High-Efficiency Steam Electrolyzer

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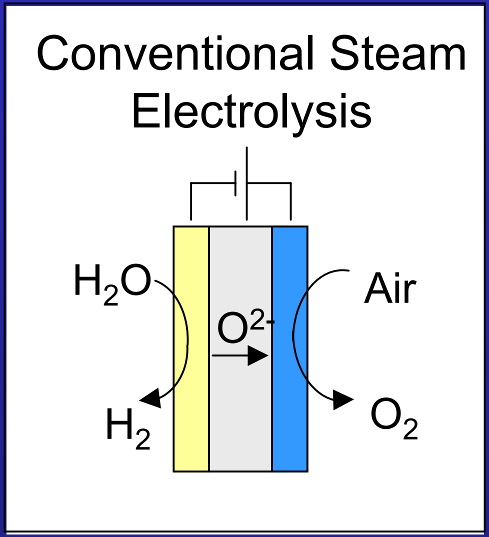
We are developing technology for distributed hydrogen production

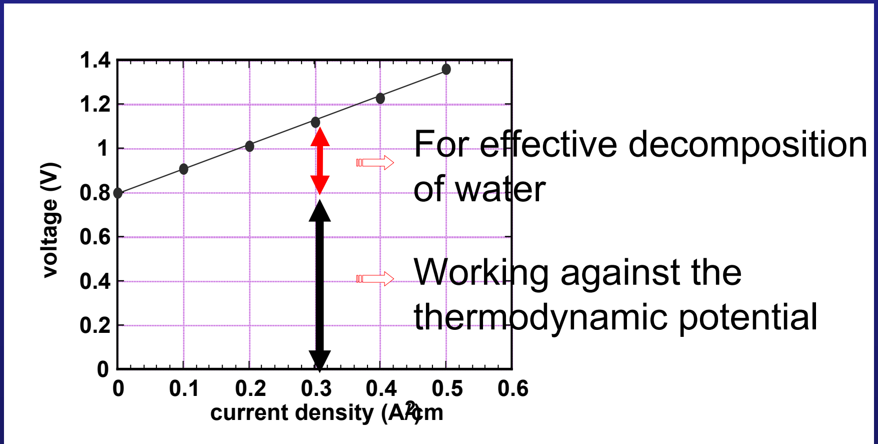
- Our goal is a hydrogen production system that is compatible with the existing, hydrocarbon-based fuel infrastructure
- We are working toward the demonstration of a 5 kW natural-gas-assisted steam electrolyzer by 2006
- For 2003, we will build and test a newly designed electrolyzer targeting 1 kW hydrogen output

There are cost and efficiency issues with conventional steam electrolysis

- In conventional systems, most of the electricity is used to overcome the thermodynamic potential
- Hydrogen production from electrolysis is expensive because of high electricity requirements



We are working toward electrolysis with reduced electricity consumption

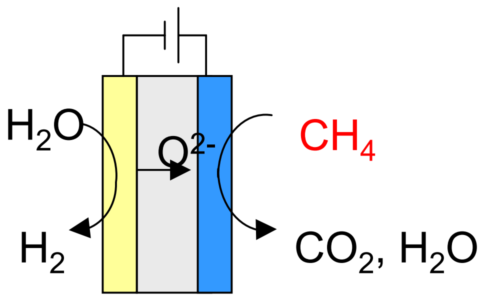


Our approach offers potential cost and efficiency advantages

Natural-Gas-Assisted Steam Electrolyzer (NGASE)

