

PROJECT facts

Advanced Research

05/2007

U.S. DEPARTMENT OF ENERGY
OFFICE OF FOSSIL ENERGY
NATIONAL ENERGY TECHNOLOGY LABORATORY



DEVELOPMENT OF PALLADIUM-SILVER COMPOSITE MEMBRANES FOR SEPARATION OF HYDROGEN AT ELEVATED TEMPERATURE

Description

The U.S. Department of Energy (DOE) Office of Fossil Energy (FE) awarded a three-year grant to the North Carolina Agricultural & Technical State University (NCAT) to fabricate thin-film palladium-silver (Pd-Ag) alloy composite membranes on microporous stainless steel (SS). The purpose of this research is to separate hydrogen (H_2) from liquid or gaseous fuels at elevated temperatures, and subsequently to demonstrate them in a membrane reactor for steam reforming of methanol to produce high-purity hydrogen for fuel cell usage. The development of a high-temperature membrane reactor is designed to combine the reforming reaction and hydrogen separation in a single operation. Successful development and demonstration of a membrane reactor is expected to increase hydrogen production by overcoming equilibrium limitations experienced in conventional reforming followed by purification reactor configurations, and will improve process economics by reducing capital costs associated with use of multiple reactors.

The grant is administered by the Advanced Research Program of the National Energy Technology Laboratory (NETL) under the Historically Black Colleges and Universities and Other Minority Institutions (HBCU/OMI) Program. HBCU/OMI encourages minority participation in fundamental research to develop technologies that promote the efficient and environmentally safe use of coal, oil, and natural gas resources.

The U.S. energy industry is undergoing a profound transformation driven by changes such as ever-increasing dependence on imported oil and natural gas, more stringent environmental standards and regulations, global climate change concerns, and other market forces. Fuel cell technology is attracting interest as one possible way of helping counter these changes, offering high power density, high efficiency, and almost zero emissions to the environment. Major potential fuel sources for fuel cells include hydrogen derived by reforming of methanol as well as liquid and gaseous light hydrocarbons derived from coal gasification processes. Membrane-based technology is a key technology for separating a pure hydrogen gas stream from fuel mixtures.

Objectives

The overall project objective is to develop an inorganic metal-to-metal composite membrane that will enable the NCAT researchers to study reforming of liquid hydrocarbons, such as methanol, while simultaneously separating a pure hydrogen gas stream. The process involves an equilibrium shift in a membrane-reactor configuration. The membrane-reactor concept is for an integrated fuel processor, which could be used in the formation and separation of hydrogen at elevated temperatures.

CONTACTS

Robert R. Romanosky

Technology Manager
Advanced Research
National Energy Technology
Laboratory
3610 Collins Ferry Road
P.O. Box 880
Morgantown, WV 26507-0880
304-285-4721
robert.romanosky@netl.doe.gov

Arun C. Bose

Project Manager
National Energy Technology
Laboratory
626 Cochran's Mill Road
P.O. Box 10940
Pittsburgh, PA 15236-0940
412-386-4467
arun.bose@netl.doe.gov

Shamsuddin Ilias

Principal Investigator
North Carolina A&T State
University
Department of Mechanical and
Chemical Engineering
1601 East Market Street
Greensboro, NC 27411
336-334-7564
ilias@ncat.edu



PROJECT DURATION

Start Date

08/05/05

End Date

08/31/08

COST

Total Project Value

\$199,996

DOE/Non-DOE Share

\$199,996 / \$0

ADDRESS

National Energy Technology Laboratory

1450 Queen Avenue SW
Albany, OR 97321-2198
541-967-5892

2175 University Avenue South
Suite 201
Fairbanks, AK 99709
907-452-2559

3610 Collins Ferry Road
P.O. Box 880
Morgantown, WV 26507-0880
304-285-4764

626 Cochran's Mill Road
P.O. Box 10940
Pittsburgh, PA 15236-0940
412-386-4687

One West Third Street,
Suite 1400
Tulsa, OK 74103-3519
918-699-2000

CUSTOMER SERVICE

1-800-553-7681

WEBSITE

www.netl.doe.gov

Technical Approach and Accomplishments

The researchers will fabricate hydrogen-selective membranes in disc (planar) and tubular configurations on a porous SS substrate. An “electroless” bath deposition technique (i.e., one that relies on chemical or autocatalytic coating rather than electrolytic plating to achieve a smooth, corrosion-resistant finish) has been modified to optimize the Pd-Ag metals deposition onto the substrate, and some membranes have been fabricated and tested. The research approach is to study the hydrogen permeation characteristics of the composite membranes using pure hydrogen and mixed gases, and to evaluate the long-term integrity and stability of the fabricated membranes under thermal cycling.

Towards the end of the project, experiments will be conducted on steam reforming of methanol using a shell-and-tube

structured membrane reactor

to produce high-purity hydrogen; and a previously developed reactor model will be validated for steam reforming of methanol. If needed, the model will be further developed to predict the reactor performance.

The membranes fabricated to date are being characterized by scanning electron microscopy (SEM) and energy dispersive X-ray (EDX) to analyze the membranes for surface morphology and structural composition. Researchers are using an existing diffusion cell setup to evaluate the H₂ permeation characteristics of the membranes, and the transmembrane pressures for pure hydrogen and mixed gases. A special shell-and-tube structured membrane module will be designed and fabricated to hold 6 inch Pd-Ag composite membranes to study H₂ permeation and transmembrane pressures.

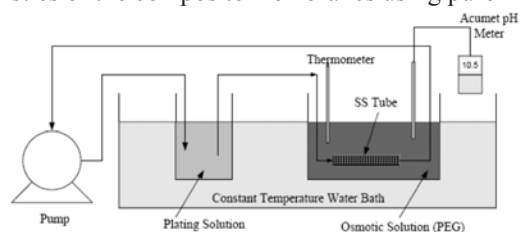
Researchers will test the best performing membranes (at least one planar and one tubular Pd-Ag composite membrane) for long-term stability under thermal cycling. Each membrane selected will be tested over six weeks for H₂ permeation from mixed gases and then will be evaluated by SEM and EDX analysis for any changes in surface morphology and structural composition.

Researchers also will conduct methanol-steam reforming experiments to study the equilibrium shifts and permeation characteristics of the membranes. For these experiments, they will use the tubular diffusion cell setup referred to above as a membrane-reactor-separator with Pd-Ag composite membranes (tubular elements). A Cu/ZnO/Al₂O₃ catalyst will be packed on the tube side of the reactor (reaction side). The produced hydrogen will be transported across the membrane wall to the shell side. The performance of the membrane reactor for the reforming reaction will be examined in terms of conversion and hydrogen production rate by varying the operating conditions.

Benefits

Successful conclusion of this research offers several technological advantages over conventional hydrogen purification methods:

- The reforming reaction is not limited by thermodynamic equilibrium — as soon as hydrogen is formed, it is transported selectively across the membrane;
- Reforming and separation are carried out in a single unit, thereby eliminating the need for separate hydrogen separation and recovery units; and
- The membrane-reactor-separator (fuel processor) is modular and compact in size,



Electroless plating bath setup using polyethylene glycol (PEG) as the osmotic solution