



# Advanced Palladium Membrane Scale-up for Hydrogen Separation

## Background

Among the options being considered to establish greater U.S. independence from foreign energy sources is to increase the use of the nation's domestic coal reserves. The Department of Energy (DOE) is committed to supporting research and development of technologies for the reliable, efficient and environmentally friendly conversion of coal to hydrogen for utilization in advanced gasification-based electric power generation systems. Gasification technologies provide an environmentally responsible and efficient way to utilize coal/biomass feedstocks as precursors of clean burning fuels. The gasification process produces a gaseous mixture of carbon monoxide (CO), carbon dioxide (CO<sub>2</sub>) and hydrogen (H<sub>2</sub>) known as synthesis gas (syngas). The carbon monoxide can be combined with steam to produce additional hydrogen and carbon dioxide (CO<sub>2</sub>). The CO<sub>2</sub> can be removed and stored to reduce greenhouse gas emissions.

One pathway for the separation of H<sub>2</sub> from the syngas is found in membrane technologies. Advances in H<sub>2</sub> membrane separation are critical to the development of advanced energy systems based on coal/biomass processing with CO<sub>2</sub> capture. To reach the technology targets set by the DOE, the National Energy Technology Laboratory (NETL) is partnering with United Technologies Research Center (UTRC) to demonstrate membrane-based separation of H<sub>2</sub> from coal-derived syngas using an improved palladium (Pd)-based membrane technology.

## Project Description

This project is a follow-up effort to *Experimental Demonstration of Advanced Palladium Membrane Separators for Central High-Purity Hydrogen Production*, which UTRC performed from 2007 to 2010. The project team will demonstrate the pilot-scale separation of H<sub>2</sub> from coal-derived syngas using a Pd-based tubular membrane separator. Project tasks include constructing the H<sub>2</sub> separators, quantifying the impact of gas composition and temperature on the membranes, comparing the performance and durability of a surface-modified, higher-H<sub>2</sub>-flux Pd-Cu membrane with the base-line Pd-Cu tubular membrane, testing durability of membrane construction materials under harsh conditions, and operating separators downstream of a coal gasifier.

The team will develop and test laboratory-scale and pilot-scale separators capable of producing two pounds per day (lb/day) of H<sub>2</sub>. The separator design is such that demonstration-scale separators capable of producing 100 lb/day of H<sub>2</sub> can be produced based on the information obtained on the pilot-scale separators. Power+Energy, Inc. will manufacture the separators and the University of North Dakota Energy and Environmental Research Center (UNDEERC) will test the separators downstream of a coal gasifier.

## NATIONAL ENERGY TECHNOLOGY LABORATORY

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## PARTNER

Power+Energy, Inc.  
University of North Dakota Energy and Environmental Research Center

## PERIOD OF PERFORMANCE

Phase 1  
10/01/2010 to 12/31/2011

## COST

Total Project Value  
\$ 1,865,223

DOE/Non-DOE Share  
\$ 1,492,178 / \$ 373,045



## Goals and Objectives

The goal of this project is to demonstrate at a pilot scale the use of Pd-based membranes to separate H<sub>2</sub> from syngas. The main objective during Phase 1 will be to construct, test, and demonstrate a surface treated palladium-copper (Pd-Cu) dense metallic tubular membrane separator capable of producing 2 lb/day of H<sub>2</sub> at a minimum of 95 percent recovery when operating downstream of a coal gasifier. Since H<sub>2</sub> separators must be able to endure the harsh conditions found in a coal gasifier system, an important additional Phase I objective will be to evaluate materials of construction for non-membrane separator parts to ensure low cost and durability. The final Phase 1 objective is an engineering analysis, following NETL guidelines, of the Power+Energy membrane design for the co-production of electric power and clean fuels, based on actual gasifier test performance. The project team will also identify a large gasification facility cost-sharing partner for Phase II of the project. The Phase II objectives will be to design, construct, and demonstrate a surface-modified Pd-Cu dense metallic tubular membrane separator capable of producing 100 lb/day of H<sub>2</sub> at a minimum of 95 percent recovery when operating downstream of a coal gasifier, and to conduct long-term tests on a 2 lb/day H<sub>2</sub> scale. The Phase III objectives will be to prepare a full engineering design of a separator system capable of producing at least four tons per day of high purity H<sub>2</sub>, and to continue testing the 100 lb/day H<sub>2</sub> separators downstream of a coal gasifier.

## Accomplishments

This project is a continuation of the UTRC-led team's approach to increase the technology readiness level of Pd-based metallic membranes for H<sub>2</sub> separation from coal/biomass gasifier product gas or similar H<sub>2</sub>-containing gas streams. Significant work (atomistic modeling and laboratory experimentation) has been done to develop an understanding of the performance of Pd-Cu systems. The impact of membrane poisons, such as carbon monoxide and hydrogen sulfide, has been studied and these poisons have been shown to be reversibly adsorbed on Pd-Cu alloys. In addition, durability experiments have established that the membranes can operate for periods of greater than 1000 hours under test protocol conditions specified by DOE. Furthermore, the design of the membrane separators has been proven to be scalable, as it is based on currently manufactured commercial hydrogen purifiers.

During this project, the team has produced the first two 0.1 square foot (ft<sup>2</sup>) separators and evaluated the gas impurity resistance of the first 0.1 ft<sup>2</sup> separator. The team has also prepared samples of eight alloys for corrosion testing and performed initial 500 hour tests on four of the alloys

## Benefits

The development of a Pd-based membrane for separating H<sub>2</sub> from coal-derived syngas will contribute to the development of advanced gasification-based power generation systems to use coal/biomass with CO<sub>2</sub> capture. The Pd-based membrane separators resulting from this project are based on existing, commercially manufactured separators, which will facilitate rapid progress toward a larger-scale demonstration of the technology. Reliable, long-term durability and corrosion data on both the Pd-based membrane and separator materials of construction will help meet the DOE's 2015 goal of a five-year separator life. The implementation of Pd-based membranes in coal gasification plants to co-produce affordable power, fuels, and chemicals in a safe and environmentally clean manner could have a major positive impact on the efficiency of the plants and the quality of the H<sub>2</sub> produced. This technology will contribute to U.S. energy security by decreasing dependence on imported fossil fuels as well as reducing negative environmental impact.



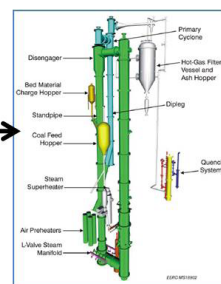
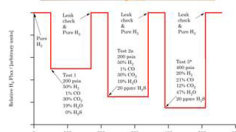
Construct laboratory-scale (<2 lb/day H<sub>2</sub>) separators



Quantify impact of gas species on performance & Materials testing and characterization



Construct pilot-scale (2 lb/day H<sub>2</sub>) Separators & DOE test protocol durability tests



Performance testing on coal gasifier syngas

*Technical approach for the project: quantify the impact of gas composition and operating conditions on durability and performance of two lb/day H<sub>2</sub> separators operating downstream of a coal gasifier.*

*Source: PI, United Technologies Research Center*

