DEVELOPMENT OF A NATIONAL CENTER FOR HYDROGEN TECHNOLOGY: A SUMMARY REPORT OF ACTIVITIES COMPLETED AT THE NATIONAL CENTER FOR HYDROGEN TECHNOLOGY FROM 2005 TO 2010

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DEVELOPMENT OF A NATIONAL CENTER FOR HYDROGEN TECHNOLOGY®

A Summary Report of Activities Completed at the National Center for Hydrogen Technology from 2005 to 2010

ABSTRACT

The Energy & Environmental Research Center (EERC) located in Grand Forks, North Dakota, has operated the National Center for Hydrogen Technology® (NCHT®) since 2005 under a Cooperative Agreement with the U.S. Department of Energy's (DOE) National Energy Technology Laboratory (NETL). The EERC has a long history of hydrogen generation and utilization from fossil fuels, and under the NCHT Program, the EERC has accelerated its research of hydrogen generation and utilization topics.

Since the NCHT's inception, the EERC has received more than $65 million in funding of hydrogen-related projects ($20 million for the NCHT project which includes federal and corporate development partner funds) involving more than 85 partners (27 with the NCHT). The NCHT project's 19 activities span a broad range of technologies that align well with the Advanced Fuels Program goals and, specifically, those described in the Hydrogen from Coal Program research, development, and demonstration (RD&D) plan.

A number of projects have been completed which range from technical feasibility of several hydrogen generation and utilization technologies to public and technical education and outreach tools. Projects under the NCHT have produced hydrogen from natural gas, coal, liquid hydrocarbons, and biomass. The hydrogen or syngas generated by these processes has also been purified to transportation-grade quality in many of these instances or burned directly for power generation. Also, several activities are still undergoing research, development, demonstration, and commercialization at the NCHT.

This report provides a summary overview of the projects completed in the first 5 years of the NCHT. Individual activity reports are referenced as a source of detailed information on each activity.

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EXECUTIVE SUMMARY

Formed in 2004 and founded upon the Energy & Environmental Research Center's (EERC's) more than 60 years of experience in hydrogen systems, the National Center for Hydrogen Technology® (NCHT®) project develops and pursues commercialization of technologies that produce and utilize hydrogen for fuels and chemicals, as a coproduct with power, and other uses. Since the NCHT's inception, the EERC has received more than $65 million in funding of hydrogen-related projects ($20 million for projects in the NCHT which includes federal and corporate development partner funds) involving more than 85 partners (27 with the NCHT).

The NCHT's 19 activities span a broad range of technologies that align well with the Advanced Fuels Program goals and, specifically, those described in the U.S. Department of Energy (DOE) Hydrogen from Coal Program research, development, and demonstration (RD&D) plan. Technical activities include development of hydrogen production technologies by gasification and gas cleanup with domestic coals, especially cheap, abundant coals; extension of novel, proprietary EERC bonding technologies to join high-temperature, hydrogen-compatible materials that enable more efficient hydrogen and power production; and development of tools to increase efficiency and reliability of gasification systems through improved fuel selection and preparation, among others. Of particular note are NCHT efforts to provide a flexible gasifier facility capable of larger-scale, more realistic testing of advanced technologies (e.g., low-cost hydrogen separation membranes), which directly address DOE "high cost" and "lack of demonstration" technical barriers. This technical breadth and close working relationship with DOE and numerous partners endow the EERC with the capacity to quickly respond to evolving DOE needs, especially in areas such as warm-gas cleanup and hydrogen purification.

To a great extent, the NCHT project pursues technologies that permit it to leverage and extend the EERC's experience and unique facilities focused upon fossil fuel and hydrogen technology development. While the project exploits the expertise of its many partners, its core capabilities are provided by the EERC's more than 345 scientists, engineers, and support staff applying a 60-year history of research in hydrogen, including gasification. The EERC has one of the largest, single-site collections of pilot-scale gasifiers in the world – a site comprising 245,000 square feet of laboratories, demonstration facilities, fabrication shops, and offices. The EERC has a long history of developing, testing, and integrating modular technologies for the production and utilization of hydrogen. Specifically, the EERC has unique, world-class experience in hydrogen production from fossil fuels, renewable fuels, and the development of on-demand hydrogen fueling systems produced from a variety of liquid fuels, as well as gas separation and purification through conventional and advanced methods, and utilization of hydrogen via direct combustion and in fuel cells. This work under the NCHT is outlined in this

5-year summary report. Individual activity reports are referenced as a source of detailed information on each activity.

DEVELOPMENT OF A NATIONAL CENTER FOR HYDROGEN TECHNOLOGY®

Summary Report of Activities Completed at the National Center for Hydrogen Technology from 2005 to 2010

INTRODUCTION

Hydrogen is generally envisioned as a primary energy carrier of the future and, as such, would play a key role in the future energy security of the United States. Using hydrogen as a fuel in a fuel cell, turbine, or internal combustion engine produces no harmful emissions, only water. However, the reality is that, with the exception of a few isolated prototypes, much of the infrastructure for hydrogen does not exist and has not been demonstrated. Significant advances are being made in laboratories throughout the world to develop technologies dealing with the various aspects of hydrogen production, including its generation, separation, purification, transportation, dispensing, and utilization. Advancing toward a hydrogen economy is not only about developing the individual components and methods, but also integrating and testing various technologies together for a complete hydrogen system.

The Energy & Environmental Research Center (EERC), working under agreement with the U.S. Department of Energy (DOE) National Energy Technology Laboratory (NETL), has founded the National Center for Hydrogen Technology® (NCHT®) to develop a broad range of technologies required to advance the opportunity for a hydrogen economy within the United States. The EERC is helping accomplish this opportunity by building on its long 60-year history in coal and fossil fuel utilization in order to generate hydrogen from fossil fuels.

Multiple activities were performed under this agreement, such as hydrogen production from coal, coal and biomass blends, and other solid feedstocks; purification of syngas; improved methods of generating hydrogen from natural gas; distributed hydrogen generation at gas stations from liquid fuels; fuel cell utilization; and utilization of hydrogen in turbines, among other tasks. Concurrently, activities were completed in the areas of education and outreach to increase public acceptance and support needed to ensure the widespread use of hydrogen. These activities have been completed at the EERC with important commercial support from industrial partners and DOE.

Table 1 is a partial listing of some of the key partners and collaborators with the EERC who have supported or been involved in NCHT projects from 2005 to 2010. In conjunction with industrial partnerships, advancement of hydrogen into the energy mix is also reliant on public support. A component of the activities under the NCHT Program is to increase public support and acceptance. In order to accomplish this task, the EERC has held public events, designed and built a demonstration hydrogen-powered ice resurfacer, and created a documentary about hydrogen with Prairie Public Broadcasting, Inc.

Table 1. Partial List of Partners of NCHT Activities

- Aboriginal Cogeneration Corporation
- Advanced Biomass Gasification Technologies, Inc.
- Air Products and Chemicals, Inc.
- Basin Electric Power Cooperative
- Bobcat Company
- ConocoPhillips Company
- Diversified Energy Corporation
- Electric Power Research Institute
- ePower Synergies, Inc.
- G F Truss, Inc.
- Great River Energy
- IdaTech LLC
- Intellection Holding Pty Ltd.
- Kraus Global Inc.
- North American Coal Corporation
- North Dakota Association of Rural Electric Cooperatives
- North Dakota Industrial Commission
- Porvair Filtration Group Ltd.
- Prairie Public Broadcasting, Inc.
- Pratt & Whitney Rocketdyne, Inc.
- Red River Valley Research Corridor
- Resurfice Corporation
- Rio Tinto
- SGL Group – The Carbon Company
- Siemens Power Generation, Inc.
- TXU Power
- U.S. Army Corps of Engineers Construction Engineering Research Laboratory
- U.S. Department of Energy
- University of Wyoming
- Verendrye Electric Cooperative, Inc.
- Western Research Institute
- Xcel Energy, Inc.
- Xethanol Corporation

This report summarizes the main results of each of the 19 completed activities of the NCHT listed in Table 2. The overarching findings of each of these pieces of research and outreach point to the fact that hydrogen and fuel cells will be a growing piece of the energy mix including use in vehicles, power generation, and energy storage. Moreover, the research indicates that, in many instances, hydrogen can be generated and utilized economically and in the near future. More detailed information can be found in the specific topical reports for each activity, referenced throughout the document.

Table 2. Listing of NCHT Projects (Years 1–5)

Activity Number	Title	Principal Investigator
1.1 (1)*	Catalytic Combustion of Hydrogen for Fuel for Next-Generation Low-NO_x Turbines	Bruce Folkedahl
1.2 (2)	Gasification of Lignites to Produce Liquid Fuels, Hydrogen, and Power	Joshua Stanislowski
1.3 (3)	High-Temperature Materials Development for Hydrogen Applications	John Hurley
1.4 (4)	Hydrogen Production and Purification from Coal and Other Heavy Feedstocks	Grant Dunham
1.5 (5)	Distributed Hydrogen Production from Gasification	Darren Schmidt
1.6 (6)	Advances in Gasification for Distributed Hydrogen Production	Nikhil Patel
1.10 (7)	Coal Ash Behavior in Reducing Environments (CABRE) III	Joshua Stanislowski
1.11 (8)	Integrated Gas Turbine–Gasifier Pilot-Scale Power Plant – Coal Design Considerations and Testing	Phil Hutton
1.12 (9)	Long-Term Demonstration of Hydrogen Production from Coal at Elevated Temperatures	Joshua Stanislowski
2.2 (10)	Engineering Analysis of Indirect Biomass Liquefaction	John Hurley
3.1 (11)	High-Density Activated Carbon for High-Pressure Hydrogen Purification	John Hurley
3.2 (12)	Demonstration of Pratt & Whitney Rocketdyne's Hydrogen Generator Technology – Phases II–III	Jay Almlie
3.5 (13)	Distributed H_2 Supply for Fuel Cell Utility Vehicles	Jay Almlie
6.2.1 (14)	Solid Oxide Fuel Cell Utilization of Coal-Derived Fuels	Phillip Hutton
6.2.2 (15)	Future Issues and Opportunities for the Hydrogen Economy Workshop	Chad Wocken
6.2.3 (16)	Hydrogen-Fueled Vehicles for Niche Markets	Jay Almlie
6.2.4 (17)	Hydrogen End-Use Demonstration and Outreach	Kirk Williams
6.2.5	Outreach and Education for Hydrogen Production and Utilization	Tera Buckley
6.2.8	Lignite Gasification Technologies Summary Report	Michael Holmes

* Numbers in parenthesis are reference numbers.

ACTIVITY 1.1 – YEAR 1 – CATALYTIC COMBUSTION OF HYDROGEN FOR FUEL FOR NEXT-GENERATION LOW-NO_x TURBINES

An investigation of the durability of two preferred catalyst materials was carried out at the EERC in Grand Forks, North Dakota (1). The objective of the investigation was to study the degradation in catalyst performance when exposed to syngas obtained from the integrated gasification combined-cycle (IGCC) process under F class gas turbine operating conditions. Operation on actual syngas provided real-world durability data with exposure to low levels of gaseous impurities such as H_2S, NH_3, HCN, HCl, and alkali. Depending on the temperature and pressure of the gasification and cleanup system, these impurities may pass through the gas-scrubbing system and foul catalyst and turbine blades. Gas-phase alkali impurities are known precursors for condensates that form on turbine blades, causing corrosion.

