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Progress in High Temperature Electrolysis at the Idaho National Laboratory

2007 Fuel Cell Seminar & Exposition

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INTRODUCTION

The United States is considering the development of a domestic hydrogen-based energy economy. Hydrogen is of particular interest as a secondary energy carrier because it has the potential to be storable, transportable, environmentally benign, and useful in many chemical processes. Obviously, before a hydrogen economy can be implemented, an efficient and environmentally friendly means for large scale hydrogen production must be identified, proven, and developed. Hydrogen is now produced primarily via steam reforming of methane. However, from a long-term perspective, methane reforming is not a viable process for large-scale production of hydrogen since such fossil fuel conversion processes consume non-renewable resources and emit greenhouse gases. The U. S. National Research Council has recommended the use of water-splitting technologies to produce hydrogen using energy derived from a nuclear reactor. For the past several years, the Idaho National Laboratory has been actively studying the use of solid oxide fuel cells in conjunction with nuclear power for large-scale, high-temperature, electrolytic hydrogen production.

Conversion to a hydrogen-based energy economy, however, will require decades. Synthetically-derived hydrocarbon fuels (synfuels) represent an interim solution and a bridge to a future hydrogen economy. Synfuel production is a mature technology, and requires syngas – a mixture of hydrogen (H_2) and carbon monoxide (CO) – as a feedstock. Traditionally, syngas has been produced via coal gasification, and more recently by steam reforming of natural gas. Both techniques consume non-renewables and emit green-house gases. More recently, it has been demonstrated that syngas can also be produced by high temperature coelectrolysis of CO_2 and H_2O simultaneously in solid oxide cells.

This provides an overview of the high temperature solid oxide electrolysis and coelectrolysis research activities at the Idaho National Laboratory (INL) and subcontractor Ceramatec, Inc. Steam electrolysis activities are being funded by the DOE, Office of Nuclear Energy under its Nuclear Hydrogen Initiative, while coelectrolysis activities are funded by the INL Laboratory Directed Research and Development (LDRD) program.

HIGH TEMPERATURE STEAM ELECTROLYSIS ACTIVITIES AT THE INL

World oil demand is increasing, while world oil production is projected to peak and decline. The United States holds ~3% of the world's oil reserves but consumes ~25% of the world's total oil output, creating a worrisome situation. Furthermore, there is the controversial issue of greenhouse gas emissions and global warming. There are several pathways to better energy security for the United States: improved energy efficiency, new oil deposits located in friendly countries such as the Athabasca Oil Sands, and the transition to a hydrogen economy. Hydrogen will be the "transportation fuel of the future" and is essential for overall CO_2 reduction, assuming CO_2-free hydrogen production. Most current hydrogen production is accomplished via steam reforming of methane. However, for large-scale future hydrogen production for the hydrogen economy, viable hydrogen production technologies must avoid consumption of fossil fuels and avoid emissions of greenhouse gases and other forms of air pollution.

To accomplish this, large scale hydrogen production must arise from water-splitting technologies using either renewable or nuclear energy. Such options include low-temperature electrolysis using renewable power sources (e.g., wind), low-temperature electrolysis using nuclear electricity, high-temperature electrolysis using nuclear electricity and process heat, and high-temperature thermochemical processes using nuclear process heat. The scale of the issue, however, is huge. The U.S. currently produces approximately 460 GWe [1]. To replace the petroleum consumed by only U.S. passenger cars would require an additional 240 GWe of electrical capacity. If wind power alone were used, 640,000 1.5-MW wind turbines (@ 0.25 capacity factor) would be required, using a land area of 71,000 square miles (equal to the states of Ohio and Indiana). Clearly, multiple energy sources will be used, including nuclear.

There are two competing technologies being considered by the U.S. Department of Energy for nuclear-powered hydrogen production: thermochemical and high-temperature electrolysis. The INL is the lead laboratory for DOE-sponsored high-temperature electrolysis research. Figure 1 shows how a new design high temperature nuclear reactor would be coupled to a high-temperature electrolysis facility for hydrogen production. Electrical power and process heat is used to split steam into hydrogen and oxygen.

