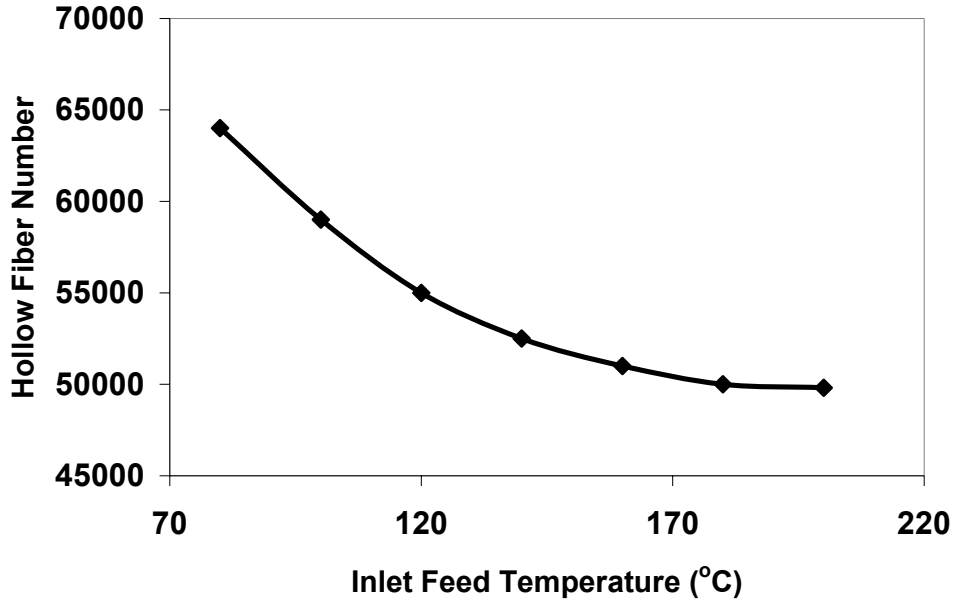

Figure 1. The effect of inlet feed temperature on required membrane area.

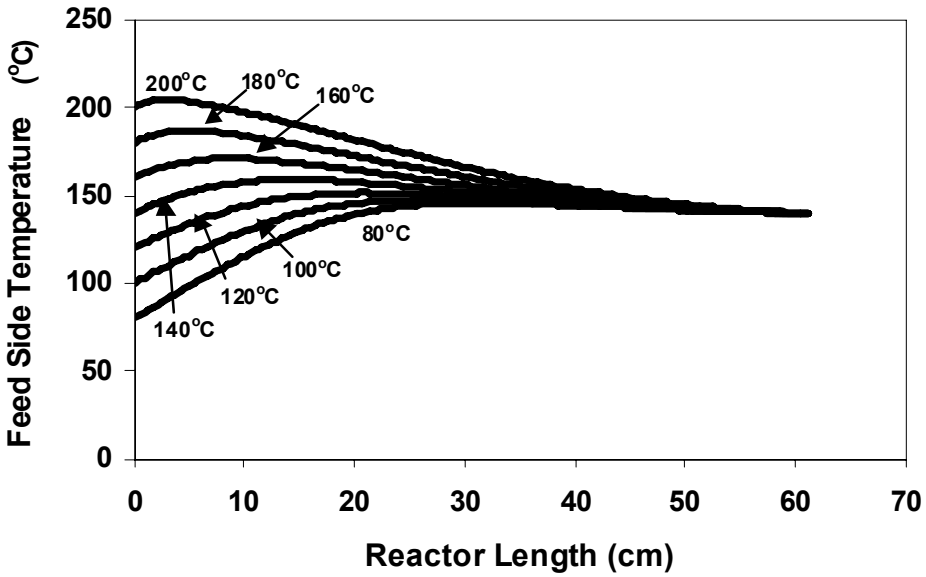

Figure 2. Feed-side temperature profiles along the length of membrane reactor with different inlet feed temperatures.