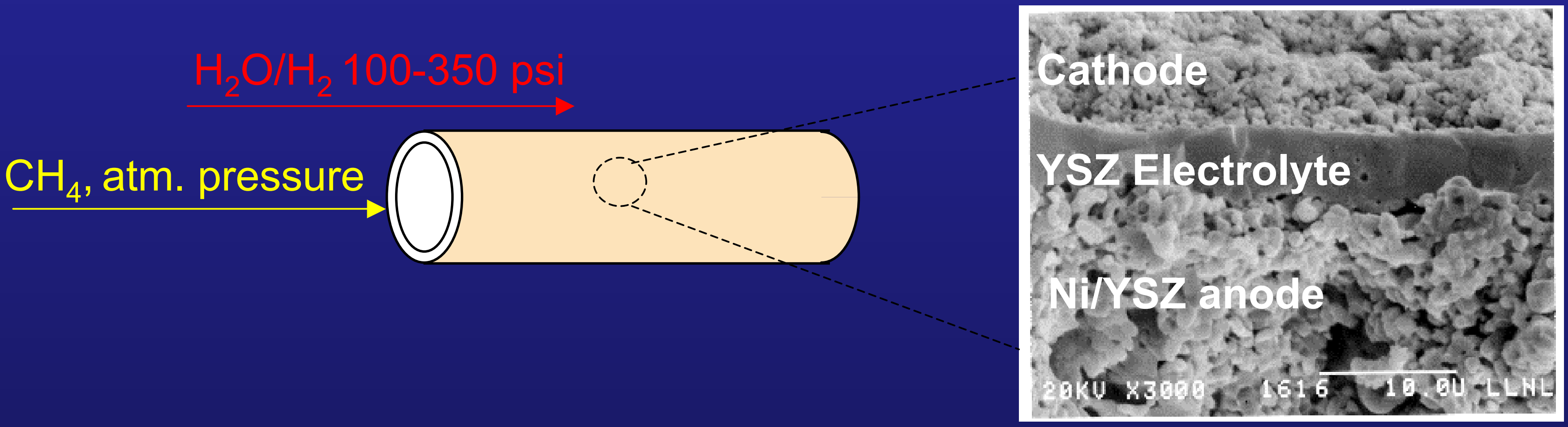
- Natural gas at the anode reduces the potential difference - lowering electricity consumption
- Hydrogen is produced at pressure at the cathode
- Current focus utilizes total oxidation mode
- Partial oxidation mode also possible, yielding additional hydrogen from methane after gas separation

Total oxidation mode



The tubular design allows pressurized operation

- Tubes best for operation with significant pressure differentials
- Size and layer thickness controlled by fabrication technique
- Low risk of contamination of hydrogen due to pressure differential
- System provides in-situ electrochemical hydrogen compression



The project has advanced from feasibility demonstration to prototype systems

FY00 - Project initiation, feasibility demonstration of reduced electricity requirement

FY01 - Improved tube fabrication, designed multiple-tube electrolyzer, initiated industrial collaboration

FY02 - Demonstrated 200 W, 35 psi electrolyzer, improved tube performance, began design of 1 kW reactor

FY03 - Improved design of 1 kW electrolyzer, assemble and test new reactor, initiated collaboration with UConn for tube fabrication, testing improved anode materials to reduce carbon deposition

FY04-FY06 - Continued testing and refinement of 1 kW system, design and assembly of 5 kW system with field demonstration in 2006

A few challenges were encountered in the first half of FY03

- Continuing resolution limited funding levels
  - Forced cuts in manpower (loss of two technicians)
  - Delayed purchase of needed big-ticket items
- Funding difficulties for industrial partner, Solid Oxide Systems, delayed system scale-up
- PI (Quoc Pham) left LLNL for industry in February

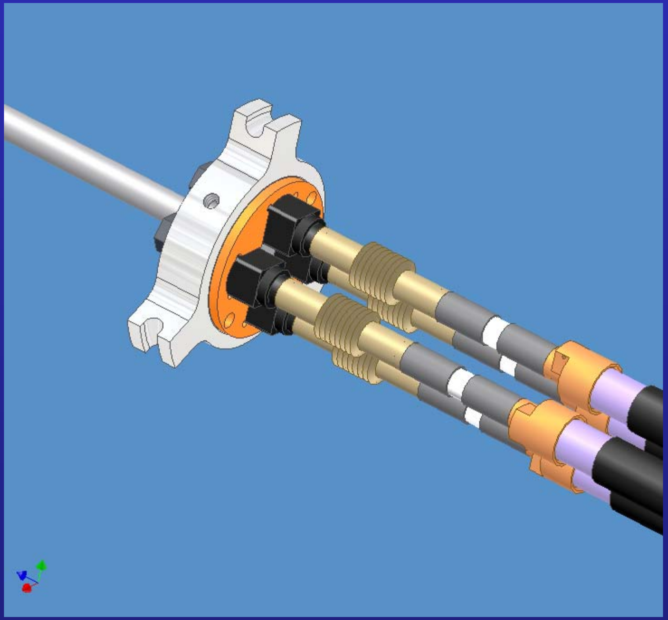
The project is back on track for FY03

We are building an improved electrolyzer and tackling materials science problems

- Significant improvements have been made in the electrolyzer design
  - Flexible and scalable to accommodate tube lengths up to 18”
  - Materials utilized to avoid problems related to operating temperature and environment
  - System will be complete in August with testing to follow
- Evaluation of potential materials for next generation systems
  - Screening of improved anode formulations
  - Materials for tube assembly (brazing alloys, interconnects, fittings)
  - Electrolyte and cathode coating methods