Two catalyst coatings, provided by Engelhard Corporation and Precision Combustion, Inc., were subjected to long-term operation on syngas produced by gasification of coal in a bench-scale fluidized-bed gasifier. Both catalysts were run for 112 hours on H_2S-scrubbed syngas and 19.5 hours on syngas with 350 ppm of H_2S.

The second objective of this study was to determine whether the addition of mineral sorbents to a fluidized-bed gasifier can reduce the gas-phase alkali concentration in the syngas below gas turbine manufacturer specifications of 24 ppb. Equilibrium calculations determined that kaolin and activated bauxite have the potential to reduce gas-phase alkali concentrations below 24 ppb under proper gasification conditions. From these calculations, bench-scale testing was performed with kaolin and activated bauxite with BNI North Dakota lignite coal in a fluidized-bed gasifier.

Results

It was found that catalyst performance on H_2S-scrubbed syngas was very stable and consistent throughout the 112 hours of operation. Both catalysts showed no change in surface temperature or gas conversion throughout the test. On 350 ppm H_2S syngas, there was an initial reduction of approximately 20% in catalyst performance within the first 2 hours of operation, as measured by conversion percentages. However, catalyst performance stabilized after the first 2 hours. Subsequent testing and visual inspection of the catalyst revealed no observable surface defects that could be attributed to the sulfur and no indication of degradation or fouling of the catalyst because of impurities in the syngas.

The EERC attempted to demonstrate the ability of several common sorbents (kaolin, calcined bauxite, and activated bauxite) to reduce alkali concentration in syngas below manufacturer specifications for gas turbines (<25 ppb) when mixed with coal during gasification. However, experimental error caused by caking of the ash on the probes was found to potentially increase the measured alkali gas concentrations on runs with kaolin sorbent because of alkali carryover from previous runs. When this was addressed for runs with activated bauxite, gas-phase alkali concentrations were reduced by an order of magnitude. While it can be concluded that these sorbents significantly reduced gas-phase alkali concentrations in the syngas, the actual amount remains unknown for the kaolin sorbent because of experimental error.

ACTIVITY 1.2 – YEARS 1–3 – GASIFICATION OF LIGNITES TO PRODUCE LIQUID FUELS, HYDROGEN, AND POWER

Coal gasification technologies will play a central role in future global electric power generation because, over the next 10 to 15 years, the technologies have the potential to double efficiency in electricity generation over today's power plant (18). In addition, coal gasification can provide synthesis gas that can be used as a feedstock for coproduction of hydrogen, fuels, and chemicals along with power. IGCC systems have relatively high efficiency and low emissions, can produce hydrogen for use in fuel cells, and are amenable to CO_2 separation and sequestration. Although IGCC systems have lower emissions and higher efficiencies (approaching 45%, as compared to 33%–35% efficiency for conventional combustion systems), conventional coal combustion systems have been selected for new electric power systems over IGCC because of IGCC's higher installation costs and uncertainties in reliability (19). However, the perspective is changing. Plans for new power systems in the United States now indicate that IGCC and circulating fluidized-bed combustion (CFBC) are both being considered for future power generation for conventional power systems (20). The 2005 Energy Act enabled DOE to provide $200 million annually from 2006 to 2014 for the development of gasification and other clean coal projects.

A consortium was developed with DOE through the NCHT, the EERC, and seven commercial sponsors to address the key technical challenges encountered during lignite gasification and warm-gas cleanup. The goal of the project was to develop a process for efficient gasification of lignites to create a clean syngas for the production of liquid fuels, electric power, and hydrogen and the capture of carbon dioxide. The overall project was split into 2 years. The first year of the project focused on bench-scale fluidized-bed gasification and warm-gas cleanup development. The second year focused on additional testing in a pilot-scale transport reactor and a bench-scale entrained-flow gasifier (EFG). The project report presents the testing results from both Year 1 and Year 2 of the project (2).

The Year 1 test results include a detailed study of the front-end gasification performance of the coals tested and their agglomeration potential in fluid-bed systems. That is followed up by detailed studies of a warm-gas cleanup system, including two-stage sulfur control, particulate control, and mercury control. The Year 1 testing concluded with gas-conditioning studies, including water–gas shift and hydrogen separation testing.

Year 2 of the consortium focused on testing lignites in the pilot-scale transport reactor demonstration unit (TRDU) and the bench-scale EFG. The gas cleanup train developed in Year 1 was used for scrubbing a slipstream from the TRDU. A hydrogen separation membrane was used on the back end of the gas cleanup train to demonstrate the production of a pure hydrogen stream from lignite coal. A bench-scale EFG was constructed as part of this program, and gasification test runs were completed on the EFG with each of the sponsor fuels. Additional equipment was acquired with NCHT funding Year 3 and used to analyze samples from Year 2.

Results

Overall, it was demonstrated that lignites are suitable feedstocks for a wide variety of gasification systems; but when a system is chosen, the fuel properties must be taken into account. It was demonstrated that lignites are suitable for fluid-bed systems because they are highly reactive and convert at low temperature. However, high-sodium lignites can present significant challenges to gasifier operation because of the potential for ash agglomeration. These issues can be mitigated through the use of the strategies developed in this test program, including introduction of dolomite and bauxite as bed materials and use of kaolin as a gettering agent. Lignites are also a good choice for transport reactors, because they take advantage of the high reactivity and the high velocity helps to disperse heat and reduce the chances for agglomeration.

Lignites can be very challenging to run in an EFG. The high moisture content necessitates a drying step, and some lignites still have a high tendency to cake even when dried. Lignites are high in ash, so managing slag flow in an EFG becomes even more difficult. However, under carefully chosen operating conditions, it was demonstrated that lignites could be successfully gasified in an entrained-flow system.

It was shown that commercially available or near-commercial technologies exist today that can clean the syngas from gasification at elevated temperatures. A slipstream of syngas was taken just after the hot-gas filter vessel at the top of the gasification tower and sent to the first floor through a heated tubing bundle. The syngas was processed through the warm-gas cleanup train, where desulfurization, shifting, chorine control, and mercury control occurred to various levels, depending upon coal type and control strategy used. For details, please refer to the detailed topical report.

Sulfur was removed to sub-ppm levels, and chlorine and trace elements were also controlled. Hydrogen and CO_2 separation was tested with a shift catalyst and a hydrogen separation membrane. The membrane used in the test was Pd–Cu-based, and after 50 hours of operation, it was shown that the membrane could achieve hydrogen purity greater than 99.9%. No adverse effects to the membrane were noted, indicating that the warm-gas cleanup train was adequately removing the contaminants from the syngas. Overall, it was demonstrated that a pure stream of hydrogen could be produced from Texas lignite while the temperature of the syngas was maintained above 400°F.

ACTIVITY 1.3 – YEARS 1–3 – HIGH-TEMPERATURE MATERIALS DEVELOPMENT FOR HYDROGEN APPLICATIONS

For this activity, the EERC worked with Siemens Energy Inc. to develop a bonding process to join the oxide dispersion-strengthened (ODS) FeCrAl alloy Kanthal APMT® to the nickel-based superalloy CM 247 LC for possible use in future high-temperature hydrogen-burning turbines (3). These alloys have very specific microstructures and compositions that give them properties suitable for use in high-temperature hydrogen environments.

Because the microstructure must be maintained for high-temperature creep resistance, consideration must be given to the bonding method used to join the materials. Conventional fusion joining methods, e.g., fusion welding, destroy the microstructure of both the ODS and DS superalloy substrates. Hence, to retain creep resistance, other methods of joining must be used, such as transient liquid-phase (TLP) bonding, which is the focus of this research.

The pieces to be joined need to be under pressure during joining, but the pressure can be provided by a simple clamp made of a material with a lower coefficient of thermal expansion than the pieces being joined. In addition, the clamp must allow for swiveling, or articulation, of the structures being joined in order to make the applied pressure evenly distributed (Figure 1).

TLP bonding creates a joint between two surfaces through diffusion, rather than melting the materials together. This can preserve the microstructure and, therefore, may retain the high-temperature properties of the material. TLP is used to dissolve small quantities of the structures into the joint; the bonding alloy then diffuses into the large volume of the structure. However, the remaining bonding alloy, even if dilute, can adversely affect the properties of the bonded structure.

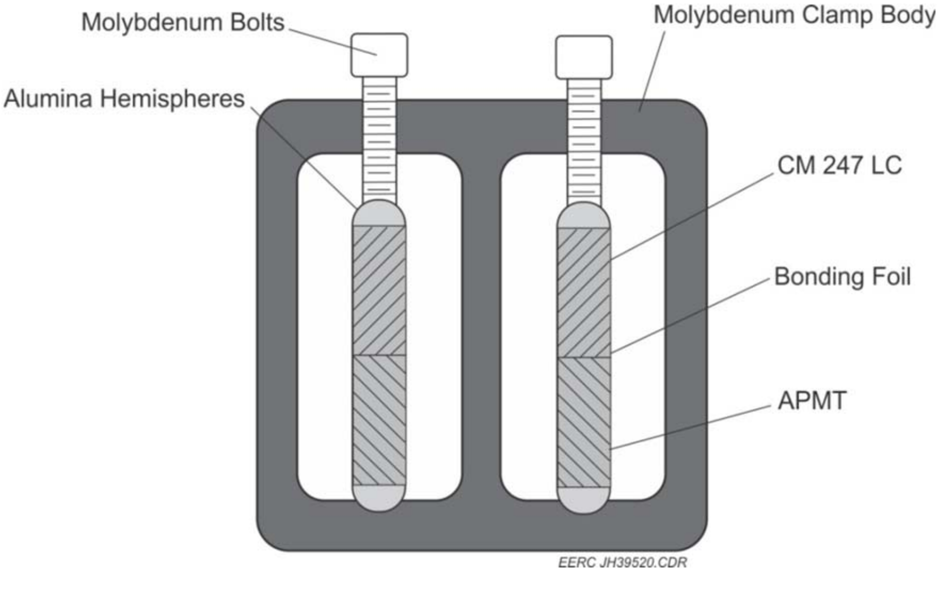

Figure 1. Clamping system for applying compressive force to the rods of APMT and CM 247 LC.

Results

The work performed consisted of two primary activities: computer modeling and physical testing. Initially, computer modeling was used to attempt to determine what pure metals could be used for joining the two alloys via TLP bonding and to estimate the heat treatments to be used during the bonding process. The modeling was able to predict times for solidifying and homogenizing the joint for a chosen interlayer and base, which was then compared to experimental tests.

Based on the results of this modeling, laboratory tests of the TLP metals and heat treatments were performed on several physical forms of the two alloys to be joined. In some cases, a non-dispersion-strengthened form of APMT designated APM was also used. The physical forms included rods joined to rods in a butt joint, thin discs of APMT and APM joined in a sandwich between rods of CM 247 LC, including both flat and curved joint surfaces, and thin plates of APM or APMT sandwiched between actual turbine ring segments made of CM 247 LC.