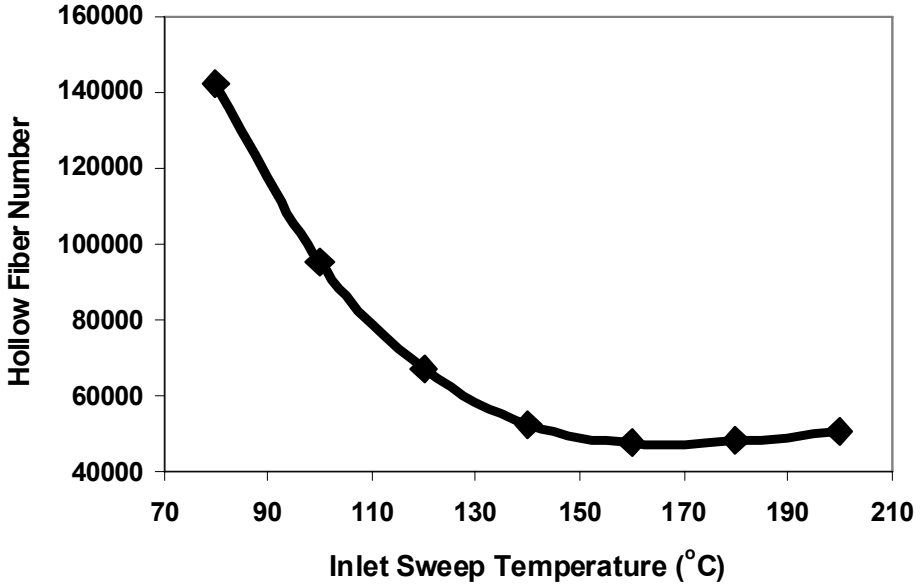

Figure 3. The effect of inlet sweep temperature on required membrane area.

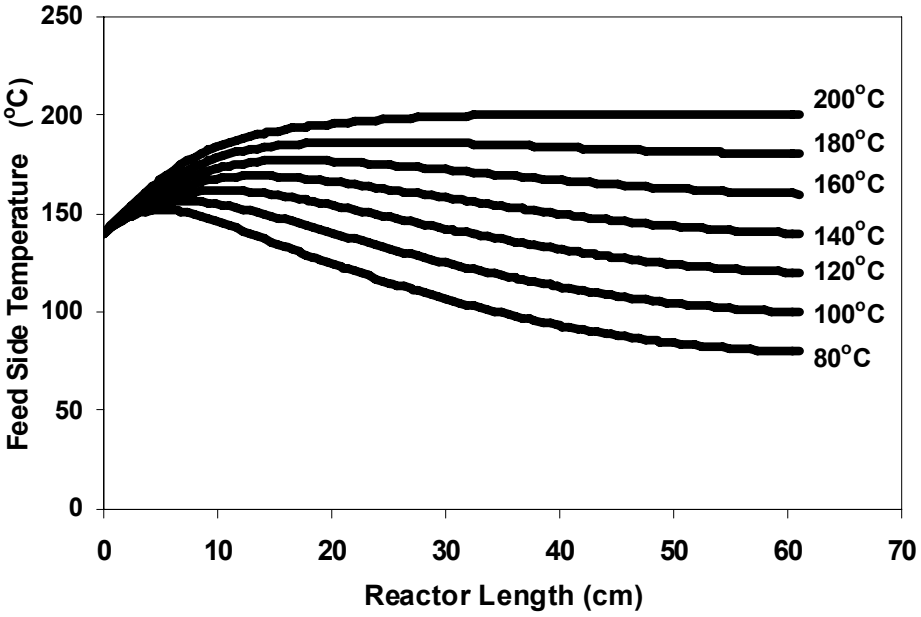

Figure 4. Feed-side temperature profiles along the length of membrane reactor with different inlet sweep temperatures.

With the increase of feed-side pressure, the membrane area requirement decreased as the result of higher WGS reaction rate and CO_2 partial pressure difference. Increasing catalyst activity made the WGS reaction faster and enhanced the permeation driving force, which was similar to the effect of feed-side pressure. In addition, the modeling study showed that the CO_2-selective membrane reactor could be used for syngases with high CO concentration, e.g. 5% or 10%.