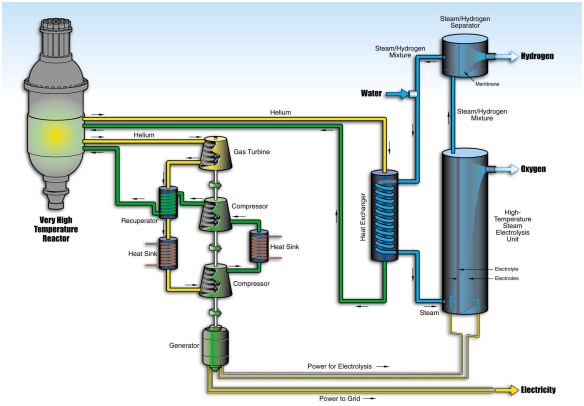

Figure 1. Nuclear-powered high-temperature electrolysis.

The INL has been conducting research into high-temperature steam electrolysis for the past 3 years [2, 3, 4]. Activities at the INL include small bench-scale experiments (see Figure 2) ranging from 1 to 500 W electrolysis power, skid-mounted integrated-laboratory scale (ILS) electrolysis tests (see Figure 3) of 15 kW power, computational fluid dynamics simulations, and flow sheet modeling of the entire process. Future plans include scale-up to pilot and demonstration-scale testing as well as continued smaller scale testing and modeling.

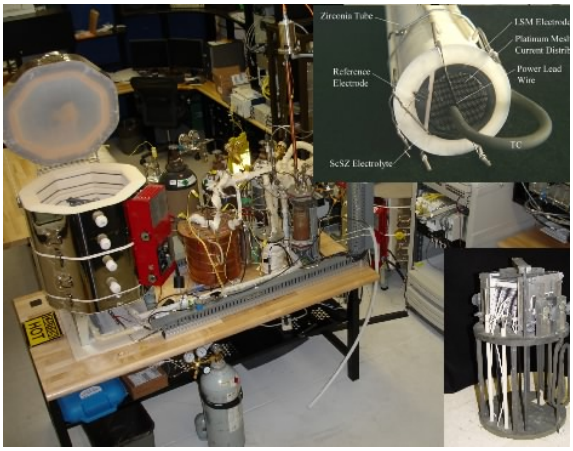

Figure 2. INL small scale testing activities.

Bench-scale testing at the INL has demonstrated hydrogen production rates of over 100 SLPH for a 10-cell stack and 180 SLPH for the 25-cell stack depicted in the Figure 2 insert. Test results from this stack are depicted in Figure 4. A Ceramatec test of a 120-cell stack showed hydrogen production rates in excess of 1200 SLPH and operated for over 2000 hours.

Figure 3. INL ILS test facility under construction..

The ILS scale facility currently under construction will incorporate a total of 720 cells in 3

modules of 4 stacks each. The Figure 3 insert shows one such module. Electrolysis in this test stand will consume 15kW electrical and produce over 5000 NLPH hydrogen. Operation will begin in September, 2007. This experimental facility is designed to address integral issues such as multiple-stack manifolding, stack electrical integration, and inlet / outlet gas handling and thermal management.

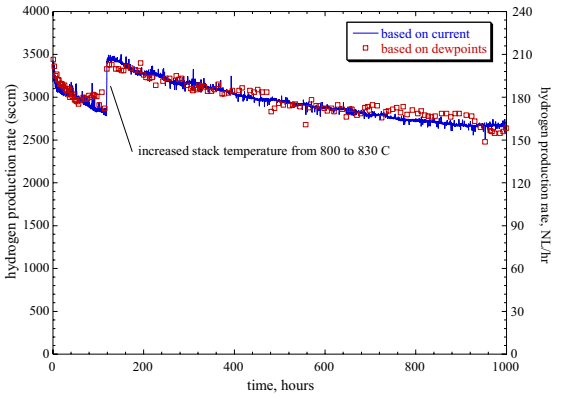

Figure 4. 25-cell stack H_2 production rate.