The testing shows that a variety of metals can be found to make the bonding foils, but one in particular is unique in that it is widely available and inexpensive and diffuses away from the joints, primarily through the ferritic ODS alloy. Once at the surface of the structure, the bonding alloy vaporizes and condenses in the cold end of the process furnace tube in a process that we call evaporative metal (EM) bonding. The creep rupture lifetime of the joint is greater than that of the APMT at 950°C. Electron microscope energy-dispersive x-ray analyses do not detect bonding alloy remaining near the joint, demonstrating that its concentration is below 0.1 wt%. Two presentations describing the process were given at technical conferences, and it was also described in a book chapter written about EERC experiences with high-temperature ODS alloy heat exchangers. A patent application for the evaporative metal bonding process has been submitted to the U.S. Patent Office (21).

ACTIVITY 1.4 – YEARS 1–5 – HYDROGEN PRODUCTION AND PURIFICATION FROM COAL AND OTHER HEAVY FEEDSTOCKS

Air Products and Chemical, Inc. (APCI) has been working on an alternative approach for rejecting CO_2 from the syngas, as well as the CO, at high temperatures (>400°C). The process is called the sorption enhanced water–gas shift (SEWGS) process which is used in conjunction with steam methane reforming. The WGS reaction is presented in Equation 1:

$$CO_{(g)} + H_2O_{(v)} \rightarrow CO_{2(g)} + H_{2(g)} \qquad (1)$$

APCI originally developed the SEWGS technology for use with natural gas-based syngas, i.e., syngas that contains essentially no sulfur. The process combines the WGS reaction with simultaneous removal of CO_2 by adsorption on a solid adsorbent. The adsorption of CO_2 drives the WGS reaction to completion, yielding low CO levels in the product gas. The APCI process utilizes an assembly of fixed-bed reactors filled with a mixture of shift catalyst (to lower the temperature of reaction) and CO_2 adsorbent. With CO and CO_2 removed, the product gas consists predominantly of hydrogen and steam at feed pressure (~20–30 atm) and high temperature (400°–550°C), which is optimally used as gas turbine fuel. A portion of this product stream can also be cooled and sent to a relatively small H_2 pressure swing adsorption (PSA) for production of 99+% pure H_2 product gas.

For a process using syngas as the feedstock, a key research need is the determination of the long-term impacts of poisons, specifically H_2S, on the integrity of the solid adsorbents. The goal of this program was to screen a number of potential ambient- and high-temperature adsorbent materials for sour syngas operation under realistic conditions of pressure, temperature, and gas composition and also demonstrate that the selected materials can effectively remove sulfur from sour syngas (4).

The major objectives of combining EERC's gasifier units with the two-bed PSA unit was to evaluate adsorbent stability to coal-derived syngas under PSA conditions, conduct H_2S breakthrough tests under controlled adsorber conditions, and obtain PSA performance data. Two major campaigns were conducted: the first with the CFBR on coal feedstock and the second with the EFG on petcoke feedstock.

Tested high-temperature adsorbents include a promoted hydrotalcite (HTC, general chemical formula: $Mg_6Al_2(CO_3)(OH)_{16}{\cdot}4H_2O$), promoted lithium orthosilicate (LO, general chemical formula: Li_4SiO_4), a supported Mg/Na double salt (general chemical formula: $NaMg(CO_3)_2$), and a supported metal oxide (MO, general chemical formula: Na_2O). Low-temperature adsorbents include activated carbon (AC), alumina (AL), silica gel (SG), polymeric resin (PR), and titanium oxide (TI). Spent adsorbent samples were returned to APCI for characterization and analysis. To gauge adsorbent stability, CO_2 capacities for the fresh and spent samples were determined by thermogravimetric adsorption (TGA) analysis. Conventional low-temperature N_2 adsorption techniques were used to quantify the adsorbent surface area and provide details on the pore volume of the samples (conducted after an initial regeneration under vacuum at 200°C).

In the first body of work, an exposure unit was constructed and operated to subject a series of these materials to simulated sour syngas for a period of up to 6 months. Analysis of the adsorption properties of the exposed samples, and comparison to those with fresh materials, identified which materials could withstand these conditions and potentially be useful in sour gas.

In the second phase of work, the preferred adsorbent was loaded into a fixed-bed sour PSA/TSA (thermal swing adsorption) unit, and H_2S breakthrough experiments were conducted to characterize equilibrium and mass-transfer parameters. The PSA was then cyclically operated with a sour syngas feed generated from coal in the CFBR and petcoke in the EFG. In all, a total of more than 4000 PSA cycles were executed. The results indicate that the PSA can reject H_2S and CO_2 from the syngas. The high tar environment of syngas generated from the fluidized-bed gasifier appears to reduce the effective H_2S capacity by about 20% compared to fresh adsorbent. More long-term PSA operation is necessary, for both types of gasifiers, to estimate adsorbent lifetime and, effectively, adsorbent replacement schedules. In the last phase of work, the impact of low levels of COS, HF, and $Fe(CO)_5$ on the preferred adsorbent were evaluated.

Results

Exposure of high-temperature adsorbents to simulated H_2S-containing syngas indicates that HTC and MO adsorbents lose 40%–60% of their original CO_2 adsorption capacity and are, therefore, of limited usefulness in sour syngas applications. The DS material loses between 10%–20% of its original capacity and shows a significant accumulation of sulfur. The LO material in CO_2-loaded form appears to be insensitive to H_2S—there is very little change in CO_2 capacity of the exposed materials—and it showed the lowest sulfur accumulation of all the adsorbents tested. It could potentially be useful in treating sour syngas streams. More tests are necessary to determine if CO_2-free LO adsorbent is also H_2S-tolerant.

Of the five different types of ambient-temperature adsorbents tested, only two, silica gel SG1 and polymeric resin PR1, appear to be useful for sour syngas applications. Accumulation of sulfur was evident in all samples, most predominantly in AC materials (9–18 wt%).

Two separate loadings of SG adsorbent were operated in the sour PSA unit for 1200 and 1500 cycles, respectively, with a feed gas of shifted sour syngas from a fluidized-bed gasifier. Periodic H_2S breakthrough experiments indicated that the H_2S adsorption capacity of the beds drops about 20% relative to the fresh adsorbent. This decline appears to occur during the first 700 cycles and then stabilizes with further cycles. The sour syngas was analyzed and shown to contain appreciable amounts of tar species (e.g., aromatics, indanes, tetrahydronaphthalenes, and naphthalenes, among others). These species were found deposited along the packed bed and presumably led to the 20% decline in H_2S capacity. In addition, higher levels of sulfur and chlorine were noted in the adsorbent at the feed end of the vessel. Future efforts must be directed toward long-term stability (multiple weeks on stream) of the adsorbent with this type of feed gas.

Similar PSA testing was conducted with syngas generated from petcoke in an EGF. This syngas contained essentially undetectable levels of the tar species. A total of 1450 PSA cycles were conducted, and periodic H_2S breakthrough experiments suggest that the adsorbent is stable in this syngas. Although the PSA on-stream time was not extensive, the results showed no evidence of a significant decline in performance over a 48-hr period. These results are encouraging signs that the H_2S in the petcoke syngas can be effectively removed by the PSA.

ACTIVITY 1.5 – YEAR 2 – DISTRIBUTED HYDROGEN PRODUCTION FROM GASIFICATION

The EERC worked in partnership with Xethanol Corporation on distributed gasification technology referred to as microgasification (5). Microgasification includes a gasification process that economically scales to the 5–50-ton/day fuel consumption range. These gasifiers can process relatively dry fuels such as biomass and stoker-grade coal, which are ideally suitable fuels for distributed applications. The process includes a downdraft gasifier, gas cleanup system, and engine generator capable of producing 50 kW of electricity.

Results

Stoker-grade lignite coal was fired in the EERC's commercial demonstration gasification system located in Grand Forks, North Dakota. Lignite stoker coal is a good candidate for distributed gasification. Lignite coal was gasified in the system to determine gas composition, determine how to clean hydrogen sulfide from the gas, and investigate potential alkali controls.

The project demonstrated gas heating values of 130–150 Btu/scf, while producing low levels of tar vapors. 50 kW of electric power was produced from lignite coal syngas at 132 scfm having a heating value of 132 Btu/scf. Maximum hydrogen composition of the syngas was measured at approximately 17%.

Sulfur treatment methods using activated carbons yielded 100% effectiveness for removing hydrogen sulfide. Carbonyl sulfide concentrations were approximately 100 ppm downstream of the AC bed. Alkali metals appear to be removed effectively by wet scrubbing (<1 ppm) and, therefore, do not require filtration.

The economics of distributed hydrogen production from gasification of lignite coal at \$4.15/lb H_2 appear competitive with electrolytic hydrogen production. However, costs not included in this study such as sulfur removal, additional compression, and processing of syngas will increase the costs of distributed hydrogen produced from microgasification.

ACTIVITY 1.6 – YEAR 3 – ADVANCES IN GASIFICATION FOR DISTRIBUTED HYDROGEN PRODUCTION

The focus of this project was to achieve small-scale, distributed hydrogen production from gasification (less than 2000 kg/day or 183 lb/h) (6). Two separate system-level designs were developed to compete with large-scale hydrogen production from natural gas: the gasification system and the syngas cleanup system. The feedstock ranged from coal to a variety of biomass and opportunity feedstocks with little to no cost. The focus of each design was low operating costs, environmental acceptability, portability, and the ability to provide hydrogen to small, distributed markets at less cost than centralized hydrogen production from natural gas.

Initial tests were performed with a commercially available downdraft gasifier. This was done as a baseline to understand the issues with current systems and the balance-of-plant systems that are needed for this type of gasification process. From those tests, an advanced fixed-bed gasifier (AFBG) design was developed at the EERC which was able to convert both coal and opportunity fuels with very little balance-of-plant complexity. The feedstocks used were Powder River Basin coal, North Dakota lignite, Illinois No. 6 coal, creosote-treated railroad ties, and common wood chips. The target fuel feed rate was 50 lb/h.

This report describes the specifications taken into consideration during the gasification design process as well as the ability of that gasifier design to provide a syngas that mated well with a low-cost syngas-to-hydrogen purification system.

Results

To assist in the design of a distributed small-scale hydrogen production unit, a significant amount of modeling was performed on various scenarios in order to reduce overall balance of plant operations and cost. The modeling showed expected gas composition results, material flow rates, temperatures, and ash generation results. This information was used to estimate equipment required for the balance of plant in each case. It was determined that the AFBG would produce hydrogen at a rate of 12% by weight of the coal input rate.

A downdraft-style Imbert-type gasifier (commercial) was operated at a local wood truss plant to produce electricity from a generator run on the syngas. The feedstocks used were railroad ties and several coals. This provided baseline test data and experience to improve the models and design for the AFBG.

The AFBG was able to achieve high fuel conversion efficiency with very low levels of syngas contaminants. Full details on conversion efficiencies and syngas contaminants can be found in the topical report. This is due to a contaminant reinjection system to decrease syngas contaminants and increase hydrogen production. This shows that small-scale production of hydrogen for gasification is possible with selected technology innovation.

