

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



## FABRICATION AND SCALE-UP OF POLYBENZIMIDAZOLE - BASED MEMBRANE SYSTEM FOR PRE - COMBUSTION CAPTURE OF CARBON DIOXIDE

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### Background

In order to effectively sequester carbon dioxide (CO<sub>2</sub>) from a gasification plant, there must be an economically viable method for removing the CO<sub>2</sub> from other gases.

While CO<sub>2</sub> separation technologies currently exist, their effectiveness is limited. Amine-based separation technologies work only at low temperatures, while pressure-swing absorption and cryogenic distillation consume significantly large quantities of energy (up to 35 percent). In contrast, polymer-based membrane separations use much less energy and typically provide low-maintenance operations. Polymer membranes have been used successfully in a number of industrial applications, including the production of high-purity nitrogen (N<sub>2</sub>), gas dehydration, removal of acid gases, and recovery of hydrogen (H<sub>2</sub>) from process streams for recycle. However, successful use of a polymer membrane in synthesis gas separation requires a membrane that is thermally, chemically, and mechanically stable at high temperature and high pressure. Unfortunately, the commercially available polymeric materials currently employed are not sufficiently stable in such demanding environments.

In general, as operating temperatures increase, gas separation is significantly reduced and gas flow declines due to membrane compaction (creep). In addition, current membrane materials are often subject to chemical degradation by impurities in the process stream, a problem that is exacerbated by elevated temperature. Consequently, there is a compelling need for membrane materials that can operate under these more demanding environmental conditions for extended periods of time while providing a level of performance that is economically sustainable by the end user.



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## PERFORMANCE PERIOD

04/01/07 to 03/31/09

## COST

**Total Project Value**  
\$5,083,854

**DOE/Non-DOE Share**  
\$4,047,695 / \$1,036,159

## Description

The system produced in this project will consist of a minimum of four membrane modules, a control system, and a skid. The system must be able to separate H<sub>2</sub> from CO<sub>2</sub> in a syngas feed stream that has undergone low-temperature water gas shift process. The size of the membrane modules will be sufficient to separate a syngas stream that has 0.25 MWth heating value. The system will be able to accommodate syngas feed streams ranging in temperature from 200 °C to 400 °C. The system will also be able to operate at feed pressures ranging up to 700 psi.

## Primary Project Goal

The overall goal of this project is to develop a polybenzimidazole (PBI)-based capture system that is capable of operation under a broad range of conditions relevant to the power industry while meeting the U.S. Department of Energy's Carbon Sequestration Program goals of 90 percent CO<sub>2</sub> capture at less than a 10 percent increase in the cost of energy services.

## Objectives

- One of the key milestones for the project will be to construct a capture system in a 3-year timeframe that can be field tested at an end user's facility.
- Acquire and test a polymer-based membrane—available commercially or reported in the literature—for separations involving hydrogen.

This achievement is validated via membrane productivity (separation factor and flux) comparisons (Figure 2). The improved performance of this technology in an application such as IGCC-integrated capture is further substantiated by the accessible operating temperature range (up to 400 °C), long-term hydrothermal stability, sulfur tolerance, and overall durability of the composite membrane materials in these challenging pre-combustion environments. Additionally, the modular, low-maintenance, and flexible design of the membrane technology, combined with the technology achievements anticipated over the course of the life of this project, make it an exceptional candidate for use for pre-combustion capture of CO<sub>2</sub>.

## Benefits

This project will produce step-change improvements in the cost and performance of gas separation technology.

## Accomplishments

- The first generation (Gen1) of PBI-based hollow fibers have been produced and are being evaluated and optimized. The hollow-fiber platform offers a means to produce an economically viable, high area density membrane system amenable to incorporation into an IGCC plant for pre-combustion CO<sub>2</sub> capture.
- The Gen1, small-scale hollow-fiber-based membrane module has been designed and fabricated.
- The impact of membrane placement on process economics is under evaluation using ASPEN.
- Preliminary specifications and protocols for evaluations of the PBI-based fiber and membrane have been established.
- Market trends that may impact the successful commercialization of the PBI-based system for pre-combustion applications have been evaluated and reported.