We are forming alliances with universities and industry

- Outsourcing for efficiency:
  - Ni-YSZ tubes to be fabricated by extrusion technique at the University of Connecticut's Global Fuel Cell Center
  - Brazing of tubes to fittings to be completed by industrial vendor
  - Industrial partner, Solid Oxide Systems, working to secure funding for manufacturing support

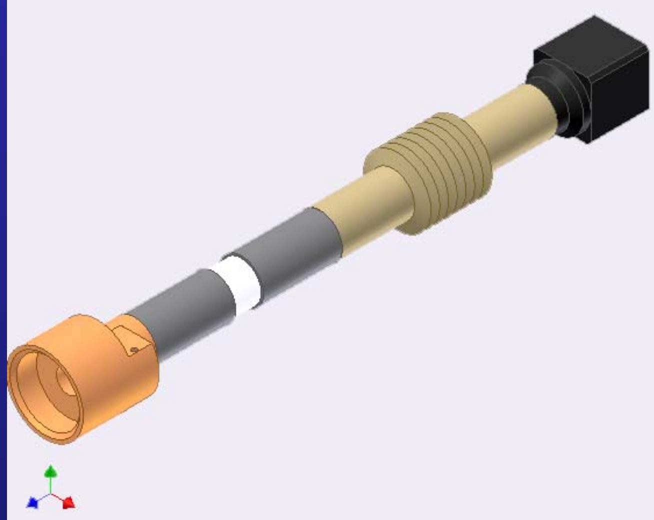


- We will focus on design, chemistry and materials issues and work to strengthen ties to other institutions (e.g. UC-Davis)

The new flexible electrical isolator design overcomes several technical issues

- CTE-matched components for 700°C
- Bellows to accommodate movement of tubes
- Insulating standoff – allows series connection
- Steam/ hydrogen environment tolerant

Electrical standoff with CTE-matched fitting for Ni-YSZ tube

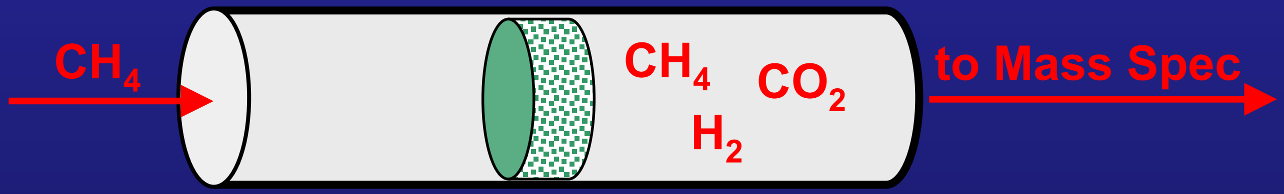


We are also addressing basic materials issues

- Carbon deposition catalyzed by nickel remains a challenge
  - Without adequate oxygen ion flow to anode, methane rapidly converts to carbon and hydrogen at operating temperature
  - Short-term solution is humidification of methane with applied potential (Liu, J.; Barnett, S.A. *Solid State Ionics*, 2003, *158*, 11)
- Long-term goal is improved material allowing use of untreated natural gas fuel
  - Processing issues must be overcome (e.g. incompatible sintering temperatures)
  - Material must be good ionic and electrical conductor