ACTIVITY 1.10 – YEARS 3–5 – COAL ASH BEHAVIOR IN REDUCING ENVIRONMENTS (CABRE) III

The goal of the CABRE Program is to obtain the fundamental data necessary to develop a software program capable of predicting the fate and behavior of inorganic species during gasification (7). To meet this goal, the program was broken into seven tasks: 1) feedstock characterization, 2) laboratory evaluation of ash behavior, 3) evaluation of ash behavior in bench- and pilot-scale systems, 4) ash impacts on gas cleanup and CO_2 capture, 5) modeling of ash behavior, 6) model validation, and 7) reporting. Tasks 1–4 were completed as part of the NCHT Years 3–5. Tasks 5 and 7 are ongoing from NCHT funding Years 3–6. Task 6 is scheduled to take place in Year 6.

The data collection phase for the building of the CABRE model has been completed. Fundamental laboratory analyses and fuel gasification data have been combined to form the building blocks of the CABRE model. Data have been gathered from gasification in an EFG and a fluid-bed gasifier on four different fuel types. The model development is well under way. The software for the CABRE model is planned to be available at the conclusion of the NCHT Year 6 funding contract.

Two fuels were selected by each sponsor for extensive characterization. Two North Dakota lignites were selected by the North Dakota Industrial Commission (NDIC): a Falkirk Mine lignite and a Center Mine lignite. An Illinois No. 6 bituminous coal and a petroleum coke were selected by ConocoPhillips. Sufficient quantities of the fuels were obtained to conduct pilot-scale testing in an EFG and a fluid-bed gasifier. Gasification testing in the EFG was performed with Falkirk Mine lignite, Center Mine lignite, and Illinois No. 6 bituminous coal. Testing in the fluid-bed gasifier was completed with Falkirk lignite. The purpose of the testing was to gather fundamental data on the fate of inorganic species during gasification, and relate the findings to viscosity modeling.

Results

During entrained-flow gasification, silicon and iron showed consistent enrichment in the slag pot deposits when compared to the filter vessel. Aluminum was to be present in similar concentrations in fly ash and slag pot. Magnesium and calcium did not show consistent relations during the entrained-flow experiments. Sodium showed slight to strong tendencies to enrichment in the fly ash. Potassium and phosphorus were highly enriched in the filter vessel ashes, while they showed signs of depletion in the slag pot. Strontium showed preference to enrichment in the filter vessel, while barium showed similar concentrations in slag and fly ash. No consistent and strong correlation was found between partitioning and the O_2/coal as well as the steam-to-coal ratio.

During fluidized-bed gasification, very consistent enrichment factors for all major elements were present in the gasification ashes of Falkirk lignite. The effect of higher temperature resulted in less deviation of enrichment. Aluminum, iron, titanium, sodium, potassium, and barium were consistently more enriched in the bed ash. Phosphorus, calcium, magnesium, and strontium were predominantly enriched in the fly ash. The O_2/coal ratio and the steam-to-coal ratio had no apparent effect on partitioning.

Several available viscosity models were extracted from literature and evaluated using six different coal ash slags that were collected from commercial facilities. A broad database of coal ash compositions and their respective viscosity measurements at different temperatures was created from the literature. All sources were carefully checked for validity. This will be of use for the creation and evaluation of viscosity models. Several structural parameters were evaluated for use in a viscosity module inside the CABRE III model.

The basic building blocks have been completed for the development of the CABRE model. The data gathered will be translated to a computer code to predict the fate and behavior of inorganic species during gasification based on the composition of the incoming fuel. The model will be based on a graphical user interface. Users will have the option to input fuel and flux data manually in the model, import formatted Excel spreadsheets, or choose from an existing database of fuels. Model outputs will include viscosity and slag flow predictions, slag production rates, fly ash production rates, and deposition potential on cooling surfaces. Model predictions will be validated using full-scale samples.

ACTIVITY 1.11 – YEARS 4–5 – INTEGRATED GAS TURBINE–GASIFIER PILOT-SCALE POWER PLANT – COAL DESIGN CONSIDERATIONS AND TESTING

Small-scale gasifiers are ideally suited for distributed power systems utilizing opportunity fuels and biomass residues. Studies have shown that up to 600 million tons of opportunity fuels are produced each year in the United States. This represents a potential renewable electricity resource of up to 120 GW_e. However, the nature of distributed resources reduces economic viability. A distributed biomass power system may be vulnerable to seasonal and cyclic availability of biomass. The ability to cofire biomass with coal decreases system downtime and increases system reliability, subsequently increasing the overall economics of the distributed power system.

This project investigated the feasibility of cofiring biomass and coal in an indirectly heated microturbine power system to obtain additional fuel flexibility (8). The use of an indirectly heated microturbine eliminates the need to compress the syngas for injection into the microturbine (Figure 2). In this system, hot syngas is fed to an atmospheric combustor which then heats high-pressure air through a high-temperature heat exchanger. Since the syngas never contacts the high-speed turbine, particulate cleanup requirements are greatly reduced. The compressor is eliminated, and the need to cool the syngas below the condensation temperature of tars is also eliminated. This eliminates tar fouling in the pipes and greatly reduces the particulate cleanup requirements. Without the need to compress the syngas, the syngas temperature can remain above the condensation temperature of the tars. In addition, contaminants within the syngas do not come in contact with the turbine expander, reducing the cleanup requirements for the syngas.

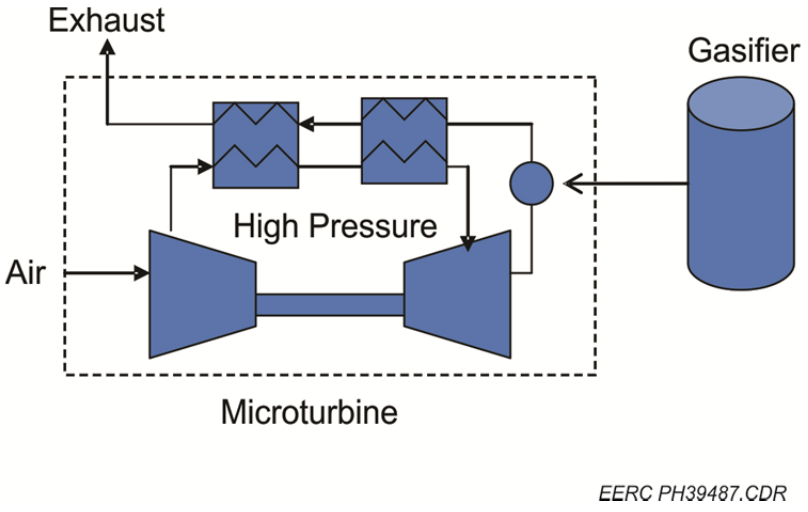

Figure 2. Indirectly heated gas turbine system for biomass power production.

Results

The gasifier successfully gasified coal char, wood char, coal, pine wood, oak pellets, corn, and wood bark. Cold-syngas composition was similar for all feedstocks and characterized by high CO content (~10%–23%), medium hydrogen content (~6%–12%), and low oxygen and methane content (~2%–4%). Carbon dioxide was approximately 10% ± 2%. The energy content was similar for all feedstocks at approximately 80 Btu/scf.

The primary difference between coal and biomass was due to the high ash content of the coal. Gasification of coal produced substantial clinker formation within the gasifier, which eventually choked mass flow through the gasifier. To overcome this, the gasifier was modified to break up clinker formation. This was done by extending the vertical auger in the gasifier above the air nozzles to break up clinker formation.

Production of electricity with both coal and biomass will require further modifications to the microturbine to convert it to an indirectly heated microturbine. Operation of the system, including the microturbine, will be demonstrated under work funded by Xcel Energy. However, the similarity in syngas composition and Btu content between coal and biomass suggests that there should be no significant difference in performance of the microturbine on either fuel.

ACTIVITY 1.12 – YEAR 5 – LONG-TERM DEMONSTRATION OF HYDROGEN PRODUCTION FROM COAL AT ELEVATED TEMPERATURES

The EERC together with DOE NETL and the state of Wyoming has completed a project to evaluate the performance of hydrogen separation membranes on coal-derived syngas (9). EERC small pilot-scale gasifiers were used to produce the syngas and solid sorbents were used for warm-gas cleanup and WGS. Three hydrogen separation membranes were exposed to this coal-derived syngas for several hundred hours (a full-stream flow membrane [FS] and two slipstream membranes [SS1 and SS2]). The FS membrane was larger and was capable of handling all of the syngas from the gasifier. The other two were smaller and took a slipstream from the gasifier. The FS membrane is a palladium–copper membrane. High hydrogen flux rates and high hydrogen purities are anticipated with this membrane, but it is also expected to have a low tolerance for impurities. The SS1 membrane is a palladium–gold-based material that is also expected to produce good flux rates and high purities. This membrane may have more resistance to impurities than a standard copper-based membrane. SS2 membrane is a palladium-based membrane that is expected to have high flux and purity rates and has shown some resistance to sulfur. Sulfur has been shown to reduce performance during exposure, but performance returns to baseline when the sulfur is removed.

The project report details the results of the gasification, warm-gas cleanup, and membrane tests. An energy efficiency analysis is also presented that provides insights into the potential economic advantages of hydrogen separation membranes over conventional low-temperature technologies (9).

Results

Overall, the membranes were shown to meet some of DOE's targets for hydrogen separation membrane development. The results of the three membranes are compared to DOE targets in Table 3.

Sulfur tolerance was not able to be specifically determined as part of this test campaign, because sulfur was kept well below 1 ppm for the duration of the testing. Undoubtedly, small levels of sulfur reached the membranes, and they will be evaluated for sulfur poisoning in the postmortem analysis being conducted by the providers. Cost of the small separators is also not relevant to a commercial-scale operation, and cost numbers were not provided by the membrane producers. The membranes did not appear to provide significant WGS activity, but this was difficult to determine in this test program because in order to achieve the highest possible partial pressure differential, the syngas was shifted as far as possible before hydrogen separation. Slipstream Membrane 2 probably had a significant leak and did not meet the purity goals. DOE lists a 5-year membrane life as the durability target for 2015. It is difficult to derive the full life of the membranes over the duration tested. Leaks developed during testing and are certainly a concern but likely easily resolved with additional engineering.

Table 3. Membrane Performance in This Test Campaign vs. DOE Targets

Performance Criteria	Units	2010 Target	2015 Target	Full-Stream Membrane	Slipstream Membrane 1	Slipstream Membrane 2
Flux (100 psi dP basis)	ft^3/(hour*ft^2)	200	300	21.3	117	29.4
Temperature	ºF	572–1112	482–932	650	750	900
S Tolerance	ppmv	20	>100	Not determined	Not determined	Not determined
Cost	\$/ft^2	100	<100	Not determined	Not determined	Not determined
WGS Activity	–	Yes	Yes	Not determined	Not determined	Not determined
ΔP Operating Capability	psi	Up to 400	Up to 800 to 1000	600	300	200
CO Tolerance	–	Yes	Yes	Yes	Yes	Yes
Hydrogen Purity	%	99.5%	99.99%	99.99	99.2	59.7
Stability/Durability	years	3	5	Not determined	Not determined	Not determined

🟩 Meets DOE 2015 goal.
🟨 Meets DOE 2010 goal.
🟧 Under DOE 2010 goal.