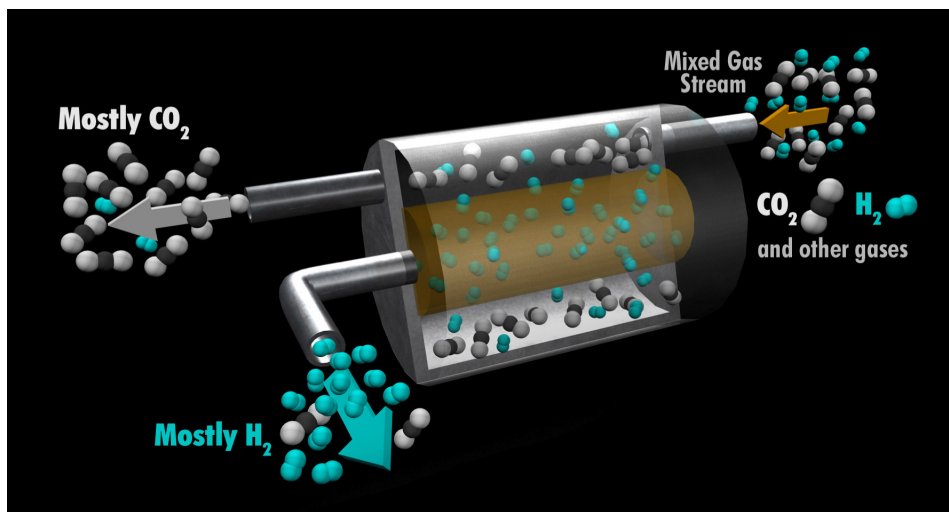


Figure 1. Simplified model of a single tube membrane module

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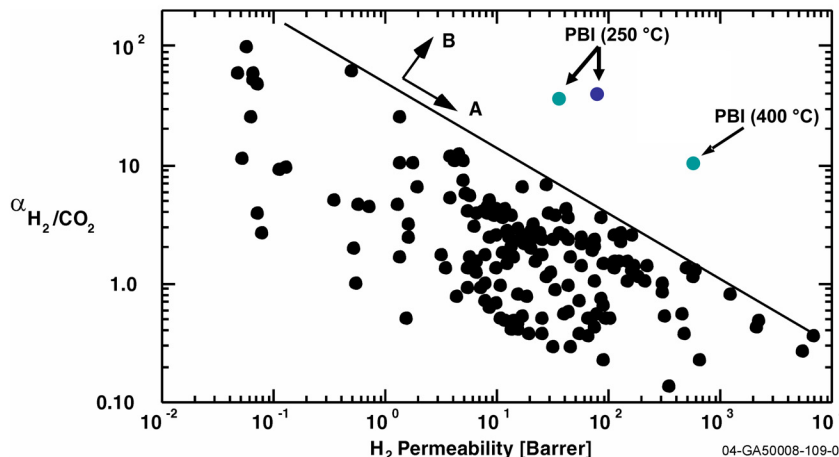
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# Selectivity vs. Permeability: H<sub>2</sub>/CO<sub>2</sub>



● = "wet" ● = "dry"

1 barrer = 10<sup>-10</sup> cm<sup>3</sup>-cm/s-cm<sup>2</sup>-cmHg

We have demonstrated improved selectivity over the state-of-the-art while operating at industrially attractive conditions



From Polymer. (23) 35, L.M. Robeson, et al., 1994.



Figure 2. Compilation of literature data of membranes for separation of H<sub>2</sub> and CO<sub>2</sub>. The line on the plot is called the "upper-bound" and is thought to be a limit of what is obtainable for a given separation with a polymeric material. The PBI in this investigation exceeds the "upper-bound" and falls into a commercially attractive regime.