Potential anode materials are screened by temperature-programmed oxidation

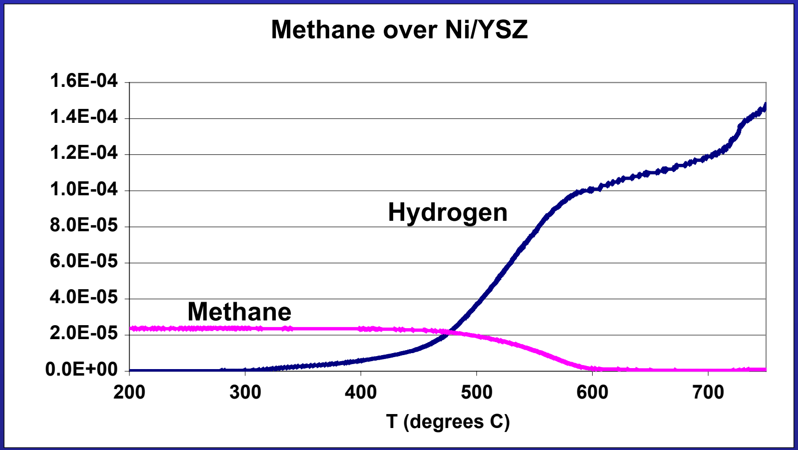
- Materials with approximate anode composition are prepared and analyzed as powder samples
- Methane flowed through heated powders in quartz tube
- Good candidates for further study will be used in single-cell testing
- Temperature-programmed oxidation-mass spectrometry (TPO-MS) useful for rapid screening of potential anode materials



TPO-MS screening reveals reaction products for powders

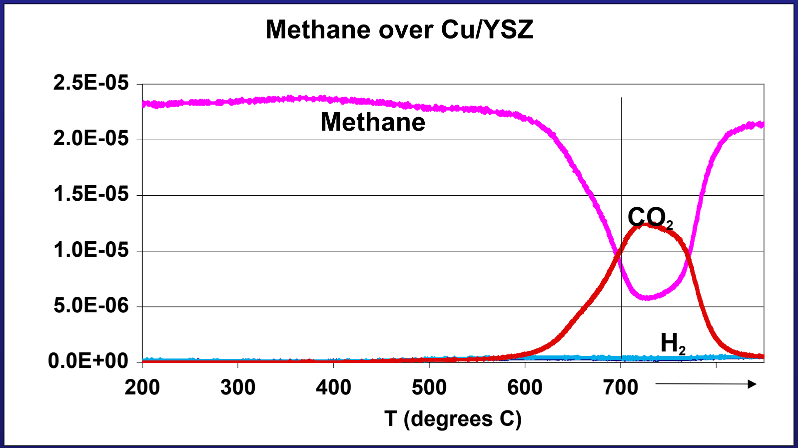
- Ni/YSZ carbon deposition shown by significant hydrogen generation

$$CH_4 \rightarrow C + 2\,H_2$$



- Cu/YSZ yields only carbon dioxide at 700°C

$$CH_4 \xrightarrow{O^{2-}} CO_2 + 2\,H_2O + 8\,e^-$$



Copper illustrative of challenges in modifying anode composition

- Well known that copper does not catalyze carbon deposition at operating temperature
  - Melting points of CuO (1326 °C) and Cu (1083 °C) below normal sintering temperature of YSZ (~1450 °C)
  - Research indicates minimization of YSZ agglomeration allows reduced sintering temperatures (Lu et al., *Solid State Ionics*, 2002, *152-153*, 393)

- Single cells will be utilized for testing of promising materials
  - Anode-supported cells for testing under electrolyzer conditions (thin electrolyte)
  - Electrolyte-supported cells to allow testing of materials without processing issues associated with anode-supported cells

Future work builds toward 5 kW system

- Remaining work for FY03
  - Complete assembly of current 1 kW system (9/03)
  - Initial testing of 1 kW system (9/03)
- FY04
  - Characterize improved anode materials (11/03)
  - Durability testing of components (1/04)
  - Long-term testing of 1 kW system (4/04)
  - Integrate interconnects and tubes (5/04)
  - Fabricate modified tubes (6/04)
  - Optimize tube composition and fabrication (8/04)
  - Define requirements for 5 kW system (9/04)

We are working toward a 1 kW electrolyzer for FY03

- After funding and personnel issues in first half of FY03, we are back on track
- We developed an improved electrolyzer design
  - Flexible, scalable system
  - Components matched to operating conditions
- Materials testing is underway
  - Exploring potential next generation tube materials
- Forming additional alliances to facilitate future work
  - Universities, industry, outside vendors

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