The full-stream membrane exhibited what appeared to be a slight degradation in performance over the 331 hours of exposure time, although the exact degradation in performance was difficult to quantify fully due to the leaks. More exposure time would be necessary to determine the full potential impact of impurities. The testing did show that the membrane could still produce significant flux over several hundred hours of operation using commercial or near commercial technologies for warm-gas cleanup. In fact, there was no conclusive reduction in flux for Slipstream Membrane 1. This is a promising result as future membrane materials are developed. While additional testing is required to answer many membrane performance questions, the initial has provided many promising results.

ACTIVITY 2.2 – YEAR 1 – ENGINEERING ANALYSIS OF INDIRECT BIOMASS LIQUEFACTION

The EERC has partnered with IdaTech, LLC, and completed a study to determine the feasibility and process economics for production and operation of a truck-mounted biomass gasification plant (10). Gasification is well suited to economically produce a clean syngas for conversion to methanol, a high-concentration hydrogen liquid fuel. The project partners' primary interest is small-scale, portable methanol production. Furthermore, the methanol would be green, transportable, and available for fuel cell utilization. Methanol could be transported to distributed sites where it will be converted to fuel cell-grade hydrogen for use in a proton exchange membrane.

Methanol is currently produced from petroleum, natural gas, and coal resources. Biomass is a relatively new addition to the existing list of possible resources. The focus of the current feasibility study is based on biomass as a feedstock for truck-mounted distributed methanol production. Since biomass has low energy density and is a highly distributed fuel source, large-scale methanol production from biomass is not economically favorable. However, the experience gained in the development of the large-scale methanol synthesis process is very valuable and applicable for small-scale distributed methanol production. This is possible since the same syngas (mixture of H_2, CO, CO_2, and CH_4) composition used as a feedstock for methanol synthesis is achievable using the appropriate biomass gasification technology.

Results

A detailed study and investigation were completed to identify the most appropriate method for gasification of biomass relative to methanol production and potential methanol production systems that can handle various compositions of syngas. The goal was to optimize both gasification and methanol production as a system for economical small-scale production (<200,000 gallons/yr).

The analyses determined that a thermally integrated downdraft gasification system (Figure 3) can produce an intermediate syngas that is appropriate for methanol synthesis at the lowest cost of the systems analyzed. The thermally integrated gasifier makes the most efficient use of heat produced for complete conversion of gas and utilization of biomass moisture, producing the best syngas composition in regard to catalytic conversion requirements at the lowest capital cost. The ability to eliminate or minimize process equipment is attractive for small-scale methanol production. The primary process components for a small-scale distributed methanol production system include a feed system, gasification, gas filtering, sulfur removal, syngas compression, methanol synthesis, and energy recovery (Figure 4).

The economic analysis revealed that the cost to produce methanol using thermally integrated gasification is $0.79/gallon, assuming a 20-year life with an estimated capital cost of $665,844 financed at 6.5%. The economics assume an 85% annual availability for a plant powered with grid electricity producing 176,967 gallons of methanol a year. The performance is favorable compared to other gasification approaches and minimizes the equipment components.

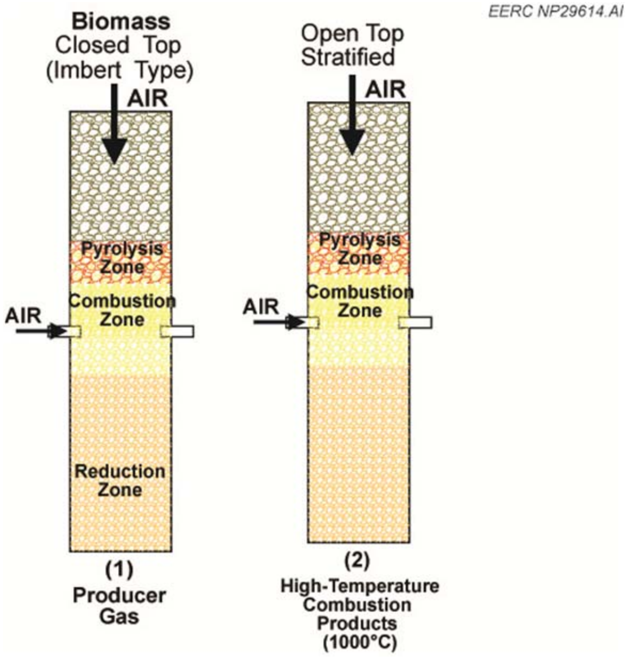

Figure 3. Schematic of downdraft gasifier: 1) Imbert type and 2) classical stratified.

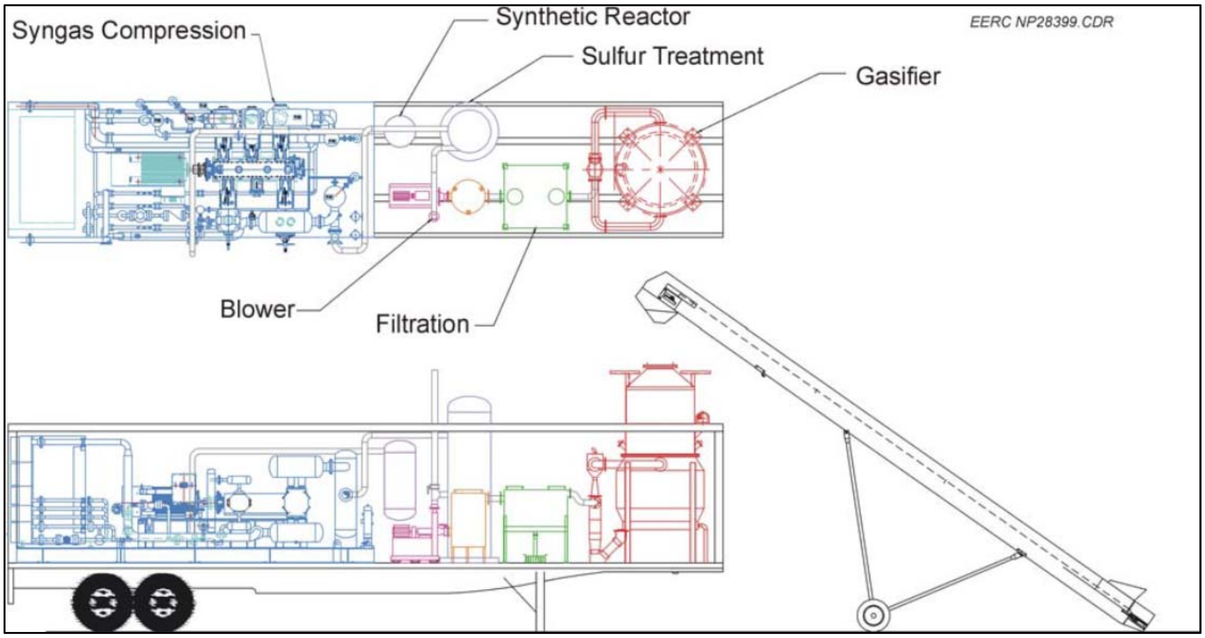

Figure 4. Process design for portable methanol synthesis from biomass gasification.

Given current methanol prices at $1.80 per gallon (at the time of the topical report writing), the potential to produce methanol from a zero-cost source of biomass could be attractive, especially if the market for carbon credit trading is realized.

ACTIVITY 3.1 – YEAR 1 – HIGH-DENSITY ACTIVATED CARBON FOR HIGH-PRESSURE HYDROGEN PURIFICATION

In this activity, the EERC worked with SGL Carbon to attempt to produce a monolithic high-density AC for use in high-pressure PSAs to separate hydrogen from a high-pressure syngas stream while maintaining the pressure of the hydrogen near that of the incoming syngas (11). Processing conditions that can swing to regenerate the adsorber include pressure, temperature, and electrical current.

Typical ACs used for purifying gas streams in either pressure or electrical swing adsorption have densities of 15%–25% of theoretical maximum carbon density (graphite $\cong$2.2 g/cm^3). During regeneration of the bed at low pressures, the mass of hydrogen lost with purge gas is significantly less than loss at high pressure and is, therefore, commercially acceptable. However, in order to produce high-pressure hydrogen for transportation uses, it is advantageous to create and purify the hydrogen at as high a pressure as possible. Unfortunately, because of the high macroporosity in current AC adsorbers and the relatively low expansion of the gas upon vaporization from the adsorber, regeneration at high pressures using modern technology would have a very low efficiency unless large masses of purge gas were used. To increase the regeneration efficiency of the adsorbers, the density of commercially available ACs must be increased without reducing the high surface area of the AC.

Results

In this activity, SGL Carbon provided the EERC with four different types of densified carbon for possible activation. The carbons are currently prepared for commercial application that do not involve activation on gas separation. Only two of the materials had adequate density, and only one of the materials demonstrated sufficient gas flow resistances, implying that it contained little continuous macroporosity that would allow channeling, which could reduce adsorption performance of the monolith for high-pressure applications. The EERC was not able to adequately activate the materials under this program. However, the materials were deemed promising enough that their activation was pursued under separate funding in a U.S. Department of Defense (DoD)-sponsored program to reform JP-8 fuel and purify the gas to make fuel cell-quality hydrogen.

ACTIVITY 3.2 – YEARS 3–5 – PRATT AND WHITNEY ROCKETDYNE'S HYDROGEN GENERATOR TECHNOLOGY – PHASE II – PERFORMANCE OPTIMIZATION AND FIELD DEMONSTRATION UNIT DESIGN

Hydrogen is a key feedstock for the production of chemicals, materials, foods, pharmaceuticals, and fuels (including some low-carbon biofuels). Currently, 40–45 million metric tonnes per year of hydrogen is produced worldwide, with about a third of that attributed to the United States. Hydrogen production is forecast to grow about 3.5% annually for the next 2 years, with the Asia/Pacific countries growing faster than other regions of the world. Conventional steam methane reforming is the most prevalent hydrogen production technology employed commercially today. Despite that popularity, the technology is expensive, has sizeable space requirements, and is a large carbon dioxide emitter. Pratt & Whitney Rocketdyne (PWR) has invented a novel, sorbent-based hydrogen production technology that promises smaller size, greater efficiency, lower costs, and reduced to no net carbon dioxide emissions compared to conventional hydrogen production technology. Development efforts at the pilot scale have addressed materials compatibility, hot-gas filtration, and high-temperature solids transport and metering, among other issues, and have provided the basis for a preliminary process design with associated economics. The process was able to achieve 93% hydrogen purity on a purge gas-free basis directly out of the pilot unit prior to downstream purification.

The purpose of Activity 3.2 was to advance PWR's technology to enable eventual design and installation of a field demonstration unit (12). This will be accomplished by way of PWR acquiring at least one partner able to host a demonstration unit at an appropriate industrial facility. The EERC hosted, tested, optimized, and collected design data from PWR's pilot-scale unit which fed the demonstration plant design efforts of a subcontractor, Zeton, Inc. Despite interest expressed by several gas supplier and petroleum companies, extended pilot-scale demonstration must be successfully accomplished before moving forward on a field demonstration. PWR and the EERC have pursued performing the extended pilot-scale demonstration, but encountered mechanical and operational issues with the unit that have delayed achieving the desired continuous hydrogen production duration target. Consequently, instead of a site-specific design, Zeton generated a generic design which provides the necessary design calculations and design documentation to enable Zeton to more quickly generate the ultimate site-specific design.

