

A HYBRID ADSORBENT-MEMBRANE REACTOR (HAMR) SYSTEM FOR HYDROGEN PRODUCTION

Aadesh Harale¹, Hyun Hwang¹, P.K.T. Liu², Muhammad Sahimi¹, and Theodore Tsotsis^{1*}
¹Mork Family Department of Chemical Engineering and Materials Science, University of Southern California, HED216, Los Angeles, CA 90089-1211, (USA)
²Media and Process Technology, Pittsburgh, PA, 15236, (USA)
*tsotsis@usc.edu

Introduction

As a result of stricter environmental regulations worldwide, hydrogen is progressively becoming an important clean energy source. For hydrogen to replace fossil fuels for mobile applications, it will require the creation of a production and delivery infrastructure equivalent to that currently existing for fossil fuels, which is an immense task. As an alternative, and an interim step towards the new hydrogen economy, various groups are currently investigating hydrocarbon steam reforming of methane (SRM) for *on-board* generation of hydrogen (for use in fuel-cell-powered vehicles), or for *on site* production, in place of compressed or liquid hydrogen gas storage [1-4]. Conventional technologies are neither convenient nor economical to apply for small-scale (on site or on-board) hydrogen generation. Reactive separation processes have, as a result, been attracting renewed interest for application in hydrogen production through SRM. They include membrane reactors (MR), and more recently adsorptive reactor (AR) processes [5, 6]. MR show substantial promise in this area and utilize nanoporous inorganic or Pd-alloy membranes. The latter are better suited for pure hydrogen production; however, they are expensive, and become brittle during reactor operation, or deactivate in the presence of sulfur or coke. Adsorptive methane steam reforming reactors also show good potential [6]. The challenge here, however, is in matching the adsorbent properties with those of the catalytic system. The development of a novel process is, therefore, needed that overcomes the shortcomings of existing processes.

Proposed Solution

What we propose for use, instead, in place of the more conventional technologies, is a novel reactor system, termed the hybrid adsorbent-membrane reactor (HAMR). The HAMR concept was originally proposed by our group [7, 8] for esterification reactions, and couples the reaction and membrane separation steps with adsorption on the reactor and/or membrane permeate side. Our focus since has shifted to using HAMR systems for hydrogen production, of potential interest to *on-board* or *on-site* hydrogen production applications. Our early studies involved the development of a mathematical model for a HAMR system for hydrogen production through SRM [9]. Our recent focus has been on experimental investigations in order to prove the feasibility of using HAMR systems for hydrogen production, and to validate the design models. We have used both microporous and metal membranes and CO₂ hydrotalcite-type adsorbents, and have studied both the water-gas shift and steam reforming reactions. Experimental data have been compared with the model predictions, and found to be consistent. The HAMR system has been shown to exhibit enhanced methane conversion, hydrogen yield, and product purity.

Results and Discussion

Figure 1 below shows the experimental data for CO conversion for a WGS HAMR reactor using a carbon molecular sieve membrane and a hydrotalcite-type adsorbent. The reactor shows complete conversion, while the adsorbent is still active. After the adsorbent saturates, the conversion settles to the value corresponding to the membrane reactor conditions, which is still significantly higher than the conversion of the conventional packed-bed reactor system and the corresponding equilibrium. Shown on the Figure are also the predictions of the HAMR model using no adjustable parameters. The agreement between model and experiments is again satisfactory. Additional experimental data for the SRM reaction will be presented at the meeting.

Significance

We describe here the development of a novel reactor system, termed HAMR, for application to *on-site* or *on-board* hydrogen production. Its characteristics have been investigated for a range of temperature and pressure conditions, relevant to the aforementioned applications, and are compared with the predictions of a mathematical model. The HAMR system is shown to exhibit enhanced methane conversion, hydrogen yield, and product purity, and good promise for reducing the hostile operating conditions of conventional methane-steam reformers, and for meeting the hydrogen purity requirements for PEM operation.

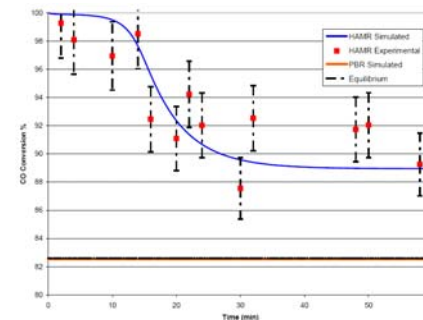


Figure 1. CO Conversion vs. time

References

- (1) Choi, Y.; Stenger, H.G. *J. Power Sources* 124, 432 (2003).
- (2) Darwish, N. A.; Hilal, N.; Versteeg, G.; Heesink, B. *Fuel* 83, 409 (2003).
- (3) Liu, Z.; Roh, H.; Park, S. *J. Power Sources* 111, 83 (2002)
- (4) Semelsberger, T. A.; Brown, L. F.; Borup, R. L.; Inbody, M. A. *Int. J. Hydrogen Energ.* 29, 1047(2004)
- (5) Elnashaie, S.S.E.H.; Adris, A.; Al-Ubaid, A.S.; Soliman, M.A. *Chem. Eng. Sci.* 45, 491(1990)
- (6) Han, C.; Harrison, D.P. *Chem. Eng. Sci.* 49, 5875(1994)
- (7) Park, B. Ph.D. Thesis, University of Southern California, Los Angeles, California, 2001.
- (8) Park, B.; Tsotsis, T. T. *Chem. Eng. Proc.* 43, 1171(2004)
- (9) Fayyaz B.; Harale, A.; Park, B.G.; Liu, P.K.T.; Sahimi, M.; and Tsotsis, T.T. *Ind. Eng. Chem. Res.*, 44 (25), 9398 (2005)