# REFORMING OF LIQUID HYDROCARBONS IN A NOVEL HYDROGEN-SELECTIVE MEMBRANE-BASED FUEL PROCESSOR

**PI:** Shamsuddin Ilias

**Student(s):** M.A. Islam

**Institution:** North Carolina A&T State University

**Address:** Department of Mechanical and Chemical Engineering
314 McNair Hall, Greensboro, NC 27411

**Telephone:** (336) 334-7564 ext. 317 **Fax:** (336) 334-7904 **E-mail:** ilias@ncat.edu

**Grant Number:** DE-FG26-01NT41361 **Performance Period:** 09/30/2001 – 09/29/2005

## ABSTRACT

The major focus on PEMFC technology is to develop fuel cell system for transportation applications, which require development of low cost cell components and reliable, high-purity $H_2$-fuel source. The PEMFC technology is attractive because of its low operating temperature and ease of start-up. Reformed methanol and liquid hydrocarbons are expected to be major fuel source in PEMFCs for terrestrial transportation application as envisioned in Vision 21 for the 21st century. The poisoning of the expensive electrocatalysts by CO in the reformed fuel is a major concern. Crossover of methanol in direct methanol PEMFC is also problematic. Thus, there is a need for developing technology for reforming of methane, methanol and liquid hydrocarbons as a source of hydrogen to the fuel cells. In this context, membrane-based technology as fuel processor appears very attractive.

We propose to develop an inorganic metal-metal composite membrane to study reforming of methane, liquid hydrocarbons and methanol by equilibrium shift in membrane-reactor configuration, viewed as fuel processor. Based on our current understanding and experience in the Pd-ceramic composite membrane, we propose to further develop this membrane to a palladium-silver alloy membrane on microporous stainless steel support to provide structural reliability from distortion due to thermal cycling and hydrogen embrittlement. Because of the metal-metal composite structure, we believe that the associated end-seal problem in the Pd-ceramic composite membrane in tubular configuration would not be an issue at all. The major objective of this research is to further develop these materials and to test them for reforming liquid hydrocarbons and methanol for simultaneous production and separation of high-purity hydrogen for PEM fuel cell applications.

The proposed membrane-reactor for steam reforming of methane and methanol would be capable of simultaneously producing and separating hydrogen (in nearly pure form due to excellent perm-selectivity of the membrane) for use in PEM fuel cell. Because of single-unit operation, the method proposed will be cost effective, in terms of both capital and energy savings when compared with the conventional reforming process. Furthermore, by selective removal of hydrogen, the reaction could be easily shifted to the right to near 100% conversion as opposed to significantly lower equilibrium conversion in conventional catalytic packed-bed reactor. Thus, the temperature limitations of the equilibrium-limited reaction can be resolved without compromising the conversion.

To investigate the potential application of Pd-ceramic and Pd-stainless steel microporous membrane in membrane-reactor configuration, we investigated the steam reforming of methane.

By electroless deposition under osmotic pressure field, thin film of palladium was deposited on tubular microporous stainless steel and ceramic substrates. The permeability and selectivity of the membranes were found to be significantly higher than many of the membranes commercially available.

To explore the usefulness of Pd-SS composite membrane in membrane-reactor configuration for simultaneous production and separation of hydrogen, we are investigating the steam reforming of methane by equilibrium shift. To have a better understanding of the membrane reactor, we developed a two-dimensional, pseudo-homogeneous reactor model for steam reforming of methane by equilibrium shift in a tubular membrane reactor. Radial diffusion was taken into account for concentration gradient in the radial direction due to $H_2$–permeation through the membrane. The reaction takes place in the shell side and the product hydrogen is permeated through the membrane radially into the tube side. With appropriate reaction rate expressions, a set of partial differential equations was derived using the continuity equation for the reaction system and then solved by finite difference method with appropriate boundary and initial conditions. An iterative scheme was used to get the converged solution. Membrane reactor performance was compared to that of a conventional fixed-bed reactor. Also conventional non-membrane reactor performance was compared with equilibrium values achievable in a conventional non-membrane reactor. The effect on the degree of conversion was studied parametrically for such factors as temperature, reactor pressure, feed and sweep flow rate, feed molar ratio, and membrane thickness to obtain optimum operating conditions.

**List of Published Journal Articles, Completed Presentations and Students Receiving Supports from the Grant:**

**Presentations:**

Khan, M.H., Rahman, M.S., and **Ilias, S**., "A Study on Steam Reforming of Methane in a Pd-Stainless Steel Tubular Membrane Reactor," Accepted for presentation at the 8th International congress on Inorganic Membranes (ICIM8), Cincinnati, Ohio, July 18 –22, 2004.

**Ilias, S.,** Rahman, M.S., and Ahmed, S.J., "A Study on Steam Reforming of Methane in Pd-Stainless Steel Composite Membrane-Reactor," Proc, 13th Symposium on separation Science and Technology for Energy Applications, Oct 27-30, 2003, Gatlinburg, Tennessee, Abstract. P. 22 (2003).

**Ilias, S**., "Reforming of Hydrocarbons in a Novel Hydrogen-selective Membrane-based Fuel Processor," 3rd Department of Defense Logistic Fuel Reforming Conference, Panama City Beach, Florida, August 27-28, 2002

**M.S. Theses:**

M.S. Rahman (2003): *Hydrogen Permselective Palladium/Porous Stainless Steel Composite Membrane Reactor for Shifting Equilibrium of Steam Methane Reforming,* North Carolina A&T State University, Greensboro, North Carolina.

Syeda J. Ahmed (2003): *Fabrication and Characterization of Palladium/Stainless Steel Composite Membrane for Hydrogen Separation,* North Carolina A&T State University, Greensboro, North Carolina.

**Students Receiving/Received Supports:**

Syeda J. Ahmed, M.S. Rahman, M.A. Islam