Mechanical and operating issues were encountered initially, but have been resolved. Lack of proper fluidization in the standpipe initially prevented sorbent from circulating through the pilot unit, but cold-flow experiments and accumulation of pilot unit operating experience have led to addition of gas jets that fluidize solids to the extent that the frequency of these problems has dropped from 3 out of 4 tests down to 1 out of 4 over the course of the third year of Activity 3.2. The high-temperature solids metering function experienced problems with binding of rotary valves, but after several valve modifications, the incidence of premature termination of tests because of valve issues decreased from 50% in the second year of this activity down to 8% in the third year. Product hot-gas filters initially experienced problems ranging from blinding to complete structural failure, but filter design changes and changes to online cleaning pulse-jet design resulted in a solution that has performed well in subsequent tests.

Results

In the third year of Activity 3.2, better pilot unit performance was realized. Evidence of this is presented in Table 4. Although fewer pilot unit campaigns and tests were performed than in previous years, the pilot unit was operated almost 20% longer and accumulated 60% longer total solids circulation time. Consequently, individual runs were more productive: the average test operated nearly twice as long in duration and maintained solids circulation more than 170% longer. Although total and average hydrogen production durations were less than those of the previous year, the unit achieved hydrogen production 70% sooner and twice as frequently than in previous years. Three consecutive tests attained hydrogen production. A major accomplishment was achieved when the unit produced a purge gas-free hydrogen product purity of 93% without downstream purification.

Table 4. Phase II (Year 4) Pilot Unit Test Performance as Compared to Phase III (Year 5)

Process State:	System Pressurized		Solids Circulation		Hydrogen Production	
	Year 4	Year 5	Year 4	Year 5	Year 4	Year 5
Total Duration, hr:min	284:26	338:32	55:33	89:58	25:04	5:41
Average Test Duration, hr:min	14:13	28:12	2:46	7:29	1:15	0:28
Runs That Achieved State, %			40%	92%	5%	25%
Circulation as % of Pressurized Test Time	20%	27%				

ACTIVITY 3.5 – YEARS 3–5 – DISTRIBUTED HYDROGEN SUPPLY FOR FUEL CELL UTILITY VEHICLES

The EERC has developed and recently scaled up and tested a high-pressure hydrogen production, purification, and dispensing system (13). The system reforms liquid reactants into a high-pressure supply of hydrogen gas. Because the system reforms liquid feedstock on-site and at high pressure, costs related to hydrogen transport and compression are reduced.

Currently, 95% of the hydrogen produced in the United States is produced via large-scale steam methane reforming (22). It has been estimated that hydrogen transportation costs \$4–\$9/gge (gallon of gasoline equivalent) and hydrogen compression, storage, and dispensing costs \$2–\$3/gge (23).

As an alternative to traditional hydrogen production and transportation methods, the EERC system reduces compression and gas transportation, virtually eliminating those costs (Figure 5).

The reactor continuously produces a high-pressure, hydrogen-rich gas stream that is purified at pressure and dispensed into vehicles. This design eliminates hydrogen transportation and hydrogen compression. Liquid feedstock would be brought to and stored in existing tanks at fueling stations, and the high-pressure hydrogen reformer would be located out of the way.

The EERC reformer system utilizes liquid hydrocarbon feedstocks and water that are pressurized at up to 700 bar (10,000 psi), introduced into a catalytic reactor, and continuously converted to a high-pressure, hydrogen-rich reformate stream that is purified at pressure and dispensed into vehicles. Although the reforming and purification are accomplished continuously,

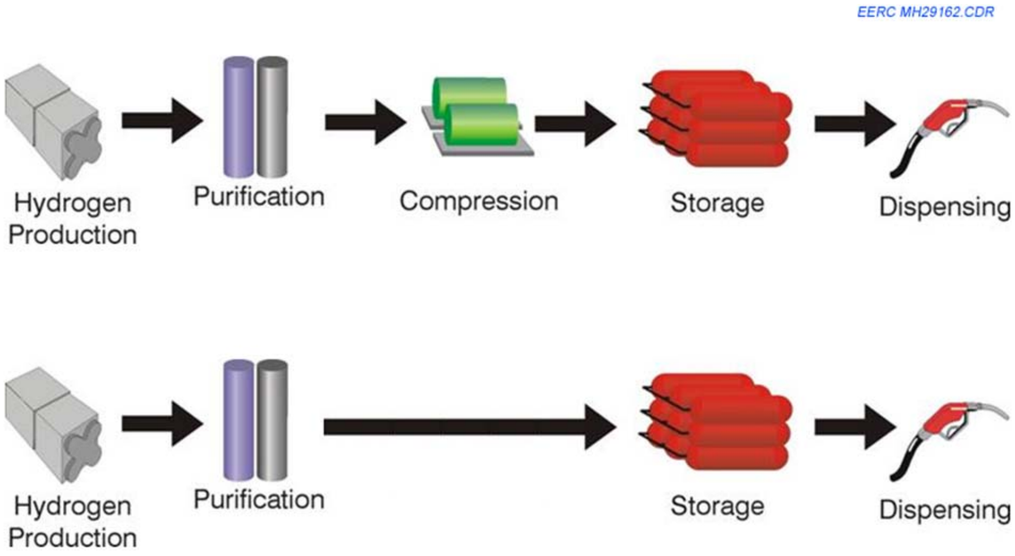

Figure 5. Traditional hydrogen production (top) vs. EERC process.

dispensing is an intermittent function. Thus a minimum amount of intermediate storage will be required. Also, the process temperatures required at these pressures create materials challenges. These challenges have been met with advanced materials selection. This design eliminates hydrogen liquefaction and/or compression and hydrogen transportation costs. The technology provides an opportunity to utilize existing liquid fuel infrastructure to provide feedstock to distributed hydrogen production and fueling stations.

Results

A scaled-up system, capable of producing 0.6 kg/hr (~15 kg/day) of purified hydrogen has been constructed and tested. Experiments using this system produced a high-purity hydrogen product from methanol and water. The product was produced at approximately 350 bar (~5000 psig) and contained no measureable amounts of nonhydrogen gases. The system is capable of producing at pressures up to 800 bar (~12,000 psig), above the 700 bar (10,000 psi) requirement for hydrogen as a transportation fuel. Production testing at 700 bar will be completed in NCHT Year 6.

High-pressure liquid reforming has certain advantages that could bring down the cost of distributed hydrogen. The technology reduces hydrogen delivery costs and has process efficiency advantages compared to other on-site hydrogen production technologies. The high-pressure liquid reforming process eliminates hydrogen compression, operates at reduced reaction temperatures, and has the ability to convert a variety of feedstocks into hydrogen. An initial capital cost assessment showed that the liquid reformer system has a similar initial investment as on-site steam methane reforming units and water electrolysis units.

Liquid reforming also holds infrastructure advantages over alternative technologies. Liquid reforming can take advantage of existing fueling supply chains. Small units can easily be distributed around the country or around a battlefield to provide clean hydrogen for transportation and distributed purposes. Initial results from this prototype hydrogen production, purification, and dispensing system are presented in the final activity report along with a description of the economic advantages of high-pressure liquid reforming (13).

ACTIVITY 6.2.1 – YEAR 1 – SOLID OXIDE FUEL CELL UTILIZATION OF COAL-DERIVED FUELS

Solid oxide fuel cell (SOFC) technology is recognized as one of the more promising technologies for converting lower-purity hydrogen fuels to electricity. The operating characteristics of SOFC technology allow the use of mixed gases with high CO, CO_2, H_2O, and CH_4 content common for coal-derived syngas. Internal reforming of the nonhydrogen components to hydrogen during use maintains the high electrical conversion efficiency typically associated with other fuel cells. Development of this contaminant-tolerant SOFC technology would accelerate the use of hydrogen on a commercial scale by decreasing the costs associated with purifying hydrogen to high-purity for conversion by the end user.

Copper–ceria anode SOFC technology, currently being developed by the project partner, Franklin Fuel Cells, has been shown to operate directly on a variety of hydrocarbon fuels (as seen in Figure 6 (14). The copper–ceria anode is less prone to carbon sooting and sulfur poisoning than typical nickel-based anode technology. These characteristics provide the potential to operate on a wider variety of producer gas compositions with lower purification requirements than nickel-based SOFC technology.

The first objective of this research was testing copper–ceria button cells on a variety of simulated producer gas compositions representing the potential coal gasification technologies. This research was done as a precursor to the future integration of gasification and fuel cell

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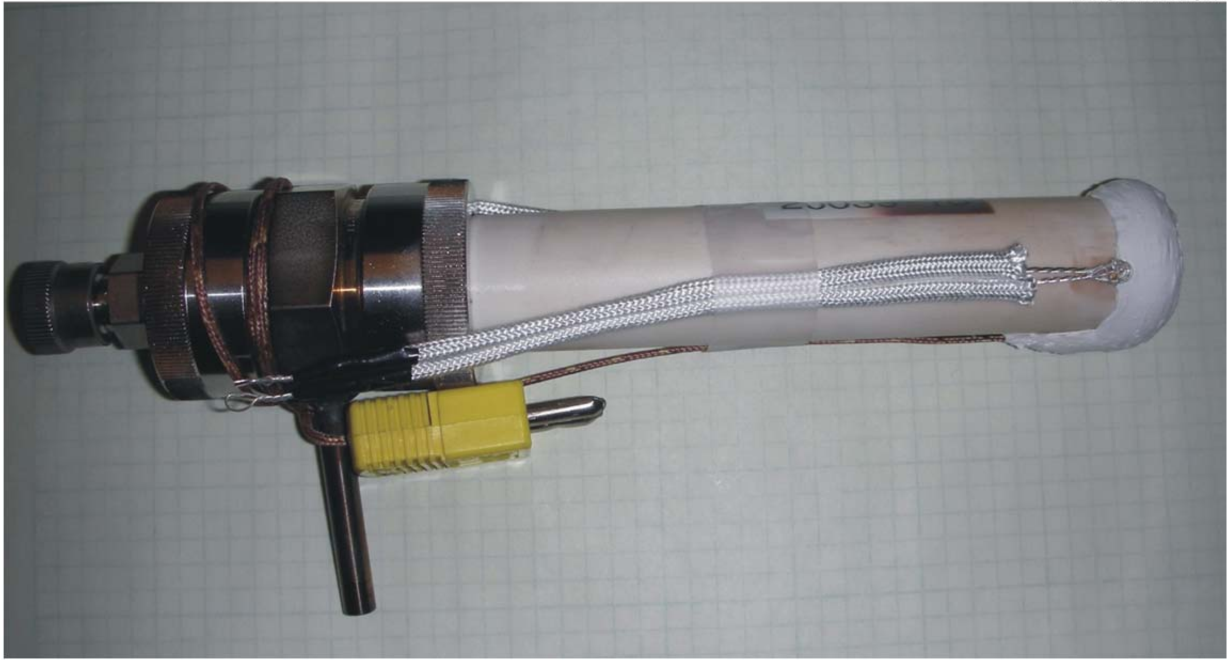

Figure 6. Test button of a Franklin Fuel Cells copper-based SOFC.

systems. The second objective completed was to test the tar tolerance levels of copper–ceria anode technology. The ability to operate directly on hydrocarbons implies a much higher tolerance to, and possibly the ability to utilize, tar in producer gas. This could reduce the tar scrubbing operations necessary for some gasification technologies and the associated waste disposal issues. If future work can also demonstrate H_2S tolerance, then copper–ceria anode SOFC technology may help accelerate future integrated gasification fuel cell systems by further decreasing overall plant cost.