To evaluate the potential benefits and relative efficiency of nuclear-driven high temperature electrolysis for hydrogen production, the UniSim process analysis software is being used to evaluate different process layouts and nuclear reactor concepts. The UniSim models of the process loops include pumps to circulate the working fluids and heat exchangers to recover heat from the oxygen and hydrogen product streams, and an in-house-developed electrolysis model. These models are being studied to improve the overall hydrogen production efficiencies.

HIGH TEMPERATURE COELECTROLYSIS ACTIVITIES AT THE INL

Complementary to investigating the feasibility of high-temperature steam electrolysis, the INL, in conjunction with Ceramatec Inc. (Salt Lake City, USA), has also been researching the use of solid oxide cells for simultaneous electrolysis (coelectrolysis) of steam/CO_2 mixtures to produce syngas:

$$H_2O + CO_2 \longrightarrow H_2 + CO + O_2 . \quad (1)$$

These activities include bench scale experimentation, modeling, and flow sheet analysis [5, 6]. When linked to a nuclear power source, this technology can provide a carbon neutral means of producing syngas while consuming CO_2.

Coelectrolysis, however, is significantly more complex than simple steam electrolysis. This is primarily due to the multiple reactions that occur: steam electrolysis, CO_2 electrolysis, and the reverse shift reaction (RSR):

$$CO_2 + H_2 \leftrightarrow CO + H_2O. \quad (2)$$

Reaction kinetics govern the relative contributions of these three reactions.

In addition to many button cells, three stacks have been tested at the INL under coelectrolysis conditions. Inlet gas composition, operating voltage, and operating temperature were varied. Stack area-specific resistance (ASR – an index of electrolysis performance) values did not vary significantly when switching from steam electrolysis to steam / CO_2 coelectrolysis. However, a given stack's ASR increased significantly when switching from steam to pure CO_2 electrolysis. This is due to the slower overall kinetics of CO_2 electrolysis and the higher overpotentials required. In coelectrolysis, the reverse gas shift reaction is relied upon for most of the CO production and therefore the overall electrical requirement is less. Stack ASR values showed trends of increasing with lower steam availability and stack age.

Figures 5 is an example test result of coelectrolysis in a 10 cell stack at 800 C. Lines represent predictions from an INL-developed computer model and symbols represent experimental gas chromatograph results. Even at zero current there was a drop in CO_2 and H_2 mole fractions from

the cold inlet values, with CO produced. This is solely due to the RSR. As the electrolysis current was increased, the yield of syngas increased linearly while the concentration of CO_2 (and H_2O, not shown in the figure) decreased. This figure also shows good agreement between experimental gas chromatograph data and results from the coelectrolysis model. These measurements and model results indicate that coelectrolysis effectively increases the syngas yield for a given steam/CO_2 process stream over that obtained from the RSR alone. Overall, the coelectrolysis process appears to be a promising technique for large-scale syngas production.

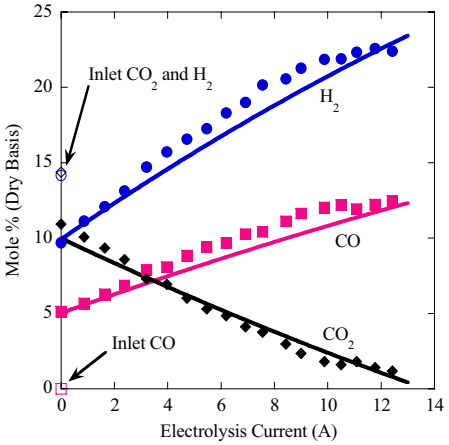

Figure 5. Experimental and modeling results for coelectrolysis stack testing..

SUMMARY

High temperature steam electrolysis and steam/CO_2 coelectrolysis experiments have been conducted during the last three years at the INL and at Ceramatec on progressively larger stacks. Both techniques appear promising for large-scale nuclear powered hydrogen and syngas production.

ACKNOWLEDGEMENTS

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