In the experiments, a rectangular flat-sheet membrane reactor using the commercial Cu/ZnO catalyst was set up. Fig. 5 shows the schematic of this membrane reactor. The gas mixture with the composition of an autothermal reforming syngas (1% CO, 45% H_2, 17% CO_2, and 37% N_2) was used as the feed gas, and Ar or air was used as the sweep gas (using Ar for the ease of GC analysis).

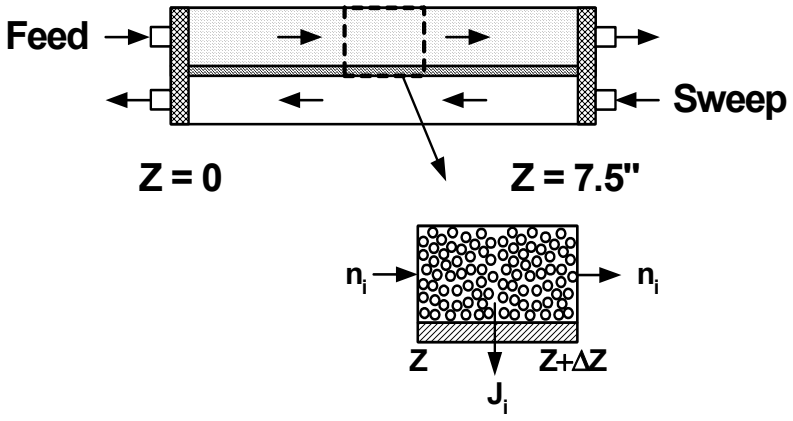

Figure 5. Schematic diagram of flat-sheet WGS membrane reactor

By varying the feed flow rate, the performance of the reactor was investigated. The experimental data agreed well with the modeling results based on the same geometrical dimensions of the reactor. A CO concentration of less than 10 ppm and a H_2 concentration of greater than 52% (on the dry basis) were achieved. The comparison between the modeling curve and experimental data is shown in Fig. 6.

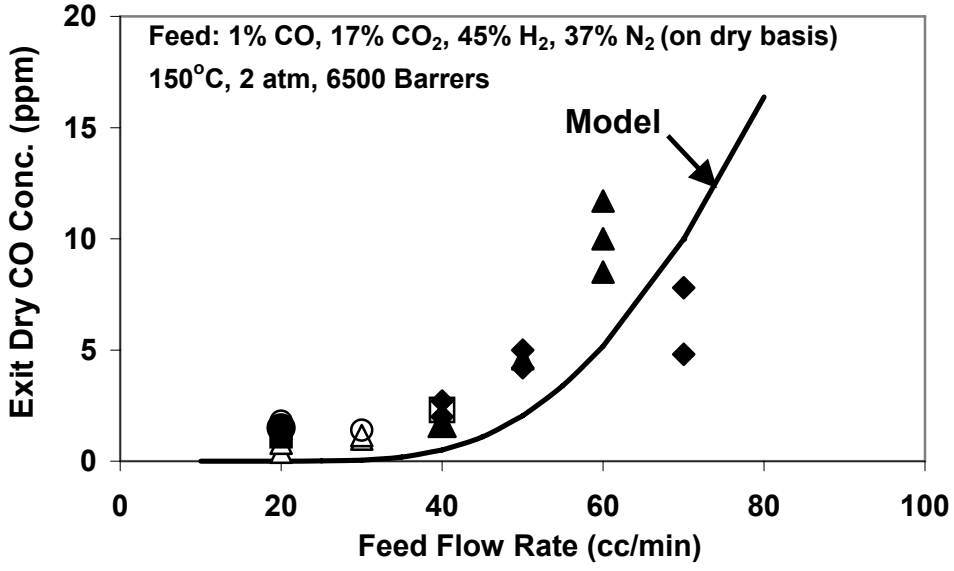

Figure 6. Experimental data agreed well with the modeling results.