Results

It was found that performance on simulated producer gas was stable and comparable to clean hydrogen (Figure 7). Power density was also found to be proportional to CO and H_2 content in the producer gas. A simulated tar mixture composed of polyaromatic hydrocarbons (tars) was injected into a producer gas stream to determine the effect on cell output. It was found that tar concentrations up to 18,000 mg/m^3 produced no discernible effect on cell output over a 30-hour period.

Copper-based SOFCs are capable of utilizing the hydrogen in low-purity, coal-derived fuels at high efficiency. These results demonstrate the potential of copper–ceria anode technology to be successfully employed in future integrated gasification fuel cell systems. Development of this contaminant-tolerant SOFC technology will accelerate the use of hydrogen on a commercial scale by decreasing the costs associated with purifying, transporting, and storing high-purity hydrogen for conversion by the end user. Results of this evaluation are provided in a final report for Activity 6.2.1 issued in October 2007 (14).

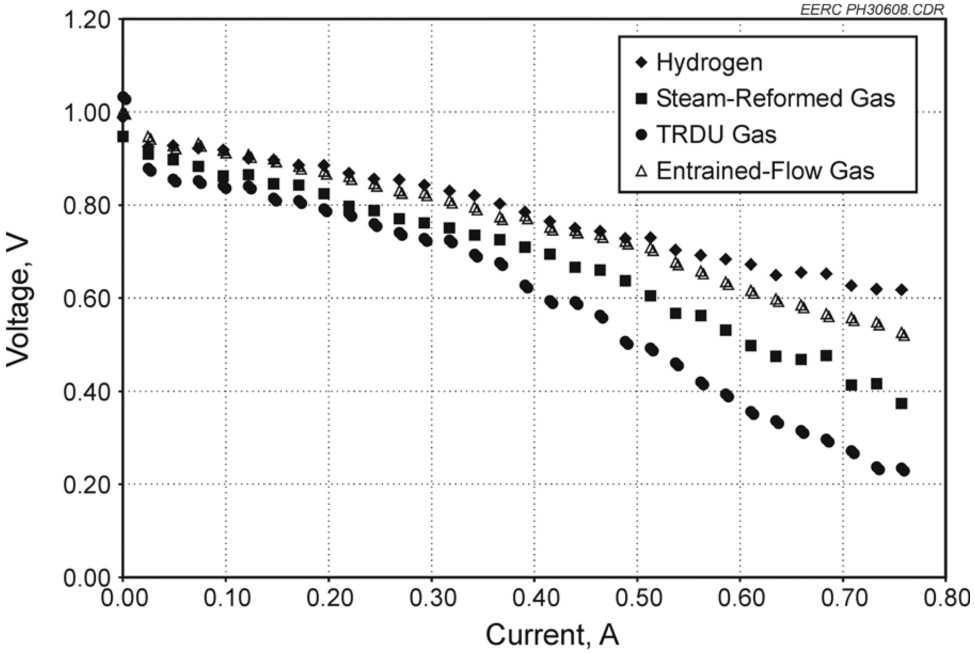

Figure 7. I–V curves for simulated producer gases.

ACTIVITY 6.2.2 – YEAR 1 – FUTURE ISSUES AND OPPORTUNITIES FOR THE HYDROGEN ECONOMY WORKSHOP

The EERC, through support from DOE NETL, the U.S. Department of Labor, the Red River Valley Research Corridor, Ballard, Xcel Energy, Air Products, and Northern Great Plains Inc. developed and hosted a 1-day workshop November 8, 2005, entitled Future Issues and Opportunities for the Hydrogen Economy (15). The workshop focused on educating people interested in the future of hydrogen energy and its potential role in satisfying our nation's energy needs. The course provided an overview of the various aspects of hydrogen-based energy technologies, including fuel cells, hydrogen production and storage, and safety considerations.

The 1-day workshop included presentations on various aspects of hydrogen technology and an opportunity for attendees to interact with each other and EERC research personnel to understand better the opportunities that exist in these developing energy fields. Topics and subtopics are listed as follows:

1) A general overview of the history of fuel cell technology and ways in which they are currently used was discussed. The presentation included the following:

    a) General concept of fuel cell operation

    b) Types of fuel cells

    c) Fuel cell uses including transportation, stationary, auxiliary, and military applications

    d) Commercial entities developing and utilizing fuel cells

    e) Fuel cell research and development challenges, both technical and logistical

    f) Summary of advantages and disadvantages of fuel cells

    g) Significant improvements needed to support further advancement of a hydrogen economy

2) Hydrogen gas does not exist naturally in the environment and, therefore, must be derived from one of many possible feedstocks, which include fossil fuels, water, biomass, and renewable fuels. The presentation focused on the following:

    a) A summary of hydrogen properties and how hydrogen compares to traditional fuels
    b) Existing uses of hydrogen
    c) A review of the various components that support a hydrogen infrastructure
    d) Production technology
    e) Separation and purification technology
    f) Storage and distribution technology

3) Energy production has a significant impact on the economy and quality of life. The use of hydrogen as a feedstock to make electricity presents a significant shift in the ways electricity has traditionally been generated—primarily from the combustion of fossil fuels. In November 2002, DOE published a document entitled "The National Hydrogen Energy Roadmap." That document outlines key issues and challenges in hydrogen energy development and suggests paths that government and industry can take to expand the use of hydrogen-based energy. A portion of the workshop focused on describing some of these issues, including the following:

    a) Hydrogen vision

    b) Methods of putting the pieces together

    c) Review of barriers and benefits to a hydrogen-based energy system

    d) Major international initiatives

    e) Federal and state incentives to support advancements in technology and implementation

    f) Business opportunities and careers in hydrogen-related areas

4) Concerns about the safety and reliability of hydrogen for energy production are widespread. Many concerns result from a lack of familiarity with hydrogen systems and historical events which have shaped perceptions. The purpose of this portion of the workshop was to discuss the science behind the risks associated with hydrogen and to discuss permitting and safety requirements for hydrogen systems. The presentation discussed the following:

    a) Popular perceptions of the dangers of hydrogen
    b) Permitting issues, requirements, and standards
    c) Safety code development, status, and requirements

Results

The workshop was attended by over 82 participants from around the region. Following the conclusion of the workshop, the presentation was revised into a 2-hour format and presented to three groups at the North Dakota State College of Science under separate funding. The information presented at this event was made available and utilized in workshops and for distribution to the public for other education and outreach activities as documented in the summary of Activity 6.2.5 that follows. Through follow-on opportunities such as this, information about hydrogen technologies will continue to be disseminated to the public and enable more broad adoption of new technologies.

ACTIVITY 6.2.3 – YEAR 1 – DEVELOPMENT AND DEMONSTRATION OF HYDROGEN-FUELED VEHICLES FOR NICHE MARKETS

It was the desire of the EERC and NCHT to demonstrate that the technology required to support the hydrogen economy is highly evolved and ready to take hold. It is recognized that infrastructure and public awareness concerns drive the change in inertia required to bring about the change to a hydrogen-based economy—a national interest indicated by President Bush's Hydrogen Fuel Initiative, announced on January 28, 2003. This initiative envisioned the transformation of the nation's transportation fleet from near-total reliance on petroleum to steadily increasing use of clean-burning hydrogen.

In order to change the inertia to initiate a global hydrogen-based economy, an alternate approach to mainstreaming hydrogen vehicles is required. Instead of attempting to change the very large automotive transportation sector, fuel cell technology may have a better chance at gaining foothold if niche vehicles such as ice resurfacers and other unique utility vehicles are demonstrated as fuel cell-powered platforms.

Therefore, the EERC and DOE partnered with ePower Synergies Inc. (ePSI) and Resurfice to develop, produce, and demonstrate the world's first and only fuel cell-powered ice resurfacer (16). This niche vehicle was labeled the "eP-ICEBEAR." ePSI is a company that develops multimodal transportation systems for communities utilizing clean, sustainable energy systems. Resurfice is an established manufacturer of Olympia brand ice-resurfacing vehicles. The EERC contracted with Kraus Global to design and manufacture an industrial hydrogen dispenser capable of filling the eP-ICEBEAR tank from either a single cylinder high-pressure hydrogen source or a cascade (multicylinder) source. This dispenser was installed at the EERC but is capable of being relocated to any demonstration venue with minimal effort.

Most ice refinishers, such as the one shown in Figure 8, are powered by propane-fueled internal combustion engines and typically utilize hydrostatic drivetrains. With these ice refinishers, engine exhaust emissions tend to concentrate near the ice. Propane-powered ice resurfacers have also been identified as the cause of carbon monoxide buildup in inadequately ventilated ice arenas. During the past 30 years, several cases of acute carbon monoxide (CO) poisoning have been reported in North America (24–35) during routine ice resurfacing. Studies from North America and Europe have documented high levels of CO in ice arenas using hydrocarbon fuel-powered ice resurfacers (36–56). Although battery and tethered electric units are available, they are expensive and have not always met the needs of ice facilities. Fuel cell-powered hybrid electric vehicles have the potential to ease operator, maintenance, air emissions, and other issues.

Results

The vehicle was developed and produced in a short 3-month span. The vehicle made its world debut at U.S. Senator Byron Dorgan's (D-ND) 2005 Hydrogen Energy Action Summit. The goals of this project were to educate the public on the readiness, practicality, and safety of fuel cells powered by hydrogen fuel and to establish a commercialization pathway in an early-

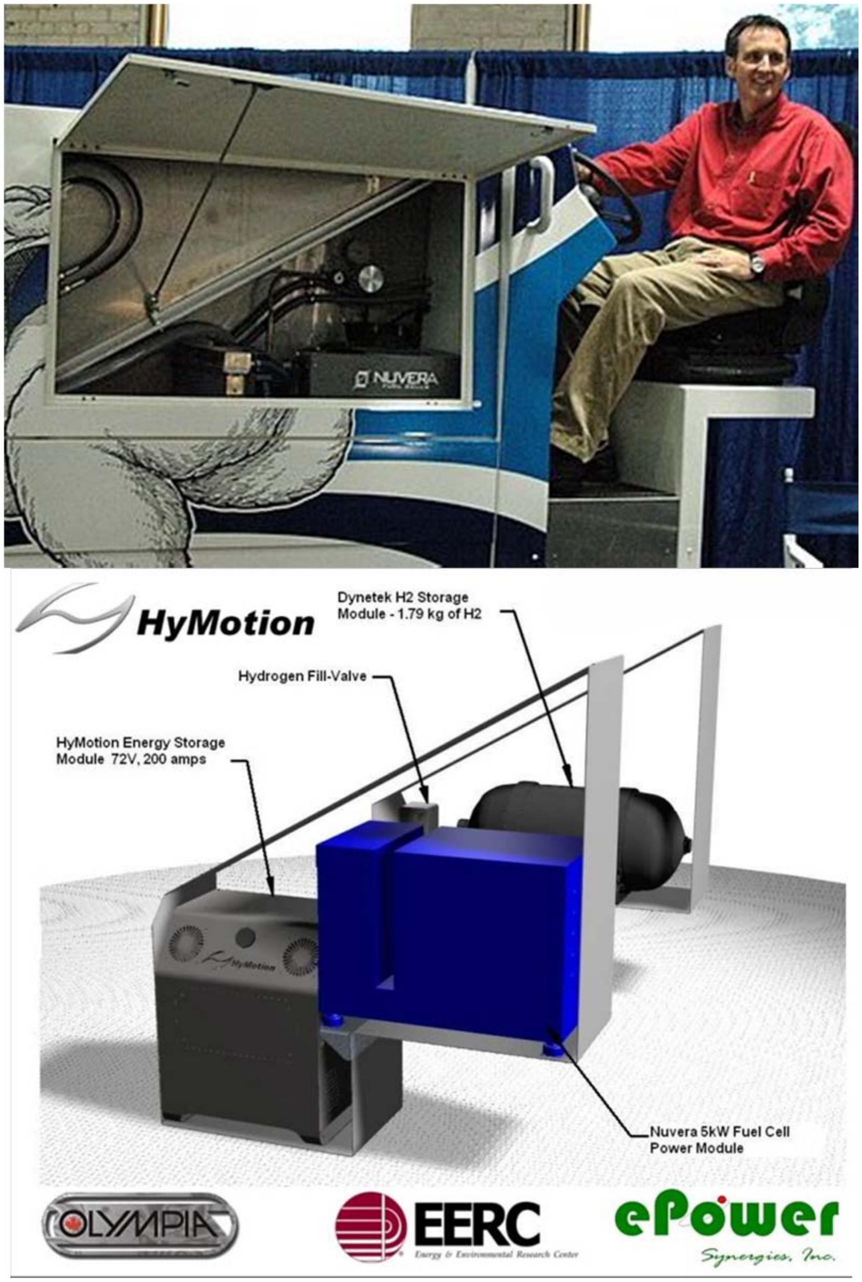

Figure 8. Powerplant configuration for ICEBEAR ice resurfacer.

adopter, niche market. A photo of the vehicle on display at this venue is shown in Figure 9. Purpur Arena was the maiden demonstration for the eP-ICEBEAR during the 2005 Hydrogen Energy Action Summit. During this demonstration, professional ice resurfacer drivers were given the opportunity to put the vehicle through its paces and give feedback on its performance. Feedback was overwhelmingly positive. The drivers, who normally drive propane-powered Zamboni ice resurfacers, were pleased with the quietness and on-ice handling of the eP-ICEBEAR. They also liked the fact that no hydrocarbon emissions were present.

Figure 9. eP-ICEBEAR at 2005 Hydrogen Energy Action Summit (EERC, Grand Forks, North Dakota).

Subsequently, the vehicle toured North America appearing at numerous public events and conferences, receiving much attention from international media outlets. During this extensive tour, it received substantial attention from many media sources, both print and television. In all, 52 stories were written about the e-P-ICEBEAR and covered in the media from 2005 to 2006.

ACTIVITY 6.2.4 – YEAR 1 – HYDROGEN END USE DEMONSTRATION AND OUTREACH

This activity created a broad set of outreach tools for NCHT to be used for outreach to the general public and decision makers regarding hydrogen in general as well as local and regional hydrogen activities (17). The outreach tools formed the central part of a hydrogen vehicle display booth visited by more than 12,000 patrons over the 9-day run of the 2006 North Dakota State Fair. The project was led by the EERC in partnership with Prairie Public Television, the North Dakota Department of Commerce Division of Community Services, Basin Electric Power Cooperative, e-Power Synergies Incorporated, and Minot area industry and commerce.

The core Hydrogen Outreach Toolkit includes an 18-minute video ("Hydrogen – Fueling Our Future…NOW!"), six fact sheets, and a set of booth display panels. The tool kit is intended to provide a basic outreach capability for the NCHT and its partners. It is also designed to inform the general public and decision makers regarding hydrogen overall as well as to inform the public regarding local and regional activities and opportunities in hydrogen. These tools provided the basis for a hydrogen press kit and supported the overall hydrogen goals of DOE and the NCHT. They address not only the potential uses of hydrogen, including applications in the transportation sector, but also provide practical information on hydrogen safety and handling issues.

The importance of providing the public with accurate, up-to-date information on hydrogen opportunities as well as safety and handling has been integral to the hydrogen efforts of the EERC's NCHT and its partners. The toolkit highlights projects at the EERC in hydrogen fuel cells, hydrogen-reforming, and hydrogen storage activities that are under way through contracts with NETL.

Results

The EERC took the lead on this activity. Primary project partners included Prairie Public Television; the North Dakota Department of Commerce Division of Community Services; Basin Electric Power Cooperative; Central Power Electric Cooperative Incorporated; Verendrye Electric Cooperative; e-Power Synergies Incorporated; North Dakota Association of Rural Electric Cooperatives; Hydrogen Engine Center; Hydrogenics Corporation; North Dakota State University – North Central Research Extension Center; the Minot Chamber of Commerce; the North Dakota State Fair; the North Dakota Army National Guard; AFVTech, Inc.; Ryan Chevrolet/GMC; and other Minot area industry and commerce.

These tools are used on the NCHT Web site, regional outreach and educational displays, as part of public display materials in the new NCHT facility, within the EERC complex, and on the University of North Dakota (UND) campus. As a result of this project, the EERC's NCHT has a consistent outreach package to advertise hydrogen in general as well as hydrogen opportunities and activities for itself, its partners, and others in the northern Great Plains region of the United States and Canada.

ACTIVITY 6.2.5 – YEARS 2–5 – OUTREACH AND EDUCATION FOR HYDROGEN PRODUCTION AND UTILIZATION

The goal of the NCHT education and outreach program was to provide objective information to researchers and the public to stimulate the development of the hydrogen economy. This goal was accomplished by activities conducted for people interested in hydrogen energy through workshops, presentations, demonstrations, and media, including print, video, audio, and Web-based material. Increased awareness about the hydrogen economy and its beneficial impact on national security, the environment, and the economy was achieved through the educational and outreach materials developed through this activity.

Results

Materials that were developed include a variety of resources that were matched to the appropriate audience. Materials produced under this activity include the following:

1. <u>60-minute Hydrogen Technology Educational Video Documentary</u> – The NCHT partnered with Prairie Public Television to produce a 1-hour broadcast-quality video documentary on hydrogen. The documentary, entitled "Hydrogen: Nature's Fuel," tells the story of hydrogen through a series of intriguing interviews from those working on cutting-edge hydrogen technologies. The documentary explains to viewers how hydrogen is created, how it is transported and dispensed, and how it is used today in fuel cell vehicles and other end uses.

2. <u>Online Course</u> – The NCHT, in collaboration with UND's Division of Continuing Education (DCE), developed an online course, entitled "Fueling the Hydrogen Economy," which provides an introduction to hydrogen. The online course is available worldwide to the public through UND's DCE and is targeted at middle and high school educators.

3. <u>Technical Briefs</u> – Two technical briefs entitled "Hydrogen Separation Membranes" and "Hydrogen On-Demand: Simplified High-Pressure Hydrogen Production" were created. The briefs are 4-page, full-color documents targeted to technical audiences.

4. <u>Hydrogen Works Training Course</u> – The NCHT hosted the Hydrogen Works Training Course at the San Diego Marriott Hotel & Marina in San Diego, California, on February 17–19, 2009. The course presented a comprehensive overview of the hydrogen economy, including production, storage, distribution infrastructure, fuel cells, alternative vehicles and fuels, and environmental impacts.

5. <u>Gasification Short Course</u> – The EERC developed a short course on gasification designed to educate potential financiers of the technology. The initial short course was presented September 9–10, 2009, at the EERC in Grand Forks, North Dakota. A second course was held September 29, 2010, at the Woodlands Waterway Marriott in The Woodlands, Texas.

6. Outreach Events – Using material contained in the online course and other outreach materials, NCHT participates in outreach events upon request.

The NCHT continues to enhance and promote the use of these materials while creating new materials for audiences not reached through traditional mediums. Most of the materials produced to date provide a unique educational format (i.e., online course, video, in-person training course).

ACTIVITY 6.2.8 – YEAR 4 – LIGNITE GASIFICATION TECHNOLOGIES SUMMARY REPORT

This activity generated a booklet which is an overview of past experience and future opportunities for gasification of North American lignites. It is not intended to be an exhaustive review of lignite gasification literature, but to describe the opportunities and challenges associated with the gasification of lignite resources. These opportunities and challenges are related to the properties of lignite that distinguish lignites from bituminous coals. The reactions of lignite coals in a gasification process differ from bituminous coals because of their distinctive chemical and physical properties. Future lignite gasification systems must consider these unique properties in order to integrate technologies that allow for installation of competitive and reliable systems for the production of power, fuels, and chemicals. The ability of lignite to be utilized in cost-competitive, zero-emission gasification systems installed in the 21st century will depend upon how we effectively use our understanding of the influence of lignite properties on critical design parameters affecting lignite preparation, feeding, carbon conversion, gas cleanup, and carbon separation and capture.

The title and authors of the summary report are “Gasification of Lignites of North America Summary Report” by Steven A. Benson and Everett A. Sondreal. The chapters and topics of the summary report are:

- Chapter 1 – Introduction: overviews the report and describes the vast global lignite reserves.

- Chapter 2 – Unique Properties of Lignite Affecting Their Utilization: describes specific chemical and physical properties that offer advantages over other fuels as well as challenges.

- Chapter 3 – The History of Gasification: overviews the technologies that have been applied to gasify lignite for over 100 years.

- Chapter 4 – Generic Types of Gasification Processes: describes the many gasification systems existing that can be utilized to gasify coal and other carbonaceous feedstocks.

- Chapter 5 – Overview of Gasification Applications: lists and describes the numerous ways that syngas or fuel gas produced from gasification can be used to produce products.

- Chapter 6 – Fundamentals of Coal Gasification: overviews the basic chemical reactions involved in coal gasification.

- Chapter 7 – Major Gasification Technologies and Their Suitability for Lignite: describes the experience associated with the use of lignite in various gasification processes and provides heat and material balances for selected gasification technologies.

- Chapter 8 – Hydrogen and Power Generation: shows how integrated lignite gasification technologies can be used to produce a source of hydrogen as well as electric power.

- Chapter 9 – Cost and Performance of IGCC with and Without Carbon Capture and Sequestration: describes CCS technologies and use in systems designed for the gasification of lignite coals.

- Chapter 10 – References: lists references cited in the report.

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